

Electron-Phonon Interactions and Electronic Transport in Correlated Metals and Moiré Systems

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ABSTRACT

Interactions between electrons and lattice vibrations (phonons) give rise to transport, superconducting, and spectral properties in many classes of materials. These properties can be predicted from a microscopic understanding of the electrons, phonons, and electron-phonon coupling, but theoretical and computational hurdles limit this ability in materials with strong electronic correlations or large unit cell sizes. In Chapters 2-4, I present developments on the prediction of electron-phonon interactions and electronic transport in correlated metals. First, I will show methodology to combine first-principles electron-phonon interactions with dynamical mean-field theory electron-electron interactions into a beyond-quasiparticle description of spectra and transport. I will show applications to transport and spectral properties in the prototypical correlated metals SrVO_3 and Sr_2RuO_4 , leading to an improved understanding of the respective roles of electron-phonon and electron-electron interactions in these systems. Then, I will present a method to compute the coupling of electrons to representative phonons within density functional theory plus dynamical mean-field theory. I will discuss the insights from this method on the role of orbital and phonon character, electron energy, and correlation in modifying the coupling and resulting physical properties. Transitioning to moiré systems with large unit cells, in Chapter 5, I will present a tight-binding based approximation to enable electron-phonon and transport calculations in twisted-bilayer graphene structures with over 5,000 atoms per unit cell. I will show trends in the phonon-limited resistivity as the twist angle is decreased, including the breakdown of transport properties associated with Dirac dispersion as the electronic energy scale is decreased. Taken together, this work makes promising strides and sets the stage for future developments in the study of electron-phonon physics in previously inaccessible regimes.

PUBLISHED CONTENT AND CONTRIBUTIONS

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D.J.A. performed first-principles Boltzmann transport calculations in collaboration with the experiment, discussed findings with all authors, and contributed to the section of the manuscript on computational results.

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Chapter 1

INTRODUCTION

1.1 Coupled Electrons and Phonons

1.1.1 Hamiltonian

Interactions between electrons and quantized lattice vibrations (phonons) are present in every material. These interactions often control essential material properties including electron and thermal transport, electronic and phononic excitation spectra, conventional superconductivity, and electronic and structural phase transitions. Because of this essential role, understanding such electron-phonon (e-ph) interactions has been a major part of condensed matter and solid state physics for nearly a century [1, 2, 3, 4, 5]. In recent decades, there is a growing field dedicated to the computational study of e-ph interactions and their consequences in real materials with minimal empirical assumptions-known as first-principles calculations [6, 7]. The Hamiltonian for a system of interacting electrons and phonons can be written as

$$\hat{H} = \sum_{n\mathbf{k}} \varepsilon_{n\mathbf{k}} c_{n\mathbf{k}}^\dagger c_{n\mathbf{k}} + \sum_{\nu\mathbf{q}} \omega_{\nu\mathbf{q}} a_{\nu\mathbf{q}}^\dagger a_{\nu\mathbf{q}} + \sum_{nm\nu\mathbf{k}\mathbf{q}} g_{mn\nu}(\mathbf{k}, \mathbf{q}) c_{m\mathbf{k}+\mathbf{q}}^\dagger c_{n\mathbf{k}} (a_{\nu-\mathbf{q}}^\dagger + a_{\nu\mathbf{q}}), \quad (1.1)$$

where $c_{n\mathbf{k}}^\dagger$ creates an electron in band n at crystal momentum $\mathbf{k}$ with energy $\varepsilon_{n\mathbf{k}}$, $a_{\nu\mathbf{q}}^\dagger$ creates a phonon in mode ν at crystal momentum $\mathbf{q}$ with energy $\omega_{\nu\mathbf{q}}$, and $g_{mn\nu}(\mathbf{k}, \mathbf{q})$ is the e-ph coupling strength. Electron-electron (e-e) interactions are also often present and, depending on the level of theory, can be included as an explicit term in the Hamiltonian. In this thesis, I will usually consider Hamiltonians with parameters reflecting values in real materials obtained from first-principles calculations. In these methods, some or all of the effects of e-e interactions are usually included as an effective renormalization of the zero- and one-electron quantities in Eq. 1.1. I discuss the formulation of e-ph interactions in different electronic structure methods in Sec. 1.2.

This Hamiltonian is written in terms of the eigenstates of the electron and phonon Hamiltonians. The electronic states are eigenstates of a purely electronic one-electron Hamiltonian

$$H_{\text{electron}}(\mathbf{k})\phi_{n\mathbf{k}} = \varepsilon_{n\mathbf{k}}\phi_{n\mathbf{k}}. \quad (1.2)$$

The phonon modes are eigenstates of the dynamical matrix $\mathcal{D}(\mathbf{q})$ describing the energetics of small atomic motions around equilibrium

$$\mathcal{D}_{(a\mu,a'\mu')}(\mathbf{q}) = \frac{1}{\sqrt{M_a M_{a'}}} \sum_{\mathbf{R}_p} \frac{\partial^2 U}{\partial u_{a\mu\mathbf{0}} \partial u_{a'\mu'\mathbf{R}_p}} \exp(i\mathbf{q} \cdot \mathbf{R}_p) \quad (1.3)$$

$$\sum_{(a'\mu')} \mathcal{D}_{(a\mu,a'\mu')}(\mathbf{q}) e_{a'\mu'}^{\nu\mathbf{q}} = \omega_{\nu\mathbf{q}}^2 e_{a\mu}^{\nu\mathbf{q}}. \quad (1.4)$$

Here, U is the total energy of the system, $u_{a\mu\mathbf{R}_p}$ denotes a small displacement from the equilibrium position of an atom a (mass M_a) in direction μ in a unit cell at lattice vector $\mathbf{R}_p$. The eigenvalues of the dynamical matrix define the phonon frequency $\omega_{\nu\mathbf{q}}$ while the eigenvalues $e_{a\mu}^{\nu\mathbf{q}}$ define the displacements associated with the modes. The e-ph coupling is given by the response of the electrons to an atomic displacement associated with a phonon mode,

$$g_{mn\nu}(\mathbf{k}, \mathbf{q}) = \langle \phi_{m\mathbf{k}+\mathbf{q}} | \partial_{\nu\mathbf{q}} \hat{V}_{\text{electron}} | \phi_{n\mathbf{k}} \rangle_{u.c.}, \quad (1.5)$$

where $\hat{V}_{\text{electron}}$ is a potential which acts on electrons (the details of which depend on the electronic structure method and are discussed more in Sec. 1.2), $\langle | \dots \rangle_{u.c.}$ denotes averaging over a unit cell and $\partial_{\nu\mathbf{q}}$ is the mode resolved phonon derivative

$$\partial_{\nu\mathbf{q}} = \sum_{\mathbf{R}_p a\mu} e^{i\mathbf{q} \cdot \mathbf{R}_p} \sqrt{\frac{\hbar}{2M_a \omega_{\nu\mathbf{q}}}} \mathbf{e}_{\nu\mathbf{q}}^{a\mu} \left. \frac{\partial}{\partial u_{\mathbf{R}_p a\mu}} \right|_{u_{\mathbf{R}_p a\mu}=0}. \quad (1.6)$$

An important aspect of the e-ph interaction is the ability to Fourier transform between real space and reciprocal space representations. For example, the e-ph coupling can be written in terms of local contributions in a basis of local orbitals $|i\mathbf{R}_e\rangle$ and real space atomic displacements

$$g_{mn\nu}(\mathbf{k}, \mathbf{q}) = \frac{1}{N_e} \sum_{ija} \sum_{\mathbf{R}_e \mathbf{R}_p} e^{i(\mathbf{k} \cdot \mathbf{R}_e + \mathbf{q} \cdot \mathbf{R}_p)} \sqrt{\frac{\hbar}{2M_a \omega_{\nu\mathbf{q}}}} \\ \times \mathbf{e}_{\nu\mathbf{q}}^{a\mu} U_{mi,\mathbf{k}+\mathbf{q}} \langle i\mathbf{0} | \partial_{u_{a\mu\mathbf{R}_p}} V | j\mathbf{R}_e \rangle U_{nj,\mathbf{k}}^\dagger. \quad (1.7)$$

This representation is often used to interpolate the e-ph interaction to arbitrary $\mathbf{k}$ and $\mathbf{q}$ grids, and plays an important role in the calculation of physical properties arising from the interaction [8]. It also enables a physical real space understanding of the e-ph coupling which can facilitate certain approximations.

1.1.2 Effect on Physical Properties

The e-ph interaction gives rise to many physical properties in materials. In this section, I discuss the contribution of the e-ph interaction to a few of these properties as illustrative examples.

First, e-ph interactions modify the properties of electronic states in a material. Compared to the non-interacting electronic states, which are eigenstates of the one-particle electronic Hamiltonian with crystal momentum $\mathbf{k}$ and band index n , electronic states interacting with phonons will acquire finite width (inverse of the relaxation time $\tau_{n\mathbf{k}}$) which in the simplest approximation is given by [9, 7, 6, 10]

$$\tau_{n\mathbf{k}}^{-1} = \frac{2\pi}{N_{\mathbf{q}}} \sum_{m\nu\mathbf{q}} |g_{mn\nu}(\mathbf{k}, \mathbf{q})|^2 \times [\delta(\varepsilon_{n\mathbf{k}} - \omega_{\nu\mathbf{q}} - \varepsilon_{m\mathbf{k}+\mathbf{q}})(1 - f_{m\mathbf{k}+\mathbf{q}} + N_{\nu\mathbf{q}}) + \delta(\varepsilon_{n\mathbf{k}} + \omega_{\nu\mathbf{q}} - \varepsilon_{m\mathbf{k}+\mathbf{q}})(f_{m\mathbf{k}+\mathbf{q}} + N_{\nu\mathbf{q}})]. \quad (1.8)$$

Here, $f_{n\mathbf{k}}$ and $N_{\nu\mathbf{q}}$ are the fermion and boson occupation numbers for electrons in state $n\mathbf{k}$ and phonons in state $\nu\mathbf{q}$. Furthermore, e-ph interactions will also modify the quasiparticle weight and dispersion by a factor $Z_{n\mathbf{k}}$, a phenomenon which gives rise to kinks in the photoemission spectra at the energy of strongly coupled phonons [6]. A lowest order approximation to $Z_{n\mathbf{k}}$ is

$$Z_{n\mathbf{k}}^{-1} = 1 - \frac{1}{N_{\mathbf{q}}} \sum_{m\nu\mathbf{q}} |g_{mn\nu}(\mathbf{k}, \mathbf{q})|^2 \left[\frac{N_{\nu\mathbf{q}} + 1 - f_{m\mathbf{k}+\mathbf{q}}}{(\varepsilon_{m\mathbf{k}+\mathbf{q}} - \omega_{\nu\mathbf{q}} + i\eta)^2} + \frac{N_{\nu\mathbf{q}} + f_{m\mathbf{k}+\mathbf{q}}}{(\varepsilon_{m\mathbf{k}+\mathbf{q}} + \omega_{\nu\mathbf{q}} + i\eta)^2} \right], \quad (1.9)$$

where η is a small positive broadening. Similarly, e-ph interactions also modify the properties of phonons. For example, the e-ph contribution to the phonon linewidth $\gamma_{\nu\mathbf{q}}$ is

$$\gamma_{\nu\mathbf{q}} = \frac{4\pi}{N_{\mathbf{k}}} \sum_{\mathbf{k}} |g_{m\nu}(\mathbf{k}, \mathbf{q})|^2 (f_{m\mathbf{k}+\mathbf{q}} - f_{n\mathbf{k}}) \delta(\varepsilon_{m\mathbf{k}+\mathbf{q}} - \varepsilon_{n\mathbf{k}} - \omega_{\nu\mathbf{q}}). \quad (1.10)$$

One of the biggest successes of modern e-ph calculations is the ability to calculate macroscopic observable properties such as transport, superconductivity, and optical absorption. Here, I provide only the simplest approximation to transport building upon the relaxation time in Eq. 1.8

$$\sigma_{ij}^{\text{RTA}} = \frac{2e^2}{AN_{\mathbf{k}}} \sum_{n\mathbf{k}} v_{n\mathbf{k}}^i v_{n\mathbf{k}}^j \left(-\frac{\partial f_{n\mathbf{k}}}{\partial \varepsilon_{n\mathbf{k}}} \right) \tau_{n\mathbf{k}}, \quad (1.11)$$

where e is the electron charge, A is the unit cell area, $v_{n\mathbf{k}}^i$ is the electron band velocity, and $f_{n\mathbf{k}}$ is the Fermion occupation function. This equation can be derived either by considering a many-body Kubo transport formalism and neglecting both vertex corrections and beyond-quasiparticle properties, or starting from a semi-classical Boltzmann transport formalism and neglecting back scattering [10].

More details about predicting different physical properties arising from the e-ph interaction can be found in the following reviews and papers [6, 11, 12, 13, 14, 15, 16, 17]. In the past two decades, a number of codes have been developed to perform these calculations [18, 19, 20, 21], including Perturbo [19], which is developed in the Bernardi group at Caltech and used throughout this thesis.

1.2 Electronic Structure Theory and Correlation

1.2.1 Many-Body Hamiltonian

Electronic structure theory methods are computational schemes to predict the properties of materials from a quantum many-body description of the electrons and atoms. The many-body Hamiltonian for such a system is

$$\begin{aligned} \hat{H}_{\text{electron-ion}} &= \hat{T}_{\text{ion}} + \hat{T}_{\text{electron}} + \hat{V}_{\text{electron-ion}} + \hat{V}_{\text{ion-ion}} + \hat{V}_{\text{electron-electron}} \\ &= \sum_a \frac{1}{2M_a} \nabla_a^2 + \sum_i \frac{1}{2m_e} \nabla_i^2 + \\ &\quad \sum_{i,a} \frac{-Z_a}{|\mathbf{r}_i - \mathbf{R}_a|} + \frac{1}{2} \sum_{a,b \neq b} \frac{Z_a Z_b}{|\mathbf{R}_a - \mathbf{R}_b|} + \frac{1}{2} \sum_{i,j \neq i} \frac{1}{|\mathbf{r}_i - \mathbf{r}_j|}. \end{aligned} \quad (1.12)$$

Here, a and b index ions with mass M_a , charge $Z_a q_e$, and position $\mathbf{R}_a$ while i and j index electrons with mass m_e , charge $-q_e$, and position $\mathbf{r}_i$. I have set $\hbar = q_e = 1$ for simplicity. This equation is almost never feasible to solve directly, so different approximations are needed.

A set of approximations deal with simplification of the treatment of the ions. For example, using the Born-Oppenheimer approximation, the electronic and ionic parts can be separated, so that an electronic Hamiltonian $H_{\text{electron}}(\{\mathbf{R}_a\}) = \hat{T}_{\text{electron}} + \hat{V}_{\text{electron-ion}} + \hat{V}_{\text{electron-electron}}$ is used, defined in terms of a set of classical atomic positions $\{\mathbf{R}_a\}$ usually chosen to minimize energy. Another approach to simplify the ionic part of the Hamiltonian is to treat the ions in terms of small displacements around their equilibrium positions, and thus the ion contributions are replaced by phonon contributions. While these simplifications are important, further approximations are still needed to treat the many-body electronic Hamiltonian due to the e-e interactions.

1.2.2 Density Functional Theory

A common approach to greatly simplify the electronic system is to replace the many-body Hamiltonian with an effective one-body Hamiltonian. This is most commonly achieved with Density Functional Theory (DFT), in which an effective one-electron Kohn-Sham Hamiltonian replaces $H_{\text{electron}}(\{\mathbf{R}_a\})$ [22, 23]. In this case, the Hamiltonian is

$$H_{\text{KS}} = \hat{T}_{\text{electron}} + \hat{V}_{\text{electron-ion}} + \hat{V}_{\text{hartree}} + \hat{V}_{\text{XC}} = \hat{T}_{\text{electron}} + \hat{V}_{\text{KS}}. \quad (1.13)$$

Here, $\hat{V}_{\text{electron-ion}}$ contains the Coulomb interaction between electrons and classical ions, $\hat{V}_{\text{hartree}}$ is the Coulomb potential felt by an electron from the mean electron density, and $\hat{V}_{\text{XC}}$ is the correction to the electronic potential caused by quantum exchange and correlation effects between electrons. Collectively, these three terms are known as the Kohn-Sham potential $\hat{V}_{\text{KS}}$. In principle, this Hamiltonian can describe the exact ground state energy of the interacting electron system [22]. In practice however, the $\hat{V}_{\text{XC}}$ term must be approximated, so the Hamiltonian is only approximate. Furthermore, because it uses an effective one-particle description of the Hamiltonian, DFT does not give access to many important properties, including dynamical and beyond-quasiparticle physics. Despite these limitations, DFT often gives an excellent description of many material properties, and is a common starting

point for many-body physics calculations of real materials [24].

DFT and its extensions have been widely applied to study the e-ph interaction in materials. Phonon energies and e-ph coupling can be calculated from linear response DFT (DFPT) [25]. In this approach, the e-ph coupling $g_{mnv}(\mathbf{k}, \mathbf{q})$ is

$$g_{mnv}(\mathbf{k}, \mathbf{q}) = \langle \phi_{m\mathbf{k}+\mathbf{q}} | \partial_{v\mathbf{q}} \hat{V}_{KS} | \phi_{n\mathbf{k}} \rangle_{u.c.}, \quad (1.14)$$

where $\phi_{n\mathbf{k}}$ is a Kohn-Sham eigenstate.

1.2.3 Many-Body Physics via Correlation Functions

So far, we have discussed two approaches to the quantum mechanical description of materials: solving a many-body Hamiltonian, which may be exact, but is incredibly difficult, and approximation via an effective one-particle Hamiltonian. What these approaches share is the choice of the Hamiltonian and its eigenstates as the chief quantities of interest. The resulting problem is that the eigenstates become vectors in vector space which grows exponentially with the number of particles. This makes the problem impossible to solve exactly, but it also complicates the physical interpretation of the resulting quantities.

An alternate route is to work not in terms of eigenstates but correlation functions, known as Green's functions (see e.g. Refs [9, 26, 27, 28]). In this case, one can probe effective one- or two- particle quantities that exist in many-body systems, without resorting to neglecting many-body effects in the Hamiltonian. An example is the retarded single particle Green's function for fermions

$$G_{ij}^R(t) = -i\theta(t) \langle c_i(t) c_j^\dagger(0) + c_j^\dagger(0) c_i(t) \rangle. \quad (1.15)$$

This Green's function describes the response to creating a single electron in state j at time zero. This can be Fourier transformed to be frequency dependent

$$G_{ij}^R(\omega) = \int_{-\infty}^{\infty} G_{ij}^R(t) e^{-i\omega t}. \quad (1.16)$$

It can also be transformed into a different basis, for example to be $\mathbf{k}$ dependent. A physical quantity related to the Green's function is the spectral function $A_{\mathbf{k}}(\omega) = -2\text{Im}G_{\mathbf{k}}^R(\omega)$, which contains the spectrum of one-electron excitations. When a one-electron description is sufficient, the spectral weight will be contained in sharp peaks near the band energy. However, when strong correlations are present between

the electrons (or between electrons and phonons), weight will be transferred away from the peak, which will in turn be broadened and possibly shifted. The spectral function can be measured through experimental methods such as angle-resolved photoemission spectroscopy, making it an important quantity for characterizing electronic structure in materials.

A strength of the Green's function approach is that it often facilitates approximations, such as expansions around the non-interacting states in terms of interaction order. For example, for the Hamiltonian in eq. 1.1, a widely used approximation to the electron Green's function is

$$G_{nn'\mathbf{k}}(\omega) = [(\omega + \mu - \varepsilon_{n\mathbf{k}})\delta_{nn'} - \Sigma_{nn'\mathbf{k}}^{\text{FM}}(\omega)]^{-1}, \quad (1.17)$$

where the lowest order effect of the e-ph interaction is contained in the Fan-Migdal self-energy [2, 6]

$$\begin{aligned} \Sigma_{nn'\mathbf{k}}^{\text{FM}}(\omega) = \frac{1}{N_{\mathbf{q}}} \sum_{m\nu\mathbf{q}} g_{mn\nu}^*(\mathbf{k}, \mathbf{q}) g_{mn'\nu}(\mathbf{k}, \mathbf{q}) \times \\ \left[\frac{N_{\nu\mathbf{q}} + 1 - f_{m\mathbf{k}+\mathbf{q}}}{\omega - \varepsilon_{m\mathbf{k}+\mathbf{q}} - \omega_{\nu\mathbf{q}} + i\eta} + \frac{N_{\nu\mathbf{q}} + f_{m\mathbf{k}+\mathbf{q}}}{\omega - \varepsilon_{m\mathbf{k}+\mathbf{q}} + \omega_{\nu\mathbf{q}} + i\eta} \right]. \end{aligned} \quad (1.18)$$

This self-energy contains many of the properties discussed in Sec. 1.1.2. For instance [6],

$$\tau_{n\mathbf{k}}^{-1} = -2\text{Im}\Sigma_{n\mathbf{k}}^{\text{FM}}(\omega = \varepsilon_{n\mathbf{k}}) \quad (1.19)$$

$$Z_{n\mathbf{k}} = [1 - \partial\text{Re}\Sigma_{n\mathbf{k}}^{\text{FM}}(\omega)/\partial\omega|_{\omega=0}]^{-1}. \quad (1.20)$$

Perturbative contributions from other interactions can also be included. Other approaches to quantum many-body systems include partial diagrammatic expansions, exact diagonalization, and evaluation via Monte Carlo integration (see for instance [28]).

1.2.4 Dynamical Mean-Field Theory

One Green's function based approximation to the electronic structure of correlated materials is dynamical mean-field theory (DMFT) [29, 30]. In this approximation,

the electron self-energy is approximated as local in a basis of local orbitals and obtained via the self-consistent solution of an "impurity problem" consisting of these orbitals interacting with a bath that approximates the effect of the lattice. DMFT works well when e-e interactions are strong but primarily confined to localized orbitals, such as partially filled d or f orbitals in transition metal compounds.

In DMFT, correlations are treated within an impurity defined by local orbitals containing local interactions and coupling to a bath. The local Green's function can be written

$$G_{\text{loc}}^{(0)}(\omega) = \left[\omega + \mu - V_{\text{loc}}^{(0)} - \Delta(\omega) \right]^{-1}, \quad (1.21)$$

where $\Delta(\omega)$ defines an effective hybridization to the bath and $V_{\text{loc}}^{(0)}$ is the local non-interacting Hamiltonian. In combination with an interaction H_{int} ¹, $G_{\text{loc}}^{(0)}(\omega)$ defines the impurity problem. Solving the impurity problem produces the interacting Green's function $G_{\text{loc}}(\omega)$ and defines the impurity self-energy

$$\Sigma(\omega) = [G_{\text{loc}}^{(0)}(\omega)]^{-1} - [G_{\text{loc}}(\omega)]^{-1}. \quad (1.22)$$

The local self-energy approximation in DMFT enables the self-energy to be upfolded to the lattice, so that the lattice Green's function is

$$G_{\mathbf{k}}(\omega) = \left[\omega + \mu - V_{\mathbf{k}}^{(0)} - U_{\mathbf{k}}^{\dagger} \Sigma(\omega) U_{\mathbf{k}} \right]^{-1}, \quad (1.23)$$

where the matrices $U_{\mathbf{k}}$ and $U_{\mathbf{k}}^{\dagger}$ rotate / project the local orbitals to the lattice states. The final piece of the DMFT approximation is self-consistent determination of the $G_{\text{loc}}^{(0)}(\omega)$ from the interacting lattice Green's function

$$G_{\text{loc}}(\omega) = \frac{1}{N_{\mathbf{k}}} \sum_{\mathbf{k}} G_{\mathbf{k}}(\omega) \quad (1.24)$$

which in combination with Eqs. 1.21 and 1.22 defines the hybridization for the next iteration.

Many variants of DMFT and its first-principles extensions exist. These include

¹Here the interaction is written as a Hamiltonian although it can in general be a frequency dependent action.

different approaches to construct the local correlated orbitals, the interaction Hamiltonian, and the embedding to lattice states; different approaches to solve the correlated impurity problem; and extensions beyond a single site. Approaches to define DMFT in combination with other Green's function methods, most notably variants of GW+DMFT [31, 32], are also promising routes to include non-local correlation and more realistic interactions and double-counting.

1.2.5 Effect of Correlation On Electron-Phonon Interactions

Evidence from a variety of experimental and theoretical sources suggest that beyond DFT correlation effects between electrons can significantly modify the interaction between electrons and phonons. In the context of Hubbard-like models, interactions generally suppress the coupling of phonons to a single orbital or total charge density [33, 34], while often enhancing the coupling of Jahn-Teller modes which couple to orbital splitting [35]. Meanwhile, electronic screening from non-local Coulomb interactions generally suppresses e-ph coupling compared to the bare (unscreened) value [5, 6], but the effect of GW level screening or beyond compared to DFT is more complicated.

Recent methods to calculate e-ph coupling with hybrid functional DFT and GW [36, 37, 38] have helped improve the understanding of e-ph correlation in moderately correlated materials. In some cases, e-ph coupling is increased because semilocal DFT has overestimated the screening. The effects of local, static correlation have also been treated with Hubbard-correct DFPT [39, 40]. These methods have led to improved predictions of superconductivity, spectral kinks, and transport compared to experiment in several cases.

Relatively less is known about how strong electronic correlation (beyond the level of GW) effects e-ph interactions in real materials. One study found that DFT+DMFT level correlation effects significantly increase the response of electronic spectra in FeSe to Γ -point A_{1g} phonon displacements [41], which were found to be large experimentally [42]. Other studies have shown an effect of dynamical correlation (DFT+DMFT compared to DFT+ U) on the energetics of breathing distortions in the moderately correlated metal SrMoO₃ [43], and an effect on optical phonon energies in SrVO₃ and LaNiO₂ [44, 45]. There is a need for further research in this direction, particularly theoretical understanding and methodology for the e-ph coupling beyond the level of DFT+ U and GW.

1.3 Outline of Thesis

In this thesis, I present work from my PhD to investigate e-ph interactions and electronic transport in two regimes beyond the reach of existing DFT based methods: strongly correlated metals and large moiré superlattices in twisted bilayer graphene.

1.3.1 Respective Roles of Interactions in Correlated Metals

First, I present work from the first half of my PhD applying a combination of DFT based e-ph interactions and DFT+DMFT treatment of correlated electrons to understand the physics of correlated metals. In Chapter 2, we turn this approach to Sr_2RuO_4 , investigating the respective contributions of e-ph and e-e interactions to resistivity and the role of correlated electronic states on e-ph scattering processes. In Chapter 3, we study the prototypical correlated metal SrVO_3 , focusing in particular on understanding the origins of experimentally observed transport and spectral properties.

1.3.2 Electron-Phonon Coupling within Dynamical Mean-Field Theory

Next, I present work investigating the important and relatively seldom studied topic of the modification of e-ph coupling strength in real materials due to strong correlation effects. Chapter 4 presents a study developing a finite difference approach to study e-ph coupling of representative phonon modes using DFT+DMFT, the application of this method to SrVO_3 and CaCuO_2 , and resulting physical insights.

1.3.3 Electron-Phonon Interactions and Transport in Moiré Twisted Bilayer Graphene

I then present work to study e-ph interactions and electronic transport in large scale moiré superlattices with 1000s of atoms per unit cell. At such scales, DFT based e-ph calculations are prohibitively expensive, so "second-principles" approaches must be used. In Chapter 5, we develop this approach to study e-ph interactions and resistivity in twisted bilayer graphene, showing the evolution of temperature- and filling-dependent resistivity as the twist angle decreases and the electronic energy scale decreases.

1.3.4 Collaborations with Experimentalists

I conclude by summarizing my contributions to two experimental-computational collaborations in Chapter 6. First, I discuss Boltzmann transport studies of electron mobility in tetragonal SrSnO_3 in collaboration with precision growth and car-

rier mobility studies. Then, I discuss calculations of the electronic structure and paramagnetic surface states in thin film NbO in collaboration with experimental observations of field-induced superconductivity in these films.

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Chapter 2

COMBINING ELECTRON-PHONON AND DYNAMICAL MEAN-FIELD THEORY CALCULATIONS OF CORRELATED MATERIALS: TRANSPORT IN THE CORRELATED METAL Sr_2RuO_4

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Abstract

Electron-electron (e - e) and electron-phonon (e -ph) interactions are challenging to describe in correlated materials, where their joint effects govern unconventional transport, phase transitions, and superconductivity. Here we combine first-principles e -ph calculations with dynamical mean field theory (DMFT) as a step toward a unified description of e - e and e -ph interactions in correlated materials. We compute the e -ph self-energy using the DMFT electron Green’s function, and combine it with the e - e self-energy from DMFT to obtain a Green’s function including both interactions. This approach captures the renormalization of quasiparticle dispersion and spectral weight on equal footing. Using our method, we study the e -ph and e - e contributions to the resistivity and spectral functions in the correlated metal Sr_2RuO_4 . In this material, our results show that e - e interactions dominate transport and spectral broadening in the temperature range we study (50–310 K), while e -ph interactions are relatively weak and account for only $\sim 10\%$ of the experimental resistivity. We also compute effective scattering rates, and find that the e - e interactions result in scattering several times greater than the Planckian value $k_B T$, whereas e -ph interactions are associated with scattering rates lower than $k_B T$. Our work demonstrates a first-principles approach to combine electron dynamical correlations from DMFT with e -ph interactions in a consistent way, advancing quantitative studies of correlated materials.

2.1 Introduction

In strongly correlated materials, characteristic behaviors such as high-temperature superconductivity [1, 2], phase transitions [3, 4], multiferroicity [5], and unconventional transport [6, 7] involve a subtle interplay of charge and lattice degrees of freedom. The atomic vibrations (phonons) couple with the strongly interacting d or f electrons, resulting in electron-electron (e - e) and electron-phonon (e -ph) interactions with nontrivial dependence on orbital, spin, and crystal symmetry [8, 9]. Although heuristic models can address this rich phenomenology [10, 11, 12, 13, 14, 15], deriving generic quantitative frameworks to combine e - e and e -ph interactions remains challenging [16, 17], especially in correlated materials.

First-principles calculations based on density functional theory (DFT) and its linear-response extension, density functional perturbation theory (DFPT), can address the electronic structure [18], lattice dynamics [19], and e -ph interactions [20] in many materials. Yet these methods often fail to describe important features of correlated systems [21], predicting incorrect ground states, lattice vibrations, and e -ph coupling [22, 23]. Recent work has focused on two directions to improve the description of e -ph interactions in correlated materials: Hubbard-corrected DFT (DFT+U), which has enabled calculations of e -ph interactions in a Mott insulator [23], and DFPT with improved electronic correlations (from GW or hybrid functionals) which has revealed correlation-enhanced e -ph interactions in metallic systems [24, 25, 26, 27].

However, while GW and DFT+U can renormalize the band structure and e -ph coupling, both methods describe the electronic states in the quasiparticle (QP) picture, mapping them to noninteracting bands. These approximations are typically used as a starting point to study equilibrium properties and nonequilibrium dynamics [23, 27]. Developing first-principles calculations of e -ph interactions beyond the QP picture remains challenging [20, 28]. Ideally, one would use the full electron Green's function renormalized by e - e interactions, which includes the renormalization of both QP weight and band dispersion, as a starting point to study e -ph interactions in correlated materials [29, 30].

Dynamical mean field theory (DMFT) and its *ab initio* variant, DFT+DMFT, can capture dynamical electronic correlations by mapping the solid to an embedded atomic site with a self-energy local in the atomic orbital basis [31, 32, 33]. These methods have been successful in describing the ground state and transport properties in materials with strongly interacting d or f electrons, including both correlated metals and insulators [34, 35, 36, 6, 37]. A key question addressed in this work

is how one can combine first-principles DMFT and e -ph calculations to explicitly treat e - e and e -ph interactions together in correlated materials.

Here we show first-principles calculations combining e -ph and DMFT e - e interactions using a Green's function approach. We compare two treatments of the e -ph interactions: the Fan-Migdal self-energy with standard approximations [20] (using DFT or DMFT-renormalized band structures) and the same self-energy computed as a convolution integral of the DMFT Green's function. As a proof of principle, we apply this method to Sr_2RuO_4 (SRO) in the normal state, a prototypical correlated metal [38, 39, 40, 41, 42, 8, 43, 44, 37, 45, 46, 47, 48, 49, 50, 51] (and unconventional superconductor below 1 K [52, 53, 54]) requiring treatment of strong e - e interactions between d orbitals [48, 51, 49, 50]. In SRO, we find that the e -ph interactions are relatively weak and momentum dependent, in contrast with the strong local e - e interactions. From Green-Kubo calculations, we find that e - e interactions account for $\sim 50\%$, and e -ph interactions $\sim 10\%$, of the experimental resistivity. Only in the e -ph calculation using the DMFT Green's function the resistivity equals the sum of the e -ph and e - e contributions; in contrast, the standard e -ph calculation with renormalized QP bands leads to an artificial enhancement of e -ph interactions and nonadditive resistivities. The origin of these trends is discussed in detail, together with possible improvements for resistivity calculations in correlated materials and future extensions of our method.

2.2 Numerical Methods

We compute the electron Green's function by combining the DMFT and e -ph self-energies:

$$G_{n\mathbf{k}}(\omega, T) = [\omega + \mu(T) - \varepsilon_{n\mathbf{k}} - \Sigma_{n\mathbf{k}}^{e-e}(\omega, T) - \Sigma_{n\mathbf{k}}^{e-ph}(\omega, T)]^{-1}, \quad (2.1)$$

where T is temperature, $\varepsilon_{n\mathbf{k}}$ are electronic band energies, ω is the electron energy, n is a band index, $\mathbf{k}$ is crystal momentum, and μ is a temperature dependent chemical potential obtained from DMFT. This Green's function includes the DMFT e - e self-energy in the band basis, $\Sigma_{n\mathbf{k}}^{e-e}(\omega, T)$, which is computed starting from DFT, and the e -ph self-energy $\Sigma_{n\mathbf{k}}^{e-ph}(\omega, T)$, which we compute using different approximations as discussed below.

Our calculations in SRO use a DMFT self-energy taken from recent work [48] and transformed from the Wannier-orbital to the band basis using [55]

$$\Sigma_{\mathbf{k}}^{e-e,b} = \mathbf{U}_{\mathbf{k}}^\dagger \Sigma^{e-e,w} \mathbf{U}_{\mathbf{k}}, \quad (2.2)$$

where $\mathbf{U}_{\mathbf{k}}$ are unitary matrices made up by eigenvectors of the Wannier Hamiltonian, while $\Sigma^{\text{e-e},w}$ and $\Sigma_{\mathbf{k}}^{\text{e-e},b}$ are respectively the self-energies in the Wannier and band basis; the former is calculated directly from DMFT and is therefore $\mathbf{k}$ -independent. As in the DMFT calculation, both the Wannier and band basis self-energies are taken to be diagonal¹.

These DMFT calculations use a 3-orbital correlated subspace with t_{2g} symmetry resulting from the hybridization of Ru $4d$ (d_{xy} , d_{yz} , d_{xz}) and O $2p$ orbitals; the orbitals interact with Hund's type Coulomb repulsion and exchange [48]. The DMFT impurity problem is solved on the imaginary-time axis with the TRIQS/CTHYB quantum Monte Carlo solver [56, 57] and analytically continued to the real-frequency axis using Padé approximants [58]. These methods are discussed and validated in greater detail in recent work [48]. Previous studies have clarified the role of spin-orbit coupling (SOC) in SRO [59, 60, 61, 46, 48, 51], which is relevant primarily near band crossings. Because such crossings occur away from the Fermi surface in the majority of the Brillouin zone, we neglect SOC in our calculations.

Our DFT and DFPT calculations are carried out using Quantum Espresso [62] with $10 \times 10 \times 10$ $\mathbf{k}$ -point and $5 \times 5 \times 5$ $\mathbf{q}$ -point grids, and then projected onto Wannier orbitals using the Wannier90 code [63]. We use the Perturbo code for Wannier interpolation of the electronic structure, phonon modes, and e -ph coupling, and for computing the e -ph self-energy, spectral functions, and transport properties [64]. The transport calculations use a $60 \times 60 \times 60$ fine $\mathbf{k}$ -point grid and 10^5 $\mathbf{q}$ -points randomly sampled in the Brillouin zone. Transport results were converged with respect to the $\mathbf{k}$ -point grid density, number of $\mathbf{q}$ -points sampled, the energy window in which the $\mathbf{k}$ -points were chosen, and the frequency grid of the self-energy and spectral functions.

2.3 Results

2.3.1 Electronic structure and e -ph coupling

The electronic structure of SRO is strongly renormalized by electron correlations. The DFT band structure near the Fermi energy consists of relatively narrow d bands [Fig. 2.1(a)]. The e - e interactions significantly decrease the d -band Fermi velocity relative to DFT, as captured by the DMFT electronic spectral functions [Fig. 2.1(b)]. In the 50–310 K temperature range studied here, the e - e interactions also reduce the spectral weight Z of the electronic states on the Fermi surface—to $Z \approx 0.2$ for

¹This assumption is often justified—in SRO, for example, the off-diagonal elements of the DMFT self-energy have been shown to be negligibly small in Ref. [48].

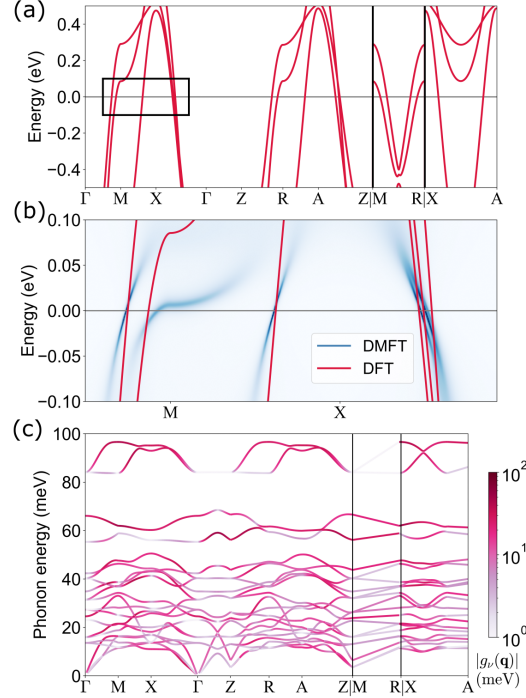


Figure 2.1: Electronic structure and e -ph coupling in SRO. (a) DFT band structure for the t_{2g} d bands included in the Wannierization. (b) DMFT spectral function at 77 K, plotted in an enlargement of the black rectangle in (a), showing significant renormalization relative to the DFT bands near the Fermi energy (shown with a horizontal line). (c) Phonon dispersion colored based on the e -ph coupling strength, which is computed as $|g_v(\mathbf{q})| = (\sum_{mn} |g_{mnv}(\mathbf{k} = M, \mathbf{q})|^2 / N_b)^{1/2}$ [64] by averaging over $N_b = 3$ bands.

the d_{xy} and $Z \approx 0.3$ for the d_{xz} and d_{yz} bands [48] —and cause a large broadening of the associated spectral functions. These effects are a signature of strong electron correlations in SRO and highlight the need to treat the electronic structure beyond the band picture.

The phonon dispersion with a color map of the e -ph coupling strength is shown in Fig. 2.1(c). The dispersions are calculated from real space interpolation of DFPT forces [64] and are in good agreement with inelastic neutron scattering measurements [65]. We find that the e -ph interactions are overall relatively weak in SRO, with coupling strength $|g| < 100$ meV for all phonon modes. These values are significantly smaller than in insulating transition metal oxides —for example, CoO and SrTiO₃ [66, 23] —where the long-range Fröhlich interaction with longitudinal optical (LO) modes [66, 23] reaches coupling strengths of order $|g| \approx 1$ eV. Due to its metallic character, such polar LO phonons are screened out in SRO, resulting in

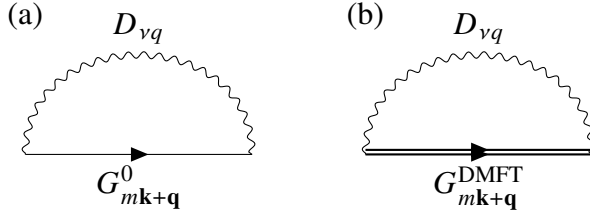


Figure 2.2: Feynman diagrams for the e -ph self-energy $\Sigma_{n\mathbf{k}}^{e\text{-ph}}$, computed with (a) the noninteracting electron Green's function and (b) the DMFT electron Green's function. The vertices carry a factor equal to the e -ph coupling, $g_{mn\nu}(\mathbf{k}, \mathbf{q})$.

short-range e -ph interactions with weaker coupling strengths.

2.3.2 Electron-phonon self-energy calculations

We describe the e - e and e -ph interactions using the corresponding self-energies. When e -ph coupling is relatively weak, the e -ph interactions are well described by the lowest-order (so called Fan-Migdal) self-energy [17]

$$\Sigma_{n\mathbf{k}}^{e\text{-ph}}(i\omega_n) = \sum_{m\nu\mathbf{q}} \sum_{i\nu_n} |g_{mn\nu}(\mathbf{k}, \mathbf{q})|^2 \times G_{m\mathbf{k}+\mathbf{q}}(i\omega_n - i\nu_n) D_{\nu\mathbf{q}}(i\nu_n), \quad (2.3)$$

written as a convolution of electron and phonon propagators, $G_{n\mathbf{k}}(i\omega_n)$ and $D_{\nu\mathbf{q}}(i\nu_n)$ respectively, using Matsubara frequencies $i\omega_n$ for electrons and $i\nu_n$ for phonons; $g_{mn\nu}(\mathbf{k}, \mathbf{q})$ are e -ph coupling matrix elements [64].

In current first-principles calculations, this expression is evaluated using noninteracting electron (and phonon) Green's functions, tacitly assuming that the electron spectral functions consist of a sharp QP peak. In this approximation, the e -ph self-energy on the real-frequency axis ω , computed as in Fig. 2.2(a) with noninteracting electron Green's functions $G_{m\mathbf{k}+\mathbf{q}}^0(z) = (z - \varepsilon_{m\mathbf{k}+\mathbf{q}})^{-1}$, becomes [17, 20]

$$\Sigma_{n\mathbf{k}}^{e\text{-ph}}(\omega, T) = \sum_{m\nu\mathbf{q}} |g_{mn\nu}(\mathbf{k}, \mathbf{q})|^2 \times \left[\frac{N_{\nu\mathbf{q}}(T) + f_{m\mathbf{k}+\mathbf{q}}(T)}{\omega - \varepsilon_{m\mathbf{k}+\mathbf{q}} + \omega_{\nu\mathbf{q}} + i\eta} + \frac{N_{\nu\mathbf{q}}(T) + 1 - f_{m\mathbf{k}+\mathbf{q}}(T)}{\omega - \varepsilon_{m\mathbf{k}+\mathbf{q}} - \omega_{\nu\mathbf{q}} + i\eta} \right], \quad (2.4)$$

where $\omega_{\nu\mathbf{q}}$ are phonon energies while $N_{\nu\mathbf{q}}(T)$ are equilibrium occupation numbers for phonons and $f_{n\mathbf{k}}(T)$ for electrons at temperature T . When DFT is used to obtain the band structure $\varepsilon_{n\mathbf{k}}$, we denote this self-energy approximation as e -ph@DFT.

In systems where DFT fails to describe the electronic structure, several methods are

used to correct the QP band dispersion and e -ph coupling, including GW [67, 68], DFT+U [23], and DFT with hybrid functionals [69]. Therefore, as previously shown in the literature, the e -ph self-energy in Eq. (2.4) can be computed using the “best available” QP band structure and e -ph couplings from one of these methods [70, 67, 69, 71, 68]. Here, for example, by fitting the QP peaks of the DMFT spectral functions we obtain an improved DMFT band structure and use it to compute the self-energy in Eq. (2.4). Below we refer to this level of theory as e -ph@DMFT bands.

However, even with these improved schemes, the standard e -ph self-energy in Eq. (2.4) misses key features of the electronic structure in correlated materials. These include the QP peak broadening, finite QP weight, and any satellite peaks, background, or spectral weight redistribution outside the QP peak, all of which are encoded in the electron spectral function. Improving the QP band structure or e -ph couplings in Eq. (2.4) addresses only part of these renormalization effects because it places the full spectral weight on the electronic band states, neglecting spectral weight redistribution from e - e interactions. The inadequacy of this approximation is apparent in correlated systems like SRO, where the QP weight of the d bands is only $Z \approx 0.2$ – 0.3 . Going beyond these limitations requires computing the e -ph self-energy directly from the electronic spectral function dressed by electron correlations.

To address this challenge, we develop a method to calculate the e -ph self-energy from the DMFT electron Green’s function, G_{nk}^{DMFT} . Starting from the Lehmann representation [17], and using a band-diagonal DMFT self-energy, we write

$$G_{nk}^{\text{DMFT}}(z) = \int d\omega \frac{A_{nk}^{\text{DMFT}}(\omega)}{z - \omega}, \quad (2.5)$$

where $A_{nk}^{\text{DMFT}}(\omega) = -\text{Im}(G_{nk}^{\text{DMFT}}(\omega))/\pi$ is the DMFT spectral function. We substitute this expression in Eq. (2.3) and follow the usual derivation of the lowest-order e -ph self-energy [72]. After analytic continuation to the real frequency axis, $i\omega_n \rightarrow \omega + i\eta$, we obtain:

$$\Sigma_{nk}^{e\text{-ph}}(\omega, T) = \sum_{m, \nu, \mathbf{q}} |g_{mn\nu}(\mathbf{k}, \mathbf{q})|^2 \int d\omega' A_{m, \mathbf{k}+\mathbf{q}}^{\text{DMFT}}(\omega') \left[\frac{(N_{\nu\mathbf{q}}(T) + f(\omega', T))}{\omega - \omega' + \omega_{\nu\mathbf{q}} + i\eta} + \frac{(N_{\nu\mathbf{q}}(T) + 1 - f(\omega', T))}{\omega - \omega' - \omega_{\nu\mathbf{q}} + i\eta} \right]. \quad (2.6)$$

This e -ph self-energy, written as an integral of the DMFT spectral function, captures on equal footing the key factors mentioned above —band renormalization,

finite QP weight and broadening, and background or satellite contributions caused by electron correlations—all of which are included in the DMFT spectral function. This approximation for the e -ph self-energy, here referred to as e -ph@ G^{DMFT} , is shown diagrammatically in Fig. 2.2(b). Its numerical evaluation is discussed in Appendix 2.6. (Note that in this work we do not renormalize the e -ph coupling, $g_{mn\nu}(\mathbf{k}, \mathbf{q})$, using DMFT; the role of that renormalization is discussed in Section 2.4.)

2.3.3 Electron-phonon and DMFT self-energies

We analyze the e -ph self-energy obtained with these different approximations and compare it with the DMFT e - e self-energy. Figure 2.3 shows these quantities on a cut of the Fermi surface in the xy -plane (the real and imaginary frequency dependent self-energy at select $\mathbf{k}$ -points is plotted in SM [73]). In each plot, the middle band with stronger e -ph and e - e coupling has d_{xy} character while the other two bands have d_{xz} and d_{yz} characters. The e -ph self-energy, which depends on both electronic orbital and momentum $\mathbf{k}$, varies by a factor of ~ 1.5 on the Fermi surface at 77 K and 310 K. This $\mathbf{k}$ -dependence in the self-energy also changes direction as a function of temperature in some parts of the Brillouin zone. For example, in the d_{xy} band it is greater near the Γ -M (Γ -X) axis at low (high) temperature, due to a change of available e -ph inter- and intra-band scattering processes. Our DMFT self-energy also depends on orbital character, but it is local and thus $\mathbf{k}$ -independent by construction; it can accurately capture electron correlations in SRO [48].

Comparing our different e -ph self-energy approximations sheds light on the physics they capture. In the picture of renormalized QP bands [72], the Fermi velocity decreases by a factor of $1/Z$ (in SRO, $1/Z$ is ~ 3 for d_{xz} and d_{yz} , and ~ 5 for d_{xy} bands [48]), and the QP weight also decreases by the same factor, leaving the density of states (DOS) at the Fermi energy unchanged. This QP picture holds reasonably well in SRO. The e -ph self-energy from DFT bands (e -ph@DFT), shown in Fig. 2.3(a) at 77 K and in Fig. 2.3(e) at 310 K, misses all these renormalization effects and treats the material as weakly correlated. Computing the e -ph self-energy in Eq. (2.4) with the renormalized DMFT band structure (e -ph@DMFT bands), which is obtained by fitting the QP peaks of DMFT spectral functions, artificially enhances the e -ph self-energy by a factor of 3–5, as shown in Fig. 2.3(b) at 77 K and in Fig. 2.3(f) at 310 K. This artifact is a consequence of using a band structure with Fermi velocity decreased by a factor of $1/Z \approx 3 - 5$, which increases the DOS at the Fermi energy, and thus also increases $\text{Im}\Sigma_{n\mathbf{k}}^{e\text{-ph}}$ in Eq. (2.4) by the same factor.

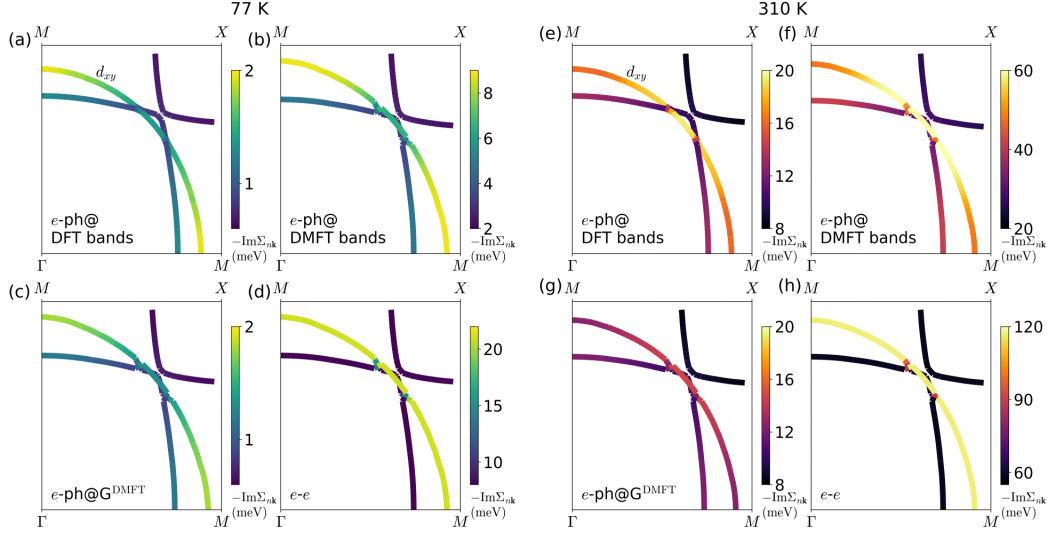


Figure 2.3: Imaginary part of the e -ph and e -e self-energies in SRO, plotted on cuts of the Fermi surface at 77 K (left) and 310 K (right). We plot the e -ph self-energy obtained using (a) the DFT electronic structure (e -ph@DFT), (b) the DMFT band dispersion (e -ph@DMFT bands), and (c) the DMFT spectral function using Eq. (2.6) (e -ph@ G^{DMFT}). (d) DMFT e -e self-energy, shown here for comparison. The corresponding self-energies at 310 K are plotted in (e)-(h) respectively.

However, the DOS at the Fermi energy is nearly unchanged in the renormalized electronic structure because the decrease in Fermi velocity is compensated by a corresponding decrease in QP weight. This physics is correctly described by Eq. (2.6), where the imaginary part of the e -ph self-energy is roughly proportional to the DOS at Fermi energy written as $D(E_F) \propto \sum_{\mathbf{n}\mathbf{k}} \int d\omega' A_{\mathbf{n}\mathbf{k}}(\omega') \delta(E_F - \omega')$, which captures changes in the QP weights. Therefore, as we show in Fig. 2.3(c) at 77 K and in Fig. 2.3(g) at 310 K, the more accurate e -ph self-energy computed from the DMFT spectral functions using Eq. (2.6) (e -ph@ G^{DMFT} method) removes the artificial enhancement introduced by using the “best-available band structure” approach.

Overall, the e -ph self-energy obtained from DFT bands in Fig. 2.3(a) and DMFT Green’s functions in Fig. 2.3(c) show similar magnitude and trends at 77 K mainly because the DOS at Fermi energy is nearly unchanged in DFT and DMFT. However, at higher temperatures the effects of broadening in the DMFT spectral functions become more pronounced; for example, at 310 K we find a decrease in $\mathbf{k}$ -dependence in the e -ph self-energy from DMFT Green’s function, as shown by comparing Fig. 2.3(e) and 2.3(g). We attribute this difference to smearing of sharp features in the electronic structure by the spectral width, which is not captured by the DFT-based calculation; this effect is more pronounced in the strongly correlated d_{xy} band. Our

results show the importance of computing the e -ph self-energy from the electron spectral functions in correlated materials to capture the subtle interplay of e -ph and e - e interactions.

2.3.4 Spectral functions

We obtain the spectral function including both e - e and e -ph interactions using the Green's function in Eq. (2.1):

$$A_{n\mathbf{k}}(\omega, T) = -(1/\pi) \text{Im } G_{n\mathbf{k}}(\omega, T). \quad (2.7)$$

To evaluate this expression, our most accurate approximation consists of first computing the DMFT e - e self-energy starting from DFT and then obtaining the e -ph self-energy from the DMFT spectral function. This e -ph@ G^{DMFT} approach is justified by the adiabatic approximation: the fast e - e interactions renormalize the electronic states, and the slow nuclear motions governing e -ph coupling occur in this renormalized ground state.

Figures 2.4(a)–(b) show the electron spectral functions computed in SRO with this approach and plotted in the $\Gamma \rightarrow M$ direction at 77 K and 310 K. The main features are consistent with photoemission and transport measurements [48, 38, 41] —in particular, the spectral functions remain fairly sharp near the Fermi energy below ~ 100 K, indicating well-defined QP excitations at low temperature [6]. The spectral broadening increases at higher temperatures, further demonstrating the importance of computing e -ph interactions beyond the band picture. Figures 2.4(c)–(d) compare the spectral functions and their QP peak broadening at a fixed $\mathbf{k}$ -point ($\mathbf{k} = 0.75M$) with and without the inclusion of e -ph interactions. We find that the e -ph interactions broaden the QP peaks only slightly, consistent with the overall weak e -ph coupling in SRO. Increasing the temperature, rather than including e -ph interactions, is the main factor responsible for broadening. We find similar trends when analyzing spectral functions in the $\Gamma \rightarrow X$ direction [73]. These results point to a dominant role of e - e interactions in SRO.

2.3.5 Transport

We study electrical transport in SRO in the characteristic bad metallic regime [34], focusing on understanding the roles of e - e and e -ph interactions. We compute the optical conductivity from our combined DMFT plus e -ph spectral function in

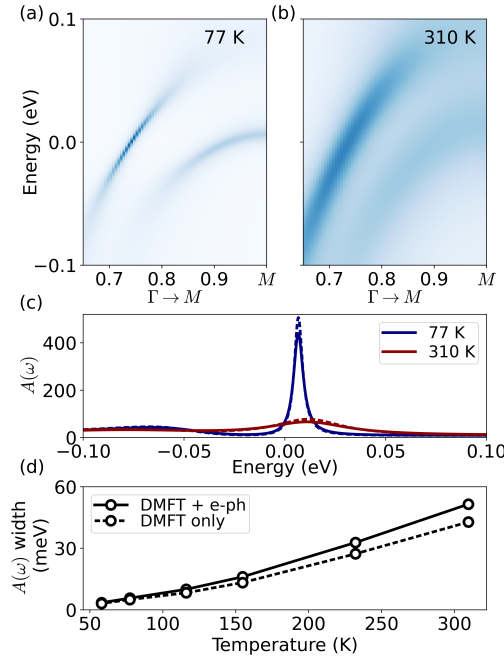


Figure 2.4: Electronic spectral functions in the $\Gamma \rightarrow M$ direction computed using the DMFT e - e plus e -ph@ G^{DMFT} self-energies at (a) 77 K and (b) 310 K. (c) Spectral functions at $\mathbf{k} = 0.75M$ showing DMFT quasiparticle peaks (dashed lines) broadened by the inclusion of e -ph interactions (solid lines) at two temperatures. (d) Calculated full-width at half maximum for the spectral functions in (c), showing quantitatively the small e -ph broadening effect.

Eq. (2.7), using Green-Kubo theory without current-vertex corrections [17]:

$$\sigma_{\alpha\beta}(\omega, T) = \frac{\pi \hbar e^2}{V_{\text{uc}}} \int d\omega' \frac{f(\omega', T) - f(\omega' + \omega, T)}{\omega} \times \sum_{n\mathbf{k}} v_{n\mathbf{k}}^{\alpha} v_{n\mathbf{k}}^{\beta} A_{n\mathbf{k}}(\omega', T) A_{n\mathbf{k}}(\omega' + \omega, T), \quad (2.8)$$

where $v_{n\mathbf{k}}$ are band velocities, α and β are Cartesian directions, and V_{uc} is the unit cell volume; the dc resistivity tensor is obtained as $\rho_{\text{dc}}(T) = \sigma^{-1}(\omega \rightarrow 0, T)$. This approximation has been used (separately) to study e -ph limited transport in oxides and organic crystals [66, 68] and e - e limited transport in correlated metals [37, 74, 75, 76]; different from these previous studies, here we focus on combining the e - e and e -ph interactions and studying their interplay.

Figure 2.5(a) shows the resistivity in the xy -plane as a function of temperature. We compare the resistivity limited by e - e interactions alone, computed from the Green's function including only the DMFT self-energy, with calculations including both e - e and e -ph interactions in the Green's function. When both interactions are

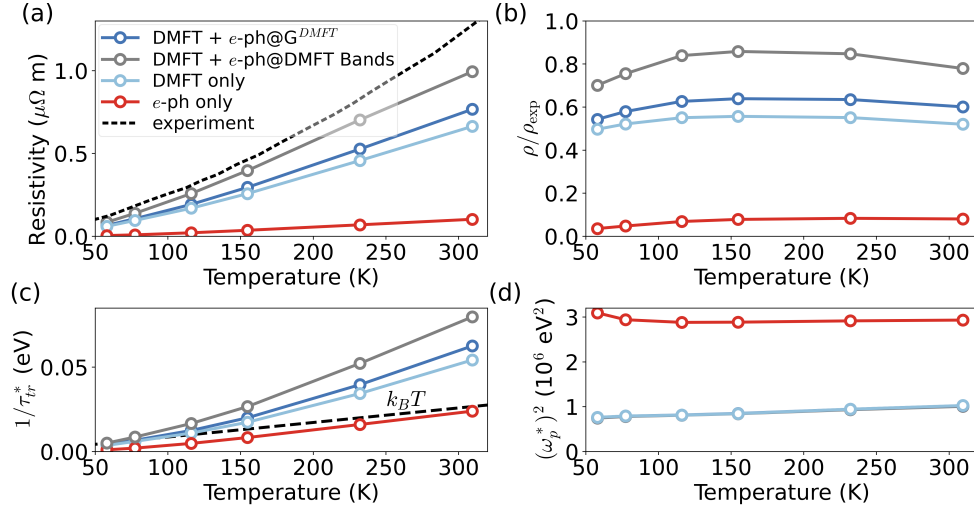


Figure 2.5: Transport properties as a function of temperature in SRO. (a) In-plane resistivity, comparing DMFT+ $e\text{-ph}$, DMFT-only, and $e\text{-ph}$ -only calculations to experimental data from Ref. [40]. (b) Same as (a), but plotted as a fraction of the experimental resistivity. (c) Effective scattering rate, and (d) effective plasma frequency. In the legend, DMFT only and $e\text{-ph}$ only refer to using, respectively, only the DMFT or $e\text{-ph}$ self-energies in the Green’s function, spectral function, and Green-Kubo formula.

included, we compare results from the more accurate $e\text{-ph}@G^{\text{DMFT}}$ approach with calculations using Eq. (2.4) with the best-available band structure ($e\text{-ph}@G^{\text{DMFT}}$ bands method). Experimental values are also shown for comparison [40]. For convenience, Fig. 2.5(b) shows the same results expressed as a fraction of the experimental resistivity.

The calculation including only DMFT $e\text{-e}$ interactions correctly predicts the order of magnitude of the experimental resistivity, providing a resistivity smaller than experiment by a factor of 2 in the entire temperature range studied here (50–310 K). This calculation is comparable with results from Deng *et al.* [37], who used a different scheme to construct the correlated subspace in the DMFT calculation and obtained somewhat larger values for the $e\text{-e}$ limited resistivity due to stronger interaction parameters U and J . In contrast, the resistivity limited by the $e\text{-ph}$ interactions alone (computed with the $e\text{-ph}@G^{\text{DMFT}}$ approach) is only $\sim 10\%$ of the experimental value.

When combining $e\text{-e}$ and $e\text{-ph}$ interactions through the $e\text{-ph}@G^{\text{DMFT}}$ approach, the resulting resistivity is the sum of the individual $e\text{-e}$ and $e\text{-ph}$ limited resistivities, a behavior known as Matthiessen’s rule [16]. Therefore, adding $e\text{-ph}$ interactions

improves the agreement with experiment and predicts a resistivity equal to $\sim 60\%$ of the experimental value between 50–310 K. In contrast, using only an improved QP band structure (*e*-ph@DMFT bands) artificially enhances the *e*-ph interactions, leading to an incorrect prediction that the *e*-ph contribution is 30% of the experimental resistivity [see Fig. 2.5(b)]. Note that our most accurate calculation still underestimates the experimental resistivity by $\sim 40\%$. The possible origin of this “missing resistivity” is discussed below.

To better understand the contributions to the resistivity, we analyze our transport results using an effective Drude model [6], writing the resistivity as $\rho_{\text{dc}} = 4\pi/[\tau_{\text{tr}}^*(\omega_p^*)^2]$, where $1/\tau_{\text{tr}}^*$ is an effective scattering rate and ω_p^* is an effective plasma frequency. We extract these quantities from our computed optical conductivity $\sigma(\omega)$ using [6]

$$\tau_{\text{tr}}^* = -\frac{2}{\pi\sigma_{\text{dc}}} \int_0^\infty \frac{1}{\omega} \frac{\partial \text{Re}(\sigma(\omega))}{\partial \omega} d\omega \quad (2.9)$$

and

$$(\omega_p^*)^2 = \frac{-2\pi^2\sigma_{\text{dc}}^2}{\int_0^\infty \frac{1}{\omega} \frac{\partial \text{Re}(\sigma(\omega))}{\partial \omega} d\omega}. \quad (2.10)$$

Figure 2.5(c) shows the effective scattering rate $1/\tau_{\text{tr}}^*$ and Fig. 2.5(d) the plasma frequency $(\omega_p^*)^2$ obtained from this analysis. These results confirm that *e*-*e* interactions dominate electron scattering and lead to a decreased Fermi velocity (and thus plasma frequency) by renormalizing the band structure. The effective *e*-ph scattering rate is significantly smaller (and the plasma frequency greater) than for *e*-*e* interactions, consistent with the small *e*-ph contribution to the resistivity and negligible band renormalization (relative to DFT) from *e*-ph interactions. In addition, most of the temperature dependence of the resistivity comes from the effective scattering rates, while the effective plasma frequency depends weakly on temperature.

Figure 2.5(c) additionally compares the effective scattering rates with the Planckian limit $\Gamma_p = k_B T$ [77]. Interestingly, the *e*-ph scattering rate is lower than Γ_p and approaches it near 310 K, whereas the scattering from *e*-*e* interactions exceed Γ_p above ~ 100 K, reaching $\sim 2k_B T$ at 310 K. The weaker *e*-ph scattering in SRO contrasts the cases of insulating oxides such as SrTiO₃ and CoO [66, 23], where pronounced polaron effects are present—both materials exhibit polaron satellites, and in SrTiO₃ the effective *e*-ph scattering rate exceeds the Planckian limit [66].

2.4 Discussion

Our most accurate transport calculation underestimates the in-plane resistivity in SRO by $\sim 40\%$ relative to experiments. The origin of this discrepancy deserves a detailed discussion. First, we verify that our lowest-order (Fan-Migdal) treatment of e -ph interactions is sufficient in SRO by recomputing the resistivity with an e -ph cumulant method that can describe higher-order e -ph interactions and transport in the presence of polarons [66, 68]. Consistent with the effective screening of polar phonons in metals, we find that the e -ph limited transport is nearly identical in the lowest-order and cumulant calculations [73].

Second, our Green-Kubo calculations neglect current-vertex corrections, which typically play a small role in e -ph limited transport in metals. While vertex corrections are difficult to compute in Green-Kubo theory [78, 79], they can be quantified in the semiclassical limit using the Boltzmann transport equation (BTE). In particular, including vertex corrections corresponds to a full solution of the BTE, obtained here with an iterative approach (ITA) [64], while neglecting vertex corrections is equivalent to the relaxation time approximation (RTA), to which the Green-Kubo formula reduces in the weak-coupling limit [66]. Therefore, we can estimate the effect of vertex corrections by comparing our e -ph limited resistivity computed with Green-Kubo to BTE calculations using the ITA and RTA, which respectively include and neglect vertex corrections [79]. From low temperature up to 310 K, the ITA resistivity is $\sim 20\%$ greater than the RTA value, while the RTA and Green-Kubo calculations agree to within 2% at 310 K [73]. These results indicate that vertex corrections have a modest effect on e -ph limited transport.

Third, most DMFT studies of transport neglect vertex corrections from e - e interactions, which is strictly correct only in the limit of infinite dimensions [74]. However, their role is difficult to quantify and often non-negligible [80]. Fourth, although our DMFT calculations produce band dispersions in excellent agreement with photoemission data [48], DMFT results can in some cases be sensitive to the method used to construct the local site [81], and neglecting nonlocal interactions may remove relevant e - e scattering mechanisms which affect the calculated spectral width. Studying these effects is an active area of DMFT research and is beyond the scope of this work.

Finally, recent work has shown that DFPT based on semilocal functionals can lead to underestimated e -ph coupling in correlated materials [24, 26]. Yet much of that work has focused on materials with nonlocal correlations, so it is difficult to estimate the size of the effect in SRO, where correlations are dominantly local [48]. It is

possible that the e -ph coupling and its role in transport are somewhat stronger than captured by our DFPT calculations. However, an enhancement of the e -ph coupling consistent with those reported to date would not change our main conclusion that the e -ph contribution to the resistivity is relatively small—for example, assuming the limit case of a doubling of e -ph coupling strength $|g|^2$ would still give only a $\sim 20\%$ e -ph resistivity contribution.

A qualitative comparison between e - e and e -ph interactions is also interesting. The $\mathbf{k}$ -dependence of the e -ph interactions clearly contrasts the local ($\mathbf{k}$ -independent) e - e interactions. Extending this comparison to other correlated materials may highlight key differences between materials with dominant e - e or e -ph interactions. We also observe that our e -ph@ G^{DMFT} self-energy generally has a weaker $\mathbf{k}$ -dependence than predicted by the e -ph@DFT method, suggesting that electron correlations suppress nonlocal e -ph interactions. This effect may have interesting implications for materials with strong or long-range e -ph interactions, including cuprates and correlated insulators.

Lastly, our development of the e -ph@ G^{DMFT} method fits within efforts in computational many-body physics to obtain accurate components of Feynman diagrams. For e -ph interactions, this includes improving the accuracy of the electron propagator G , phonon propagator D , and e -ph coupling g . When viewed this way, there is a clear analogy with the use of different levels of theory in the GW method, where the accuracy can be improved by using the “best G ” and “best W ”. In e -ph calculations, methods for Hubbard- [23] or DMFT-corrected phonons [82, 83], as well as anharmonic lattice dynamics [84, 66], address the phonon propagator D , while Hubbard-corrected DFPT [23], GW perturbation theory (GWPT) [26], and DFT+DMFT deformation potentials [85] aim to improve the e -ph coupling g . These effects can be treated at a suitable level of theory for each material; for example, if the lattice is strongly anharmonic, the phonon propagator can be computed using renormalized phonon frequencies, or one can use a full frequency dependent phonon propagator analogous to our use of the DMFT Green’s function.

Our work improves the accuracy of the electron propagator G entering the e -ph self-energy diagram, by dressing it with the DMFT self-energy to capture dynamical electron correlations. This provides a scheme where the e -ph self-energy is computed from the DMFT spectral functions. Our results highlight the importance of using the spectral functions to compute e -ph interactions in correlated materials, and cautions against using just the best available QP band structure—if the dispersion is renormalized but the QP weight is not adjusted, the e -ph self-energy

becomes inaccurate as we have shown for SRO.

2.5 Conclusion

We demonstrated a method to combine strong dynamical correlations described by DMFT with first-principles e -ph calculations. This advance enables a quantitative treatment of both e - e and e -ph interactions and their effect on transport in correlated materials. Our approach computes e -ph interactions from the DMFT Green's function, which captures band structure and spectral weight renormalization from strong electron correlations. In SRO, where e -ph interactions are relatively weak, we have shown that transport is governed by e - e interactions, with e -ph interactions contributing only $\sim 10\%$ of the experimental resistivity in a wide temperature range. Our work expands the reach of first principles e -ph calculations to strongly correlated materials by treating the electronic structure at the level of the spectral function rather than non-interacting bands. Note that our calculations can also use more accurate versions of the phonon propagator (e.g., Hubbard-corrected DFT, DMFT, or anharmonic lattice dynamics) and e -ph coupling (e.g., from GWPT or DFPT+U) as an input for calculating interactions, and thus are complementary to these approaches. The method presented in this work enables studies of a range of correlated materials, particularly when combined with emerging approaches to treat e -ph interactions in materials with spin-orbit coupling [64], magnetism, and Mott insulating gaps [23]. Considering the wide variety of physical parameters in correlated materials—number of relevant orbitals, conducting or insulating, type of magnetic ordering, strength of spin-orbit coupling, etc.—one expects that the interplay of e - e and e -ph interactions will vary greatly in these systems, opening wide-ranging possibilities for future work.

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2.6 Appendix A: Numerical Evaluation of the Electron-Phonon Self-Energy

Computing the e -ph self-energy from DMFT spectral functions using Eq. (2.6) requires an additional frequency integral compared to the standard e -ph self-energy in Eq. (2.4). We develop an efficient approach to compute this e -ph@ G^{DMFT} self-energy. Using the relation

$$\frac{1}{\omega + i\eta} = P\left(\frac{1}{\omega}\right) - i\pi\delta(\omega), \quad (2.11)$$

where P denotes the principal part, allows us to write the imaginary part of the e -ph self-energy as

$$\begin{aligned} \text{Im}\Sigma_{n\mathbf{k}}^{e\text{-ph}}(\omega) = & -\pi \sum_{m\nu\mathbf{q}} |g_{mn\nu}(\mathbf{k}, \mathbf{q})|^2 \\ & [(N_{\nu\mathbf{q}} + f(\omega + \omega_{\nu\mathbf{q}}))A_{m\mathbf{k}+\mathbf{q}}(\omega + \omega_{\nu\mathbf{q}}) \\ & + (N_{\nu\mathbf{q}} + 1 - f(\omega - \omega_{\nu\mathbf{q}}))A_{m\mathbf{k}+\mathbf{q}}(\omega - \omega_{\nu\mathbf{q}})]. \end{aligned} \quad (2.12)$$

This way, the calculation has a computational cost similar to the standard e -ph self-energy in Eq. (2.4), but it additionally requires the shifted DMFT spectral functions

$$A_{n\mathbf{k}}^{\text{DMFT}}(\omega' = \omega \pm \omega_{\nu\mathbf{q}}) = -\frac{1}{\pi} \text{Im} \left(\frac{1}{\omega' - \varepsilon_{n\mathbf{k}} - \Sigma_{n\mathbf{k}}^{e-e}(\omega')} \right). \quad (2.13)$$

In DMFT, $\Sigma_{n\mathbf{k}}^{e-e}(\omega)$ is local in the Wannier basis and thus it can be rapidly computed using Eq. (2.2) and interpolated to the shifted energies $\omega' = \omega \pm \omega_{\nu\mathbf{q}}$. The real part of the e -ph@ G^{DMFT} self-energy, which is not computed in this work, can be obtained from the imaginary part using the Kramers-Kronig relations without repeating the sum over phonon modes and momenta.

In our calculations, we obtain the e -ph@ G^{DMFT} self-energy using a fine energy grid with 0.5 meV spacing in a window of ± 100 meV around the Fermi energy (E_F), together with a coarser grid with 20 meV spacing in the $E_F \pm 2$ eV energy window. We obtain the spectral functions in Eq. (2.13) using Wannier-interpolated band energies and the DMFT self-energy interpolated in frequency from a grid with 4 meV spacing.

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RESPECTIVE ROLES OF ELECTRON-PHONON AND ELECTRON-ELECTRON INTERACTIONS IN THE TRANSPORT AND QUASIPARTICLE PROPERTIES OF SrVO_3

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Abstract

The spectral and transport properties of strongly correlated metals, such as SrVO_3 (SVO), are widely attributed to electron-electron (e - e) interactions, with lattice vibrations (phonons) playing a secondary role. Here, using first-principles electron-phonon (e -ph) and dynamical mean field theory calculations, we show that e -ph interactions play an essential role in SVO: they govern the electron scattering and resistivity in a wide temperature range down to 30 K, and induce an experimentally observed kink in the spectral function. In contrast, the e - e interactions control quasiparticle renormalizations and low temperature transport, and enhance the e -ph coupling. We clarify the origin of the near T^2 temperature dependence of the resistivity by analyzing the e - e and e -ph limited transport regimes. Our work disentangles the electronic and lattice degrees of freedom in a prototypical correlated metal, revealing the dominant role of e -ph interactions in SVO.

Introduction— Strontium vanadate, SrVO_3 (SVO), is a perovskite oxide widely studied as a prototypical correlated metal [1, 2, 3]. Experiments have measured transport and spectral functions in detail in SVO, owing to advances in growth of clean samples [4, 5] and characterization by angle-resolved photoemission spectroscopy [6, 7, 8]. There are clear spectroscopic signatures of strong electron interactions in SVO, including kinks in the quasiparticle dispersion [9, 8] and mass enhancement with quasiparticle weight $Z \approx 0.5$ [6]. In addition, transport measurements have found a near T^2 -dependent resistivity in broad temperature ranges below 300 K [10, 11, 12, 13, 14].

These findings are often attributed to strong electron-electron (e - e) interactions. As a result, SVO serves as a testbed for theoretical methods treating strongly correlated materials, including first-principles variants of dynamical mean field theory (DMFT) such as density functional theory (DFT)+DMFT [15], GW+DMFT [16, 17, 18, 19], and linear response DMFT [20], and the dynamical cluster [21] and dynamical vertex approximations [22].

However, one can question whether the transport properties and spectral features observed in SVO are the result of purely electronic interactions. In particular, electron-phonon (e -ph) interactions may also play a role in SVO, as they do in other correlated metals where experiments [23, 24, 25] and theory [26, 27, 28] have highlighted the importance of e -ph coupling for spectral kinks [23, 29] and electronic transport [30]. A quantitative study combining e - e and e -ph interactions in SVO is needed to clarify the microscopic origin of its electronic behavior.

In this Letter, we show calculations of spectral and transport properties in SVO combining first-principles e -ph interactions with DFT+DMFT e - e interactions [30]. We find that e -ph interactions govern the resistivity and its temperature dependence above ~ 30 K, and account for the experimentally observed kinks and for most of the linewidth broadening of the spectral functions. In contrast, the e - e interactions control the resistivity below 20 K, and are responsible for most of the quasiparticle mass renormalization. We also find that the e - e interactions lead to an enhancement of the effective e -ph coupling. Our results provide a blueprint for quantifying electronic and lattice contributions to the properties of correlated metals.

Electronic Structure and Electron-Phonon Coupling— We calculate the electronic structure, phonon dispersions, and e -ph coupling using DFT and density functional perturbation theory (DFPT) with the Quantum Espresso package [31, 32, 33, 34]. We use the experimental lattice parameter of 3.842 Å [2, 13] and project the electronic structure onto the t_{2g} d -orbitals of vanadium [35]. We use Perturbo to compute the e -ph interactions, e -ph self-energy, spectral functions, and transport [36]. The e - e self-energy is obtained with DFT+DMFT using the TRIQS code with a continuous-time quantum Monte Carlo solver [37, 38, 39, 40, 41, 42] and Padé analytical continuation [38]. We use Hubbard-Kanamori interactions with $U = 4.5$ eV and $J = 0.675$ eV to produce spectra in agreement with experiment [6, 7, 8]. Additional computational details are provided in the Supplemental Material (SM).

As shown in Fig. 3.1(a), our DFT calculations predict a bandwidth of 2.5 eV for the t_{2g} electronic bands, which is renormalized by a factor $Z \approx 0.5$ by DMFT, in agree-

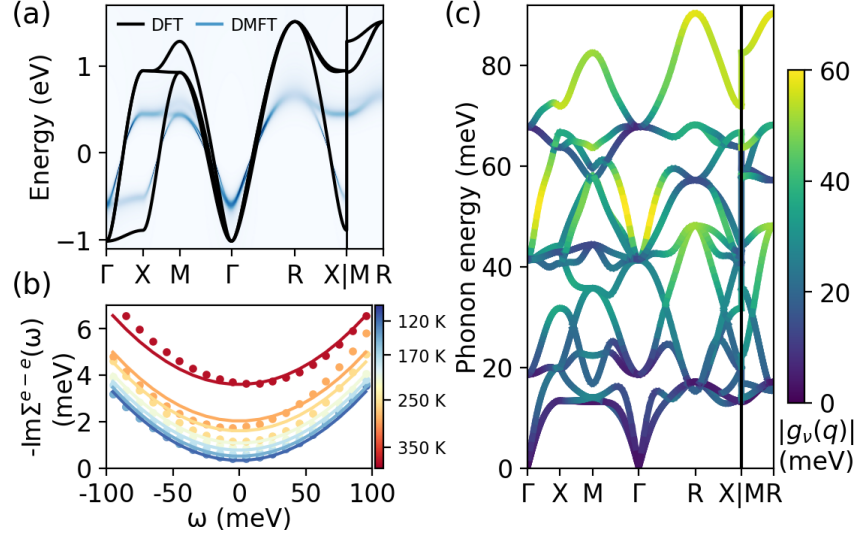


Figure 3.1: (a) DFT electronic band structure (black) and the spectral function computed with DMFT at 290 K (blue), showing renormalization by a factor $Z \approx 0.5$. (b) Imaginary part of the electron self-energy due to e - e interactions, computed with DMFT. The lines show a fit to the Fermi liquid form, $\text{Im}\Sigma^{e-e}(\omega, T) = -c((\hbar\omega)^2 + \pi^2(k_B T)^2)$ with $c \approx 0.33\text{eV}^{-1}$. (c) DFPT phonon dispersions in SVO, with colors showing the e -ph coupling strength $|g_\nu(\mathbf{q})|$ averaged on the Fermi surface.

ment with experiments [6] and previous DMFT results [15, 17]. In the temperature range we study (~ 115 – 390 K), the imaginary part of the e - e self-energy, $\text{Im}\Sigma^{e-e}$, follows a Fermi liquid behavior. Figure 3.1(b) shows that $\text{Im}\Sigma^{e-e}(\omega, T)$ within 100 meV of the Fermi energy can be fit closely by a Fermi liquid parameterization [43], $\text{Im}\Sigma^{e-e}(\omega, T) = -c((\hbar\omega)^2 + \pi^2(k_B T)^2)$ with $c \approx 0.33\text{ eV}^{-1}$ ¹. Therefore, based on the Kramers-Kronig relations, $\text{Re}\Sigma^{e-e}(\omega, T)$ and the quasiparticle dispersion near the Fermi energy depends weakly on temperature.

Figure 3.1(c) shows the phonon dispersions in SVO computed with DFPT and color-coded according to the e -ph coupling strength $|g_\nu(\mathbf{q})|$, for each phonon mode ν and momentum $\mathbf{q}$, averaged over the Fermi surface (see SM). The e -ph coupling is stronger for the six highest-energy modes, which involve distortions of the VO_6 octahedra, such as Jahn-Teller modes.

Spectral Properties— We investigate the contributions of e -ph and e - e interactions in SVO by computing the corresponding self-energies [30]. The real and imaginary parts of the e - e and e -ph self-energies at 115 K are shown in Fig. 3.2(a)-(b), respectively. The e - e interactions dominate quasiparticle renormalization, as seen from the greater derivative of $\text{Re}\Sigma^{e-e}$ compared to $\text{Re}\Sigma^{e-ph}$ within 150 meV of the Fermi

¹In the following, we set $\hbar = k_B = 1$

energy. Accordingly, extracting quasiparticle weights $Z = (1 - \frac{\partial \text{Re}\Sigma(\omega)}{\partial \omega}|_{\omega=0})^{-1}$ with a fit near the Fermi surface, gives a weak contribution to renormalization for e -ph interactions ($Z_{e\text{-ph}} = 0.81$) and a dominant contribution for e - e interactions, with $Z_{e\text{-}e} = 0.53$ and $Z_{\text{both}} = 0.47$.

The imaginary part of the self-energy shows an opposite behavior: $\text{Im}\Sigma^{e\text{-ph}}$ is much greater than $\text{Im}\Sigma^{e\text{-}e}$, and thus the e -ph interactions account for the majority of electron scattering and spectral width at low energy. The dominant role of e - e interactions on quasiparticle renormalization in SVO, despite their small effect on low energy scattering consistent with a Fermi liquid, can be rationalized using the Kramers-Kronig relations [43, 44]: due to the larger energy scales involved, the e - e interactions dominate the imaginary part of the self-energy at higher energies (see SM), leading to a greater magnitude (and energy derivative) of $\text{Re}\Sigma^{e\text{-}e}(\omega)$ compared to $\text{Re}\Sigma^{e\text{-ph}}(\omega)$ at low energy.

We compute the spectral function $A_{n\mathbf{k}}(\omega) = -\frac{1}{\pi}\text{Im}G_{n\mathbf{k}}(\omega)$ from the Green's function

$$G_{n\mathbf{k}}(\omega, T) = [\omega - \varepsilon_{n\mathbf{k}} + \mu - \Sigma_{n\mathbf{k}}(\omega, T)]^{-1} \quad (3.1)$$

at energy ω for electron band n and momentum $\mathbf{k}$. Here, $\varepsilon_{n\mathbf{k}}$ is the DFT band energy, μ is the Fermi energy, and $\Sigma_{n\mathbf{k}}(\omega, T)$ is the electron self-energy. Following our previous work [30], in separate calculations we compute this Green's function using the self-energy from DMFT e - e interactions, the lowest-order self-energy from e -ph interactions, and their sum², obtaining corresponding spectral functions capturing different combinations of interactions [Fig. 3.2(c)-(e)]. The spectral functions from e -ph, and those from e -ph plus e - e interactions, show a kink around 60 meV from the Fermi energy which has been observed in experiments [9, 8]. There is a corresponding sharp change in the derivative of $\text{Re}\Sigma_{n\mathbf{k}}^{e\text{-ph}}(\omega)$ at this energy [Fig. 3.2(a)], whereas this feature is absent in $\text{Re}\Sigma_{n\mathbf{k}}^{e\text{-}e}(\omega)$. This result shows that the 60 meV kink observed experimentally in SVO is caused by e -ph interactions.

Transport.— Numerous experiments have measured a near T^2 temperature dependence of the resistivity in SVO below 300 K [45, 46, 2, 4, 11, 47, 48, 49, 13, 14]. Due to the strong electronic correlations in SVO, several studies have attributed this resistivity to e - e interactions in the Fermi liquid regime [45, 11, 49], where T^2 behavior is expected. An exception is recent work by Mirjolet *et al.*, who argued that the temperature dependence is better explained by e -ph limited resistivity with strong coupling to a dominant phonon mode [12]. Recently, the growth of ultra-

²The e -ph self-energy calculation uses the DFT electron propagator. Unlike in Sr_2RuO_4 [30], using the DMFT electron propagator has a negligible effect in SVO.

clean samples has enabled detailed measurements of the resistivity with reduced defect scattering [4, 13, 14]. In these samples, Ahn *et al.* [13] and Brahlek *et al.* [14] find a near- T^2 resistivity below 25 K and between about 100–300 K, together with a stronger than T^2 temperature dependence at intermediate temperatures.

To understand the microscopic origin of this behavior, we compute the resistivity arising from e -ph and e - e interactions using the Green-Kubo formula [50, 30]:

$$\rho_{\alpha\beta}^{-1}(T) = \frac{\pi\hbar e^2}{V_{uc}} \int d\omega (-f'(\omega, T)) \sum_{nk} v_{nk}^\alpha v_{nk}^\beta A_{nk}(\omega, T)^2, \quad (3.2)$$

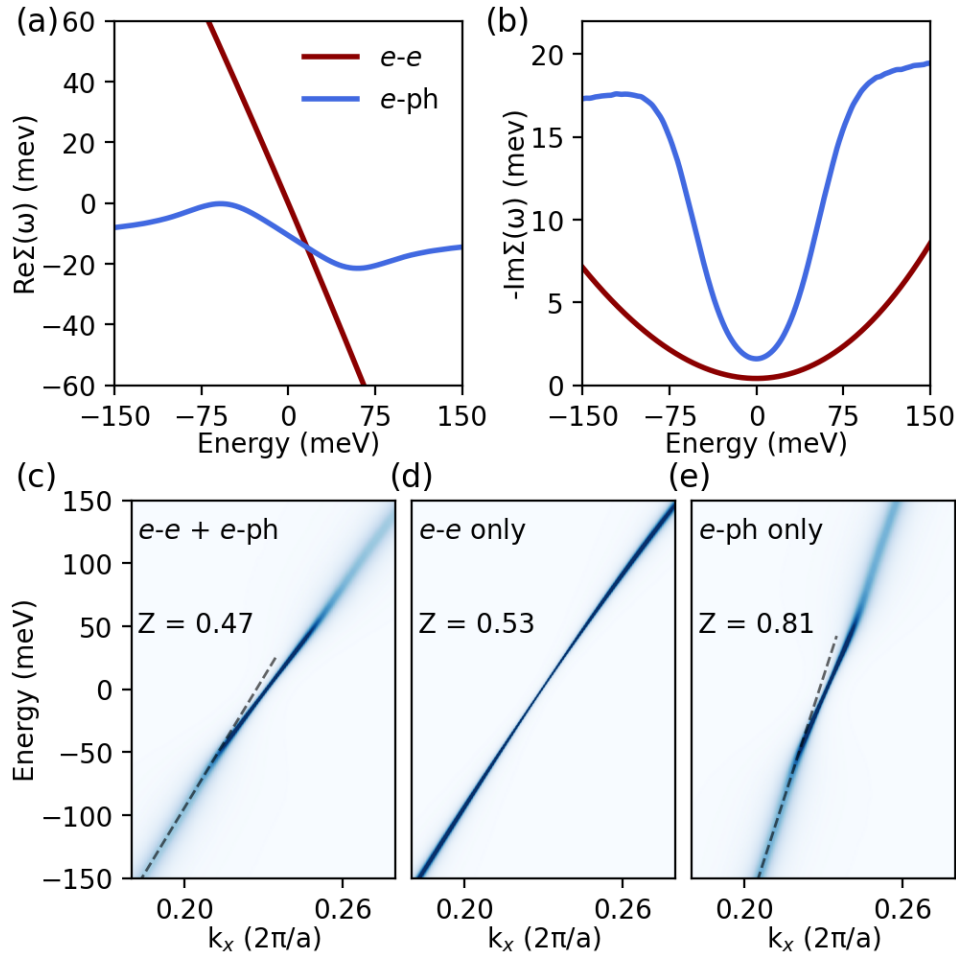


Figure 3.2: (a) Real and (b) imaginary parts of the self-energy at $T = 115\text{K}$ and $\mathbf{k} = (0.23 \times 2\pi/a, 0, 0)$, showing contributions from e - e and e -ph interactions. (c) Spectral functions including both e - e and e -ph interactions, (d) e - e interactions only, and (e) e -ph interactions only. Quasiparticle weights Z are indicated for each spectral function, and the dashed lines in (c) guide the eye to the quasiparticle dispersion near the kink. All spectral functions are shown along the Γ -X direction near the Fermi surface at 115 K.

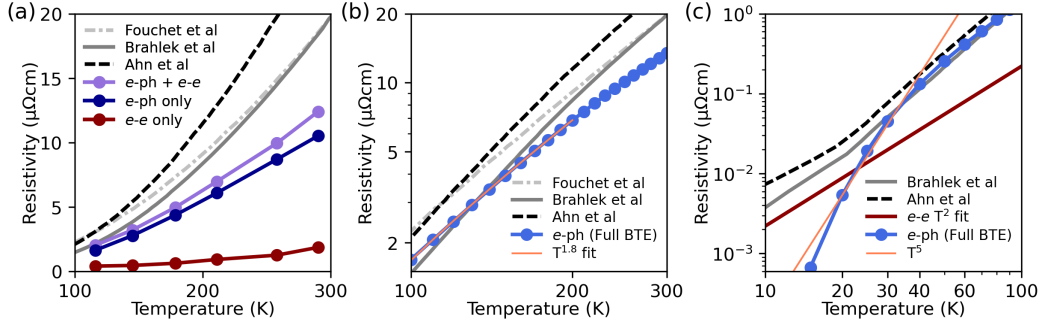


Figure 3.3: (a) Resistivity as a function of temperature calculated using the Green-Kubo formalism with e - e interactions, e -ph interactions, and their combination. (b) Temperature dependence of the e -ph limited resistivity calculated using the full (iterative) solution of the BTE. (c) Low-temperature e -ph and e - e limited transport. Experimental data (from which the $T = 0$ residual resistivity was subtracted) are from Refs. [11, 13].

where $\rho_{\alpha\beta}$ is the resistivity tensor, α and β are Cartesian directions, $f'(\omega, T)$ is the energy derivative of the Fermi occupation factor, $v_{n\mathbf{k}}^\alpha$ is the band velocity, and $A_{n\mathbf{k}}(\omega, T)$ is the spectral function. The resistivity for different combinations of interactions is shown in Fig. 3.3(a) and compared with experimental data [11, 13, 14].

Surprisingly, we find that the resistivity is governed by the e -ph interactions in SVO. The e -ph limited resistivity is an order of magnitude greater than the e - e limited resistivity, with the latter accounting for only $\sim 10\%$ of the experimental value. This result is in contrast with the conventional wisdom that transport properties in SVO are governed by purely electronic interactions. In addition, the contributions are opposite to another prototypical strongly correlated metal, Sr_2RuO_4 , where the e -ph interactions account for only $\sim 10\%$ of the resistivity [30]. In SVO, the e -ph contribution is similar in magnitude to the experimental value, and the total resistivity including both interactions is in good agreement with experiments. Below, we show that taking into account the electron correlation induced enhancement of the e -ph interactions increases the resistivity and brings the results in even better agreement with experiments.

The temperature dependence of the e -ph limited resistivity is analyzed in more detail in Fig. 3.3(b), where we show our results on a log-log plot and compare them with experiments [11, 13, 14]. In that plot, the resistivity is computed with the full (iterative) solution of the Boltzmann transport equation (BTE) [36] to include backscattering and improve the treatment of acoustic phonons. The computed e -

ph limited resistivity follows a $T^{1.8}$ temperature dependence between 100–200 K, in excellent agreement with the $T^{1.8-2}$ dependence found in experiments in that temperature range [11, 13, 14], and falls to $T^{1.5}$ at 300 K. We identify the origin of this nearly T^2 temperature trend of the e -ph limited resistivity by analyzing the contribution of different phonon modes. Our calculations show that the increasing contribution of strongly coupled optical phonons at higher temperatures is responsible for the T^2 dependence of the resistivity between 100–200 K (see SM).

Next, we focus on transport at low temperature, where the e -ph contribution is expected to be weaker. While DMFT calculations become difficult at low temperatures, we obtain the e - e limited resistivity by extrapolating our higher-temperature DMFT calculations with a T^2 fit. This approach is justified because the e - e scattering is in the Fermi liquid regime below at least 400 K. Figure 3.3(c) shows the computed e -ph and DMFT e - e limited resistivities below 100 K. We find a clear crossover between 20–30 K from e -ph to e - e dominated transport. The e -ph limited resistivity becomes much smaller than the e - e limited resistivity below 20 K, showing that e - e scattering governs transport at low temperature. This result indicates that e - e interactions are the origin of the T^2 resistivity observed experimentally below 25 K, with the e -ph contribution causing deviations from a T^2 behavior above 25 K. Note that our DMFT resistivity underestimates the experimental value below 25 K by a factor of 2-3. We attribute this discrepancy to limitations in the single site DMFT impurity model, as transport can be sensitive to the methods used to downfold orbitals and effective interactions [51, 52, 53, 30], the lack of non-local contributions to the DMFT self-energy [54], and corrections to the current-current vertex [55, 56, 57, 58].

Correlation-corrected electron-phonon interactions.— Strong electronic interactions are known to significantly modify e -ph interactions [59, 60, 61, 62]. In correlated metals, e -ph coupling is often enhanced. For example, calculations using hybrid functionals and the GW method found correlation-enhanced e -ph coupling in unconventional superconductors, attributing the enhancement to decreased electronic screening [26]. Similarly, in multiband d -electron systems such as FeSe, treating correlations with DMFT enhances e -ph coupling, in this case by increasing the orbital polarization response to phonon perturbations [27].

To study the role of correlations in SVO, we compute the e -ph interactions using Hubbard-corrected DFPT (DFPT+ U) [34, 33]. DFT+ U can be viewed as a static approximation of DFT+DMFT [63]; DFT+DMFT e -ph coupling calculations are not currently possible. Like DFT+DMFT, DFT+ U captures the strong local interactions

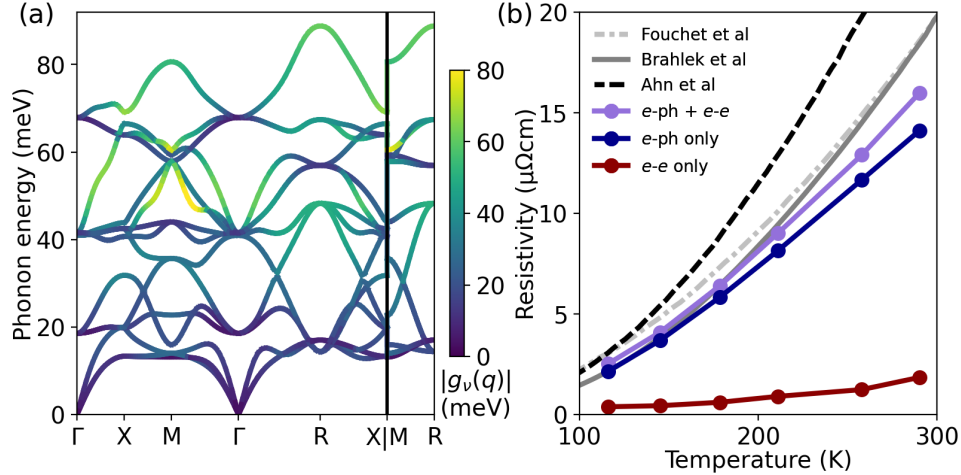


Figure 3.4: (a) Phonon dispersions and e -ph coupling as in Fig. 3.1(c) but calculated with DFPT+ U . Note the change in e -ph coupling scale. (b) Transport as in Fig. 3.3(a) but calculated with DFPT+ U phonons and e -ph couplings, showing improved agreement with experiments [11, 13, 14].

between d -orbitals and the resulting change in orbital polarization response, which modifies e -ph coupling. We use a Hubbard- U parameter of 3 eV in the atomic orbital basis, which is somewhat empirical but provides orbital polarization and band structure responses to phonon perturbations similar to a calculation including a Hubbard- U parameter from linear response and a reasonable value of Hund's J . The orbital occupation responses are also similar to those found using DMFT with the same orbitals and interaction parameters used throughout (see SM).

Adding the Hubbard correction has a small effect on the phonon dispersions, but it enhances the e -ph interactions, as shown in Fig. 3.4(a). The enhancement is mode-dependent and is generally higher for strongly-coupled optical phonons involving VO_6 distortions. The enhancement is also higher for phonons with momenta away from the Γ point, suggesting a more important role of correlations for distortions breaking lattice-translation symmetry. The spectral and transport properties computed with enhanced e -ph coupling from DFPT+ U give results qualitatively similar to those from DFPT, but with stronger e -ph effects. Notably, the e -ph limited resistivity increases by $\sim 35\%$ at room temperature [Fig. 3.4(b)], bringing the resistivity computed with both e -ph and e -e interactions into very good agreement with experiments. For example, the computed resistivity at 290 K is $16 \mu\Omega\text{cm}$, versus an experimental value of $19\text{--}25 \mu\Omega\text{cm}$ [11, 13, 14].

Discussion— The origin of the temperature dependence of the resistivity merits further discussion. While the T^2 trend for e -e interactions is expected based on Fermi

liquid theory [50], the origin of the near- T^2 behavior of the e -ph limited resistivity is less clear. At very low temperatures, the e -ph limited resistivity in metals is expected to exhibit a T^5 temperature dependence [64] when scattering is dominated by acoustic phonons with momentum $q \propto k_B T$. In our calculations, we find a T^5 e -ph limited resistivity below ~ 30 K, but the overall resistivity becomes e - e limited in this temperature range, explaining the experimental T^2 resistivity below 25 K.

In the high-temperature limit, based on the temperature dependence of the phonon occupations, one expects a T -linear e -ph limited resistivity [64]. However, this requires that all phonon modes contribute equally to e -ph scattering, with no mode frozen out. While our computed e -ph limited resistivity becomes nearly T -linear well above 300 K, it is close to a T^2 behavior between ~ 100 –200 K, in agreement with experiments. As discussed above, this T^2 trend is due to the increasing contribution of higher-energy optical phonons with strong e -ph coupling for increasing temperatures. Note also that e -ph scattering above ~ 50 K in SVO involves phonons with all momenta, ruling out momentum-dependent mechanisms resulting in T^2 behavior [65].

Finally, we analyze two approximations made in the e -ph transport calculations (see results in SM). First, we examine the use of the lowest-order e -ph self-energy [66, 67] by computing the resistivity with a cumulant diagram-resummation method capable of treating delocalized polarons [68]. Including polaron effects leads to a small increase in the resistivity, showing that lowest-order e -ph interactions are adequate to describe SVO. Second, we examine the effect of vertex corrections to the current-current correlation function in the Green-Kubo formalism [50, 43]. Vertex corrections improve the description of backscattering and the momentum dependence of e -ph scattering, which is particularly important at low temperature [50]. We assess their role above 100 K in the semiclassical limit by comparing the full solution of the BTE, which includes vertex corrections, to the relaxation time approximation, which neglects them. We find that vertex corrections in the BTE give only a small increase in the resistivity and its temperature dependence. This analysis shows that higher-order e -ph interactions and vertex corrections play a minor role in SVO and do not affect our conclusions.

Conclusion— In summary, we have shown that e -ph interactions play an essential role in the transport and spectral properties of a prototypical correlated metal, SVO. In this material, electronic correlations control other aspects of the low-energy physics, including the quasiparticle mass renormalization and transport at low temperature. We also found that electronic correlations lead to an effective enhancement

of the e -ph interactions. This suggests that SVO may serve as a testbed for investigating the interplay between electron correlations and e -ph interactions. Our results highlight the potential of first-principles calculations combining e - e and e -ph interactions in a consistent way as an emerging tool to study correlated materials. This work paves the way for a quantitative description of transport and spectral properties in broad classes of correlated quantum materials.

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Chapter 4

ELECTRON-PHONON COUPLING IN CORRELATED METALS: A DYNAMICAL MEAN-FIELD THEORY STUDY

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Abstract

Strong electron-electron interactions are known to significantly modify the electron-phonon coupling relative to the predictions of density functional theory, but this effect is challenging to calculate with realistic theories of strongly correlated materials. Here we define and calculate a version of the EPC applicable beyond band theory by combining first principles density functional theory plus dynamical mean-field theory with finite difference phonon perturbations, presenting results for several representative phonon modes in two materials of interest. In the three-orbital correlated metal SrVO_3 , we find that intra- $\text{V-}t_{2g}$ -band correlation significantly increases the coupling of these electrons to a Jahn-Teller phonon mode that splits the degenerate orbital energies, while slightly reducing the coupling associated with a breathing phonon that couples to the charge on each V atom. In the infinite layer cuprate CaCuO_2 , we find that local correlation within the $d_{x^2-y^2}$ orbital derived band has a modest effect on coupling of near-Fermi surface electrons to optical breathing modes. In both cases, the interaction correction to the electron-phonon coupling predicted by dynamical mean-field theory has a significant dependence on the electronic frequency, arising from a lattice-distortion dependence of the correlated electron dynamics, showing the inadequacy of the simple picture in which correlations change static local susceptibilities. We also show that the electron-phonon scattering and phonon lifetimes associated with these phonon modes are modified by the electronic correlation. Our findings shed light on the material- and mode-specific role of dynamical electronic correlation in electron-phonon coupling and highlight the importance of developing efficient computational methods for treating electron-phonon coupling in correlated materials.

4.1 Introduction

Electron-phonon (e -ph) interactions play an essential role in the physics of metals [1, 2]. A current focus is quantum materials with electronic properties controlled by strong correlations arising from electron-electron (e - e) interactions. However, there is growing evidence that e -ph interactions contribute in important ways to phenomena observed in these materials. For example, e -ph interactions have been argued to cause the "kinks" seen in angle-resolved photoemission studies of high- T_c cuprates [3, 4], phase transitions in rare earth nickelates [5, 6, 7, 8], important contributions to transport and optical lifetimes in some correlated metals [9, 10, 11, 12, 13], and contributions to novel superconductivity [14, 15, 16, 17, 18, 19, 7]. Experiment and theory indicate that the strength of the e -ph interactions may be strongly affected by electronic correlations [20, 4, 14, 15, 17, 18, 19, 7].

The current standard for e -ph calculations in real materials obtains electron-phonon coupling (EPC) from the dependence of the Kohn-Sham potential in density functional theory (DFT) on atomic displacements. However, EPC is sensitive to the treatment of electronic correlations, and DFT is not always sufficient. For example, calculations using DFT with hybrid or advanced functionals [21, 14, 22, 23], "+ U " extensions [24, 25, 12, 26], and GW [27, 28, 29, 15, 4, 30] have found modified EPC compared to semi-local DFT in a range of materials. However, these methods are perturbative and/or employ static correlation, and as a result, do not necessarily describe basic electronic properties of strongly correlated quantum materials (such as the large dynamical mass enhancement owing to the frequency-dependent self-energy).

Methods applicable to strong electronic correlation, such as dynamical mean-field theory (DMFT) [31], have been used to study the physics of electron-lattice coupling in model systems [32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42]. It is natural to ask whether the combination of density functional and dynamical mean-field theory (DFT+DMFT), which has proven a powerful and effective method of calculating electronic properties of correlated materials [43, 44], can be used to study EPC in correlated systems. First principles DFT+DMFT calculations have been shown to strongly modify proxies for EPC, such as spectral shifts in response to atomic displacements in FeSe [17, 19], and the energetics of octahedral tilting in SrMoO₃ [45] and optical phonons in LaNiO₂ [46]. However, the EPC has never been calculated directly in real materials with DFT+DMFT, and its dependence on the microscopic nature of the relevant electronic states and phonon modes has not been investigated. Here, we present a method to calculate the EPC for selected phonon modes using

DFT+DMFT calculations with finite phonon perturbations obtained from first principles Density Functional Perturbation Theory (DFPT) [47] calculations. We use the method to investigate the effect of correlation on EPC and related properties for representative phonon modes in two correlated materials of interest: SrVO_3 (SVO), a multi-orbital correlated metal where the interesting physics arises from bands derived from the V t_{2g} levels and phonons may couple to the orbital splitting, and CaCuO_2 (CCO), a member of the high- T_c cuprate family where the correlation physics primarily resides in a single Cu $d_{x^2-y^2}$ -derived band.

In SVO, our calculations show that local correlation significantly increases the coupling of t_{2g} electrons to a Jahn-Teller optical phonon mode which couples to the orbital degree of freedom, while slightly decreasing the coupling associated with a breathing mode which couples to the total V-site charge density. The frequency dependence of the coupling is non-negligible. We use the computed EPC to calculate temperature-dependent scattering rates of electrons due to these phonon modes. In the infinite layer cuprate CaCuO_2 (CCO), our calculations show that correlation within the $d_{x^2-y^2}$ orbital has a moderate effect on the coupling of Fermi surface electrons to full- and half-breathing modes. However, we find that the correlation effect depends strongly on electronic frequency. We investigate this behavior at different interaction strengths and carrier concentrations, and calculate phonon linewidths.

The rest of this paper is organized as follows: In the next section (Sec. 4.2), we present our theoretical method, based on finite difference DFT+DMFT calculations. Then we present the results of these calculations in SVO with the application to the temperature-dependent electron scattering (Sec. 4.3), and in CCO investigating the filling and U dependence and the phonon linewidth (Sec. 4.4). We next discuss in Sec. 4.5 the interpretation of our method and results in terms of the auxiliary impurity model used in the DFMT calculations and then compare to DFPT+ U and other research on correlation-modified e -ph interactions in Sec. 4.6. Finally, we summarize our findings in Sec. 4.7.

4.2 Theory and Methods

4.2.1 Definition of Coupling

In this paper, as in most treatments of EPC in solids, we make the Born-Oppenheimer approximation, valid when the ratio of a typical ionic velocity to a typical electronic velocity is very small. In this case the EPC may be approximated by the response of the electronic system to static ionic displacements. In DFT-based EPC calculations the EPC is defined as the scattering amplitude between two Kohn-Sham

states induced by a small atomic displacement. In a strongly interacting system, the electronic physics should instead be described in terms of a Green's function (propagator), and the EPC is correspondingly defined in terms of changes to the electron Green's function induced by changes in atomic position.

The frequency-domain electron Green's function $\hat{G}(r', r; \omega)$ describing the propagation of an electron from r to r' is defined in terms of a reference non-interacting system, which in this paper we take to be the Kohn-Sham Hamiltonian $\hat{T} + \hat{V}_{KS}$ with $\hat{T}$ the free electron kinetic energy operator and $\hat{V}_{KS}$ a Kohn-Sham potential. The differences between the physical propagation and the propagation defined by the reference non-interacting system are parametrized by the self-energy operator $\hat{\Sigma}(r, r'; \omega)$, and thus

$$G(r', r; \omega) = [\omega + \mu - \hat{T} - \hat{V}_{KS} - \hat{\Sigma}(\omega)]^{-1}, \quad (4.1)$$

where μ is the chemical potential and the carat^ denotes an operator acting on spatial indices. A displacement $u_{\mathbf{R}_p a \mu}$ of atom a in unit cell at $\mathbf{R}_p$ in direction μ changes both $\hat{V}_{KS}$ and $\hat{\Sigma}$. A formal expansion to first order in $u_{\mathbf{R}_p a \mu}$ (using the identity $\delta G = -G[\delta G^{-1}]G$) gives

$$\begin{aligned} G(r', r; \omega) &= G^{cn}(r', r; \omega) \\ &+ \int dr'' dr''' u_{\mathbf{R}_p a \mu} G^{cn}(r', r''; \omega) \times \\ &\quad \frac{\partial}{\partial u_{\mathbf{R}_p a \mu}} [\hat{V}_{KS} + \hat{\Sigma}(\omega)] G^{cn}(r''', r; \omega) + O(u^2). \end{aligned} \quad (4.2)$$

Here and below, we use *cn* (for "clamped nuclei") to denote quantities in the absence of atomic displacements. The object sandwiched between the two unperturbed Green's functions, $\frac{\partial}{\partial u_{\mathbf{R}_p a \mu}} [\hat{V}_{KS} + \hat{\Sigma}(\omega)]$ is the many-body definition of the EPC. In the absence of many-body effects, the self-energy operator vanishes so that the Green's function describes propagation of Kohn-Sham eigenstates and the definition reduces to the standard DFT EPC.

In our calculations, we use a basis of DFT eigenstates, denoted $|\phi_{n\mathbf{k}}\rangle$ for band n at electron crystal momentum $\mathbf{k}$, and express atomic displacements in terms of normal mode phonons with index ν and crystal momentum $\mathbf{q}$. Represented this way, our definition of the EPC $g_{mn\nu}^{\mathbf{kq}}(\omega)$ in a correlated system is

$$g_{mn\nu}^{\mathbf{kq}}(\omega) = \langle \phi_{m\mathbf{k}+\mathbf{q}} | \partial_{\nu\mathbf{q}} [\hat{V}_{KS} + \hat{\Sigma}(\omega)] | \phi_{n\mathbf{k}} \rangle_{u.c.} \quad (4.3)$$

which now depends on the electronic frequency due to the frequency dependence of the Green's function and here the carat^ denotes an operator in the momentum

indices. $\partial_{\nu\mathbf{q}}$ denotes the standard phonon mode-resolved derivative

$$\partial_{\nu\mathbf{q}} = \sum_{\mathbf{R}_p a\mu} e^{i\mathbf{q}\cdot\mathbf{R}_p} \sqrt{\frac{\hbar}{2M_a\omega_{\nu\mathbf{q}}}} \mathbf{e}_{\nu\mathbf{q}}^{a\mu} \left. \frac{\partial}{\partial u_{\mathbf{R}_p a\mu}} \right|_{u_{\mathbf{R}_p a\mu}=0}, \quad (4.4)$$

where a indexes an atom with mass M_a , $\mathbf{R}_p$ denotes a unit cell, and $\omega_{\nu\mathbf{q}}$ and $\mathbf{e}_{\nu\mathbf{q}}^{a\mu}$ are respectively the phonon energy and normalized eigenvector. Similar forms of EPC have been used for model systems [32] and first-principles *GW*-based calculations [15].

4.2.2 Evaluation with Dynamical Mean-Field Theory

The central challenge is to reliably evaluate the change in the self-energy due to the phonon perturbation $\partial_{\nu\mathbf{q}}\Sigma(\omega)$. We evaluate this using the density functional plus dynamical mean-field theory (DFT+DMFT) approximation. In DFT+DMFT, one combines a band structure calculated with DFT with a dynamical mean-field treatment of strong correlations within a set of local orbitals $\phi_i(r - \tau_a - \mathbf{R}_e)$ that are constructed around an atom a located at position τ_a within unit cell $\mathbf{R}_e$ using Wannier or projector methods. The DMFT assumption is that the electronic self-energy is site-local in the orbital basis (but may be a matrix in the on-site orbital indices ij), so that the unfolded self-energy is given by

$$\Sigma(r, r'; \omega) = \sum_{\mathbf{R}_e aij} \phi_i^*(r - \tau_a - \mathbf{R}_e) \Sigma_{ij}^a(\omega) \phi_j(r - \tau_a - \mathbf{R}_e). \quad (4.5)$$

The self-energy $\Sigma_{ij}^a(\omega)$ for the correlated site is calculated from the solution of a quantum impurity model defined by the DFT band structure, the local orbitals, a site-local inter-orbital interaction representing the effective screened Coulomb interaction projected onto these orbitals, and a "double-counting correction", each of which can be treated in several approximations.

In this paper we use the "frontier orbital" DFT+DMFT methodology in which the correlated orbitals are defined by a Wannierization of bands crossing the Fermi level, we neglect full charge self-consistency, and we subtract a double-counting term from the self-energy. We use empirical interaction parameters and neglect changes to the interaction parameters associated with phonon displacements for simplicity. Similar approximations are commonly used in DFT+DMFT studies of structural deformations [17, 48, 45, 6].

The materials studied in this paper have a single correlated site (a transition metal) in the undistorted unit cell, with a small number of relevant correlated orbitals

(partially filled d -shells). We consider phonon modes corresponding to lattice distortions which reduce the translational symmetry of the unit cell, resulting in a supercell with multiple correlated atoms. We apply the phonon perturbation of magnitude α to the supercell by adding $\alpha \times \langle i, \mathbf{R}_e | \frac{1}{2} (\partial_{v\mathbf{q}} \hat{V}_{KS} + \partial_{v-\mathbf{q}} \hat{V}_{KS}) | j, \mathbf{R}_e' \rangle$ to the supercell Wannier Hamiltonian. Here, α represents the ratio of the effective atomic displacements in the finite perturbation to the phonon eigen-displacements, and the term in the bra-ket is the Wannier basis perturbation caused by a set of real atomic displacements¹ associated with the phonon in question calculated with DFPT [47] and Wannier interpolation [49].

We perform DFT+DMFT calculations with the perturbed and unperturbed Hamiltonians and extract the self-energies corresponding to the inequivalent sites in the perturbed supercell ($\hat{\Sigma}^{ph}(\omega)$) and unperturbed supercell ($\hat{\Sigma}^{cn}(\omega)$). The self-energy term of the EPC in Eq. 4.3 is then calculated from the difference between the two aforementioned sets of site-dependent self-energies, up-folded into the band basis as

$$\begin{aligned} \langle \phi_{m\mathbf{k}+\mathbf{q}} | \partial_{v\mathbf{q}} \hat{\Sigma}(\omega) | \phi_{n\mathbf{k}} \rangle_{u.c.} &= \frac{1}{\alpha N_e} \sum_{\mathbf{R}_e} e^{-i\mathbf{q} \cdot \mathbf{R}_e} \\ &\times U_{mi}^\dagger(\mathbf{k} + \mathbf{q}) \langle i, \mathbf{R}_e | \hat{\Sigma}^{ph}(\omega) - \hat{\Sigma}^{cn}(\omega) | j, \mathbf{R}_e \rangle U_{jn}(\mathbf{k}), \quad (4.6) \end{aligned}$$

where $U(\mathbf{k})$ are the transformations between the Wannier and Bloch basis and α is chosen to be small enough to generate a good approximation to the derivative. Note that in this procedure the Wannier basis is held fixed.

Here we have used standard e -ph Fourier transform conventions [49] but with the simplification that the DFT+DMFT self-energy is local and therefore non-zero only for $\mathbf{R}_e = \mathbf{R}_e'$. The DFT+DMFT EPC is then calculated with Eq. 4.3 using Eq. 4.6 and $\langle \phi_{m\mathbf{k}+\mathbf{q}} | \partial_{v\mathbf{q}} \hat{V}_{KS} | \phi_{n\mathbf{k}} \rangle_{u.c.}$ from DFPT (which is interpolated from the Wannier function form in the standard way [49]). The workflow for performing finite difference DFT+DMFT calculations is further explained in the Supplemental Material (SM) [50].

¹The phonon modes studied in this paper have $\mathbf{q} = -\mathbf{q}$ and thus the corresponding atomic displacements $u_{\mathbf{R}_p a \mu} = \sqrt{\frac{\hbar}{2M_a \omega_{v\mathbf{q}}}} e^{i\mathbf{q} \cdot \mathbf{R}_p} \mathbf{e}_{v\mathbf{q}}^{a\mu}$ can be written as fully real. However, for a general phonon, real atomic displacements $\sqrt{\frac{\hbar}{2M_a \omega_{v\mathbf{q}}}} \frac{1}{2} (e^{i\mathbf{q} \cdot \mathbf{R}_p} \mathbf{e}_{v\mathbf{q}}^{a\mu} + e^{-i\mathbf{q} \cdot \mathbf{R}_p} \mathbf{e}_{v-\mathbf{q}}^{a\mu})$ must be used to obtain a physical perturbed Hamiltonian. This is described as the real valued displacement field in Ref. [49].

4.2.3 Electron-Phonon Scattering

The EPC calculated in our approach can be used to calculate quantities such as the electron and phonon scattering rates due to the e -ph interaction, which are related to the imaginary part of the e -ph contribution to the electron and phonon self-energies. To lowest order, the electron self-energy is a single phonon exchange, which can be schematically written $\Sigma = ig^2GD$, where g is the EPC, G is the electron Green's function, and D is the phonon Green's function. When evaluated with the frequency-dependent DFT+DMFT EPC, DFT+DMFT electron Green's function, and the non-interacting DFPT phonon Green's function, the imaginary part is

$$\begin{aligned} \text{Im}\Sigma_{\mathbf{k},nn'}^{e\text{-ph}}(\omega) = & \frac{-\pi}{N_{\mathbf{q}}} \sum_{\mathbf{q} \nu m} \left[g_{mn\nu}^{\mathbf{kq}} \left(\omega - \frac{\omega_{\nu\mathbf{q}}}{2} \right)^* g_{mn'\nu}^{\mathbf{kq}} \left(\omega - \frac{\omega_{\nu\mathbf{q}}}{2} \right) \right. \\ & \times A_{m\mathbf{k}+\mathbf{q}}^{\text{DMFT}}(\omega - \omega_{\nu\mathbf{q}}) [n(\omega_{\nu\mathbf{q}}) + 1 - f(\omega - \omega_{\nu\mathbf{q}})] \\ & + g_{mn\nu}^{\mathbf{kq}} \left(\omega + \frac{\omega_{\nu\mathbf{q}}}{2} \right)^* g_{mn'\nu}^{\mathbf{kq}} \left(\omega + \frac{\omega_{\nu\mathbf{q}}}{2} \right) \\ & \left. \times A_{m\mathbf{k}+\mathbf{q}}^{\text{DMFT}}(\omega + \omega_{\nu\mathbf{q}}) [n(\omega_{\nu\mathbf{q}}) + f(\omega + \omega_{\nu\mathbf{q}})] \right]. \quad (4.7) \end{aligned}$$

where $A(\omega)$ is the spectral function and f and n are respectively the fermion and boson occupation numbers. Eq. 4.7 reduces to the conventional DFT Fan-Migdal expression if $A(\omega)$ is taken to be a Dirac delta function at the band energy and the EPC is defined from $\hat{V}_{KS}$ only [51, 2]. Previous work also studied e -ph interactions in correlated materials using the full DMFT spectral function but the DFPT EPC [52]. Similarly, to lowest order in g the e -ph contribution to the phonon linewidth is given by a bubble diagram formed from two e -ph vertices and two electron propagators (schematically, $\Pi = g^2G^2$) calculated as

$$\begin{aligned} \text{Im}\Pi_{\nu\mathbf{q}}^{e\text{-ph}}(\omega_{\nu\mathbf{q}}) = & \frac{-2\pi}{N_{\mathbf{k}}} \sum_{mn\mathbf{k}} \int d\omega \left| g_{mn\nu}^{\mathbf{kq}} \left(\omega + \frac{\omega_{\nu\mathbf{q}}}{2} \right) \right|^2 \\ & \times A_{n\mathbf{k}}^{\text{DMFT}}(\omega) A_{m\mathbf{k}+\mathbf{q}}^{\text{DMFT}}(\omega + \omega_{\nu\mathbf{q}}) [f(\omega) - f(\omega + \omega_{\nu\mathbf{q}})]. \quad (4.8) \end{aligned}$$

If $A(\omega)$ is set to be a Dirac delta function at the band energy and the DFPT EPC is used, Eq. 4.8 is the conventional expression for the leading order electronic contribution to the phonon linewidth [53, 2]. The importance of using a beyond-DFT (non-delta function) A was noted in a recent work (which however used the DFPT-derived EPC) [54].

In Eqs. 4.7 and 4.8, the EPC depends on one frequency because we have neglected the dependence on the phonon frequency due to the Born-Oppenheimer approximation. Since the coupling connects two electronic states that have different frequencies, we evaluate it at the average of the two electronic frequencies (i.e. ω and $\omega \pm \omega_{\nu\mathbf{q}}$) to avoid biasing the calculation. Because the two frequencies are separated by $\omega_{\nu\mathbf{q}}$, this approximation disappears in the $\omega_{\nu\mathbf{q}} \rightarrow 0$ limit. In our calculations, we calculate scattering rates using EPC derived from the real part of $\partial_{\nu\mathbf{q}}\Sigma(\omega)$ because the imaginary part is small near the Fermi surface.

We also note that these forms of self-energy have two vertex corrected e -ph interactions as they come from perturbative expansions in phonon displacements. This differs from the self-energy in the Hedin-Baym equations, due to the latter's self-consistency [55, 2]. A similar approximation using two screened vertices has been found to be accurate when considering electronic screening of e -ph interactions [56] and shown to be physically correct for the phonon lifetime [34, 57].

The derivation and computational evaluation of these self-energies is further discussed in the Supplemental Material (SM) [50], as is the effect of the frequency averaging approximation, the use of the DFT vs. DFT+DMFT electronic Green's functions, and other approximations.

4.2.4 Computational Methods

We calculate the electronic structure, phonons, and EPC using DFT and DFPT with the Quantum Espresso code [58]. We convert the EPC to the real-space Wannier basis and interpolate using Perturbo [59]. We use Wannier90 [60] to construct Wannier orbitals, which are used as the DMFT impurity orbitals. More details on the parameters used for DFT and DFPT are provided in the SM [50].

All DMFT calculations use TRIQS [62] with the TRIQS/DFTTools and solid_dmft first-principles packages [63, 64]. The lattice Hamiltonian is calculated by interpolating the Wannier Hamiltonian and summing over a sufficiently large $\mathbf{k}$ -grid to converge the hybridization. All calculations use the continuous-time quantum Monte Carlo-hybridization expansion solver (CTHYB) [65, 66], the Held formula for the double counting correction [44], and Padé analytic continuation. In the finite difference DFT+DMFT calculations, we use a scale factor of $\alpha = 1/2$ for the phonon perturbation for all modes. More details on the DFT+DMFT calculations are provided in the SM [50].

For SVO we use the experimental lattice parameter of $a = 3.842 \text{ \AA}$ [67, 68] in the $Pm\bar{3}m$ cubic structure. We compute maximally-localized Wannier functions for the

three t_{2g} -derived orbitals. For the DMFT calculations, we use a Hubbard-Kanamori interaction Hamiltonian with $U = 4.5$ eV and $J = 0.675$ eV. Our DFT, DFPT, and unperturbed DFT+DMFT calculations in SVO largely follow previous work [12]. For the finite difference DFT+DMFT calculations, we use a $2 \times 2 \times 2$ supercell for both modes.

For CCO, we use the tetragonal $P4/mmm$ structure with a hole doping fraction of 0.15 holes per stoichiometric unit cell and relaxed lattice parameters of $a \times a \times c = 3.742 \text{ \AA} \times 3.742 \text{ \AA} \times 3.051 \text{ \AA}$. We use an impurity consisting of the $d_{x^2-y^2}$ Wannier orbital and a Hubbard interaction. As described below, we perform calculations with a variety of Hubbard U parameters (3.1, 3.9, and 4.7 eV), and consider different fillings in the rigid band approximation. For the finite difference DFT+DMFT calculations, we use a $2 \times 2 \times 1$ supercell for both modes.

4.3 Results: SrVO₃

4.3.1 Modes and Coupling

SVO is a correlated metal with a cubic perovskite structure and one electron in the V- t_{2g} orbitals often used as a prototypical example for many-body methods. SVO stands out as a promising system to study the interplay of e - e and e -ph interactions, as demonstrated by recent calculations showing e -ph-induced photoemission kinks and phonon-limited resistivity above ~ 25 K [12] and experiments showing transport properties beyond Fermi liquid e - e scattering [69]. In the present context, the multi-orbital electronic structure of the t_{2g} orbitals (respective bands shown in Fig. 4.1(a)) is of interest because it enables investigation of both "Jahn-Teller"-type phonons that lift the orbital degeneracy and "Holstein"-type phonons that couple mainly to the on-site charge.

We focus on two representative phonon modes in SVO: an **M**-point Jahn-Teller mode which couples to the orbital degree of freedom on each site, and an **R**-point breathing mode which couples to the charge degree of freedom on alternating sites. These modes are indicated on the DFPT phonon dispersion in Fig. 4.1(b), where color indicates the EPC (averaged over electronic states on the Fermi surface as in Ref. [12]). Both modes are high-frequency ($\omega_{\mathbf{M}} = 64$ meV and $\omega_{\mathbf{R}} = 90$ meV) optical phonon modes with fairly strong EPC, owing to stretching V–O bonds. The atomic displacements associated with each mode are illustrated in Fig. 4.1(c,d). The pattern of O displacements in the **M**-point Jahn-Teller mode (c) splits the three t_{2g} orbitals on each V site while maintaining equivalence between all sites (under

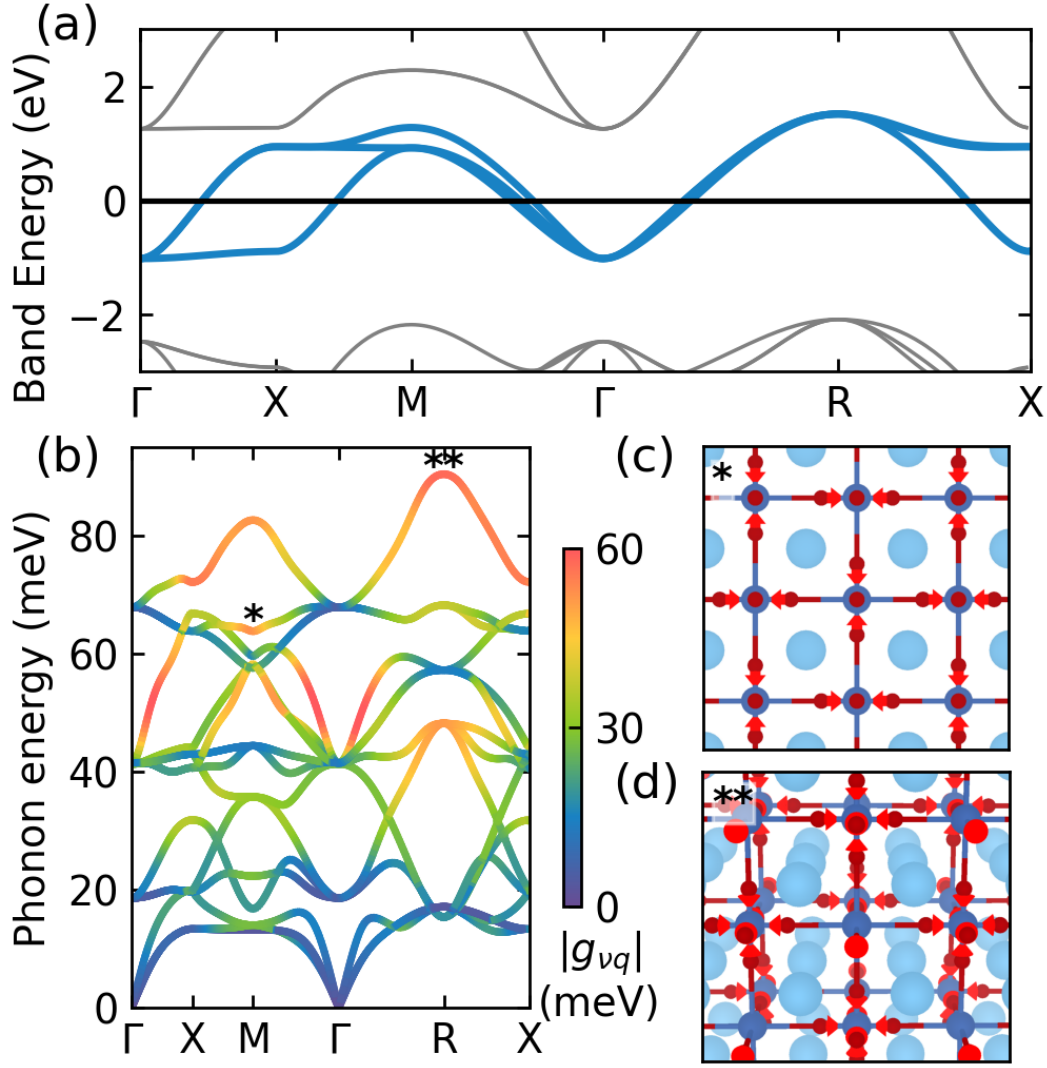


Figure 4.1: (a) Band structure of SrVO₃, highlighting in blue the Wannierized t_{2g} bands used in the DMFT calculations. (b) Phonon dispersion of SrVO₃, with color showing EPC averaged on the Fermi surface as in [12]. The M and R -point phonon modes studied are indicated by single and double stars, respectively. Visualizations of (c) the M -point bond stretching mode and (d) the R -point breathing mode (right) where red atoms are O, light blue are Sr, and dark blue are V.

the combination of translation and $\pi/2$ rotation), whereas in the R -point breathing mode (d), alternating V sites are split but the orbitals on each site remain equivalent.

We perform finite difference DFT+DMFT EPC calculations for both modes and compare to DFPT. Fig 4.2(a) shows the Wannier basis local EPC $\langle i, 0 | \partial_{vq} [V_{KS} + \Sigma(\omega)] | j, 0 \rangle$ – where i, j label orbital indices and the site is $\mathbf{R}_e = 0$; this quantity provides a natural comparison between DFT and DFT+DMFT because the DMFT con-

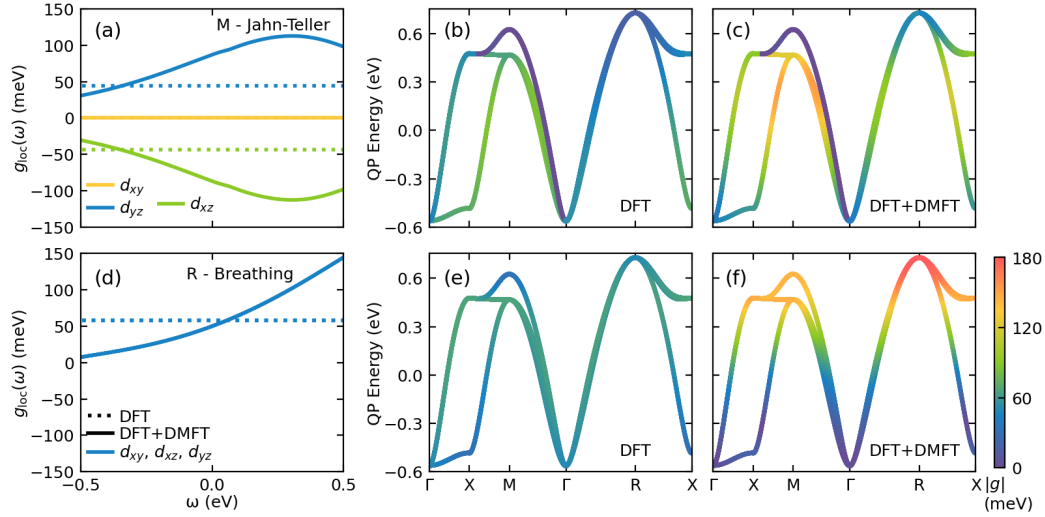


Figure 4.2: EPC in SVO with DFT and DFT+DMFT. (a) Local EPC in the Wannier basis for the **M**-point mode calculated with DFT (dashed lines, $\partial_{v\mathbf{q}}V(\mathbf{R}=0)$) and DFT+DMFT (solid lines, $\partial_{v\mathbf{q}}[V(\mathbf{R}=0) + \Sigma(\omega, \mathbf{R}=0)]$). (b) DFT and (c) DFT+DMFT EPC projected onto the quasi-particle bands $|g| = (\sum_m |g_{nmv}^{\mathbf{kq}}|^2)^{1/2}$. (d) Local EPC and band-resolved (e) DFT and (f) DFT+DMFT EPC for the **R**-point breathing mode. Calculations shown are for $\beta = 80$ / eV.

tribution is local in the Wannier basis. For the **M**-point Jahn-Teller mode, we find significantly increased EPC to electrons near the Fermi surface. As expected, the mode splits the d_{yz} and d_{xz} orbitals while leaving the d_{xy} orbital unchanged. The Wannier basis local coupling strength is nearly doubled from DFT (44 meV) to DFT+DMFT at $\omega = 0$ (87 meV). The coupling also exhibits a fairly strong dependence on the electron frequency, increasing in strength with increasing frequency. The band-resolved EPC calculated with DFT and DFT+DMFT is plotted on the quasi-particle bands (defined as the solutions of $\det[\omega + \mu - \varepsilon_{n\mathbf{k}}\delta_{nn'} - \text{Re}\Sigma_{nn'\mathbf{k}}^{\text{DMFT}}(\omega)] = 0$) in Fig. 4.2(b) and (c), respectively. The DFT EPC shows clear band dependence arising from the orbital character of the bands and non-local terms in the coupling. This dependence is maintained in the DFT+DMFT EPC, but there is a clear enhancement of the coupling strength as well as frequency dependence from the self-energy term.

In contrast to the **M**-point mode, the **R**-point breathing mode shows a slight decrease in the magnitude of the local EPC, from 58 meV with DFT to 50 meV with DFT+DMFT at $\omega = 0$ eV (Fig. 4.2(d)). Like the **M**-point mode, the coupling shows a frequency dependence which tends to increase coupling at higher frequencies, but exhibits less band and $\mathbf{k}$ dependence due to the symmetry of the displacements. The band-resolved DFT and DFT+DMFT EPC are plotted on the quasi-particle bands

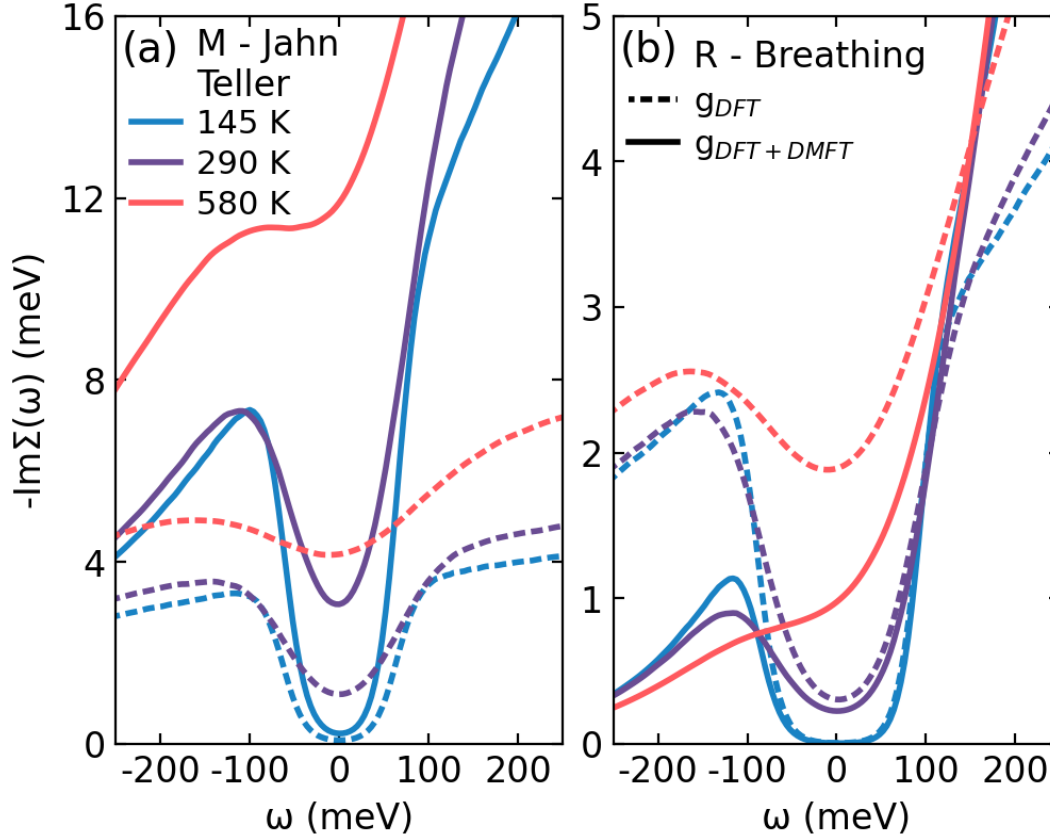


Figure 4.3: Contribution to the electron scattering rates in SVO calculated using the DFT+DMFT couplings (solid lines) and DFPT coupling (dashed lines) from (a) scattering by the **M**-point bond stretching mode and (b) **R**-point breathing mode. The frequency-dependent scattering rates calculated using DFT and DFT+DMFT EPC are shown as the BZ averaged $-\text{Im}\Sigma(\omega)$ in the Wannier orbital basis for the $d_{xz/yz}$ orbital for the **M**-point mode and the $d_{xy/xz/yz}$ orbital for the **R**-point mode.

in Fig. 4.2(e) and (f), respectively.

In summary, DMFT significantly increases the coupling of the **M**-mode which couples to orbital splitting, but slightly decreases the coupling of the **R**-mode which couples to charge disproportionation, and introduces a strong frequency dependence to the coupling, absent in DFPT or its $+U$ and hybrid functional extensions. The difference in correlation effects on the EPC for the two modes may be understood in context of local interactions enhancing orbital fluctuations and suppressing charge fluctuations in correlated systems. The strong frequency dependence is another important consequence of correlation physics and will be discussed in more detail below.

4.3.2 Electron Scattering

Next, we calculate the scattering of electrons by each of the two phonon modes studied. The scattering is proportional to the imaginary part of the lowest order self-energy (Eq. 4.7); we calculate the contributions from the phonon modes studied using the EPC calculated above. We plot the Brillouin Zone (BZ) averaged part of this self-energy in the Wannier basis $\Sigma_{ii}(\omega) = \sum_{\mathbf{k}} U_{in}(\mathbf{k}) \Sigma_{nn'}(\mathbf{k}, \omega) U_{n'i}(\mathbf{k})^\dagger$ in Fig. 4.3 at several temperatures (the temperature applies to both the DFT+DMFT calculation and the electron and phonon occupation numbers in Eq. 4.7).

We find that the electron scattering near the Fermi energy by the **M** mode phonon (Fig. 4.3(a)) is significantly increased when calculated with DFT+DMFT EPC compared to DFT EPC, while scattering by the **R** mode phonon (Fig. 4.3(b)) is slightly suppressed, consistent with our calculated coupling strengths. We also find a significantly increased asymmetry between scattering above and below the Fermi energy due to the frequency dependence of the EPC, with the scattering rate at positive frequencies enhanced, consistent with the frequency dependence of the EPC. For both modes, the scattering rate obeys a temperature dependence consistent with the electron and phonon occupation numbers, with the temperature dependence of the DFT+DMFT EPC having a minor effect (See SM [50]). We note that these calculations used the DFPT phonon frequency; the phonon frequency enters the results both via the convention for g (Eq. 4.4) and via the phonon propagator. From these formulae the effects of substituting the physical phonon frequencies may be deduced. Based on a previous study [48], correlation results in a moderate hardening of the optical phonons in SVO, which could suppress scattering independent of the coupling used.

4.4 Results: CaCuO₂

4.4.1 Modes and Coupling

Next, we investigate the EPC in the DFT+DMFT theory of calcium copper oxide (CCO), a member of the widely studied family of cuprate superconductors. In cuprate materials, e - e interactions are believed to dominate the physics. These interactions have been extensively studied with DMFT, both in the context of minimal models with a single $d_{x^2-y^2}$ orbital per Cu site [70, 71, 72, 73, 74, 75, 76] and more advanced calculations incorporating additional orbitals, dynamically screened interactions, and/or GW [77, 78, 79, 80]. The effect of e -ph interactions in cuprates have been studied in relation to superconductivity [81, 82], quasi-particle properties [83, 26] including as a potential origin of photoemission kinks [3, 84, 20, 4], and phonon

softening associated with metal-insulator transition or charge density waves [85, 86, 87, 88, 89, 90]. The cuprates thus provide a useful computational platform to study many-body effects on the EPC in a single-orbital strong correlation context.

CCO is the structurally simplest cuprate-based superconductor. The basic unit is a flat Cu-O₂ plane with a C_4 symmetry and a single Cu ion per unit cell. The interplane coupling is very weak so that the $d_{x^2-y^2}$ electronic band is nearly two-dimensional [91, 92]. CCO is insulating in the undoped normal state, but upon hole doping transitions from insulating to metallic, where a range of doping values in the metallic phase also show superconducting behavior.

We base our DMFT calculations on a minimal model of the electronic structure composed of the $d_{x^2-y^2}$ Wannier orbital, which dominates the electronic structure near the Fermi energy as shown in Fig. 4.4(a). We calculate the phonon dispersion and the EPC with DFPT, as shown in Fig. 4.4(b). The two high-symmetry optical phonon modes which couple most strongly to the $d_{x^2-y^2}$ band at the Fermi surface are the **X**-point half-breathing mode (panel (c)), and an **M**-point full-breathing mode (panel (d)) marked by single and double stars in panel (b), respectively. Both modes involve movement of O atoms in the Cu-O plane as depicted in Fig. 4.4(c) and (d). We perform finite difference DFT+DMFT EPC calculations on these modes.

4.4.2 U -dependent EPC at 0.15 hole doping

Fig. 4.5 shows the EPC at several U values for 0.15 hole doping, a carrier concentration at which high T_c superconductivity occurs in some of the cuprate compounds. We begin by considering the Wannier basis local EPC for $U = 3.1$ eV, shown in light blue in Fig. 4.5(a) for the **X**-point half-breathing mode and Fig. 4.5(e) for the **M**-point full-breathing mode. For both phonons we observe that the difference in the EPC at the Fermi level between DFT and DFT+DMFT is small, but that the DMFT-derived EPC has an extremely strong frequency dependence over a few hundred meV scale. The highly frequency-dependent EPC combines with the $\mathbf{k}$ dependence already present in DFPT and yields a strong frequency dependence when projected onto the quasiparticle bands shown in panels (c,d) (**X**-point mode) and (g,h) (**M**-point mode).

We now consider higher U -values. The Wannier local EPC at $U = 3.1, 3.9$, and 4.7 eV is shown in Fig. 4.5(a) for the **X**-point half-breathing mode and Fig. 4.5(e) for the **M**-point full-breathing mode. While the zero frequency EPC shows moderate changes compared to the DFT value for all values of U , the frequency dependence

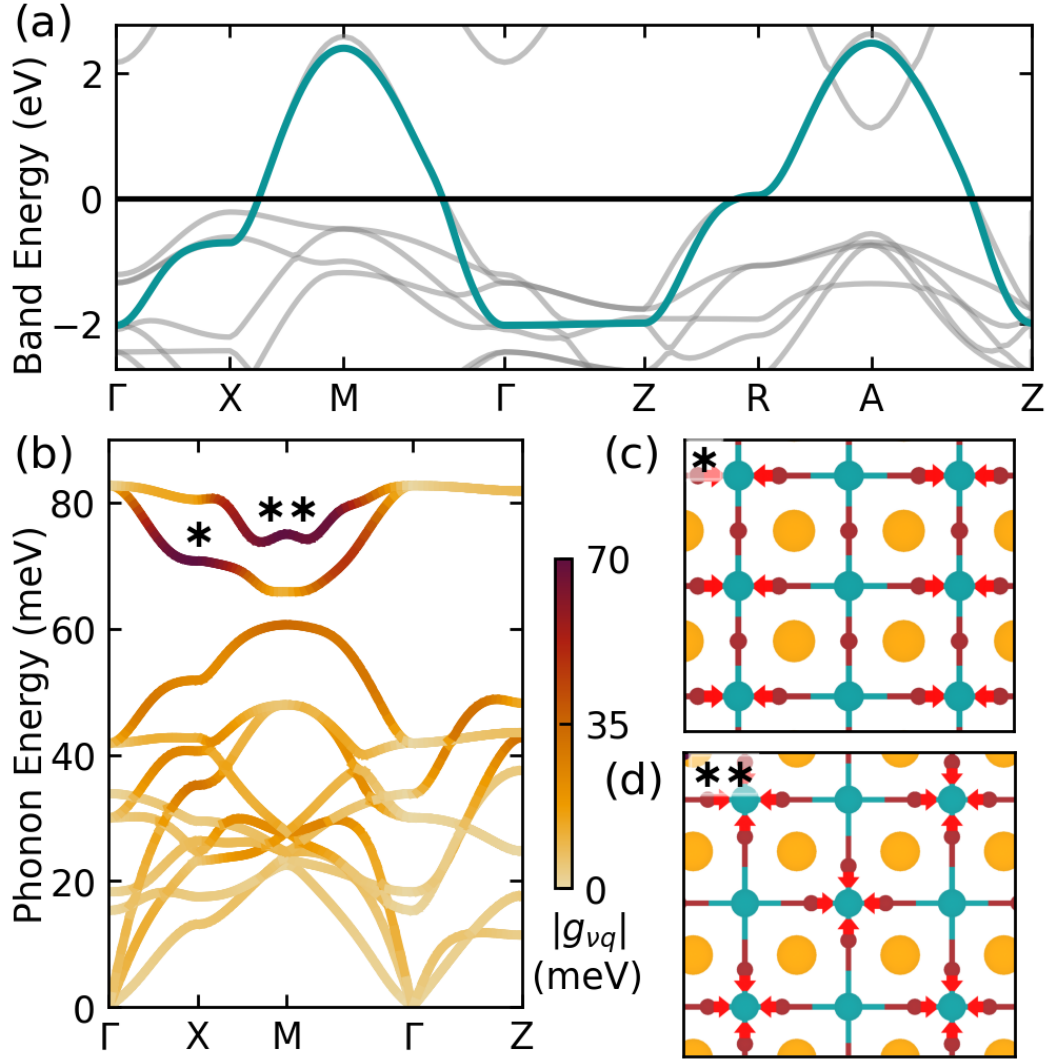


Figure 4.4: (a) Band structure of CaCuO_2 , showing the Wannierized $d_{x^2-y^2}$ band used in the DMFT calculations (teal) and other bands in the DFT calculation (grey). (b) DFPT-calculated phonon dispersion of CaCuO_2 , with color showing EPC averaged on the Fermi surface and studied half breathing (*) and full breathing (**) phonon modes indicated. Visualizations of (c) the X-point half breathing mode and (d) the M-point full breathing mode. Cu atoms are shown in teal, O in red, and Ca in orange.

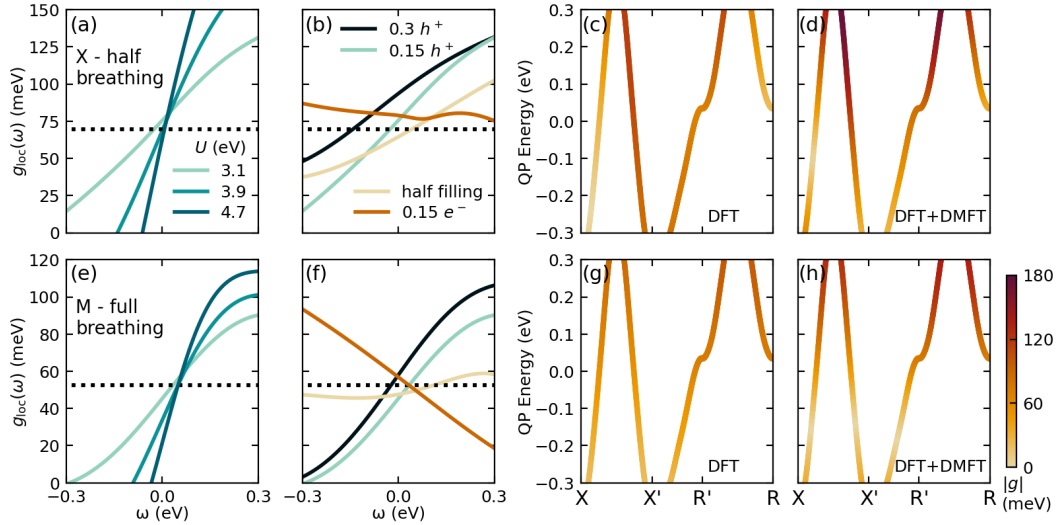


Figure 4.5: EPC in CCO with DFT and DFT+DMFT. (a) Local EPC in the Wannier basis for the **X**-point half breathing mode calculated with DFT (dashed lines, $\partial_{\mathbf{v}\mathbf{q}}V(\mathbf{R}=0)$) and DFT+DMFT (solid lines, $\partial_{\mathbf{v}\mathbf{q}}[V(\mathbf{R}=0) + \Sigma(\omega, \mathbf{R}=0)]$) as a function of electron frequency ω at U -values indicated at 0.15 hole doping and (b) at the filling values indicated and $U = 3.1$ eV. (c) DFT and (d) DFT+DMFT EPC at 0.15 hole doping and $U = 3.1$ eV projected onto the quasi-particle bands $|g| = (\sum_m |g_{nmv}^{\mathbf{k}\mathbf{q}}|^2)^{1/2}$ (e-h). Same as (a-d) but for the **M**-point full breathing mode.

increases sharply with increasing U . This frequency dependence becomes so strong that for $U = 4.7$ eV, the $g_{\text{loc}}(\omega)$ varies from 0 to $\sim 2 \times g_{\text{loc}}(\omega = 0)$ in a range of $\pm \omega_{\mathbf{v}\mathbf{q}} \sim 70$ meV from the Fermi energy. This indicates the potential for large adiabaticity in the e -ph interaction at large U , i.e. because the e -ph interaction varies significantly on the scale of the phonon frequency, the approximation of a static phonon for the purposes of calculating coupling may be insufficient.

The increasing frequency dependence of g can be connected with increasing correlation strength; Z decreases from ~ 0.50 at $U = 3.1$ eV to ~ 0.28 at $U = 4.7$ eV corresponding to an increased slope in the real part of the self-energy (see SM [50]). The increasing frequency dependence indicates that the coupling of the phonon to correlation increases with the degree of correlation. We note that our calculations show only a moderate change in the $\omega = 0$ coupling, although the change might be larger in more advanced calculations including more orbitals or dynamical screening (this is discussed in Sec. 4.6).

4.4.3 Filling-dependent EPC at $U = 3.1$ eV

The dependence of the Wannier-local EPC on carrier concentration is shown for $U = 3.1$ eV in panels (b,f) of Fig. 4.5. The zero frequency coupling shows some variation as a function of filling, with generally larger DFT+DMFT couplings further away from half-filling. The striking behavior is that for both phonon modes the frequency dependence has a strong dependence on carrier concentration, with a large, positive slope at 0.3 and 0.15 hole-doping, then flattening as the carrier concentration moves across half filling and then changing sign for increasing electron doping.

The observed interplay of the carrier concentration and the magnitude and sign of the frequency dependence suggests a close link between the frequency dependence of g and the frequency dependence of the self-energy. For a Fermi liquid, the dominant feature in the low frequency electron self-energy is a linear frequency dependence, $\text{Re}\Sigma(\omega) = -(Z^{-1} - 1)\omega$, with (in the DMFT approximation) the coefficient $Z < 1$ giving the inverse mass enhancement and the quasiparticle weight and Z^{-1} defining an effective correlation strength. In the metallic state of a Mott-Hubbard-like system, the correlation strength increases and Z decreases with increasing U . For U values near the Mott transition critical U , correlation is strongest (and Z is smallest) near half-filling and decreases with either electron or hole doping. This trend is evident in our calculated $\text{Re}\Sigma(\omega)$ in CCO (see SM [50]). Because the phonon mode couples to the site occupancy, it is natural to suppose that the main contribution to $\partial_{\nu\mathbf{q}}\Sigma(\omega)$ is via the dependence of Z on orbital occupation n , suggesting a contribution to $g \sim \frac{\partial Z^{-1}\omega}{\partial n} \frac{\partial n}{\partial u_{\nu\mathbf{q}}}$. The derivative increases with increasing Z^{-1} (and hence with U), is small near half-filling where Z passes through a minimum, and has a sign that depends on the sign of the doping.

This explanation may also apply to SVO, in which the t_{2g} orbitals are at 1/6 filling and there is significant particle hole asymmetry.

4.4.4 Phonon Linewidth

As an application of the EPC in CCO, we calculate the U - and filling-dependent phonon linewidths $\gamma_{\nu\mathbf{q}}$ for the phonons studied. We evaluate $\gamma_{\nu\mathbf{q}} = 2\text{Im}\Pi_{\nu\mathbf{q}}(\omega_{\nu\mathbf{q}})$ with the phonon self-energy from Eq. 4.8 based on the lowest order phonon self-energy diagram. The resulting linewidths are plotted in Fig. 4.6(a) as a function of U and Fig. 4.6(b) as a function of filling.

The dependence of the linewidth on correlation (U) is opposite for the two phonon modes. We see that for the **M**-mode (dark blue), increasing correlation suppresses

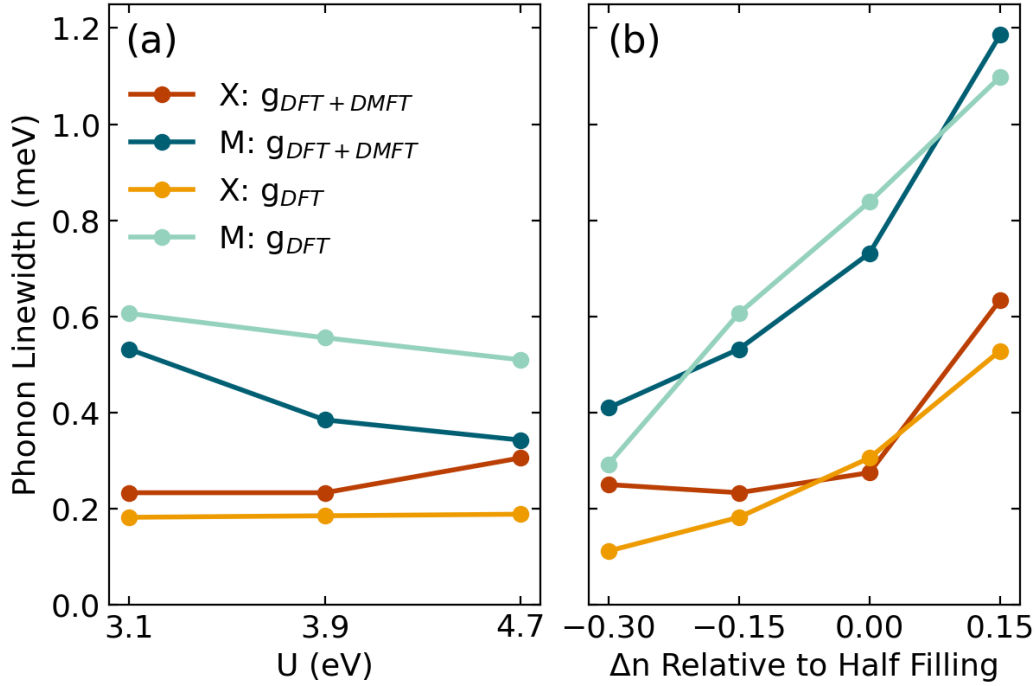


Figure 4.6: Phonon linewidth in CCO (a) as a function of U at 0.15 hole doping and (b) as a function of filling at $U = 3.1$ eV.

the linewidth at fixed doping. However, for the **X**-mode (dark orange) an increase with U is found. Here the linewidth reflects that the correlation increases the coupling of the **X**-mode slightly and decreases that of the **M**-mode slightly.

The linewidth of both phonons increases significantly with filling for the DFT EPC (light blue and orange lines) due to the increasing joint density of states at the wavevectors $\mathbf{q}$ as the Fermi energy is raised. When the DFT+DMFT EPC is used (dark blue and red lines), the trend remains but the linewidths increase or decrease slightly depending on the change to g near the Fermi energy. Experimentally, an anomalously large phonon linewidth has been observed in $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ at a $\mathbf{q}$ -point between Γ and **X** [89], which was not found in DFT based calculations [20]. Future studies with DFT+DMFT may be able shed light on this observation, which appears to be related to strong correlation.

4.5 Electron-Phonon Coupling in the Impurity Problem

While the main focus of this paper is on the correlation-induced changes in the lattice EPC, in this section we consider the EPC from the perspective of the quantum impurity model that determines the self-energy within DMFT. We first analyze the hybridization between the impurity and lattice $\Delta(i\omega_n)$ and its phonon-induced

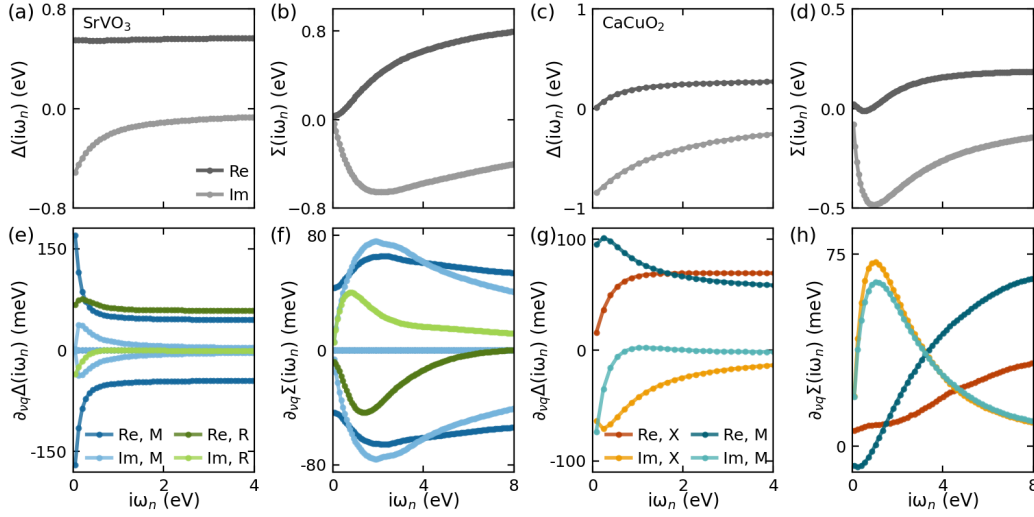


Figure 4.7: Impurity problem self-energy and hybridization on the Matsubara axis. (a) Impurity hybridization and (b) impurity self-energy in SVO at $\beta = 80/\text{eV}$. (c-d) Same for CCO at 0.15 hole doping and $U = 3.1 \text{ eV}$. (e) phonon perturbations to the impurity hybridization and (f) impurity self-energy in SVO. (g-h) Same for CCO. In all cases, the on-site energy shift is added into the hybridization, and the double counting is subtracted from the self-energy.

change $\partial_{v\mathbf{q}}\Delta(i\omega_n)$ in order to understand how lattice EPC translates to a coupling to the correlated local orbitals. Then, we analyze the self-energy $\Sigma(i\omega_n)$ and its phonon-induced change $\partial_{v\mathbf{q}}\Sigma(i\omega_n)$ to understand the interaction contribution to EPC in terms of changes to dynamical correlation. We work with the Matsubara-frequency quantities that are direct outputs from our calculations and draw mainly qualitative conclusions.

In the impurity model, the local orbitals couple to a non-interacting bath of lattice states via a hybridization function $\Delta(i\omega_n)$. The hybridization is a frequency-dependent matrix in the local orbital basis determined through a self-consistency condition between the impurity and lattice Green's functions. A phonon-induced change to the non-interacting lattice states $\partial_{v\mathbf{q}}V_{KS}$ will therefore translate to a change to the hybridization $\partial_{v\mathbf{q}}\Delta(i\omega_n)$. The change in the hybridization function defines the EPC in the impurity model. The infinite frequency limit of $\partial_{v\mathbf{q}}\Delta(i\omega_n)$ is a real matrix whose diagonal elements are phonon induced changes in the on-site energies², equal to the Wannier-local part of the DFPT EPC $\langle i, 0 | \partial_{v\mathbf{q}}V_{KS} | j, 0 \rangle$. The

²In some formulations of DMFT the hybridization function is defined to vanish in the infinite frequency limit and the infinite frequency limit is included in the static Hamiltonian of the impurity model. In the present discussion we consider V_{loc} to be part of the hybridization as both it and the frequency-dependent part of $\Delta(i\omega_n)$ contribute an EPC in the impurity model.

frequency dependence of the change in $\Delta(i\omega_n)$ found in our calculations reflects changes to the hybridization with surrounding sites due to a phonon distortion with a definite wavevector. These changes arise from phonon-induced changes to the intersite hopping, to in on-site energies on different sites, and to phonon-induced changes to correlation effects. The frequency dependence indicates that the physics of EPC to a specific $\mathbf{q}$ -dependent phonon in the DFT+DMFT formalism is in general richer than models of local EPC.

Fig. 4.7 panels (a) and (e) respectively show the Matsubara-axis hybridization function for SVO and the changes due to $\mathbf{M}$ -mode and $\mathbf{R}$ -mode phonon distortions. For the SVO $\mathbf{M}$ -mode, which couples to level splitting between local orbitals, panel (e) shows that impurity EPC $\partial_{v\mathbf{q}}\Delta(i\omega_n)$ is primarily real. We observe that $\text{Re}(\partial_{v\mathbf{q}}\Delta(i\omega_n))$ is frequency dependent only below ~ 0.5 eV. The high frequency constant value is the DFPT Jahn-Teller coupling that relates atomic motions to the level splitting; the increase in amplitude at low frequency expresses the physics that band structure and correlation effects enhance the tendency towards orbital ordering at the $\mathbf{q} = \mathbf{M}$ wavevector. For the SVO $\mathbf{R}$ -mode, which couples to the total on-site charge density, $\partial_{v\mathbf{q}}\Delta(i\omega_n)$ is almost frequency-independent, suggesting that the charge susceptibility in SVO has only a weak momentum dependence.

Panels (c) and (g) of Fig. 4.7 respectively show the Matsubara-axis hybridization function for CCO and the changes due to $\mathbf{X}$ and $\mathbf{M}$ -mode phonon distortions. Panel (g) shows that for the CCO $\mathbf{M}$ -mode (which does not break the point symmetry) the bare coupling is enhanced at low frequencies, indicating that lattice effects and non-local EPC increases the tendency to charge ordering at the $\mathbf{M}$ -wavevector. For the $\mathbf{X}$ -mode (which reduces point symmetry from C_4 to C_2), the Matsubara frequency dependence is opposite, indicating that these effects suppress the ordering tendency at this wavevector. In CCO, $\partial_{v\mathbf{q}}\Delta(i\omega_n)$ is moderately renormalized from its bare DFT value by correlation via the self-consistency condition (see SM [50]).

We now consider the impurity self-energy $\Sigma(i\omega_n)$ and its phonon-induced change $\partial_{v\mathbf{q}}\Sigma(i\omega_n)$. The self-energy is the sum of a real, frequency-independent contribution (the Hartree (Σ_H) and double counting (Σ_{DC}) terms) and a frequency-dependent part that originates from the beyond-Hartree many-body dynamics and vanishes in the infinite frequency limit. The correlation contribution to the EPC is defined in terms of the changes to the self-energy; in what follows we analyze our results to obtain insights into the nature of changes.

It is useful to consider the spectral representation linking the imaginary part of the real frequency self-energy $\text{Im}\Sigma(\omega)$ to the Matsubara axis self-energies,

$$\text{Im}\Sigma(i\omega_n) = -i\omega_n \int \frac{d\omega}{\pi} \frac{\text{Im}\Sigma(\omega)}{\omega_n^2 + \omega^2} \quad (4.9)$$

$$\text{Re}\Sigma(i\omega_n) = \int \frac{d\omega}{\pi} \frac{\omega \text{Im}\Sigma(\omega)}{\omega_n^2 + \omega^2} + \Sigma_H - \Sigma_{DC}. \quad (4.10)$$

Note that the $i\omega_n$ dependence in $\text{Re}\Sigma(i\omega_n)$ requires particle-hole asymmetry: if $\text{Im}\Sigma(\omega) = \text{Im}\Sigma(-\omega)$, $\text{Re}\Sigma(i\omega_n)$ in Eq. 4.10 is frequency independent.

Eqs. 4.9,4.10 show that changes to $\Sigma(i\omega_n)$ can arise from both changes to the Hartree/DC terms and from changes to $\text{Im}\Sigma(\omega)$. Frequency-independent beyond-DFT methods including DFT+ U and hybrid functionals give frequency-independent EPC renormalizations related to those arising here from changes to $\Sigma_H - \Sigma_{DC}$. These changes may be thought of as coming from correlation-induced changes in average local susceptibilities (for example suppression of on-site charge fluctuations by a repulsive U or correlation-induced enhancements of orbital splitting relating to correlation-induced orbital order).

Lattice distortion-induced changes to $\text{Im}\Sigma(\omega)$ lead to frequency dependence of the EPC. To understand the nature of these changes, it is useful to review the basic properties of $\text{Im}\Sigma(\omega)$, which may roughly be characterized by an over-all amplitude, a frequency scale, and a particle-hole asymmetry. For simple moderately correlated metals at low temperature, we expect $\text{Im}\Sigma(\omega)$ to vanish at zero frequency, to grow $\sim \omega^2$ at low frequency, pass through maxima at positive and negative frequency near $\omega \sim \pm\omega_0$, and then to decay to zero at high frequency. In this picture, the particle-hole asymmetry is encoded in different weights at the peaks at $\pm\omega_0$ and the overall scale of the self-energy comes from the peak amplitudes. A simple model that encodes this physics is

$$\text{Im}\Sigma(\omega) = (A + dA)\delta(\omega - \omega_0) + (A - dA)\delta(\omega + \omega_0), \quad (4.11)$$

where A is an amplitude with dimension of energy squared, ω_0 is a characteristic frequency scale, and dA parametrizes the particle-hole asymmetry. Particle-hole asymmetry may also manifest as a difference in the absolute value of the peak positions for positive and negative frequency; as will be seen, introducing different frequencies is not necessary for our analysis. The parameters can be fit from the Matsubara axis data in Fig 4.7, with ω_0 linked to the frequency scale and A and dA setting the magnitudes of $\text{Im}\Sigma(i\omega_n)$ and $\text{Re}\Sigma(i\omega_n)$ respectively. A rough measure of the effective low-frequency correlation strength is the low-energy mass enhancement/quasiparticle renormalization $Z^{-1} = 1 - \frac{\partial \text{Im}\Sigma}{\partial i\omega} \sim \frac{A}{\omega_0^2}$, so that vanishing

ω_0 causes the effective low-frequency correlation strength to diverge. Though simplistic, this model qualitatively accounts for the calculated frequency dependence of $\partial_{\mathbf{v}\mathbf{q}}\Sigma(i\omega_n)$ and its simple shape is conceptually confirmed by preliminary data from real frequency solvers to be published elsewhere.

For SVO, the self-energy of the undistorted structure, shown in panel (b) of Fig. 4.7 is consistent with this simple picture, with an $\text{Im}\Sigma$ characterized by a single frequency scale $\omega_0 \sim 1.8$ eV and a high degree of particle-hole asymmetry (since the change in real part of $\Sigma(i\omega_n)$ from high to low frequency $\int \frac{d\omega}{\pi} \frac{\omega \text{Im}\Sigma}{\omega^2} \sim \frac{dA}{d\omega_0}$ is very similar in magnitude to the maximum value of $\text{Im}\Sigma(i\omega_n = \omega_0) = \int \frac{d\omega}{\pi} \frac{\text{Im}\Sigma\omega_0}{\omega_0^2 + \omega^2} \sim \frac{A}{\omega_0}$). Now consider $\partial_{\mathbf{v}\mathbf{q}}\Sigma(i\omega_n)$ for SVO shown in Fig. 4.7(f), beginning with the **M**-mode. For this mode, there is no change in charge density on the impurity so $\partial_{\mathbf{v}\mathbf{q}}\Sigma_{\text{DC}} = 0$. We see that $\partial_{\mathbf{v}\mathbf{q}}\text{Re}\Sigma(i\omega_n)$ has only a weak $i\omega_n$ dependence, meaning that the phonon-induced change in the self-energy is nearly particle-hole symmetric. The $\partial_{\mathbf{v}\mathbf{q}}\Sigma_H$ is large, about the same size as the static part of $\partial_{\mathbf{v}\mathbf{q}}\Delta$, and acts to amplify the effect of the lattice distortion as expected from the Hartree-level enhancement of the orbital susceptibility. However, the $\partial_{\mathbf{v}\mathbf{q}}\text{Im}\Sigma(i\omega_n)$ is of comparable magnitude to the Hartree shift, and has a frequency dependence very similar to that of $\text{Im}\Sigma(i\omega_n)$, indicating that $\partial_{\mathbf{v}\mathbf{q}}\Sigma(i\omega_n)$ is in effect an increase in A plus a similarly sized Hartree shift.

The effect for the SVO **R**-mode distortion is different. Because this phonon couples to the total charge density, the double counting term is affected. The real part of the change in the Matsubara axis self-energy essentially vanishes at high frequency, meaning that $\partial_{\mathbf{v}\mathbf{q}}\Sigma_{\text{DC}}$ and $\partial_{\mathbf{v}\mathbf{q}}\Sigma_H$ cancel out. The change in the real part also nearly vanishes at low frequency, so the change in $\text{Im}\Sigma$ must be composed of two counterbalancing effects which add in the imaginary part and cancel in the real part: a change in the particle-hole asymmetry and a change in the characteristic frequency. That $\partial_{\mathbf{v}\mathbf{q}}\text{Im}\Sigma(i\omega_n)$ decays much more rapidly in $i\omega_n$ than does $\text{Im}\Sigma$ also shows that a change in frequency ω_0 is an important effect. These qualitative considerations are further supported by the more detailed modeling of the self-energy and its changes presented in the SM [50].

For CCO, the self-energy of the undistorted structure, shown in Fig. 4.7(d) is also mainly consistent with this simple picture. The weak low frequency structure evident in $\text{Re}\Sigma(i\omega_n)$ indicates a small amplitude low-frequency contribution to the self energy, which we do not consider further here. The main feature is a $\text{Im}\Sigma$ characterized by a single frequency scale $\omega_0 \sim 1$ eV and a lower degree of particle-hole asymmetry than in SVO (since the change in $\text{Re}\Sigma(i\omega_n)$ from

high to low frequency $\int \frac{d\omega}{\pi} \frac{\omega \text{Im}\Sigma}{\omega^2} \sim \frac{dA}{\omega_0}$ is about half of the maximum value of $\text{Im}\Sigma(i\omega_n = \omega_0) = \int \frac{d\omega}{\pi} \frac{\text{Im}\Sigma\omega_0}{\omega_0^2 + \omega^2} \sim \frac{A}{\omega_0}$.

Last we consider $\partial_{\nu\mathbf{q}}\Sigma(i\omega_n)$ for CCO shown in Fig. 4.7(h), beginning with the **M**-mode. The large amplitude of the change in the real part, and the slower frequency decay (cf the discussion of the **R**-mode in SVO) indicates that an important effect of the phonon distortion is a change in particle-hole asymmetry. The more rapid decrease of the change in the imaginary part indicates that a particle-hole symmetric change in the basic frequency scale is also important. The **X**-mode exhibits a smaller but still present change in particle hole asymmetry as well as a change in the frequency scale. Consistent with the discussion in Sec. 4.4, we see that the phonons couple to dynamical correlation including particle hole asymmetry and proximity to a Mott transition.

This qualitative analysis of the self-energy reveals that the phonon-distortion-induced changes to the self-energy (and therefore the correlation-induced changes to the EPC) have a dynamical structure that depends on the physics of the material and the nature of the distortion. Because the phonon distortions can modify the correlation strength, particle hole asymmetry, and characteristic frequency of the self-energy in addition to inducing a static shift, a static (Hartree-type) theory of EPC misses important physical effects.

4.6 Comparison to DFPT+ U and Previous Studies on Correlation-Modified EPC

In this section we compare our results to DFPT+ U and other previous work, beginning with SVO. Our calculations show increased EPC and e -ph scattering for the **M**-point Jahn-Teller phonon mode which couples to the orbital degree of freedom, while a slight suppression is found for the **R**-point breathing mode. These effects can be qualitatively compared to those found in SVO using DFPT with a Hubbard correction [12] (differences in the interactions, orbitals, and charge self-consistency between the two calculations mean that the comparison is only qualitative). The Wannier-local EPC for these modes calculated with DFPT, DFT+DMFT, and DFPT+ U are shown in Table I.

DFPT+ U predicts some trends consistent with our DFT+DMFT calculations, such as significantly increased EPC for the **M**-point mode compared to DFPT. However, DFPT+ U does not capture the frequency dependence of the coupling. One consequence of this can be seen in the **R**-point mode: DFPT+ U and the $\omega \rightarrow \infty$ Hartree limit of DFT+DMFT both predict slight increases in the coupling strength,

g_{loc}	M Jahn-Teller	R Breathing
DFPT	44 meV	58 meV
DFPT+ $U = 3$ eV [12, 93]	73 meV	61 meV
DMFT ($\omega = 0$)	87 meV	50 meV
DMFT ($\omega \rightarrow \infty$)	93 meV	60 meV

Table 4.1: Wannier-local EPC in SVO in different approximations. $g_{ij\nu}^{\mathbf{R}_e=0,\mathbf{q}}$ is a diagonal matrix of $(0, g, -g)$ for the **M**-point mode and (g, g, g) for the **R**-point mode, where g is the value shown. DFT+DMFT calculations shown are at $\beta = 80$ / eV. DFPT+ U calculations use atomic orbitals with $U = 3$ eV and $J = 0$ [12].

but DFT+DMFT predicts a slight decrease at $\omega = 0$ (which is more relevant to transport). This indicates that the frequency dependence is important because the EPC of physical interest near the Fermi energy may differ from that in the static limit.

In CCO, we find that e - e interactions have a moderate effect on EPC at the Fermi energy and induce a strong frequency dependence. Some of these results are consistent with previous work in model systems, for example, our finding that increasing U decreases the $\omega = 0$ EPC for the Holstein-like **M**-point full breathing mode agrees with work in a Hubbard-Holstein model using quantum Monte Carlo [32] and DMFT [42]. Table II lists the Wannier-local EPC in CCO in different approximations, including a DFPT+ U calculation. In DFPT+ U , the coupling is slightly increased for both phonon modes compared to DFPT, which can be compared with the $\omega \rightarrow \infty$ Hartree limit of DFT+DMFT where we find large increases for both modes. In contrast, at $\omega = 0$ in DFT+DMFT, the **X**-mode coupling is slightly increased while the **M**-mode coupling is slightly decreased, further demonstrating the importance of a dynamical treatment of the EPC.

These results also highlight the effect of the double counting term in the self-energy.

g_{loc}	X Half-Breathing	M Full-Breathing
DFPT	70 meV	53 meV
DFPT+ U [93]	79 meV	65 meV
DMFT ($\omega = 0$)	76 meV	45 meV
DMFT ($\omega \rightarrow \infty$)	117 meV	137 meV

Table 4.2: Wannier local EPC in CCO in different approximations. All calculations have $U = 3.1$ eV and 0.15 hole doping. The DFPT+ U calculations use atomic orbitals.

The Hartree contribution to the self-energy from the Hubbard interaction suppresses

local charge fluctuations and thus decreases the effect of "breathing mode" lattice distortions on the electrons, as observed in model-system studies [32, 42]. However, the double counting term present in DFT+DMFT substantially changes the infinite frequency limit of the self-energy, partially canceling or in some cases outweighing the Hartree term, consistent with findings that the double counting correction in DFT+DMFT can promote charge ordering [94]. Further explanation of the double counting correction as well as tables indicating the contribution of the double counting potential to the Wannier-local g , and values of the Wannier-local g for other temperatures in SVO and U and doping values in CCO are provided in SM [50]. In cases where the Wannierization contains both correlated and uncorrelated orbitals, the double counting corrections and charge self-consistency may play a more important role, making these important topics for future investigation.

Recently, first-principles calculations using GW -based perturbation theory (GWPT) have found significantly increased EPC in hole doped La_2CuO_4 [4] compared to DFPT, and both GWPT and hybrid functional calculations have found significantly increased EPC in single orbital bismuthates [14, 15]. These results, which have been attributed to decreased electronic screening in GW and hybrid functional DFT compared to semi-local DFT, contrast our findings in CCO. While we expect our DFT+DMFT calculations to better capture local, dynamical correlation, they are limited by the single orbital impurity approximation and the DFT starting point. More research is needed to disentangle these effects on the e -ph interactions, and more realistic impurity construction schemes or methods like GW +DMFT [95, 96] may help clarify this by including both strong correlation and accurate electronic screening.

4.7 Conclusion

We have presented a method for calculating the EPC in a correlated material using DFPT and finite difference DFT+DMFT calculations. We applied this method to several representative optical phonon modes in two materials of interest: the 3-band cubic perovskite SVO, for which we compared phonons that couple to orbital splitting and to on-site density, and the infinite layer cuprate CCO, a representative Mott-Hubbard system.

In SVO, we find that electronic correlation from DMFT significantly enhances the EPC at the Fermi level of an **M**-point bond stretching phonon which couples to the orbital degree of freedom, while slightly suppressing that of an **R**-point breathing mode which couples to the charge degree of freedom. We find the same trend in the

calculated electron scattering rates due to these phonon modes. These findings help to explain the lower resistivity in DFPT-based transport calculations compared with experiment [12, 69], and highlight the strong effect of electronic correlation in the e -ph interactions of multi-orbital correlated metals.

In CCO, we find that correlation within the $d_{x^2-y^2}$ orbital has a modest effect on the coupling of Fermi surface electrons to full- and half-breathing modes but induces a very strong dependence of the EPC on electron frequency. A further surprise is that even though the basic EPC coupling in CCO is Holstein-like (coupling to density) for both breathing modes, which competes with correlation (acting to suppress density fluctuations), the net effect of the correlations on the EPC at the Fermi energy is a slight suppression for the **M**-full breathing mode but a slight enhancement for the **X**-half breathing mode. These differences observed between the two modes suggest that the physics of EPC in the actual material is richer than implied by simple Holstein-Hubbard-type models.

A remarkable finding is the significant frequency dependence of the EPC, which leads to significant differences between the value of the near-Fermi surface EPC and the infinite frequency (Hartree) limit and produces variation of the EPC over scales of the order the phonon frequency especially for CCO. This strongly frequency-dependent EPC is absent in $+U$ and hybrid extensions of DFT and may have important consequences for physical properties. For example, the associated particle-hole asymmetry in quasiparticle relaxation times could impact properties like thermoelectric transport. For instance, a recent experiment unexpectedly found a positive Seebeck coefficient in a hole-doped cuprate [97], which the authors explained by suggesting a greater scattering rate above the Fermi energy than below. This would be consistent with our findings of EPC in hole-doped CCO. Other studies in hole-doped cuprates [98, 99] have found a transition from negative to positive Seebeck coefficient as temperature increases, which were explained by Fermi surface reconstruction, but could plausibly involve thermal activation of particle-hole asymmetric e -ph scattering processes. In this context, our findings that correlation can induce a strong frequency dependence in the EPC motivate further studies on the role of this effect on scattering and transport properties.

Taken together, our results highlight the importance of electronic correlation in the e -ph physics of correlated metals, which leads to significant renormalization of the couplings at the Fermi surface, a strong electron frequency dependence, and modifications to physical properties. We also demonstrate the dependence of these effects on the microscopic nature of the electronic states and phonon modes considered.

A strength of our method is the ability to Wannier interpolate the calculated EPC to a complete set of bands and $\mathbf{k}$ -points. However, it is limited to a small number of phonon modes and $\mathbf{q}$ -points. This indicates the importance of more efficient computational schemes to calculate EPC at the DMFT level. Furthermore, our findings call for investigation into the effect of different impurity construction and double counting correction schemes and the development of EPC calculations using methods such as charge self-consistent DFT+DMFT or *GW*+DMFT.

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Chapter 5

PREDICTING ELECTRON-PHONON COUPLING AND ELECTRONIC TRANSPORT AT THE MOIRÉ SCALE IN TWISTED BILAYER GRAPHENE

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Abstract

First-principles calculations can accurately describe electron-phonon (e-ph) interactions and electronic transport in a wide range of materials, but are currently limited to unit cells with up to ~ 100 atoms due to computational cost. Here, we develop an atomistic electronic potential with Holstein- and Peierls-like terms for modeling e-ph interactions and phonon-limited electronic transport that enables the study of moiré systems with thousands of atoms per unit cell. This method can accurately reproduce first-principles e-ph coupling and resistivity in graphene and large-angle twisted bilayer graphene (TBG). Using this approach, we study TBG over a range of twist angles down to 1.6° (5044-atom unit cell), and report the evolution of e-ph interactions and phonon-limited resistivity with twist angle. The predicted resistivity increases by two orders of magnitude between 13.2° and 1.6° , driven by the progressive reduction of the electronic energy scale. Our calculations can predict key experimental trends in 2.0° and 1.6° TBG, including the resistivity and its dependence on temperature and band filling. Our work establishes a scalable approach for quantitative studies of e-ph interactions and transport in moiré materials and other systems with previously inaccessible length scales.

First-principles calculations of electronic transport achieve predictive accuracy in classes of materials ranging from metals [1] and semiconductors [2, 3, 4, 5, 6, 7, 8, 9] to complex insulating [10, 11, 12] and metallic oxides [13, 14, 15]. These calculations combine first-principles electronic structure, phonons, and electron-phonon (e-ph) interactions [16, 17, 18], building on the framework of density functional theory (DFT) and linear-response DFT [19]. Studies of electronic transport in

graphene and related systems illustrate these developments: following early analytic models [20, 21, 22], first-principles calculations have studied band renormalization and electron scattering rates [23, 24], temperature dependent resistivity [25, 26], and more recently Hall mobility [27], twisted bilayer structures [28], and coupled electron and phonon phenomena [29, 30, 31].

Electronic transport in twisted bilayer graphene (TBG) has been studied intensely as the emergent moiré physics leads to unconventional behaviors. Experimental studies have reported a large resistivity in small-angle TBG over a range of temperatures, electron fillings, and twist angles [32, 33, 34, 35]. Measurements of $\sim 2^\circ$ TBG have shown power-law resistivity and attributed this behavior to acoustic phonon scattering [32]. Near the magic angle, observations of T -linear resistivity [33, 34, 35] have been explained in terms of mechanisms including Planckian scattering and strange metallicity [34, 35], e-ph scattering reminiscent of monolayer graphene (MLG) [33, 36, 37], and dominant electron-electron scattering [38].

The electronic structure of TBG is challenging to describe [39] due to the combination of large system size, sensitivity to the atomic structure [40, 41], and correlation effects [42, 43, 44, 45, 46, 47]. Electronic transport poses additional challenges, including the sensitivity of e-ph interactions to the electronic structure and phonons, and the need to account for a large number of scattering processes. Although theoretical studies in the Boltzmann picture have proposed phonon-limited transport as a viable explanation for the observed resistivity, these works rely on simplifying approximations such as Dirac electronic states [36, 48], heuristic e-ph coupling constants [36, 37, 48], and the inclusion of only intralayer [37, 36, 48] or breathing and flexural phonons [49].

However, a treatment of e-ph interactions and electronic transport at the moiré scale with first-principles accuracy is still lacking. Despite the development of scalable approaches for e-ph interactions [50, 51, 52, 53, 54], first-principles e-ph calculations are currently limited to systems with 50–100 atoms in the unit cell [28, 9]. A relevant example is a recent study of e-ph interactions and transport in large-angle TBG, which showed that layer-breathing phonons can substantially modify the temperature dependence of the resistivity from the T -linear behavior characteristic of MLG [28]. Extending these calculations to low twist angles by brute force is not possible due to the computational cost of linear-response DFT.

In this chapter, we show scalable calculations of e-ph interactions and phonon-limited resistivity that can reach the moiré scale while retaining near-first-principles accuracy. This is achieved through an electron-lattice potential which generates

the e-ph coupling, with Holstein- and Peierls like terms parameterized using linear-response DFT. This method relies on a small number of short-range terms that depend on the local atomic positions, and can accurately reproduce first-principles e-ph interactions and resistivity in MLG and large-angle TBG, used here as benchmarks. Our approach enables the study of e-ph coupling and electronic transport in TBG for twist angles as small as 1.61° , corresponding to moiré unit cells with thousands of atoms. Our predicted resistivity in TBG, computed in the Boltzmann picture, increases substantially for decreasing twist angles, mainly driven by a corresponding decrease in the electron band velocity. Our calculations achieve very good agreement with the experimental resistivity at low twist angles, indicating that the resistivity of TBG is largely phonon-limited down to $\sim 2^\circ$ twist angles despite the substantial interlayer electronic coupling at such low angles. These findings shed light on the microscopic mechanisms of electronic transport in TBG at a previously inaccessible length scale, and our approach provides a promising route to study transport phenomena in emerging moiré material platforms.

Electron-Phonon Coupling in TBG— The e-ph coupling $g_{mn\nu}(\mathbf{k}, \mathbf{q})$ is often calculated as a Fourier transform of local real-space contributions [55, 56, 17, 18]:

$$g_{mn\nu}(\mathbf{k}, \mathbf{q}) = \frac{1}{N_e} \sum_{ija} \sum_{\mathbf{R}_e \mathbf{R}_p} e^{i(\mathbf{k} \cdot \mathbf{R}_e + \mathbf{q} \cdot \mathbf{R}_p)} \sqrt{\frac{\hbar}{2M_a \omega_{\nu\mathbf{q}}}} \times \mathbf{e}_{\nu\mathbf{q}}^{a\mu} U_{mi, \mathbf{k}+\mathbf{q}} \langle i\mathbf{0} | \partial_{u_{a\mu\mathbf{R}_p}} V | j\mathbf{R}_e \rangle U_{nj, \mathbf{k}}^\dagger, \quad (5.1)$$

where m and n index electronic bands, i and j index local orbitals, a indexes an atom with mass M_a , μ is a Cartesian direction, $\mathbf{k}$ and $\mathbf{q}$ are crystal momenta of electrons and phonons respectively, $\mathbf{R}_e$ and $\mathbf{R}_p$ are corresponding lattice vectors, $U_{nj, \mathbf{k}}$ transforms from the orbital to the Bloch basis, $\omega_{\nu\mathbf{q}}$ is the phonon frequency, and $\mathbf{e}_{\nu\mathbf{q}}^{a\mu}$ is the phonon eigenvector. The change in Kohn-Sham potential with respect to atomic motions, $\partial_{u_{a\mu\mathbf{R}_p}} V$, is typically computed using density functional perturbation theory (DFPT). When the e-ph interactions are short-ranged, the sum converges with only a few terms [55, 56].

To obtain a scalable approach that can reach the moiré scale, we formulate a tight-binding-style approximation where the e-ph coupling is generated by an electronic potential, $\hat{V}_{\text{e-ph}}$, written as an operator in the atomic-orbital basis, with onsite and hopping matrix elements depending on a small number of atomic positions:

$$\hat{V}_{\text{e-ph}}(\{\mathbf{r}_a\mathbf{R}\}) = \sum_i \epsilon_i(\{\mathbf{r}_a\mathbf{R}\}) c_{i\mathbf{0}}^\dagger c_{i\mathbf{0}} + \sum_{i,j,\mathbf{R}_e} (t_{ij\mathbf{R}_e}(\{\mathbf{r}_a\mathbf{R}\}) c_{i\mathbf{0}}^\dagger c_{j\mathbf{R}_e} + h.c.). \quad (5.2)$$

Here, $c_{i\mathbf{R}_e}^\dagger$ creates an electron in a localized orbital $\phi_{i\mathbf{R}_e}(\mathbf{r}) = \varphi_i(\mathbf{r} - \tau_i - \mathbf{R}_e)$, and the onsite energy $\epsilon_i(\{\mathbf{r}_a\mathbf{R}\})$ and hopping matrix elements $t_{ij\mathbf{R}_e}(\{\mathbf{r}_a\mathbf{R}\})$ are differentiable functions of the atom positions $\{\mathbf{r}_a\mathbf{R}\}$. The real-space e-ph couplings are then calculated as

$$\langle i\mathbf{0} | \partial_{u_{a\mu\mathbf{R}_p}} \hat{V}_{\text{e-ph}} | j\mathbf{R}_e \rangle = \partial_{u_{a\mu\mathbf{R}_p}} [\epsilon_i \delta_{i\mathbf{0},j\mathbf{R}_e} + t_{ij\mathbf{R}_e}]. \quad (5.3)$$

In our approach (see end matter), the onsite terms in $\hat{V}_{\text{e-ph}}$ are Holstein-like [57], while the hopping terms are Peierls-like [58] and include 1st-4th nearest intralayer neighbors and short-ranged interlayer hoppings. Both onsite and hopping terms depend on the positions of nearby atoms in both graphene layers. This provides a versatile ansatz for short-range e-ph coupling, which we parameterize from DFPT calculations; in polar materials, a long-range Frölich term could also be included [59]. In combination with a tight-binding Hamiltonian [40] for electronic structure and a force-field for lattice dynamics [60, 41], this approach enables e-ph coupling and transport calculations in moiré systems with more than 5,000 atoms. Our Holstein-Peierls ansatz differs from previous calculations of e-ph coupling in TBG based on derivatives of a Slater-Koster tight-binding Hamiltonian employed to study superconductivity and phonon linewidths [61, 62, 63].

Large Angle TBG— First, we benchmark our approach against DFPT-based e-ph calculations in large angle (21.8°) TBG [28]. Figures 5.1(a) and 5.1(b) compare the e-ph coupling strength, color-coded on the phonon dispersion, obtained with our approach and with DFPT. We find very good agreement for both optical and acoustic modes. Importantly, the e-ph coupling is accurately reproduced for the longitudinal acoustic (LA), transverse acoustic (TA), and layer breathing (LB) modes, which play a key role in transport in MLG [25] and TBG [28]. The model also captures the momentum dependence of the coupling, including the reduced e-ph coupling near Γ for the 20 meV LB mode and the differences between inter- and intravalley couplings, $g(\mathbf{K}, \mathbf{K})$ and $g(\mathbf{K}, \mathbf{K}')$.

We additionally compute the temperature dependent resistivity in MLG and in two large-angle TBG structures (21.8° and 13.2°). Figure 5.1(c) compares results obtained from our approach with calculations using first-principles band structure,

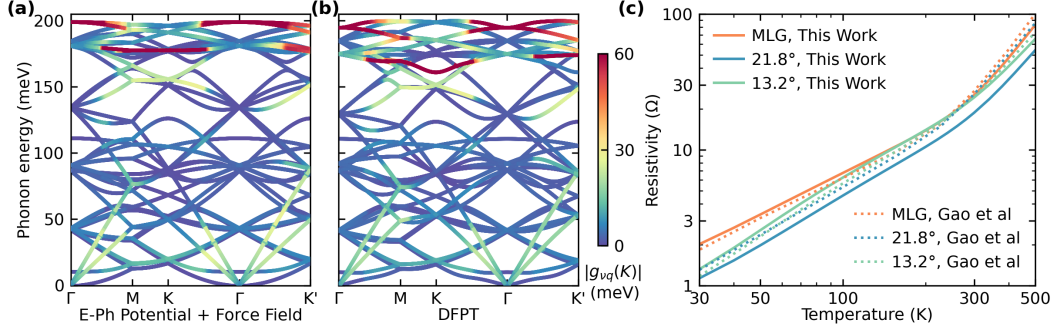


Figure 5.1: (a) E-ph coupling computed in this work, $g_{\nu}(\mathbf{k} = \mathbf{K}, \mathbf{q}) = (\frac{1}{N_b} \sum_{nm} |g_{nm\nu}(\mathbf{k} = \mathbf{K}, \mathbf{q})|^2)^{1/2}$ averaged over Dirac-cone bands, plotted on the phonon dispersion for 21.8° TBG. (b) Same plot as in (a), but with phonons and e-ph coupling from DFPT. (c) Resistivity in MLG, calculated with our approach and from DFPT e-ph coupling [28], in both cases using the RTA.

phonons, and e-ph coupling from Ref. [28], computed in both cases using the Boltzmann equation in the relaxation time approximation (RTA) [16, 64] for a Fermi energy 100 meV above the Dirac point. The resistivity curves for the two methods are in close agreement for all systems, including the overall magnitude as well as key features in the temperature dependence, such as the T -linear resistivity in MLG, activation of LB phonons in the TBG structures, and activation of optical phonons around 300 K.

Small Angle TBG— The efficiency of our approach enables the study of low-angle TBG structures with several thousand atoms. We first calculate the temperature and filling dependent resistivity in 2.0° TBG (3268-atom unit cell) and compare it with experiments [32] in Fig. 5.2. Our calculations capture key trends observed in experiments on 2.0° TBG, predicting resistivity values in very good agreement with experiments below 100 K and correctly describing the dependence of the resistivity on the electron density and band filling. Above 100 K, our calculations underestimate the temperature dependence, leading to a resistivity at 300 K lower by approximately a factor of two relative to experiment. We attribute this discrepancy to the known underestimation of e-ph coupling for optical modes in graphene within DFT [65, 66], which results in a similar discrepancy in first-principles resistivity calculations in MLG [26, 67, 68]. Note also that there is significant variation in the available experimental data. For example, Ref. [33] reports a 2° TBG sample with a 300 K resistivity of over twice that of Ref. [32] and near- T -linear temperature dependence.

We analyze the filling dependence of the resistivity in Fig. 5.2(b), which can be qual-

itatively compared to longitudinal resistance measurements in Ref. [32]. Consistent with experiment, we find a greater resistivity below half filling than above, together with maxima associated with the positions of the van Hove singularities (VHSs) in the upper and lower moiré bands, and peaks near $n = \pm 10^{13} \text{ cm}^{-2}$ corresponding to completely filled or empty moiré bands, respectively. These peaks are smaller in our calculation than in experiment, a trend we attribute to the smaller band gaps between the moiré and dispersive bands in our calculations.

Twist angle dependence— Next, we study the resistivity as a function of twist angle, reporting results for seven twist angles in Fig. 5.3(a). Our results reveal a substantial increase in resistivity with decreasing twist angles, with 1.61° TBG exhibiting an approximately 40 times greater resistivity than 13.2° TBG at 100 K. This trend is due primarily —but not entirely —to a decrease in band velocity with twist angle (see end matter). We also examine the twist angle dependence of the Eliashberg e-ph coupling constant λ [16] [see Fig. 5.3(b)]. We observe a pronounced decrease in λ with twist angle from 13.2° to 2.65° , followed by a moderate increase at lower twist angles. The dependence of λ on twist angle and Fermi energy closely follows the electronic density of states (DOS) [see Supplemental Material (SM) [69]], demonstrating the role of band structure effects in the e-ph interaction.

Analysis of transport trends— We analyze in detail the dependence of the resistivity on temperature and electronic filling for three representative twist angles: 13.2° , a large angle where the layers are nearly decoupled, 3.15° , an intermediate value where the bands are substantially flatter but still have an energy scale several times larger than acoustic phonons, and 1.61° , a small twist angle where the energy scales of flat-band electrons and acoustic phonons become comparable. The band structures for these twist angles, shown in Fig. 5.4(a)-(c) respectively, exhibit this evolution of the electronic energy scale.

For the same sequence of twist angles, the resistivity as a function of temperature and electronic filling are shown in Fig. 5.4(d)-(f) and Fig. 5.4(g)-(i), respectively. For the largest twist angle considered, 13.2° , the resistivity of TBG resembles that of MLG. The temperature dependence is approximately linear over the range of 50-200 K [Fig. 5.4(d)], with some deviation due to LB phonons, as reported previously [28]. The variation of the resistivity with electronic filling [Fig. 5.4(g)] is small over the range of -0.5 to 0.5 electrons per unit cell, where the main trend is a decrease in resistivity with increasing filling due changes in the band velocity. As in MLG, this transport behavior is related to the Dirac band dispersion: the band velocity varies slowly over the energy range associated with transport, and the electronic density of

states increases linearly with Fermi energy (see further analysis in the SM [69]).

Some of these features are also present in the resistivity of 3.15° TBG, but significant deviations emerge [Figs. 5.4(e) and 5.4(h)]. The resistivity remains approximately T -linear over the 50-200 K temperature range for fillings within ± 1 -2 electrons per unit cell, but it exhibits different trends when the moiré bands are close to empty or full. This behavior arises from features in the electronic structure of the moiré bands, including VHSs around ± 150 meV that produce maxima in the density of states and minima in the band velocity. These sharp features modify the temperature dependence of the resistivity when their energy scale becomes comparable to $k_B T$ and the phonon energy.

At the smallest twist angle considered, 1.61° , the resistivity deviates significantly from the Dirac-dispersion behavior, and the temperature dependence of the resistivity becomes strongly non-linear [Fig. 5.4(f)]. For electronic fillings near ± 1 electron per unit cell, a nearly linear regime appears below ~ 100 K, followed by a pronounced deviation at higher temperatures. In 1.61° TBG, features in the electronic structure occur on an energy scale of ~ 20 meV, so the electronic states contributing to transport vary with temperature, leading to a more complex temperature dependence. When the moiré bands are completely filled or empty, the system becomes insulating due to the small gaps between the moiré and dispersive bands, as observed experimentally at similar twist angles [32]. The filling dependent resistivity [Fig. 5.4(i)] reflects the low moiré energy scale, with peaks appearing near the VHSs at 50 K. These features broaden as $k_B T$ becomes comparable to the electronic energy scale, and the dominant feature in the filling dependence becomes a maximum around -1 electron per unit cell, corresponding to a Fermi energy near the lower VHS.

Taken together, our results indicate that the phonon-limited Boltzmann transport picture remains valid for twist angles down to $\sim 1.5^\circ$ over a relatively wide temperature range. The comparison with the experiments of Ref. [32] in Fig. 5.2(a) supports this conclusion. In addition, measurements of 1.59° TBG at half filling of the lower moiré bands (-2 electrons per unit cell) report a T -linear resistivity with a value of $1.6 \text{ k}\Omega$ at 300 K and a slope of $\sim 5 \text{ }\Omega/\text{K}$. Our calculations for 1.61° TBG predict a T -linear regime below 100 K and a slope of $2.5 \text{ }\Omega/\text{K}$ at 300 K, in agreement with the experiment within a factor of two. Our computed resistivity exhibits a weaker temperature dependence than experiment above 100 K for both 1.61° and 2.0° . As discussed above, this discrepancy originates from the known underestimation of optical-phonon coupling in DFT, previously identified in studies of MLG, which limits the accuracy of the predicted resistivity at higher temperatures.

Discussion— As in most tight-binding methods, the electronic Hamiltonian and e-ph coupling in this work do not include effects of modified screening and electronic correlation as a function of twist angle, electronic filling, or temperature. Electronic correlations induce filling-dependent band flattening in TBG at small twist angles [45]. This could increase resistivity via reduced band velocity, as well as change the temperature and filling dependence of resistivity via direct changes to the electronic states involved in transport.

Modified screening and correlation may also significantly effect the e-ph coupling. In MLG, semilocal DFPT underestimates the e-ph coupling compared to *GW* [65, 66]. This leads to roughly a twofold underestimate of experimental mobility by DFT-based calculations [26, 68, 67], which as discussed above is similar to the discrepancy between our calculations and experiment in some cases. Therefore, parameterizing our e-ph model with *GW* might increase e-ph coupling and improve agreement with experiment. In TBG, the e-ph coupling may be further modified due to modified electronic screening by the moiré bands. Near the magic angle, the TBG system becomes strongly correlated [43, 47, 45], which can have a strong effect on electronic structure and e-ph coupling [70, 15, 71, 72]. In TBG, this physics is further complicated by non-local Coulomb interactions [73, 46, 73, 74] and the low energy scale of the correlated moiré bands compared to phonons. Therefore, correlation and screening effects in the e-ph coupling deserve investigation in future studies of TBG.

The electronic transport in small twist angle TBG could also be investigated beyond the phonon-limited Boltzmann regime. The low electronic energy scale of the moiré bands suggest that the physics of the e-ph interaction should be investigated beyond the Migdal approximation [75] made in RTA transport (because the majority of bands remain dispersive, the impact of beyond-lowest-order e-ph interactions is unclear). Scattering mechanisms other than e-ph coupling may also contribute, such as electron-electron scattering at small twist angles [38], and non-quasiparticle transport may become important near the magic angle and at high temperatures.

Conclusion— In summary, we have developed an approach to calculate e-ph coupling and electronic transport at the moiré scale in TBG using an atomistic e-ph coupling model. We study resistivity in TBG across twist angles, finding that resistivity increases in magnitude and gains additional features in the temperature- and filling-dependence as the moiré energy scale decreases. Our results reproduce many of the experimental features in the resistivity of 2.0° and 1.61° TBG, including the magnitude of resistivity in several regimes and key aspects of the filling depen-

dence, and highlight areas for further investigation. Our atomistic approach to e-ph coupling can serve as a starting point for investigating the effects of correlation and screening on the e-ph interaction in TBG. This could shed light on e-ph interactions near the magic angle, relevant to transport and superconductivity [76, 61]. Beyond TBG, our work develops a new approach to understand e-ph coupling and resulting properties at previously inaccessible scales.

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¹Much of my prior knowledge about tight-binding representations of electron-lattice interactions came from a project in Liang’s group [77]

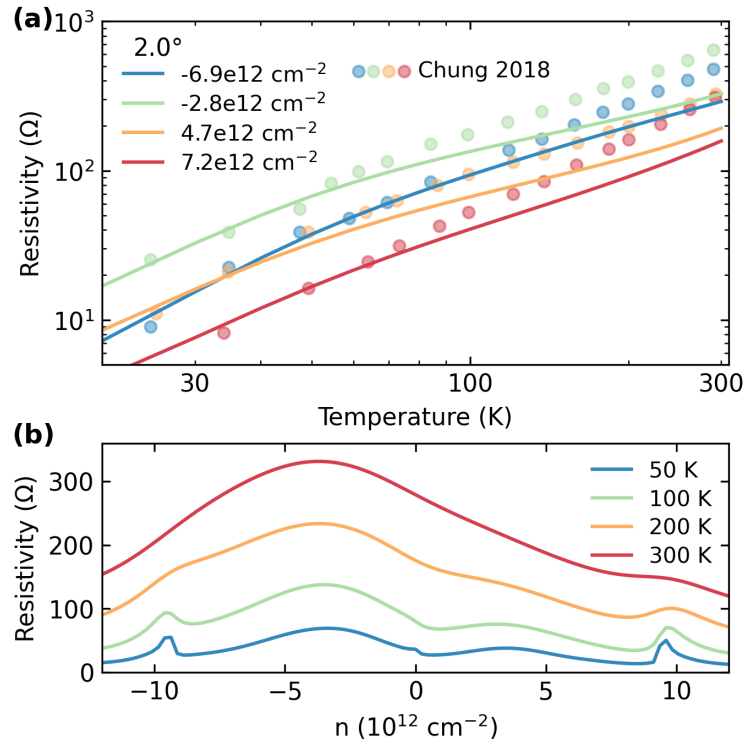


Figure 5.2: (a) Temperature dependent resistivity of 2.0° TBG computed for several electronic fillings (solid lines) and compared to experiments (symbols) [32]. (b) Filling-dependent resistivity in 2.0° TBG at several temperatures. The moiré bands are completely filled / empty at $n = \pm 9.3 \times 10^{12} \text{ cm}^{-2}$.

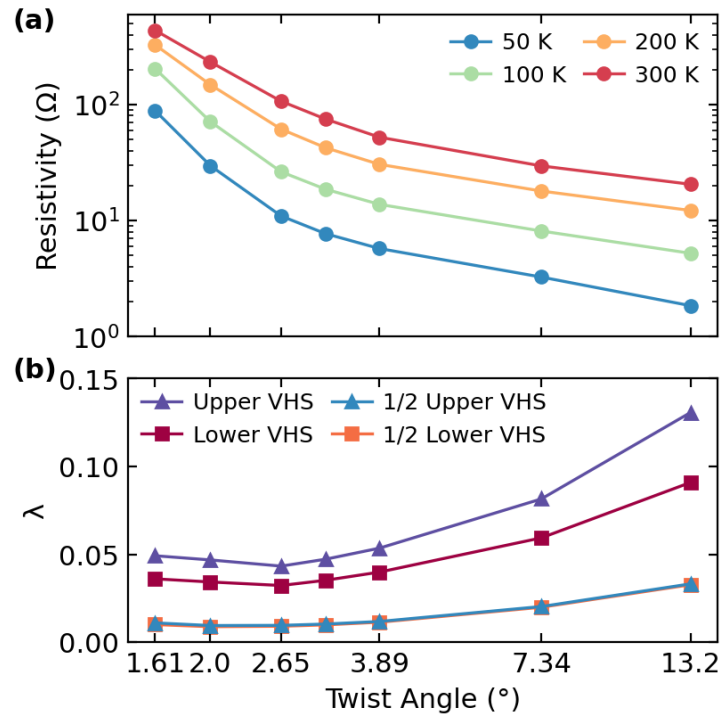


Figure 5.3: (a) Twist angle dependence of the resistivity at several temperatures for a Fermi energy located halfway to the upper VHS. (b) Twist angle dependence of the e-ph coupling strength λ at four fillings.

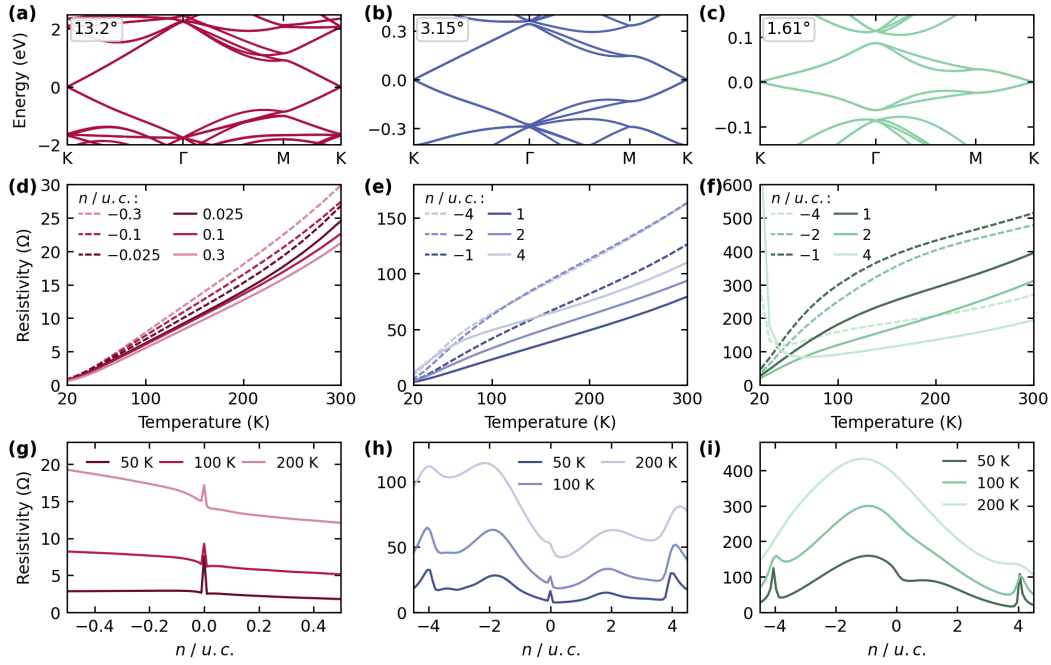


Figure 5.4: Calculated band structure of (a) 13.2°, (b) 3.15°, and (c) 1.61°TBG, respectively. For the same sequence of twist angles, we show the temperature dependent resistivity at several electronic fillings in panels (d)-(f), and the resistivity as a function of electron filling at several temperatures in panels (g)-(i), respectively.

End Matter

5.1 Electron-Phonon Model for TBG

For the e-ph coupling, we design a model for $\hat{V}_{\text{e-ph}}$ which is illustrated in Fig. 5.1. The potential includes first through fourth nearest-neighbor intralayer electronic hopping terms, which depend on the associated bond length and distances and angles between other nearby atoms, giving rise to e-ph coupling. These hopping terms and their dependence on nearby atoms is illustrated in Fig. 5.1(a)-(d). The model also includes an electronic onsite term which depends on positions of first through fourth nearest neighbors as shown in Fig. 5.1(e). The interlayer coupling terms (Fig. 5.1(f)) include coupling to the orbital onsite energy from atomic displacements in the opposite layer and coupling to interlayer hopping in a two-site approximation.

We fit this model so that the real space, orbital basis, atom resolved e-ph coupling predicted agrees with that of Wannier basis DFPT e-ph coupling calculations of MLG and AA stacked, AB stacked, and 21.8° twisted bilayer structures from previous work [28]. From the parameterized $\hat{V}_{\text{e-ph}}$, the real space e-ph coupling is extracted by evaluating Eq. (5.3) using automatic differentiation. For comparison with previous studies [61, 63], we also test $\hat{V}_{\text{e-ph}}$ given by a Slater-Koster model [78] based on the parameterization by Moon and Koshino [79] as well as a local environment tight-binding model [40]. More information on the parameterization of the tight-binding model is provided in the SM [69].

For the electronic structure, we use a local environment tight-binding model [40], which we adjust to bring the magic angle into agreement with experiment at 1.08° by increasing all interlayer hopping Hamiltonian matrix elements by 20%. For intralayer terms in the force field, we use a valence force model [60] which provides both accurate phonon dispersion and eigenmodes compared to DFPT. We use an interlayer potential based on a quantum Monte Carlo based parameterization [41] of the Kolmogorov-Crespi model [80].

5.2 Computational Methods

First-principles results reproduce previous work [28] and use Quantum Espresso [81], Wannier90 [82], and Perturbo [64] with the same numerical parameters.

The tight-binding electronic structure, force-field lattice relaxation and phonons, model e-ph coupling, and resistivity are calculated with a standalone code in the Julia language [83], and use packages for numerical automatic differentiation [84, 85] and linear algebra. We relax the lattice in all systems using the force-field such

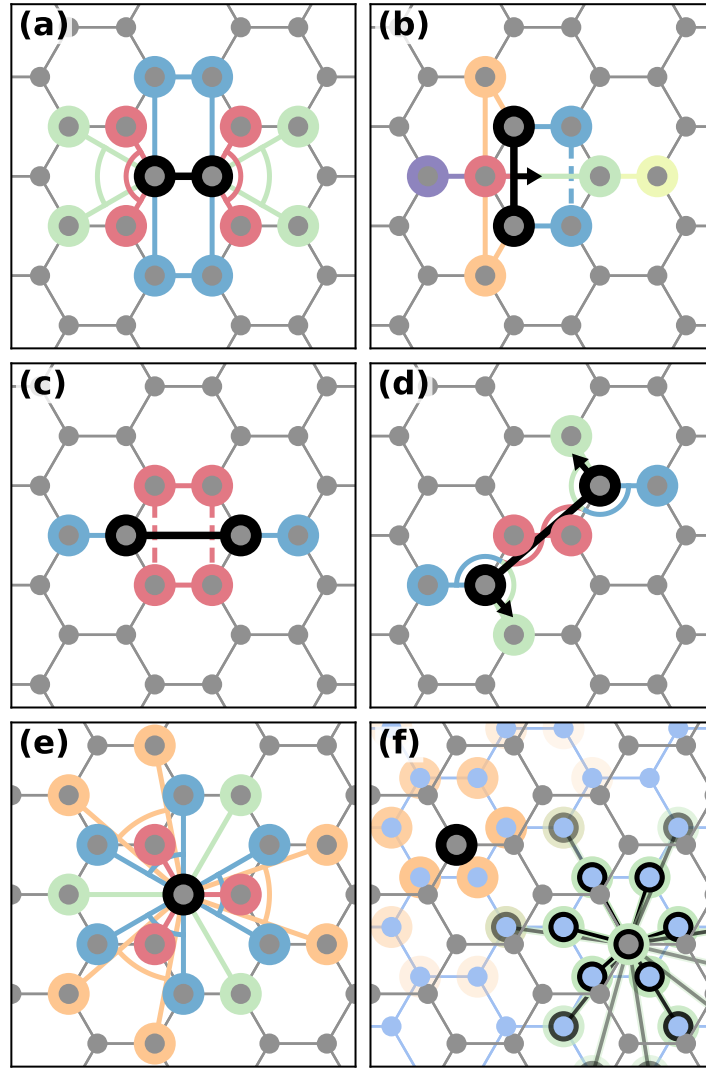


Figure 5.1: Illustration of the terms in the model for $\hat{V}_{e-ph}$. (a) 1-NN (b) 2-NN (c) 3-NN, and (d) 4-NN hopping terms. The matrix element is between orbitals outlined in black, and depends on the bond length (and bond displacement or rotation as denoted by arrows in the 2-NN and 4-NN terms, respectively) and displacements of nearby atoms outlined in colors. Solid lines, arcs, and dashed lines respectively indicate fits to a distance, angle, and perpendicular displacement. (e) Onsite term for the orbital outlined in black. (f) Interlayer terms, with atoms outlined in yellow in the bottom layer affecting the onsite energy of the orbital outlined in black in the top layer, and the bonds outlined in black and green showing hopping matrix elements which depend on bond displacement from the atom in the top layer to nearby atoms in the bottom layer.

that the norm of the matrix of atomic forces is less than 10^{-8} eV / Å, which eliminates most of the dynamical instability of the TBG structures. For all calculations, we exclude phonons with an energy below 0.1 meV.

The resistivity calculations described below use equal $\mathbf{k}$ and $\mathbf{q}$ grids with respectively 1440, 540, 300, 150, 72, 60, 48, 42, and 30 points along each dimension for the MLG and 21.8° , 13.2° , 7.34° , 3.89° , 3.15° , 2.65° , 2.0° , 1.61° TBG structures. We evaluate the Dirac delta functions in band energy using adaptive Gaussian smearing [86], and use a $\pm 8k_B T$ window around the highest / lowest Fermi energy for the highest temperature for bands included in the conductivity calculation, and windows ± 200 meV larger for the sum over bands used to calculate scattering. The electron filling is calculated by integrating the electronic density of states and defined relative to charge neutrality, taken to be the filling at zero temperature with the Fermi energy at the Dirac point.

The calculations of the coupling strength λ use $\mathbf{k}$ grids with respectively 480, 240, 120, 96, 72, 60, and 48 points along each dimension for the 13.2° , 7.34° , 3.89° , 3.15° , 2.65° , 2.0° , 1.61° TBG structures, and half as many $\mathbf{q}$ points along each dimension. These calculations include only the 4 moiré bands closest to half filling. We use static Gaussian smearing of 5 meV for the 13.2° structure going down to 2 meV for the 1.61° structure for the electronic Delta functions and 1 meV for the phonon Delta functions. More information on the computational methods, lattice relaxation and dynamical stability, and numerical parameters is provided in the SM [69].

5.3 Origins of Twist Angle Dependence and Scattering

As observed in the previous section, smaller twist angles exhibit a large decreased in band velocity and corresponding increase in resistivity. From the expression for RTA conductivity, the resistivity scales as $\rho \propto [\nu(E_F)^2 \tau(E_F) N(E_F)]^{-1}$. Because the scattering rate τ^{-1} is roughly proportional to the density of states N , the change in band velocity is the primary factor behind the increased resistivity at small twist angles, with the remainder of the twist angle dependence encoding non-trivial modification to scattering and the electronic states involved in transport.

Figure 5.2(a) shows the temperature-dependent resistivity scaled by the square of the Fermi-surface-averaged velocity at a range of twist angles. The square of the velocity is also shown as an inset. For consistency, at each twist angle, the Fermi energy is set to halfway between the energies of the Dirac point and the upper VHS. Below ~ 100 K, this quantity increases consistently with decreasing twist angle by

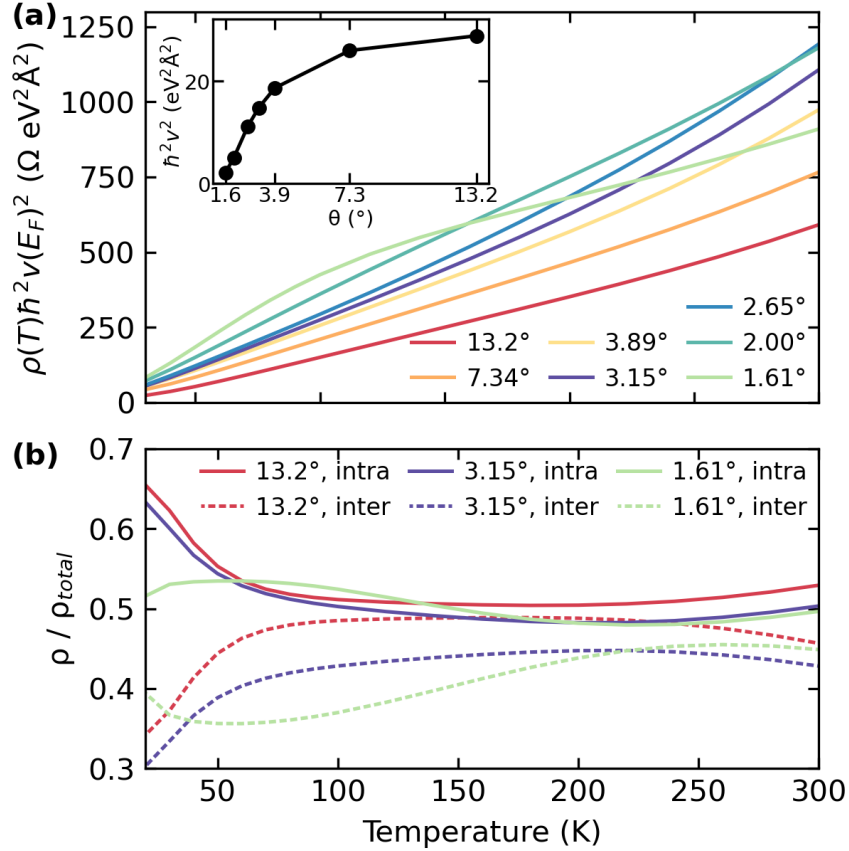


Figure 5.2: (a) Resistivity scaled by the square of the Fermi velocity as a function of temperature for twist angles between 13.2° and 1.61° . This quantifies the change in resistivity due to scattering effects. The Fermi energy is set to halfway between the energies of the Dirac point and the upper Van Hove singularity. (b) Ratio of resistivity calculated with only intra- or interlayer e-ph coupling divided by the full resistivity.

approximately a factor of 3 over the range studied. This indicates that changes in the e-ph scattering processes contribute significantly to the observed increase in resistivity as the twist angle is decreased, possibly because a lower moiré energy scale increases the phase space for e-ph scattering. The resistivity is seen to exhibit similar temperature dependence across twist angles above 2.0° , but deviates significantly for the 1.61° system above 100 K, likely due to higher energy electrons participating in conductivity.

Finally, we investigate the respective contributions of intra- and interlayer e-ph coupling to phonon-limited resistivity. To do this, we respectively set all terms in $\hat{V}_{\text{e-ph}}$ relating to intralayer coupling (all electrons and atoms in the same layer) or interlayer coupling (involving electrons or atoms in opposite layer) to zero. The resulting

resistivities, divided by the total, are plotted in Fig. 5.2(b). We see that both intra- and interlayer coupling contribute strongly to resistivity across all twist angles and temperatures studied. In the 13.2° and 3.15° structures, the effect of interlayer coupling increases between 30 and 100 K, owing to the thermal activation of LB modes as discussed in previous work [28]. An increase in the intralayer contribution is also seen around 300 K as optical phonons begin to contribute to resistivity. These trends appear somewhat modified in the 1.61° system, which may have more complicated scattering processes because the moiré bandwidth is comparable in energy to the phonons.

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CONTRIBUTIONS TO COLLABORATIONS

6.1 Overview

In this chapter, I describe my contributions to two experiment-computation collaborations. The first is on a transport study in the wide band gap semiconductor SrSnO_3 , in which the experiments were led by Fengdeng Liu and Zhifei Yang in the group of Bharat Jalan at the University of Minnesota. This work has been published as

- [1] F. Liu, Z. Yang, D. Abramovitch, S. Guo, K. A. Mkhoyan, M. Bernardi, and B. Jalan. “Deep-ultraviolet transparent conducting SrSnO_3 via heterostructure design”. In: *Science Advances* 10.44 (2024), eadq7892. DOI: [10.1126/sciadv.adq7892](https://doi.org/10.1126/sciadv.adq7892). URL: <https://www.science.org/doi/abs/10.1126/sciadv.adq7892>.

The second project is a study on field induced superconductivity in thin film NbO . Here, experiments were led by Sandra Glotzer in the group of Joseph Falson at Caltech, and the computational work is a collaboration between myself and Tommaso Chiarrotti, also in the group of Marco Bernardi at Caltech. This work has not yet been published.

In this chapter, I show mainly my own computational contributions to each study plus some discussion of the context within each study.

6.2 Deep-Ultraviolet Transparent Conducting SrSnO_3 via Heterostructure Design

Ultrawide bandgap semiconductors are important for high-power electronics and deep-ultraviolet optoelectronics. In this study, a thin heterostructure of SrSnO_3 / La:SrSnO_3 / GdScO_3 (110) was grown, facilitating high carrier mobility due to the low electron mass and absence of defects in the top SrSnO_3 (SSO) layer. The heterostructure design allows for separating electrons for La dopants, achieving phonon-limited transport. The heterostructure design also stabilizes the tetragonal structure of SSO via strain. Below, I discuss a computational study of phonon-limited carrier mobility performed in collaboration with the experiment.

6.2.1 First-Principles Calculations

In the undoped SSO layer, ionized impurity scattering should not contribute significantly to mobility, and the mobility is intrinsic. We carry out first-principles calculations of the phonon-limited mobilities [1] with the 3D carrier concentrations ranging from 5×10^{18} to $1 \times 10^{20} \text{ cm}^{-3}$ at temperatures larger than 100 K. A detailed analysis of e-ph interactions in the orthorhombic phase of SSO has been discussed in our previous work [2]. In this study, all films exhibit strain-stabilized tetragonal structure, so we perform calculations in the tetragonal structure. Fig. 6.1(a) shows the phonon dispersion in tetragonal SSO along several high-symmetry lines in the Brillouin zone. The color scale on the curves indicates the e-ph coupling strength between different phonon modes and the electronic state at the conduction band minimum $|g_v(\mathbf{k} = \Gamma, \mathbf{q})|$. Fig. 6.1(b) shows the band structure of the conduction band used in the calculations.

Using these interactions, we compute the phonon-limited mobility, including

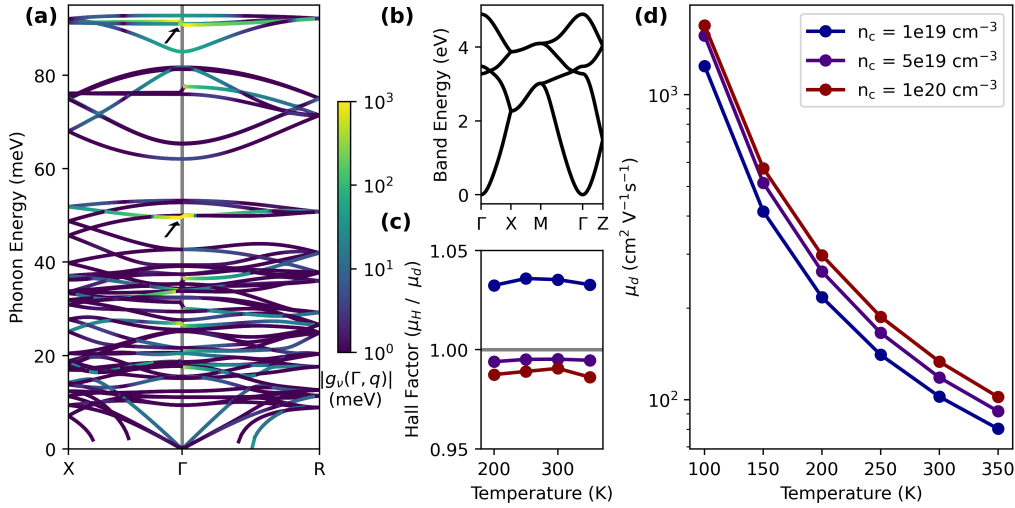


Figure 6.1: First principles calculations of electron-phonon limited carrier mobility in SSO. (a) Phonon dispersion with electron-phonon coupling between phonon modes and the conduction band minimum $|g_v(\mathbf{k} = \Gamma, \mathbf{q})|$ is shown as color. Arrows indicated strongly coupled longitudinal optical phonon modes. (b) Band structure of the conduction band used in calculations. (c) Calculated Hall factor μ_H/μ_d . In all carrier concentrations and temperatures shown, the Hall factor is near 1, so it is reasonable to consider drift mobility as an approximation of Hall mobility. (d) Phonon limited electron drift mobility as a function of temperature and carrier concentration.

scattering from all phonon modes on the same footing. In the calculations, the

carrier density can be modified by tuning the chemical potential. Note here that the calculated mobility is drift mobility. To confirm it is reasonable to compare the calculated mobilities with the experimental values which are Hall mobilities, we also calculate the Hall factor μ_{Hc}/μ_d where μ_{Hc} is the calculated Hall mobility and μ_d is the drift mobility, at selected carrier concentrations $n_{3D} = 1 \times 10^{19}$, 5×10^{19} , and $1 \times 10^{20} \text{ cm}^{-3}$ between 200 and 350 K, shown in Fig. 6.1(c). In the range of carrier concentrations and temperatures shown, the Hall factor is nearly 1, so it is reasonable to consider drift mobility as an approximation of Hall mobility.

Fig. 6.1(d) shows the calculated phonon-limited mobility at a series of carrier concentrations between 100 and 350 K. The calculated phonon-limited mobility increases with carrier concentration and reaches values of $118 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at 300 K and $263 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at 200 K for $n_{3D} = 5 \times 10^{19} \text{ cm}^{-3}$. These values are in very good agreement with the experimental mobility range of 115 to $166 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at 300 K and 260 to $289 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ at 200 K for an electron density of $\sim 5.4 \times 10^{19} \text{ cm}^{-3}$. Below 200 K, e-ph scattering does not appear to be the limited mechanism. However, the relatively good match between the experimental data and the independent theoretical calculations gives provides confidence that the mobility values we extracted truly reflects the mobility of the electron-doped SSO layer, which is mainly limited by e-ph scattering above 200 K and at room temperature due to the absence of charged defects

6.2.2 Computational Methods

We performed ab initio calculations on SSO in the strained tetragonal I4/mcm phase with lattice parameters ($a = b = 5.615 \text{ \AA}$, $c = 8.310 \text{ \AA}$). The in-plane lattice parameters were set from experiment to match the GdScO_3 substrate [3], while the out-of-plane lattice parameter was relaxed to minimize energy. We computed ground state electronic structure with Density Functional Theory (DFT) and lattice dynamics and electron-phonon coupling with Density Functional Perturbation Theory (DFPT) using the Quantum ESPRESSO code [4]. All calculations used the PBEsol-GGA exchange-correlation functional and norm conserving pseudopotentials from PseudoDojo [5] using a plane-wave basis with a 90 Ry kinetic energy cutoff. The DFPT calculations computed e -ph coupling matrix elements on $8 \times 8 \times 6$ electronic $\mathbf{k}$ -point and $4 \times 4 \times 3$ phonon $\mathbf{q}$ -point coarse grids. We used the Perturbo code [1] for transport calculations using interpolations to fine $\mathbf{k}$ - and $\mathbf{q}$ -point grids of $120 \times 120 \times 90$ ($160 \times 160 \times 120$ for Hall mobility) at temperatures above 100 K and $200 \times 200 \times 150$ at 100 K and below. Phonon-limited drift and

Hall mobility was computed by iteratively solving the Boltzmann transport equation with e -ph collisions [6].

We find an imaginary phonon mode in the phonon spectrum of the strained tetragonal SSO structure, which we neglect in transport calculations. This mode corresponds to a dynamic instability in the zero temperature system and may be related to the lack of an explicit GdScO_3 substrate in the calculation or stabilization of the tetragonal phase at finite temperature. Such modes are common in higher symmetry phases of materials with complex structures and lattice dynamics, including many perovskite oxides, and are often neglected in transport calculations due to weak coupling to electrons compared to polar LO modes [7, 8].

6.3 Field Induced Superconductivity in NbO: Paramagnetism and Surface States

In most superconductors, superconductivity is suppressed by magnetic fields, with the superconducting transition temperature T_c decreasing as an external magnetic field is applied. In less common cases, a small magnetic field may promote superconductivity, increasing T_c or even inducing superconductivity in a system which is not superconducting in the absence of a magnetic field. One mechanism for this effect involves the presence of paramagnetic impurities, which exhibit superconductivity suppressing spin fluctuations [9, 10, 11]. In materials with these paramagnetic impurities, an external magnetic field can decrease spin fluctuations and promote superconductivity [12, 13]. A recent example is in thin-film LaSb_2 doped with paramagnetic Ce impurities [14].

This study measured the magnetic field dependence of superconductivity in epitaxially grown thin film NbO [15, 16] of varying thickness. At slab thickness below ~ 10 nm, the T_c was observed to decrease with slab thickness, and at these film thicknesses, magnetic-field promoted superconductivity begins to emerge. This prompted the hypothesis of paramagnetic impurities in the thin film slabs, which increasingly suppress T_c as slab thickness is decreased, an effect which is in turn suppressed by an external magnetic field. However, the nature of these paramagnetic impurities is not known from experiment.

We performed a density functional theory (DFT) computational study of electronic structure and paramagnetism in NbO to investigate which states might give rise to paramagnetic impurities. We first investigate the electronic structure of NbO slab structures stacked along the 111 plane, corresponding to the epitaxial growth in experiment. Fig. 6.2 shows the electronic density of states (DOS) near the Fermi

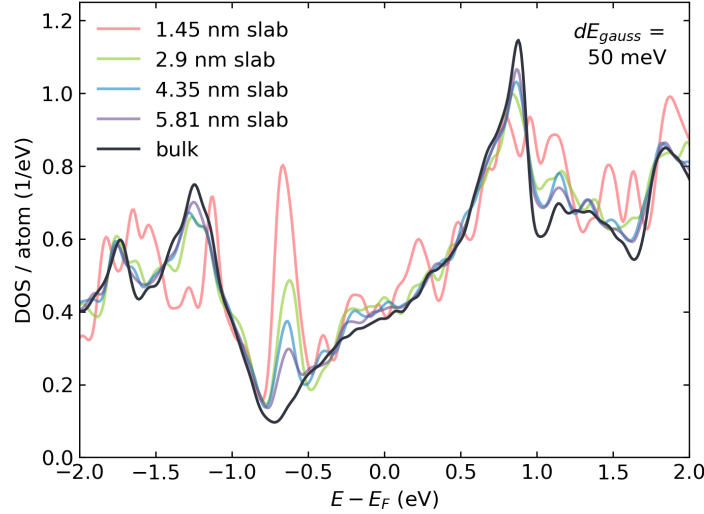


Figure 6.2: Electronic DOS near the Fermi energy in NbO slab structures of varying thickness compared to bulk NbO.

energy in slab structures of varying thickness from 1.45 to 5.81 nm, compared to the DOS of the bulk structure. The DOS quickly converges to that of the bulk by a thickness of around 4 nm, the thickness of the thinnest film considered experimentally (projected DOS calculations show that the peak around -0.7 eV is from surface states). This calculation demonstrates that there is no significant difference in metallicity between the thin films and bulk, and thus the suppression of superconductivity in the films is not due to loss of metallicity.

Next, we investigate the paramagnetic susceptibility different impurity states. After verifying that the structures are non-magnetic in the ground state, we perform spin-polarized DFT calculations with a constrained total magnetization. We then project the wavefunctions onto atomic orbitals to assign spin-resolved occupation and magnetization to each atom in the structure. The atom-resolved magnetization provides information about the relative magnetic susceptibility of each atom.

Fig. 6.3(a) shows the magnetization of Nb atoms on the Nb- and O- terminated surfaces and bulk as a function of total magnetization. The inset shows the relative magnetic susceptibility of the surface atoms and Nb atoms near defects, computed as the ratio of the slope of the magnetization. The most striking result is the high magnetic susceptibility of Nb atoms on the Nb terminated surface. These atoms exhibit 7.2 times the magnetization of bulk Nb atoms (considered all Nb atoms not in the surface layer). In contrast, Nb atoms on the O terminated surface exhibit

essentially the same susceptibility as Nb atoms in the bulk, and Nb atoms at the site of point defects studied exhibit a maximum of twice the susceptibility.

To better understand the electronic structure and magnetization of the surface

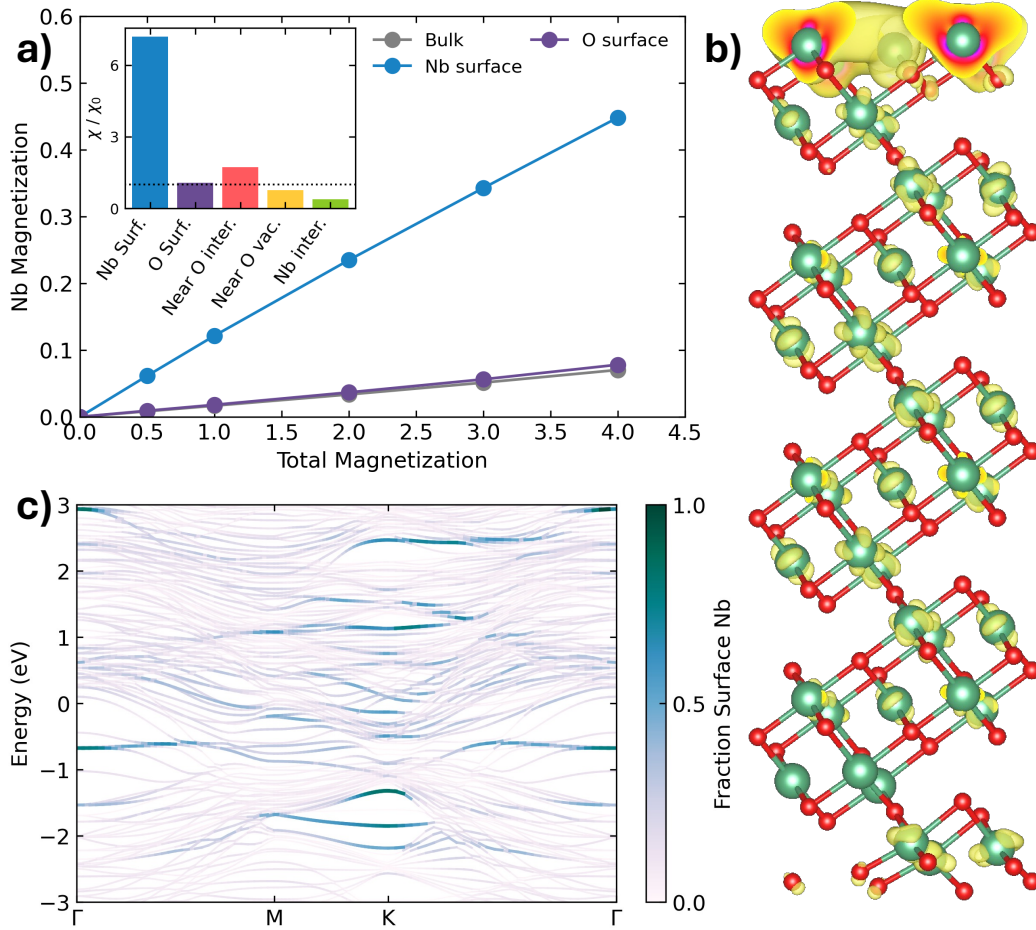


Figure 6.3: (a) Local magnetization of Nb atoms associated with the Nb- and O-terminated slab surfaces as a function of total magnetization. The inset shows the relative magnetic susceptibility of surface and defect Nb atoms relative to bulk Nb, computed from the relative slopes. (b) Band structure of a 2.9 nm slab projected onto the Nb atoms on the Nb terminated slab surface. The weak dispersion of these states indicates localization. (c) Magnetization density for a slab with a constrained total magnetization of 2 bohr. The surface magnetizes much more readily than the bulk.

states, we plot the band structure of the slab across the 2D Brillouin zone and use color to indicate the projection of the bands onto the Nb atoms on the Nb terminated surface (Fig. 6.3(b)). The bands with large projections are seen to have very weak dispersion, indicated localization. We also plot the magnetization density as an isosurface on the structure of the 2.9 nm slab with a total magnetization of 2 bohr

in Fig. 6.3(c). It is clear that the greatest magnetization density is on the Nb atoms on the Nb terminated surface, consistent with projection calculations.

These calculations establish the surface of the NbO slab as the most likely source of paramagnetic impurities. Because the surface accounts for a greater fraction of the atoms in the thinner films, it is a reasonable explanation for the experimental observations. Due to the greater role of the surface, spin-fluctuations increase as film-thickness decreases, which suppresses superconductivity, an effect which can in turn be suppressed by an external magnetic field.

6.3.1 Computational Methods

We use Quantum Espresso for DFT calculations [4] using a PBEsol functional with norm-conserving pseudopotentials. We use a relaxed lattice parameter $a_0 = 4.19$ Å for the primitive cell. We use $3 \times 3 \times 3$ supercells for the structures containing defects and structures stacked along the 111 axis for the slab (which results in a hexagonal unit cell in the xy plane with a lattice parameter of $\sqrt{2}a_0$). We relax atomic positions and lattice parameter for the defect structures, and relax the z coordinate of atomic positions for the slab structures.

For all SCF and NSCF calculations, we use a planewave cutoff of 86 Ry and occupations set via Marzari-Vanderbilt smearing with a width of 0.02 Ry. For lattice relaxation and SCF calculations, we use a $\mathbf{k}$ -grid of $12 \times 12 \times 12$ for bulk NbO, $4 \times 4 \times 4$ for defect supercells, and $9 \times 9 \times 1$ for slab structures. For NSCF and projected DOS calculations, we use Gaussian smearing with a width of 50 meV and $\mathbf{k}$ -grids of $48 \times 48 \times 48$, $10 \times 10 \times 10$, and $24 \times 24 \times 1$, respectively. Magnetization calculations use collinear magnetism and constrain the total magnetization to a fixed value.

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