Weak-coupling analysis of quasiparticle excitations in Sr_{2}RuO_{4} along the Γ-M cut

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Weak-coupling analysis of quasiparticle excitations in Sr_2RuO_4 along the Γ-M cut

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We examine normal state quasiparticle excitations along the Γ-M cut in momentum space for the putative p-wave superconductor Sr_2RuO_4 on the basis of Fluctuation Exchange Approximation calculations. We take as input first-principles derived parameters for the bandstructure and spin-orbit and electron-electron interactions. The numerical results are in excellent agreement with data from photoemission experiments and provide insight into the underlying quasiparticle properties. We find that despite the correlation induced effective mass increase near the Fermi surface, the full β and γ bandwidths are, if anything, increased by correlations. Further, for the γ band we find anomalous lifetime broadening and a significant temperature of variation of unoccupied state quasiparticle energies for temperatures between 25 and 100 K, both of which are accounted for by the momentum dependence of the electron self-energy. In addition to aiding our understanding of experimental data, these results point to the challenge of assigning appropriate Fermi-liquid parameters or momentum-independent self-energies for schemes that require such approximations in order to model Sr_2RuO_4.

I. INTRODUCTION

Nearly twenty years after the discovery of superconductivity in Sr_2RuO_4[1], the development of new as well as the continued enhancement of previously utilized experimental probes and modeling tools have led to an increasingly refined understanding of the normal and superconducting states of this compound. For example, more recent photoemission studies were able to resolve spin-orbit and surface effects[2–5]. At the same time, realistic representations of the Fermi surface using first-principles-based three-band tight-binding Hamiltonians are being used along increasingly sophisticated many-body approximations such as the fluctuation exchange approximation[6], dynamical mean-field theory[7–9] and renormalization group-inspired techniques[10,11] with atomically local electron-electron interaction containing intra- and inter- Coulomb and Hund's rule exchange terms.

Nonetheless, more than ten years after Mackenzie and Maeno's review[12] of an already extensive body of experimental and theoretical work the understanding of the superconducting and normal state properties remains incomplete. In fact, two renormalization-group-based schemes lead to entirely different conclusions with respect to the symmetry of the quasiparticle bands which dominate superconducting pairing[10,11] leading to completely different perspectives in interpreting seminal experimental results. For example, the polar Kerr effect observed for $T < T_c$[13], which suggests a superconducting state with broken time-reversal symmetry, may either be intrinsic to the superconducting state[14,15] or a consequence of impurity effects[16]. At the same time, the failure to observe edge currents[17] that are expected with broken time reversal symmetry for $T < T_c$ is explained as due to the absence of topological protection for such modes[11] or weakness of this protection on account of gap anistropy[10].

Most microscopic modeling of the superconducting state in Sr_2RuO_4 is built upon the assumption that pairing in the superconducting state is driven by electron-electron interactions. While these interactions may be key to understanding superconductivity in Sr_2RuO_4, they impact normal state properties as well. For example, the measured effective masses of Fermi surface quasiparticles are strongly enhanced with respect to density functional theory (DFT) values, presumably due to dynamical correlations that are not accounted for by DFT[18]. Consequently, models for the normal state of Sr_2RuO_4 that accurately reproduce experimental results may impact the understanding of the superconductivity in Sr_2RuO_4 as well.

With that in mind we focus on a many-body analysis of normal state quasiparticle excitations with the goal of understanding the correlation effects that are revealed in photoemission spectroscopy. The many-body analysis used is based on the fluctuation exchange approximation (FLEX) applied to a three-band Hamiltonian constructed using first principles results for the bare band-structure, electron-electron interactions and spin-orbit coupling constant and includes all particle-hole and particle-particle fluctuation diagrams. We focus on the excitations along the Γ-M cut in momentum space, a region that includes the largest observed dynamical mass renormalizations in the vicinity of a low-energy van Hove singularity. We find that we are able to reproduce key features of photoemission results such as bandwidth renormalization, lifetime broadening and the doping dependence of quasiparticle energies. However, our results suggest interpretations of the photoemission data that, in some cases, are at odds with what has been previously proposed. Further, we find significant temperature renormalization of low-energy unoccupied states in the vicinity of the van Hove singularity as well as an appre-

ciable momentum dependence for the electron self-energy for quasiparticle states in this band. While such effects are challenging to extract in photoemission spectroscopy, they may play a key role in the evolution of the superconducting state.

II. MODEL

Our calculation starts with a tight-binding representation of an LDA band-structure for Sr_2RuO_4 as developed by Pavarini and Mazin[19,20]. Low energy electronic states are represented with a basis consisting of a d_{xy}, d_{xz} and d_{yz} orbitals for each Ru ion a square lattice of side $a = 3.87$Å. In undoped Sr_2RuO_4 these orbitals are 2/3 filled on average. This three-orbital basis set describes well the three quasiparticle bands that cross the Fermi surface and are designated as α, β and γ[21,22]. In our calculations weak interplanar couplings corresponding to the z direction are ignored. For the Ru atomic spin-orbit interaction, $\lambda\ \vec{s}\cdot\vec{l}$, we use the first-principles derived value of $\lambda = 93$ meV[23] although larger values have been proposed[24].

Correlation effects on quasiparticle quantities are characterized through the electron self-energy, $\Sigma_{\nu\sigma;\nu'\sigma'}(\mathbf{k}, E)$ where ν and σ are orbital and spin indices respectively. These dynamical correlations are modeled on the basis of an atomically local bare interaction, $\Gamma^{(0),\mathrm{cRPA}}$, consisting of band-dependent intraorbital ($U_{ii} \sim 2.6$ eV) and interorbital Coulomb ($U'_{i\neq j} \sim 1.9$ eV) terms and exchange interaction ($J_{i\neq j} \sim 0.25$ eV) determined through constrained random phase approximation calculation[25]. The layered Sr_2RuO_4 crystal structure lifts the three-fold degeneracy maintained by the d_{xy}, d_{xz} and d_{yz} in cubic symmetry which is reflected in the bare interaction. For example, $U_{xy,xy} = 2.72$ eV $\neq U_{xz,xz} = 2.48$ eV. These first-principles derived interactions along with the DFT bandstructure suggest that Sr_2RuO_4 is in the intermediate coupling regime, i.e. $U \lesssim W_0$ where W_0 is the unrenormalized bandwidth. Consequently, it is possible that weak-coupling schemes such as low-order perturbation theory[26,27] and self-consistent perturbation theory[28,29] might capture electron-electron correlation effects, a possibility that is supported by what appears to be Fermi-liquid-like properties at low temperature, albeit with effective masses that are strongly renormalized in comparison to DFT values. Energy renormalization and lifetime broadening of quasiparticle excitations are calculated via the quasiparticle equation

$$\sum_{\nu'\sigma'}\left(H^{(0)}_{\nu\sigma;\nu'\sigma'}(\vec{k}) + \Sigma_{\nu\sigma;\nu'\sigma'}(\mathbf{k}, E_{qp})\right)\psi_{\nu'\sigma'}(\mathbf{k}, E_{qp}) = (E_{qp} + i\Gamma_{qp})\,\psi_{\nu\sigma}(\mathbf{k}, E_{qp}) \tag{1}$$

As FLEX has been demonstrated to overestimate the self-energy, at least on the basis of one-band model calculations in the $d \to \infty$ limit[30], we apply a single scale factor to the cRPA vertex, *i.e* $\Gamma^{(0),FLEX} = g_{FLEX}\Gamma^{(0),cRPA}$. Thus, $g_{FLEX} < 1$ becomes the single adjustable parameter in these calculation and represents the effect of missing vertex corrections in the self-energy. Since all elements of the bare vertex are scaled by the same factor, the bare vertex retains its symmetries and the conserving properties of FLEX are preserved[31]. This highly simplified treatment of screening necessarily neglects physical processes that are incorporated through momentum- and energy-dependent vertex corrections and so, consequently, we do not produce features that are normally not observed in FLEX[32]. Further, we will assume the same scale factor as a function of doping even though we might anticipate screening changes as portions of the Fermi surface pass through a van Hove singularity. Although these are limiting assumptions, they allow us to assess the extent to which experimental results can be captured by a weak to intermediate coupling theory with a minimal set of input parameters. Meanwhile improvements beyond these results might be achieved using other versions of the FLEX or related algorithms[32–34]. We set g_{FLEX} so that the FLEX calculation of the quasiparticle linewidth along the symmetry cut Γ-M is comparable in scale to experimental results[35] for binding energies between 0 and 60 meV. In doing this we obtain $g_{FLEX} \simeq 2/3$ which we will use in all calculations which follow.

To make the scheme computationally feasible, the dynamical cluster approximation (DCA)[36] is used to reduce dynamical correlations to a range of $L_c << L$ where L is the physical lattice dimension and we numerically demonstrate that our results well-describe the large L_c limit. The results shown here are for $L = 256a$ and $L_c = 16a$. The DCA allows us to relax the assumption that the electron self-energy is atomically local/momentum independent. Indeed, we will demonstrate significant momentum dependence of the electron self-energy in the vicinity of a van Hove singularity in the γ-band.

III. BANDWIDTH-WIDE POLARIZED PHOTOEMISSION

Iwasawa, *et al.*[3] utilized angle-resolved photoemission spectroscopy (ARPES) to deduce the nature of occupied states along the Γ-M cut for energies that range from 2.5 eV below up to E_F. DFT calculations sug-

gest this range is sufficient for capturing the β and γ band minima[19]. Photon polarization enabled separation of the β and γ bands which are dominated by d_{xz} and d_{xy} orbitals respectively along the Γ-M cut. Our FLEX calculations of the orbital, ν, projected spectral function from the orbital and spin, σ, diagonal components of the Green's function, $A_\nu(\mathbf{k}, E) = (-1/\pi)\mathrm{Im}G_{\nu\sigma;\nu\sigma}(\mathbf{k}, E)$, combined with the Fermi occupancy function allows for a comparison to the intensity maps produced by Iwasawa, *et al.*. However, photon-energy-dependent final state effects are not accounted for in our FLEX calculations and, consequently, caution must be taken in relating the experimental and model results.

Our results are shown in Fig. 1. For ease of comparison with experimental results, we use color maps that are similar to those used by Iwasawa, *et al.*. Results for p-polarization, which projects d_{xz} orbitals and are dominated by the β-band, are shown in the top of Fig. 1 and results for s-polarization, which projects d_{xy} orbitals and are dominated by the γ-band, are shown in the bottom of Fig. 1. Unrenormalized quasiparticle excitation energies from DFT are shown as filled circles and renormalized FLEX quasiparticles are displayed as open circles.

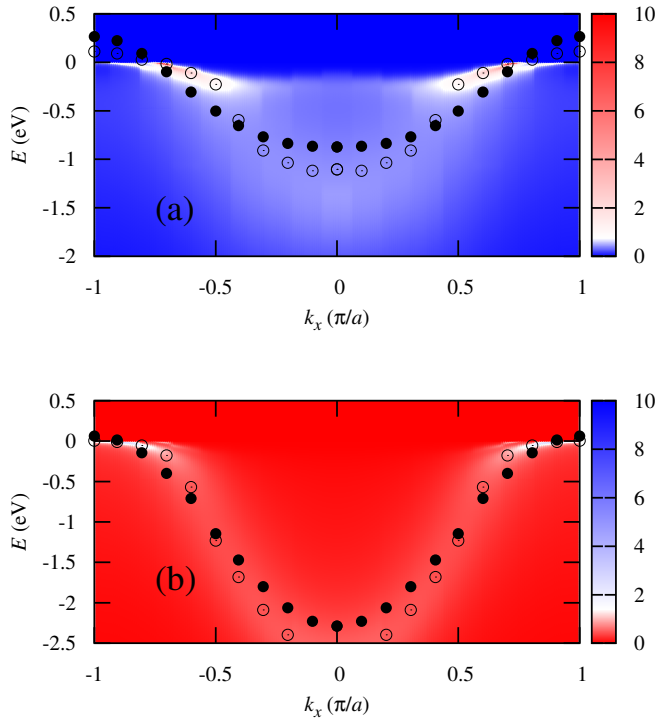

FIG. 1. (Color online) Calculated momentum and energy-dependent photoemission spectral intensities, $A_\nu(\vec{k}, E)$, for $\mathbf{k} = (k_x, k_y = 0)$ at $T = 25K$ for (a) β-band dominant $\nu = d_{xz}$ orbitals and (b) γ-band dominant $\nu = d_{xy}$ orbitals.

First, we focus on the β-band results in the top of Fig. 1. As expected, in the vicinity of the Fermi level we find that the slope, dE/dk_x, of the quasiparticle excitations is reduced for the FLEX results in comparison to DFT, a reflection of dynamical enhancement of the effective mass at the Fermi surface. However, the overall bandwidth of the renormalized band is, if anything, a bit larger than that for DFT. If the overall bandwidth is larger, but dE/dk_x is reduced near E_F, then a region where dE/dk_x is enhanced must appear as well. In our calculations, this enhanced slope appears in the form of a "kink" at approximately 0.5 eV below the Fermi level for the β band.

The calculated intensity map is in good agreement with that obtained by Iwasawa, *et al.* when using a 62 eV photon source. In the 62 eV results, the intensity map is essentially featureless for energies that are more than 0.4 eV below the Fermi level. We see similar behavior in the FLEX results. However, we do computationally identify a quasiparticle band at much lower energies using the quasiparticle equation and we attribute the lack of structure in this energy region as being due to large lifetime broadening.

The experimental 88 eV photon energy results show an enhanced intensity for low momentum values at about 0.4 eV below the Fermi level. Iwasawa, *et al.* interpret this feature as being the highly-renoramlized, substantially flattened β-band minimum. This would represent a nearly 50% reduction to the energy of the band minimum as obtained by DFT and is in contradiction to our results which shows that minimum being pushed to higher binding energy. We note that the FLEX results do display significant intensity near 0.4 eV due to incoherent features of the spectral functions. For low momentum values a very weak peak appears in the incoherent intensity at approximately 0.3 eV below E_F. We suggest that the 0.4 eV feature observed in photoemission may not represent the actual β-band minimum. Possible explanations for its appearance include the enhancement of moderate incoherent spectral weight by final state effects or the introduction of folded bands due to surface reconstruction[5,37,38].

The FLEX-based calculated intensity map for the d_{xy} orbitals, shown in the top of Fig. 1 and which is dominated by the γ band, agrees well with the experimental result. As we observed with the β band, the renormalized quasiparticle band is significantly flattened in the vicinity of the Fermi surface in comparison to the DFT band reflecting large dynamical contributions to the effective mass. Nonetheless, we once again find that the overall bandwidth is actually increased with respect to the DFT result. In this case, this completely agrees with the data analysis of Iwasawa, *et al.* who put the band minimum at a binding energy greater than 2.5 eV which is below the DFT value of approximately 2.3 eV.

Thus, we find qualitatively similar behavior for the predominantly d_{xz}-derived β band that is probed with p-polarized light and for the predominantly d_{xy}-derived γ band that is probed with s-polarized light. Namely, we find the expected dynamically induced flattening of the bands near the Fermi level, *i.e.* effective mass in-

crease, but a deepening of the band minima with respect to DFT values. Such overall structures necessarily require a steepening of the dispersion, *i.e* kinks, at some intermediate binding energy. Quasiparticle broadening is significant at large binding energies and our calculated results for the intensity maps suggests that extraction of the experimental quasiparticle dispersion can be challenging as non-intrinsic or incoherent features in the quasiparticle spectrum may appear to be larger in intensity than the broadened quasiparticle excitations at higher binding energy.

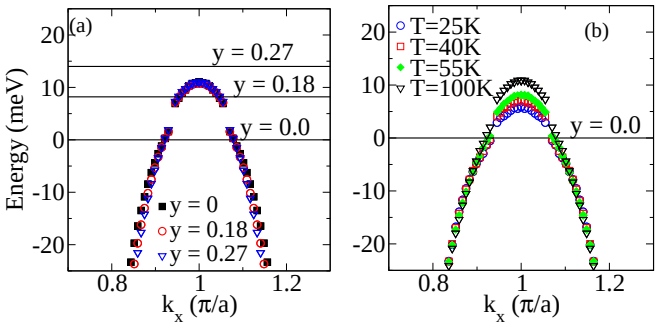

FIG. 2. (Color online) Quasiparticle dispersion as a function of doping (a) and temperature (b). The doping dependence of the dispersion is largely accounted for by a shift in the Fermi level, E_F. The energy of the van Hove singularity and the Fermi surface effective mass have significant temperature dependence even though a similar dependence is not apparent for the states below E_F.

IV. DOPING DEPENDENCE OF γ-BAND QUASIPARTICLE EXCITATION ENERGIES NEAR E_F

Shen, *et al.*[2] used ARPES to examine the γ-band quasiparticles, focusing on states in a much narrower energy window about E_F. Through thermal occupation (at T=100K) and doping, normally unoccupied $E > E_F$ quasiparticle states were resolved to energies up to about 20 meV above E_F, which encompasses a low-energy van Hove singularity. The γ-band effective mass is dramatically renormalized at E_F along this momentum cut. Thus, these quasiparticle excitations are particularly good targets for understanding the dynamical correlations in this system.

In the left panel of Fig. 2 we show the FLEX results for the dispersion of γ-band quasiparticles for doping values of $y = 0$, $y = 0.18$ and $y = 0.27$ doped electrons per unit cell. Just as was observed experimentally by Shen, *et al.*, we find that once we account for a shift in E_F the energy vs. momentum dependence of these highly renormalized states is essentially unaffected by doping even as the E_F passes through the van Hove singularity. As shown in the figure, we are able to make the dispersion curves collapse by moving the $y = 0.18$ and $y = 0.27$ Fermi levels to about 8 and 14 meV above the $y = 0$ Fermi level in the plot, analogous to what was done by Shen, *et al.* although in their case they observed Fermi level shifts of about 13 and 19 meV for these doping values.

This result suggests that the correlations that drive the large mass enhancement observed for this band are largely unaffected by the occupancy of the band itself. That is, it seems likely that γ-band excitations are strongly renormalized by interband correlations as the α and β bands do not undergo substantial changes (such as passing through a van Hove singularity) in this doping range. More specifically, strong spin fluctuations observed near $q = (2\pi/3a, 2\pi/3a)$[39,40] in the undoped compound, which are believed to originate from the α and β bands, provides a strong collective mode that drives the strong renormalizations observed in the γ band along the Γ-M cut.

While the nominally $E > E_F$ states are only accessible in photoemission through doping or through thermal occupancy with $k_BT \simeq E - E_F$, FLEX can be used to model the evolution of these states as temperature is decreased. Experimentally, it was observed that quasiparticle dispersions for $E < E_F$ in the undoped compound are nearly temperature independent, suggesting that the same would likely be found for the $E > E_F$ if it were possible to observe these states directly at low temperature. However, our FLEX results, shown on the right side of Figure 2 indicate the potential for a significant temperature dependence for these states. Namely, as temperature is lowered from 100 K to 25 K, the energy of the band maximum shifts downward from 11 meV to approximately 5.5 meV above E_F. Meanwhile, consistent with experiment, the occupied state dispersion has the appearance of being largely temperature independent over the displayed energy range. This behavior is potentially significant as the lowering in energy of the $E > E_F$ states for the γ-band along this cut might allow for greater condensation energy gains for pairing of these states than might be expected from a model that presumes temperature-independent quasiparticle excitations for the γ-band. Also, it was recently observed that Sr_2RuO_4 exhibits a peak in T_c as a function of strain along the [100] direction[41] and it was proposed that the peak may coincide with strain dependent van Hove singularities passing through E_F making it important to understand the low temperature behavior of these singularities.

V. QUASIPARTICLE LINEWIDTHS

In Fig. 3 we show our FLEX results for low-energy quasiparticle linewidths along the Γ-M cut taking into account their band, energy and temperature dependencies. The displayed results are in excellent qualitative agreement with the experiments of Kidd, *et al.*,[35] captur-

ing the significant differences between the β and γ band lifetimes. Excellent quantitative agreement is achieved by adding band-dependent, but energy and temperature independent shifts of approximately $\Gamma_{\gamma,imp} \sim 0.016$eV and $\Gamma_{\beta,imp} \sim 0.044$eV to the FLEX results to account for impurity scattering effects not directly included in the FLEX calculations. To illustrate this agreement, we note that our calculated results of $\Gamma_\gamma(E = 0.06\text{ eV}, T = 25\text{ K}) - \Gamma_\gamma(E = 0, T = 25\text{ K}) \simeq 0.09$ eV, $\Gamma_\beta(E = 0.06\text{ eV}, T = 25\text{ K}) - \Gamma_\beta(E = 0, T = 25\text{ K}) \simeq 0.05$ eV and $\Gamma_\gamma(E = 0\text{ eV}, T = 55\text{ K}) - \Gamma_\gamma(E = 0, T = 25\text{ K}) \simeq 0.015$ eV are in excellent agreement with their experimental counterparts. Konik and Rice[27] were also able to obtain good agreement using second-order perturbation theory, although their calculated lifetimes somewhat underestimated their increase as a function of energy in co

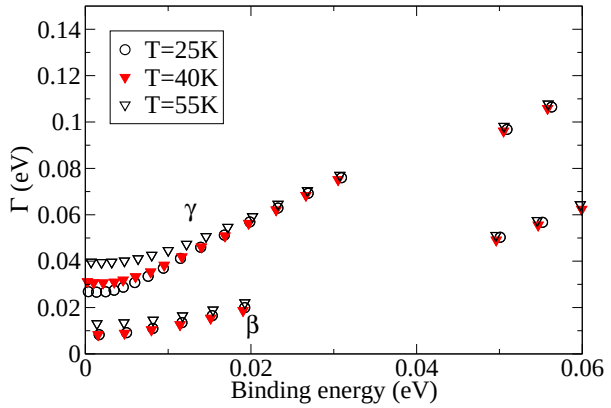

FIG. 3. (Color online) FLEX results for the energy dependence of the the β and γ band lifetimes at three different temperatures. Scattering is significantly larger for the γ band which also displays a stronger temperature dependence near $E = 0$.

Perhaps the most striking feature in the FLEX results, which might not be immediately apparent from the experimental data, is the large intrinsic linewidth for the γ-band at low energies. Indeed, we have $\Gamma_\gamma(E = 0, T = 25\text{K}) \simeq 0.025\text{ eV} \gg k_B T \simeq 0.002$eV. Thus, it is questionable as to whether the γ-band excitations along this cut in momentum space should be considered to be Fermi liquid-like quasiparticle excitations, at least for the temperatures considered here. It should be noted that the Fermi liquid regime only extends up to 25 K[42]. The linewidth observed for the β band is appreciably smaller as is the linewidth for the γ band itself for points on the Fermi surface that do not lie along or near the Γ-M and symmetry equivalent points (results are not shown here). As relatively large dynamical enhancements are observed in the effective mass of the γ band[18], it is perhaps not surprising to find relatively large lifetime broadening in the γ band as well.

The results of this and the previous section point to strong renormalizations and significant temperature variation for the γ band even below 100 K. Consequently, it may be challenging to assess the relative stability of superconducting states that are dominated by pairing of these quasiparticles vs. pairing involving other bands or regions of momentum space which exhibit more typical Fermi liquid behavior.

VI. MOMENTUM DEPENDENCE OF THE SELF-ENERGY

While FLEX is not expected to accurately represent effects due to strong couplings, such as a Mott transition, it has the advantage of enabling a numerical assessment of the momentum dependence of the electron self-energy. Such a momentum dependence might be anticipated in a two-dimensional system where electrons couple to strong collective fluctuations at low temperature. Dynamical mean-field theory (DMFT) has proven to be a powerful tool for modeling correlation effects in Sr_2RuO_4[7] and a host of other materials[43]. Nonetheless, the momentum dependence of the self-energy is ignored in DMFT and so it is important to determine if and what manner the assumption of momentum independence is limiting.

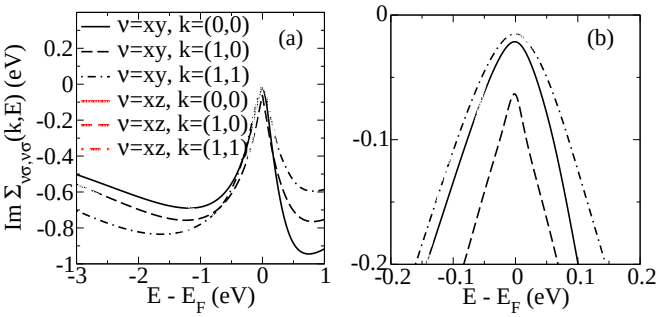

FIG. 4. (Color online) Imaginary part of the orbital- and spin-diagonal parts of the self energy, $\text{Im}\,\Sigma_{\nu\sigma;\nu\sigma}(\mathbf{k}, E)$, for the $\nu = d_{xy}$ and d_{xz} orbitals and three different momentum values (momentum given in units of π/a. In (a) the energy range spans that bandwidth and in (b) we zoom in on energies near E_F. The d_{xy} orbitals, which dominate the γ band, have anomalous behavior for momentum values near the M point, $\mathbf{k} = (\pi/a, \pi/a)$.

In Fig. 4 we display the energy dependence of the orbital- and spin-diagonal ($\nu = \nu', \sigma = \sigma'$) parts of the imaginary part of the electron self-energy, $\text{Im}\,\Sigma_{\nu\sigma;\nu'\sigma'}(\mathbf{k}, E)$, for the d_{xy} and d_{xz} orbitals at the Γ, M and X points in momentum space. In the left pane we show the energy dependence over an energy range that covers the full bandwidth while in the right pane we focus on the behavior in the vicinity of the Fermi surface. While there is both substantial orbital and momentum dependence over the scale of the bandwidth, it appears that curves coalesce in the vicinity of the Fermi surface, suggesting the potential momentum dependence of the

self-energy is not as substantial for the most important states of the system.

However, the right panel shows that while the self-energy is for the most part nearly momentum independent near E_F, there is a significant exception. Namely, we find that a strong momentum dependence develops for the d_{xy} orbital part of the self-energy, which dominates the γ-band self-energy, in the vicinity of M point. Here we find that the imaginary part of the self-energy is increased by a factor of three in comparison to the results for other orbitals (bands) or for d_{xy} orbitals at other points in the the Brillouin zone. This again illustrates the unusual behavior associated with the γ-band excitations along the Γ-M cut and shows that this behavior is described through the momentum dependence of the self-energy.

VII. SUMMARY AND FUTURE DIRECTIONS

We have presented Fluctuation Exchanged Approximation (FLEX) based calculations for the quasiparticle excitations along the Γ-M cut in momentum space for Sr_2RuO_4. The results are in excellent agreement with several recent experiments. Key aspects of our analysis include the following. First, the full bandwidth of for the β and γ band excitations are, if anything, slightly increased by dynamical correlation even though both bands are flattened (i.e. effective mass increased) near E_F. Second, while the γ-band near E_F is essentially rigidly displaced even as doping pushes the Fermi level through the van Hove singularity in the band, we do find strong temperature renormalizatons for the nominally unoccupied states lying between E_F and the van Hove singularity as the temperature is lowered below 100 K. Third, the intrinsic linewidths of γ-band quasiparticles are anomalously large for temperatures above the Fermi liquid regime, a possibility that may be obscured experimentally by uncertainty in impurity scattering contributions. Finally, the unusual behavior of the γ band excitations along the Γ-M cut is accounted for by the momentum dependence of the self-energy, specifically in the vicinity of the van Hove singularity in this band. Thus, assessing the relative stability of pairing states as a function of the bands and momenta of the electron pairs may be problematic if the momentum of the electron self-energy is assumed to be negligible.

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