Perturbation theory analysis of the strain-dependent superconducting phase diagram for Sr_2RuO_4

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Previously, it was shown that a superconducting state dominated by singlet $d_{x^2-y^2}$ intraband pairs emerges from the fluctuation exchange approximation (FLEX) applied to a realistic model for Sr_2RuO_4, a result that is increasingly aligned with experimental data. Here we apply FLEX to model the strain-dependent phase diagram of Sr_2RuO_4 and show that we are able to reproduce its unusual features. This adds weight to the argument that a predominantly $d_{x^2-y^2}$ singlet pairing state represents a reasonable starting point for describing the superconducting properties of Sr_2RuO_4.

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I. INTRODUCTION

Thirty years since the discovery of superconductivity in strontium ruthenate [1], Sr_2RuO_4, a microscopic theory that convincingly explains a range of experimental results for this material remains elusive. The possibility of spin-triplet pairing was immediately recognized on account of the observed enhancement ferromagnetic correlations in the normal state [2] and the apparent absence of Knight shifts in NMR as temperature passes through the superconducting transition temperature [3]. Additionally, muon spin rotation [4] and Kerr effect [5] experiments provided evidence of time-reversal symmetry breaking in the superconducting state. These, and other confirming results, are consistently explained with a triplet pairing state having a $p_x + ip_y$ orbital structure. The possibility of such a pairing state has been supported by several model calculations using realistic Hamiltonian for Sr_2RuO_4 and various approximate treatments of the electron-electron interactions [6–9].

However, other experimental data appears to be inconsistent with $p_x + ip_y$ spin-triplet pairing. For example, the specific heat for temperatures below T_c is suggestive of the existence of line nodes for the superconducting gap function on the Fermi surface [10]. Unexpectedly, reexamination of the Knight shift demonstrated the suppression of electron polarizability below T_c, in line with expectations for a spin-singlet superconductor [11]. Further, the application of strain along the [1,0,0] crystalline axis, ε_{xx}, fails to produce a split in the superconducting transition that would be expected for $p_x + ip_y$ pairing [12]. Indeed, the dramatic superconducting phase diagram for Sr_2RuO_4 under strain provides an excellent target to enable microscopic models to finally provide clarity on the underlying pairing state for Sr_2RuO_4 [13,14].

In this manuscript, we describe model results for the strain dependent phase diagram for Sr_2RuO_4 obtained using a realistic microscopic Hamiltonian and correlations approximated using the fluctuation exchange approximation (FLEX). Generically, we find that FLEX is able to reproduce two key results of the strain-dependent phase diagram: (1) there is no splitting of the superconducting transition as they symmetry between the x and y crystal axes is lifted and (2) T_c plummets rapidly just after the strain exceeds the critical value where a van Hove singularity passes through the Fermi level, $\varepsilon_{xx} = \varepsilon_{\rm vH}$. Additionally, when the coupling strength is manually adjusted so that the calculated unstrained T_c approaches that for experiment, FLEX accurately reproduces the strain-induced peak structure in T_c. Thus, we argue, the pairing symmetry generated by FLEX represents a strong candidate for describing the dominant pairing correlations in the superconducting state of Sr_2RuO_4.

II. MODEL AND NUMERICAL METHODS

We use a three atomic orbitals per unit cell basis, corresponding to the $4d_{xy}$, $4d_{xz}$ and $4d_{yz}$ orbitals of the ruthenium atoms, to account for the three distinct Fermi surface sheets (denoted α, β and γ) observed experimentally [15,16]. Tight-binding hopping matrix elements, $t_{\nu,\nu'}(\mathbf{R})$, where ν and ν' are orbital indices, are taken from Pavarini and Mazin's [17,18] fit to the density functional theory band structure for the baseline unstrained case. An atomically local spin-orbit interaction, $\lambda\,\vec{s}\cdot\vec{l}$, is assumed and we use the first-principles derived value of $\lambda = 93$ meV [19]. The chemical potential is adjusted to maintain an average filling of $2/3$. Finally, the relatively small interplanar hopping terms are ignored consistent with the quasi two-dimensional behavior of Sr_2RuO_4.

Electron correlations are modeled starting with an atomically local electron-electron interaction vertex, $\Gamma^{(0),\rm cRPA}$, parameterized with band-dependent intraorbital (U_ν) and interorbital ($U'_{\nu\neq\nu'}$) Coulomb and exchange ($J_{\nu\neq\nu'}$) interaction terms evaluated for Sr_2RuO_4 using the constrained random phase approximation calculation [20]. The largest interaction parameters, i.e., $U_{xy} = 2.72$ eV and $U_{xz} = 2.48$ eV, are comparable in size to the unrenormalized bandwidth suggesting that Sr_2RuO_4 is in the intermediate-coupling regime. Energy renormalization and lifetime broadening of quasiparticle excitations are calculated via the quasiparticle equation

$$\sum_{\nu'\sigma'}\left(H^{(0)}_{\nu\sigma;\nu'\sigma'}(\mathbf{k}) + \Sigma_{\nu\sigma;\nu'\sigma'}(\mathbf{k}, E_{\rm qp} + i0^+)\right)\psi_{\nu'\sigma'}(\mathbf{k}, E_{\rm qp}) = (E_{\rm qp} + i\Gamma_{\rm qp})\,\psi_{\nu\sigma}(\mathbf{k}, E_{\rm qp}), \quad (1)$$

where $\Sigma_{\nu\sigma;\nu'\sigma'}(\mathbf{k}, E)$ is the electron self-energy and ν and σ are orbital and spin indices, respectively.

The self-energy is approximated using the self-consistent fluctuation exchange approximation (FLEX) develop by Bickers, White, and Scalapino [21] supplemented by the dynamic cluster approximation (DCA) [22] and generalized to explore the superconducting state below T_c [23]. We use a 256×256 unit cell/momentum space with a 4×4 DCA coarse-graining of the self-energy to keep calculations computationally feasible while preserving the momentum dependence that is needed to describe correlation induced features that are unique for excitations near the van Hove singularity [24]. As FLEX tends to overestimate the magnitude of the self-energy [25], we introduce a single scale factor for the cRPA interaction vertex, i.e., $\Gamma^{(0),\mathrm{FLEX}} = g_o\,\Gamma^{(0),\mathrm{cRPA}}$ with $g_o < 1$.

Strain along the [1,0,0] direction drives a van Hove singularity in the γ-band from its unstrained energy of $E_{vH} \sim 20$ meV above the Fermi level, through and below the Fermi level. This evolution of a high density-of-states feature in the bandstructure is the likely source of the dramatic concurrent variation in T_c [13,14]. We use a dimensionless parameter, η, to simply represent the strain-driven changes in the tight-binding parameters most relevant for this process. We have

$$\tilde{t}_{xy,xy}(\pm 1, 0) = (1 - \eta)\, t_{xy,xy}(\pm 1, 0), \tag{2}$$

$$\tilde{t}_{xy,xy}(0, \pm 1) = (1 + \eta)\, t_{xy,xy}(0, \pm 1), \tag{3}$$

where $\tilde{t}$ and t are the strained and unstrained hopping values, respectively. Effectively, this is consistent with tensile strain along the [1,0,0] direction As η becomes sufficiently large, the van Hove singularity at $\mathbf{k} = (0, \pm\pi)$ passes through the Fermi level at some η_{vH} and will be pushed increasingly below the Fermi level for $\eta > \eta_{\mathrm{vH}}$. Assuming that tight-binding parameters vary linearly with strain, then our model parameter should track experiment via $|\eta|/\eta_{\mathrm{vH}} = |\varepsilon_{xx}|/\varepsilon_{\mathrm{vH}}$.

III. RESULTS WITHOUT STRAIN

When FLEX is applied to this model of Sr_2RuO_4 in the unstrained limit, the pairing symmetry that results [26] *is not* chiral, spin-triplet p-wave. In the absence of spin-orbit coupling ($\lambda = 0$) FLEX generates a superconducting state consisting of singlet $d_{x^2-y^2}$ pairs dominated by intraband pairing of quasiparticles in the quasi-two-dimensional γ-band. When the spin-orbit interaction is included ($\lambda = 93$ meV), the pairing state acquires triplet components representing interorbital pairing across all three bands. However, these triplet-pairing contributions remain small in comparison to the dominant singlet $d_{x^2-y^2}$ pairing terms.

The quasiparticle excitations at and near the van Hove singularity in the γ-band clearly appear to be essential drivers of the strain dependence of T_c for Sr_2RuO_4 and, perhaps, for superconductivity in the material more generally. A nontrivial representation of these quasiparticles is a strength of the FLEX method. Indeed, FLEX results for these key γ-band excitations [24], obtained with a coupling strength of $g_o = 0.67$, are in excellent quantitative agreement with experimental results for the temperature and frequency dependencies of quasiparticle lifetimes [27]. Further, FLEX generates a downward shift of the van Hove energy from its DFT value of 90 meV to approximately 20 meV at $T = 100$ K, in good agreement with the experimental result of 14 meV [28]. Although FLEX results for quasiparticle excitations demonstrate Fermi-liquid like behavior for *nearly all* of the Fermi surface, there are hints of non-Ferm liquid behavior in the γ-band near the van Hove singularity, a possibility that would be consistent with recent experimental results for the Seebeck coefficient [29].

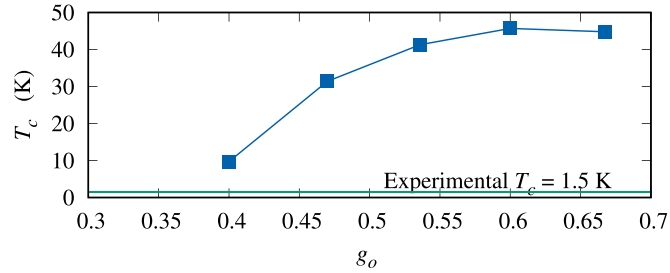

FIG. 1. Calculated FLEX results for T_c vs dimensionless coupling strength parameter, g_o. While experimental quasiparticle renormalizations are most accurately represented with $g_o \sim 0.67$, a g_o value near 0.4 is more consistent with the observed T_c for Sr_2RuO_4.

In Fig. 1, we show FLEX results for T_c versus coupling strength g_o. The coupling strength most consistent with describing normal state quasiparticle excitatons, $g_o = 0.67$, produces a T_c near 43 K, clearly much too high in comparison to the experimental value of approximately 1.5 K. This discrepancy is expected as some of the higher-order processes that are neglected in the FLEX approximation, such as fluctuations of a d-wave superconducting order parameter, can be numerically insignificant for normal state quasiparticles, but essential for calculating the superconducting T_c. The FLEX results for the unstrained T_c is in alignment with experiment when $g_o \simeq 0.4$. Indeed, we will find that FLEX generates a strain-dependent phase diagram that accurately mimics experimental results when this lower coupling strength is used and strain is varied.

IV. RESULTS WITH STRAIN

In the absence of strain, the van Hove singularity in the γ band appears at symmetry equivalent $\mathbf{k}$ points $M_x = (\pm\pi, 0)$ and $M_y = (0, \pm\pi)$. The dispersion near these points along the Γ-M cut is displayed in Fig. 2 for the unstrained, $\eta = 0$ (open symbols), and strained cases, $\eta = 0.002$ (closed symbols), at a coupling strength of $g_o = 0.67$. The energy at the van Hove singularity ($k/\pi = 1.0$) is 20 meV in the unstrained case. Finite strain splits the Γ-M_x and Γ-M_y dispersion curve with the M_x van Hove energy lowered to 10 meV and the M_y van Hove energy increased to 40 meV. The strain-lowered van Hove energy at M_x is key as it passes through the Fermi level, $E_F = 0$, with sufficient tensile strain along [1,0,0].

In Fig. 3(a), we show the M_x quasiparticle energy as a function of strain, η, at different coupling strengths. For each coupling strength, a critical value of strain, η_{vH} is identified where the van Hove energy crosses the Fermi level. Since the γ-band is increasingly flattend and Hove energy is driven

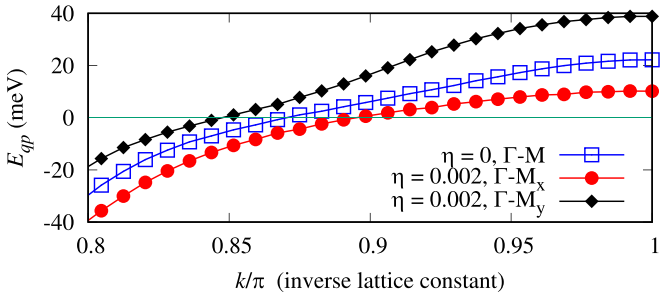

FIG. 2. Quasiparticle energies $E_{\rm qp}$ vs momentum k/π along the cuts Γ-M_x and Γ-M_y. In the unstrained case, $\eta = 0$, these cuts are degenerate, i.e., $x = y$. In the strained case, $\eta = 0.002$, excitations along the x (y) are lower (higher) in energy in comparison the unstrained case, with the van Hove singularity at M_x driven toward the Fermi level where $E_{\rm qp} = 0$.

downward with an increase in the coupling strength, $\eta_{\rm vH}$ decreases as the coupling strength increases. Figure 3(b) shows a scaled version of the same plot. Here $E_{\rm vH}(\eta)$ is divided by $E_{\rm vH}(\eta = 0)$ and η is expressed in terms of $\eta_{\rm vH}$; the plots then become similar for the coupling strengths considered here.

Superconducting properties of this model are described in FLEX via the equal-time anomolous Green's function:

$$< c_{\nu'\sigma'}(\mathbf{r} = 0)c_{\nu\sigma}(\mathbf{r}) > \equiv m_p(T)\,\psi_{\nu\sigma;\nu'\sigma'}(\mathbf{r}). \qquad (4)$$

Here ψ is the normalized pair wave function and $m_p(T)$ is the pair amplitude which becomes finite below T_c. As described in a previous work [26], in the limit of zero strain the FLEX wave function for this model describes a superconducting state dominated by singlet, $d_{x^2-y^2}$ pairing of γ-band quasiparticles with minority triplet components induced by spin-orbit coupling, albeit in a manner that does not account for the breaking of time-reversal symmetry that is observed experimentally.

In Fig. 4, we show m_p vs T curves for several different strain values at a coupling strength $g_o = 0.40$. The impact of strain on these curves is evident with the largest pair amplitudes and T_c occurring at the critical strain value of

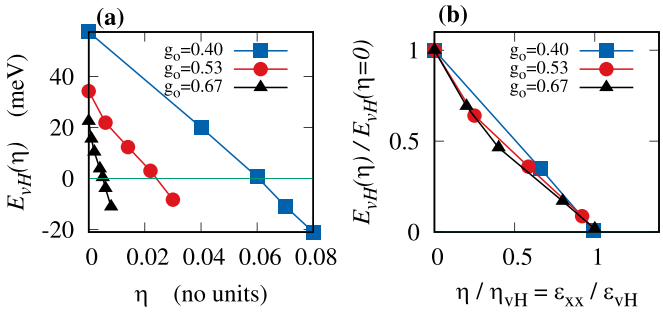

FIG. 3. (a) Quasiparticle energy at the van Hove singularity $E_{\rm vH}$ vs strain parameter η at various coupling strengths g_o, and (b) $E_{\rm vH}$ normalized by its zero strain value vs normalized strain parameter, $\eta/\eta_{\rm vH} = \varepsilon_{xx}/\varepsilon_{\rm vH}$. Stronger coupling leads to a downward dynamical renormalization of $E_{\rm vH}(\eta)$ and, consequently, the critical strain value $\eta_{\rm vH}$, where $E_{\rm vH} = 0$, is reduced as well. However, $E_{\rm vH}$ vs η follows the same trends at all couplings as observed in the scaled plot.

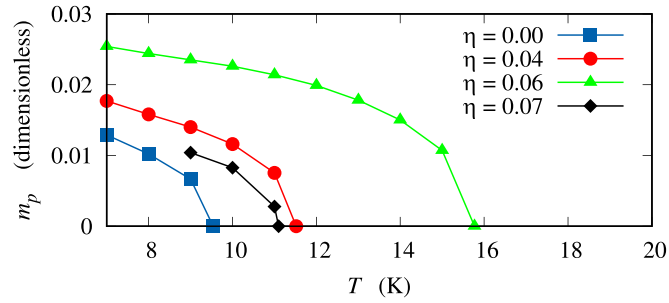

FIG. 4. Superconducting order parameter m_p vs temperature T for several strain parameters at a coupling strength $g_o = 0.40$ for which $\eta_{\rm vH} \sim 0.06$. The unstrained result, $\eta = 0.00$, shows a typical second order transition at $T_c \sim 9.5$ K. For the intermediate strain value of $\eta = 0.04 < \eta_{\rm vH}$ an elevated $T_c \sim 11.5$ K is observed and smooth, single-phase behavior for $T < T_c$. At $\eta = 0.06 \sim \eta_{\rm vH}$, a maximum $T_c \sim 15.8$ K is obtained and single phase behavior persists below the transition. The subsequent curve for $\eta = 0.07 > \eta_{\rm vH}$, shows a dramatic reduction in both T_c and the overall strength of the order parameter.

$\eta_{\rm vH} = 0.06$. Superconductivity is quickly surpressed as η increases beyond $\eta_{\rm vH}$.

This method is able to explore the model for $T < T_c$ and, thus, address the possibility of an emergent second superconducting transition should a nearly degenerate pairing symmetry become activated [30]. Numerical evidence for a second transition consists of a discontinuous slope in the m_p versus T curves. These FLEX results, which appear to be analytic for $T < T_c$, are therefore consistent with a single-component superconducting state at all strain values. This result is consistent with heat capacity, elastocaloric effect and superfluid density measurements [12,31,32], but is inconsistent with μsr results that suggest a strain-induced splitting between superconducting states with and without time-reversal symmetry breaking [33].

The main result is shown in Fig. 5 where T_c as a function of strain at different coupling strengths is displayed.

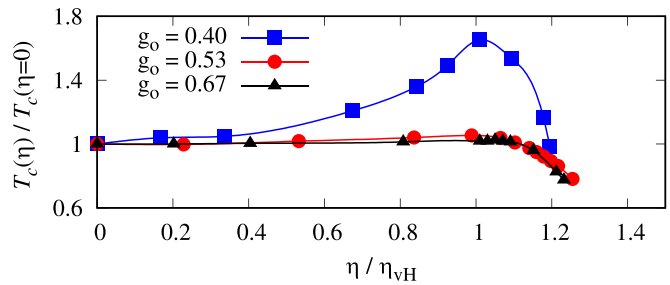

FIG. 5. Relative superconducting transition temperature $T_c(\eta)/T_c(\eta = 0)$ vs normalized strain $\eta/\eta_{\rm vH}$ for different coupling strengths g_o. For all g_o, T_c drops off rapidly for $\eta/\eta_{\rm vH} > 1$, consistent with what is observed for Sr_2RuO_4. For $g_o = 0.40$, which corresponds to $T_c(\eta = 0) \sim 9.5$ K, we observe a pronounced peak in T_c at or near $\eta/\eta_{\rm vH} \sim 1$ in alignment with experimental results for Sr_2RuO_4. Thus, when the coupling strength is set to values consistent with the low T_c values observed in Sr_2RuO_4, FLEX-based three-band Hubbard model results accurately represent the unusual strain-dependent phase diagram for Sr_2RuO_4.

For all coupling strengths, the FLEX T_c curves drop rapidly as strain increases beyond the critical value reproducing experimental results. The observed peak structure in T_c versus strain for Sr_2RuO_4 is also obtained with FLEX, but only for the lower coupling strength of $g_o = 0.40$. However, this coupling strength is most consistent with the low T_c values observed for Sr_2RuO_4. Apparently, the appearance of the peak in T_c versus strain only emerges in these FLEX results when quasiparticle excitations at the van Hove singularity are sufficiently narrow in energy.

Yuan, Bern, and Kivelson showed that when using a BCS-like Hamiltonian with an assumed d-wave pairing interaction, mean-field theory also generates a peaked structure in T_c versus strain when a similar model for the band structure of Sr_2RuO_4 [34]. Concurrent with the completion of this work, Hauck *et al.*, reported functional renormalization group results that suggest of d-wave pairing correlations, as measured through calculation of $T = 0$ pairing eigenvalues, follow a similar trend [35]. Together with the results presented here, this suggests that the peaked structure in T_c vs strain may be a generic feature of models with sharp quasiparticles in the γ-band forming d-wave singlet pairs. The results presented here are of added significance because (1) finite temperature and quasiparticle lifetime effects are included and (2) the pairing correlations emerge from a realistic Hamiltonian and we are able to track the $T < T_c$ physics unlike what is currently reported for functional renormalization group calculations.

Rømer *et al.* used an RPA treatment of a model for strained Sr_2RuO_4 [36]. In their investigation, they varied the spin-orbit coupling constant, λ, as well as the Coulomb and exchange parameters, U and J. Even though a peak in the DOS as a function of strain appears generically as these parameters are varied, a concurrent peak in the eigenvalues of the linearized gap equations only appears when $J/U = 0.1$ and $U/\lambda \ll 1$. Further, although a peak occurs in the eigenvalues for A_{1g} symmetry pairing, in alignment with our results, in the RPA results this symmetry is no longer the dominant pairing symmetry for all strain values, unlike what we find with FLEX. We should also note that in our case λ and J/U are set from first-principles calculations and the minimum value of U in our calculations still places us in the $U/\lambda \gg 1$ limit which is characteristic of the physical system where $U/\lambda \sim 20$. The emergence of a peak in our results for T_c versus strain as the coupling constant, g_o, is most likely due the reduction of lifetime broadening of the key states near the van Hove singularity, an affect that is not incorporated into RPA-based calculations.

V. DISCUSSION

Starting from a realisitic microscopic Hamiltonian and an unbiased treatment of the electron correlations driving a superconducting transition, FLEX results account for (1) the apparent absence of a splitting of the superconducting transition in strained samples and (2) the peak structure observed in T_c versus strain in Sr_2RuO_4. This result adds to the body of evidence that the underlying pairing in Sr_2RuO_4 is dominated by quasiparticles in the γ-band bound in spin-singlet pairs having $d_{x^2-y^2}$ orbital symmetry. While FLEX results for the unstrained limit were published previously, experimental evidence has increasingly shifted to support this picture.

These FLEX results do not yet account for evidence of time-reversal symmetry breaking in unstrained samples below $T_{tr} = T_c$ and in strained samples below $T_{tr} < T_c$. There are many potential explanations for this discrepancy. The additional pairing symmetry (or symmetries) needed to account for time-reversal symmetry breaking in a simple way may be quite small in comparison the dominant $d_{x^2-y^2}$ term. If so, then the physical and numerical approximations of this method will make it difficult for these small terms to be resolved. The momentum resolution we use in our numerical evaluation of the generalized self-energy (incorporating both normal and anomolous components) may be insufficient for accurately representing high angular momentum pairing components. Also, fluctuations of the dominant $d_{x^2-y^2}$ order parameter are missing in FLEX, a significant shortcoming for a quasi two-dimensional system. Further, as is typically done for this system, the possibility of translational symmetry breaking in the superconducting state is not considered in our calculation scheme [37]. Finally, while our FLEX calculation of the superconducting order parameter as a function of temperature effectively includes particle-particle vertex corrections that are absent in linearized Eliasberg calculations whose effective interaction only incorporates RPA bubbles [38], there are nonetheless other vertex corrections that may impact results as well. Notably, dynamical mean-field theory can be used to extract the full-frequency dependence of the two-particle vertex [39–41], but at the expense of neglecting the momentum-dependence that we are able to retain in our calculation.

A complete picture of superconductivity in Sr_2RuO_4 indeed does not emerge in these results, but they perhaps suggest a new path forward. Empirically it may prove fruitful to treat the underlying superconductivity in Sr_2RuO_4 as emerging through a dominant single-component $d_{x^2-y^2}$ pairing state with time-reversal symmetry breaking being a secondary feature in this complex, multiband system.

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