Electronic structure correspondence of singlet-triplet scale separation in strained Sr$_2$RuO$_4$

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At a temperature of roughly 1 K, Sr$_2$RuO$_4$ undergoes a transition from a normal Fermi liquid to a superconducting phase. Even while the former is relatively simple and well understood, the superconducting state is not even after 25 years of study. More recently it has been found that critical temperatures can be enhanced by application of uniaxial strain, up to a critical strain, after which it falls off.

In this work, we take an ‘instability’ approach and seek for divergences in susceptibilities. This provides an unbiased way to distinguish tendencies to competing ground states. We show that in the unstrained compound the singlet and triplet instabilities of the normal Fermi liquid phase are closely spaced. Under uniaxial strain electrons residing on all orbitals contributing to the Fermiology become more coherent while the electrons of Ru-$d_{xy}$ character become heavier and electrons of Ru-$d_{xz,yz}$ characters become lighter. In the process, Im $\chi(q,\omega)$ increases rapidly around the incommensurate vector $q=(0.3, 0.3, 0)\pi/a$ while it gets suppressed at all other commensurate vectors, in particular at $q=0$, which is essential for spin-triplet superconductivity. Thus the triplet superconducting instability remains the lagging instability of the system and the singlet instability enhances under strain, leading to a large energy-scale separation between these competing instabilities. At large strain an instability to a spin density wave overtakes the superconducting one.

The analysis relies on a high-fidelity, ab initio description of the one-particle properties and two-particle susceptibilities, based on the Quasiparticle Self-Consistent GW approximation augmented by Dynamical Mean Field theory. This approach is described and its high fidelity confirmed by comparing to observed one- and two-particle properties.

Introduction

The origin of superconducting pairing in Sr$_2$RuO$_4$ (SRO) has been one of the most debated topics in materials research over last two decades \[1\]. Until recently the superconductivity was believed to be of spin-triplet character. A series of recent experimental findings, including strain dependent enhancement in the critical temperature $T_c$ \[2, 3\] and the pronounced drop in O$^{17}$ NMR \[4\] measurements, observation of momentum-resolved superconducting energy gaps of Sr$_2$RuO$_4$ from quasiparticle interference imaging \[5\], direct observation of Lifshitz transition \[6\], jump in $c_{66}$ shear modulus \[7\] and high resolution $\mu$-SR studies \[8\] have challenged the existing beliefs and demand a fresh look into the enigmatic problem of superconductivity in SRO.

Strongly correlated electronic systems have a multiplicity of closely packed phases, owing to the small energy scale of the different kinds of correlations. Strain is an effective tool to tune correlations in bulk crystalline systems, as it makes small but significant changes to the one-particle spectrum, which in turn modifies two-particle properties such as superconductivity. It can lift degeneracies and separate out energy scales of competing phases, which sheds light into the underlying mechanisms that lead to different orders. Sr$_2$RuO$_4$ is a particularly salient example: as noted a recent study showed that uniaxial strain induces a two-fold enhancement $T_c$ up to a critical strain, after which it falls off rapidly \[2, 3\]. This study generated huge interest in the community and it was followed by a series of careful experimental and theoretical works, including work by Steppke et al. \[3\] which attributed the increase to a van Hove singularity inducing a Lifshitz transition just around the critical point. In the unstrained case, Sr$_2$RuO$_4$ has tetragonal symmetry, with three bands present at the Fermi level. These bands are
composed predominantly of three Ru $d$ orbitals: the $d_{xy}$ and the symmetry-equivalent $d_{xz}$ and $d_{yz}$ pair. Under strain the $d_{xz}$ and $d_{yz}$ equivalence is broken, and the Fermi surface undergoes a topological transition at a critical strain $\epsilon_x \sim 0.6\%$.

A series of theoretical studies [9–12] followed to explain the observations related to enhancement and later suppression of $T_c$ under strain. Some studies rely on a starting electronic band structure from density functional theory (DFT); more often they are phenomenological and based on low energy minimal model Hamiltonians. The latter typically employ model parameters for the Hubbard $U$ and $J$, and often rely on DFT eigenvalues to parameterise the one-body part. Such approaches are justified by the observation that superconductivity is a low energy phenomena, and should be well described if starting from a good underlying one-body part. Nevertheless, Kivelson et al., [13] recently argued that while much is known about the normal phases of Sr$_2$RuO$_4$, understanding the nature of superconductivity in Sr$_2$RuO$_4$ continues to be one of the most enigmatic problems in unconventional superconductivity even after 25 years [14]. This is indeed a remarkable observation considering that the normal phase of Sr$_2$RuO$_4$ is a relatively simple normal Fermi liquid, which is one of the better understood phases of correlated electronic materials.

In a recent work [15], we performed a thorough analysis of Sr$_2$RuO$_4$ with and without uniaxial strain, using a new high-fidelity $ab$-initio approach [16] to be described shortly. It uses an instability analysis: we monitor two-particle instabilities (points where a susceptibility diverges) in all particle-hole and particle-particle channels, starting from high temperature and decreasing it. This a significant departure from the ground state low energy model Hamiltonian approach noted above, but we believe, it is key to addressing the right questions for unconventional superconductivity, namely “can we reliably compute all finite temperature instabilities in the normal phase that on lowering of temperature would become unstable to a certain order?” As Kivelson et al. noted, [13] we believe, one key reason why superconductivity in Sr$_2$RuO$_4$ seems so difficult to explain stems from the inability of theoretical schemes to calculate all possible two-particle instabilities in the normal phase. This is particularly difficult to accomplish in a parameter free fashion. The instability analysis we use allows for possible competing phases in an unbiased manner. Further because the theory is both $ab$ initio and has very high fidelity, it has unprecedented predictive power [15] [17–20]. In this way we are able to circumvent the difficulties Kivelson et al. noted.

Our $ab$ initio approach starts from a one-particle Hamiltonian calculated from the quasiparticle self consistent $GW$ ($QSGW$) approximation [21]. It plays the role of DFT as a bath for the many-body problem to embedded in, but its fidelity is vastly superior. The one-particle Green’s function is generated from dynamical mean field theory (DMFT) [22], using QSGW as a bath. This is accomplished with a Continuous Time Quantum Monte Carlo (CTQMC) solver [23]. This framework [18, 25] is extended by computing the local vertex from the two-particle Green’s function by DMFT [26, 27], which is combined with nonlocal bubble diagrams to construct a Bethe-Salpeter equation [15, 19]. The latter is solved to yield the essential two-particle spin and charge susceptibilities $\chi^d$ and $\chi^m$ — physical observables which provide an important benchmark. Moreover they supply ingredients needed for the Eliashberg equation, which yields eigenvalues and eigenfunctions that describe instabilities to superconductivity in both singlet and triplet channels. We will denote $QSGW^{++}$ as a shorthand for the four-tier $QSGW$+$DMFT$+$BSE$+$Eliashberg$ theory. The numerical implementation is discussed in Pashov et al. [16] and codes are available on the open source electron structure suite Questaal [28].

$QSGW^{++}$ has high fidelity because QSGW captures non-local dynamic correlation particularly well in the charge channel [16, 29], but it cannot adequately capture effects of spin fluctuations. DMFT does an excellent job at the latter, which are strong but mostly controlled by a local effective interaction given by $U$ and $J$. For Sr$_2$RuO$_4$ in particular the QSGW Fermi surface is practically indistinguishable from a recent high-resolution ARPES measurement [30], and the spin susceptibility is in excellent agreement with Inelastic Neutron Scattering (INS) measurements [15] (measured only for the unstrained case when this work was published).

The present work reviews this prior study [15], which was our first attempt to use instability analysis with the full machinery of $QSGW^{++}$. It computed spin, charge and superconducting susceptibilities resolved in both energy and momenta and both in the singlet and triplet channels. We showed how the singlet instability increases under strain, while the triplet one does not,
and explained why $T_c$ increases. Here we extend that initial work to include wider excursion in strain to emphasize the trends, and provide a more detailed description of the connection between the single-particle and two-particle properties. In particular we establish the following:

1. show how strain modifies both one- and two-particle properties in a markedly orbital-dependent manner; strain enhances the role of the $d_{xy}$ orbital relative to the $d_{xz+yz}$ orbitals

2. show how the system becomes a better Fermi liquid with decreasing temperature. At low temperature, $J$ becomes the dominant factor, and the increase in coherence is orbital specific, on account of the van Hove singularity

3. show how the system becomes a better Fermi liquid with increasing strain while at the same time $d_{xy}$ becomes heavier and $d_{xz}$ and $d_{yz}$ lighter. Strain enhances the role of the $d_{xy}$ orbital relative to the $d_{xz+yz}$ orbitals, and enhances spin singlet superconductivity

4. Use instability analysis to clarify how the relative strength of competing phases evolve with strain, and compare against a spin density wave (SDW) (the latter eventually overtake the instability towards superconductivity at a strain larger than the critical one)

5. Show how spin-orbit coupling affects superconductivity.

In our original work we took $U$ and $J$ from constrained RPA [31] calculations obtained from DFT [32] which yielded $U=$4.5 eV and $J=$1.0 eV, and $J/U=0.22$. We have recently discovered from a recent implementation of C-RPA [28] that $U$ and $J$ computed from DFT are too large to be used in a QSGW framework: in the Hund’s metals $U$ and $J$ decrease in proportion to the bandwidth renormalisation, while $J/U$ remains fixed. For Sr$_2$RuO$_4$ in particular, QSGW renormalises the DFT bandwidth by about 0.6. Thus for the present study we use $U=3.0$ eV and $J=0.67$ eV; $J/U=0.22$. This reduction does not change anything qualitatively, but important details change, the most important being that with the DFT-C-RPA estimates $U$ and $J$, the leading triplet eigenvalue was found to be slightly larger than the singlet eigenvalue in unstrained Sr$_2$RuO$_4$, inconsistent with recent experimental findings [5, 7, 8]. In the present study we use the newer parameterisation of $U$ and $J$.

Before turning to the results, we note that our original work emphasised the interplay between charge and spin susceptibility. Those conclusions remain unchanged in the present work. As we have nothing new to report on this aspect, we focus on analysis spin susceptibility, which we denote as $\chi(q, \omega)$, and label spin and charge susceptibilities as $\chi^m$ and $\chi^d$ only where a description of both is needed. The superconducting instabilities we present here include both spin and charge susceptibilities.

**Results**

*Single-particle properties near the Fermi surface:*

As noted earlier, the Fermi surface produced by QSGW is essentially distinguishable from experiment (see SM, Ref. [15]). Augmentation with DMFT minimally affects the shape of the Fermi surface, but it does affect the spin-orbit splitting. QSGW+DMFT yields 90-100 meV, much larger than what had been widely thought, but in excellent agreement with revised estimate of 100 meV from a recent high-resolution laser ARPES measurement [30, 33].

Further, the ability QSGW or QSGW+DMFT to yield a nearly perfect Fermi surface, to accurately predict the spin-orbit splitting, and the critical $\epsilon_x \sim 0.6\%$ where the Fermi surface undergoes a topological transition, properties which DFT or DFT+DMFT do far less well, highlights the superior fidelity of QSGW+DMFT.

Fig. 1 (a) shows how the orbitally resolved electronic masses and single-particle scattering rates evolve with strain $\epsilon_x$. The single-site DMFT $\Im \Sigma(i\omega)$ is fit to a fourth order polynomial in $i\omega$ for low energies (first 6 Matsubara points at $\beta=40$ eV$^{-1}=290$ K) [34]. The mass enhancement, related to the coefficient $s_1$ of the linear term in the expansion $m_{\text{DMFT}}/m_{\text{QSGW}}=1+|s_1|$ [35], and the intercept $|s_0|\Gamma_{\text{DMFT}}/m_{\text{QSGW}}$ with $m_{\text{DMFT}}/m_{\text{QSGW}} = Z^{-1}$, is resolved in different intra-orbital channels. Both the masses and $\Gamma$ are orbital-dependent, and this differentiation is a signature of a Hund’s metal [19].

Electrons in the $d_{xy}$ orbital become heavier while the $d_{xz, yz}$ electrons become lighter with $\epsilon_x$. Beyond a critical $\epsilon_x \sim 0.6\% m_{\text{DMFT}}/m_{\text{QSGW}}$ becomes heavier than $m_{\text{DMFT}}/m_{\text{QSGW}}$ (see Fig. 1(b)). The trend is similar at lower temperatures: the $d_{xy}$ mass increases under strain while decreasing on $d_{xz}$ and $d_{yz}$. On the other hand, all orbitals become more coherent under strain, as seen in the reduction of the scattering rate $\Gamma$ (see Fig. 1(c)).

*Spin fluctuations: incommensurability and coherence:*
Figure 1: **Effective masses and scattering rates:** (a) The mass enhancement factors in DMFT (relative to the QSGW mass) are plotted in Ru-$d_{xy}$,$d_{yz}$,$d_{xz}$ channels. While the unstrained compound finds the heaviest electron mass for the electrons in $d_{xz}$ orbital, under strain the $d_{xy}$ mass becomes the heaviest. (b) We show the relative DMFT mass enhancement for $d_{xy}$ orbital in comparison to the $d_{xz}$ for all temperatures. (c) Scattering rates $\Gamma$ are orbitally anisotropic, but under strain it decreases in all orbital channels. For very large strains the system becomes a better Fermi liquid metal, nevertheless, the orbital anisotropy which is a typical signature of Hund’s metals survive for the entire range of strain.

![Figure 1](image1)

Figure 2: **Orbital components of real part of static spin susceptibility $\text{Re} \chi(q, \omega = 0)$:** We orbitally resolve the static spin susceptibility along some high-symmetry directions of the Brillouin Zone. The susceptibility at the ferromagnetic (FM) vector $q^{(0)}_{\text{FM}}=(0, 0, 0)$, $\chi$ is dominated by the intra-orbital fluctuations in the $d_{xy}$ channel, while at the incommensurate (IC) vector $q^{(1)}_{\text{IC}}=(0.3, 0.3, 0)$ (we use units $2\pi/a$ throughout) the three orbitals contribute almost equally. The antiferromagnetic (AFM) vector $q^{(0)}_{\text{AFM}}=(0.5, 0.5, 0)$ is fully gapped. Under strain the IC peak rapidly increases, and $d_{xy}$ emerges as the leading component of total spin susceptibilities along all high symmetry directions.

![Figure 2](image2)

$\chi(q, \omega)$ is computed from the momentum dependent Bethe-Salpeter equations in the magnetic channel.

$$
\chi_{\nu, \omega}^{m_1, m_2}(i\nu, i\nu')_{q, \omega} = [(\chi^0)^{-1}_{q, \omega} - \Gamma^{\text{irr}, m_2}_{\text{loc}}]^{-1}_{m_1, m_2}(i\nu, i\nu')_{q, \omega}.
$$

(1)

$\chi^0$ is the non-local ($k$-dependent) polarisation bubble computed from single-particle QSGW Green’s functions dressed by the local DMFT self-energy, and $\Gamma$ is the local irreducible two-particle vertex function computed in the magnetic channel. $\Gamma$ is a function of two fermionic frequencies $\nu$ and $\nu'$ and the bosonic frequency $\omega$. $\chi(q, \omega)$ is computed by closing $m(d)$ $\chi_{\nu, \omega}^{m_1, m_2}(i\nu, i\nu')_{q, \omega}$ with spin bare vertex $\gamma$ and summing over frequencies $(i\nu, i\nu')$ and orbitals $(\alpha_1, \alpha_2)$.

We compute the real part of the static susceptibility $\text{Re} \chi(q, \omega = 0)$ and resolve it in different inter- and intra- orbital channels to develop a systematic understanding of which orbitals dominate the spin susceptibilities at different q-vectors. In the vicinity of the ferromagnetic (FM) vector $q^{(0)}_{\text{FM}}=(0, 0, 0)$, $\chi$ is dominated by the intra-orbital fluctuations in the $d_{xy}$ channel (Fig. 2(a)), while at the incommensurate (IC) vector $q^{(1)}_{\text{IC}}=(0.3, 0.3, 0)$ (we use units $2\pi/a$ throughout) the three orbitals contribute almost equally. The antiferromagnetic (AFM) vector $q^{(0)}_{\text{AFM}}=(0.5, 0.5, 0)$ is fully gapped.
Figure 3: Strain and temperature dependence of susceptibilities: (a)-(e) Imaginary part of the dynamic spin susceptibility $\chi(q, \omega)$ at some high symmetry points in the Brillouin zone for different strains $\epsilon_x$. The unstrained compound shows a spin fluctuation spectrum strongly peaked at $(0.3, 0.3, 0)$. With increasing strain fluctuations become more strongly peaked at $(0.3, 0.3, 0)$ while it gets suppressed at the ferromagnetic vector and remains fully gapped at the anti-ferromagnetic vector. (f) With lowering temperature the IC peak at $q=(0.3, 0.3, 0)$ start to diverge for strains $\epsilon_x>0.6\%$, signaling an instability towards a spin density wave order.

When strain is applied we find that the IC peak rapidly increases, and $d_{xy}$ emerges as the leading component of total spin susceptibilities along all high symmetry directions (see Fig. 2 (b-e)). This is consistent with the fact that under strain, $d_{xy}$ becomes the most strongly correlated orbital. Nevertheless, the AFM vector remains fully gapped for strains up to $\epsilon_x=2.4\%$. We compute both real and imaginary parts of spin and charge susceptibilities by solving the BSE in respective channels. These equations are solved in the Matsubara representation with local dynamic vertex functions (which are functions of three Matsubara frequencies) and the non-local polarisation bubble which also has the Matsubara frequencies. After summing over all internal Fermionic Matsubara frequencies and orbital indices, we are left with $\chi(q,i\omega)$. Further, it needs to be analytically continued to real bosonic frequencies. One way is to analytically continue $\chi(i\omega)$ at each momentum, which is tremendously expensive. To understand the precise nature of the spin fluctuations at finite energies, it was imperative in this work that we extract $\text{Im} \chi(q, \omega)$ for finite $\omega$. For low energies, which is the focus here, the vertex $\Gamma_{\text{irr}}^{\text{loc}}$ is analytically continued by a quasiparticle-like approximation. We replace the frequency-dependent vertex with a constant, i.e., $\Gamma_{\text{loc}}^{\text{irr}}(i\nu, i\nu', i\omega)_{\alpha_{2} \sigma_{2}, \alpha_{4} \sigma_{4}} \sim U_{\alpha_{2} \sigma_{2}, \alpha_{4} \sigma_{4}}^{\text{eff}}$ which satisfies the constraint that $\chi(q,i\omega=0)=\chi(q,\omega=0)$.

This “quasiparticlized” vertex $U^{\text{eff}}$ contains all the important spin, orbital dependence. This approximation for analytic continuation works remarkably well for spin susceptibilities at low energy as shown in previous works [15, 19, 26, 27]. We compute the dynamic susceptibility $\text{Im} \chi(q, \omega)$ and observe that the intensity drops at $q_{\text{FM}}^{\text{FM}}=(0, 0, 0)$ under strain (see Fig. 3 (a)). The energy dispersion of $\text{Im} \chi(q, \omega)$ at $q_{\text{FM}}^{\text{FM}}=(0, 0, 0)$ remains almost invariant up to $\epsilon_x=1.6\%$, but for much
larger strains the branch loses both intensity and dispersion simultaneously. The reverse happens at $q^{IC}= (0.3, 0.3, 0)$ where under strain both the intensity and dispersion of the branch increase (see Fig. 3 (b)). For all strains, the $q^{AFM}= (0.5, 0.5, 0)$ remains fully gapped (see Fig. 3 (c)), while the peaks at $q=(0.25, 0, 0)$ and $q=(0, 0.25, 0)$ lose intensity, but in a very anisotropic manner (see Fig. 3 (d-e)). We also show in Fig. 3 (f) how the IC peak starts diverging with lowering temperatures at $\epsilon_s=0.8$%, signaling an instability towards an SDW order. However, whether the Fermi liquid phase will become unstable to an SDW phase or a superconducting phase, can only be confirmed from further investigation of superconducting pairing instabilities.

**Superconducting pairing: nodal character and dimensionality:**

The superconducting pairing susceptibility $\chi^{p-p}$ is computed by dressing the non-local pairing polarisation bubble $\chi_{pp}(k, iv)$ with the pairing vertex $\Gamma^{p-p}$ using the Bethe-Salpeter equation in the particle-hole channel (see Supplementary Figure 5 for Feynman diagram representation).

$$\chi^{p-p} = \chi_{pp} \cdot [1 + \Gamma^{p-p} \cdot \chi_{pp}]^{-1}$$  

$\Gamma^{p-p}$ in the singlet (s) and triplet (t) channels are obtained from the magnetic (spin) and density (charge) particle-hole reducible vertices by

$$\Gamma_{a_1a_2, a_3}^{p-p, s,t} (k, iv, k', iv') = \Gamma_{a_1a_2, a_3}^{p-p, s,t} (iv, iv')$$

$$\Gamma_{s}^{p-p} = \sum_{0} \frac{1}{1 - \lambda} \cdot (\sqrt{\chi_{pp}(k)} \cdot \phi^s(k))$$

$$\cdot (\sqrt{\chi_{pp}(k')} \cdot \phi^s(k'))$$

The pairing susceptibility diverges when the leading eigenvalue $\lambda$ becomes unity. The corresponding eigenfunction represents the momentum structure of $\chi^{p-p}$. Unconventional superconductivity in SRO is multi-orbital in nature with multiple competing instabilities. In our previous work [15], we performed a thorough analysis of all possible singlet and triplet instabilities in SRO and associated with a particular symmetry group. We showed that the leading eigenvalue in the singlet channel had a $d_{x^2-y^2}$ instability (B_{1g} symmetry) while the leading eigenvalue in the triplet channel was of an extended nodeless $s$-wave 2$\delta_0 + \cos k_x + \cos k_y$ gap structure with $A_{1g}$ irreducible representation in the $d_{x^2-y^2}$ basis.

A subsequent Bogoliubov quasiparticle scattering interference visualization of the gap structure at milli-Kelvin temperatures was measured to be of $B_{1g}-d_{x^2-y^2}$ nature. [5] We observe that for all strains (and without strain) the eigenvalue corresponding to the singlet instability remains the leading one and the relative strength of the singlet to triplet eigenvalues ($\lambda_s/\lambda_t$) keep increasing under strain. The enhancement in $\lambda_s/\lambda_t$ under strain, becomes more apparent at lower temperatures (see Fig. 4 (a))). This is concomitant with the mass becoming heavier in the $d_{xy}$ channel while the masses relax on other orbitals. Further, this is a direct consequence of the spin fluctuations getting suppressed at $q^{FPM}= (0, 0, 0)$ and rising steeply at $q^{IC}= (0.3, 0.3, 0)$. It is understandable that the system can undergo a spin density wave order mediated primarily via the fluctuations at and around $q^{IC}= (0.3, 0.3, 0)$. Once the spin susceptibility diverges, at lower temperatures, under large strains, the system will encounter the density wave phase and the superconducting channel will be suppressed. To check that we extract the leading eigenvalue ($\lambda_{SDW}$) in the density wave channel, by diagonalising the susceptibility matrix. We observe that while for $\epsilon_s=0.0$, the $\lambda_{SDW}$ and $\lambda_s$ show a very similar temperature dependence ($\lambda_s$ is slightly more steeper than $\lambda_{SDW}$), for finite and large strains ($\epsilon_s>0.6$%) $\lambda_{SDW}$ acquires a steeper temperature dependence than $\lambda_s$ (see Fig. 4 (b)). This suggests that although the $\lambda_s/\lambda_t$ continues to
We observe that all our essential conclusions for both spin and superconducting instabilities remain qualitatively invariant once the spin-orbit coupling (SOC) is included in the calculations. We observe that under strain, with SOC the singlet and triplet eigenvalues get further removed from each other, making the scale separation clearer for all strains. The FM spin fluctuations go down under strain, in presence of SOC and the IC becomes steeper, making a SDW instability likely for larger strains.

Summary

We have performed a detailed analysis of the single-particle and two-particle response of Sr$_2$RuO$_4$ under large strains. The instability approach allows us to compare different kinds of instabilities of the normal phase. By performing excursions in temperature or external parameters such as strain we can identify which ground states are preferred instabilities of the normal phase, distinguishing among multiple closely spaced many-body ordered phases. Key to the success of this approach is the ab initio QSGW++ machinery, whose high fidelity (which is essential) is confirmed by the excellent agreement with observed one- and two-particle properties, as we have shown.

We find that while the singlet and triplet instabilities are similar in the unstrained Sr$_2$RuO$_4$, the ratio of eigenvalues $\lambda_s/\lambda_t$ under uniaxial strain $\epsilon_x$ keeps increasing at all temperatures, leading to a clear separation between the singlet and the triplet superconducting pairing instabilities. Its emergence can be traced to the orbital-selective evolution in single-particle properties under strain: particularly $d_{xy}$ acquires a heavy mass while $d_{xz}$ and $d_{yz}$ become lighter. This directly modifies the two-particle susceptibilities; the spin susceptibility at $q_{FM}$ is suppressed under strain and at $q_{IC}$ it diverges, leading to the relative suppression of the triplet instability. Finally, the rapid divergence of $\chi$ temperature at $q_{IC}$ leads to enhancement in both $\lambda_s$ and $\lambda_{SDW}$. The latter has a steeper temperature dependence, and thereby, for large strains the superconducting phase is suppressed by an emergent SDW phase.

Methods

We use a recently developed quasi-particle self consistent $GW +$ dynamical mean field theory (QSGW+DMFT) [16, 17, 25], as implemented in the all-electron Questaal package [28]. Paramagnetic DMFT is combined with nonmagnetic QSGW via local projectors of the Ru 4$d$ states on the Ru augmentation spheres to form the correlated subspace. We carried out the QSGW calculations in the tetragonal and strained phases of Sr$_2$RuO$_4$ with space group.
139/I4mmm. DMFT provides a non-perturbative treatment of the local spin and charge fluctuations. We use an exact hybridisation expansion solver, namely the continuous time Monte Carlo (CTQMC) to solve the Anderson impurity problem.

The one-body part of QSGW is performed on a $16 \times 16 \times 16$ k-mesh and charge has been converged up to $10^{-6}$ accuracy, while the (relatively smooth) many-body static self-energy $\Sigma^0(k)$ is constructed on a $8 \times 8 \times 8$ k-mesh from the dynamical $GW$ $\Sigma(k, \omega)$. $\Sigma^0(k)$ is iterated until convergence (RMS change in $\Sigma^0<10^{-5}$ Ry). $U=3.0$ eV and $J=0.67$ eV were used as correlation parameters for DMFT. The DMFT for the dynamical self energy is iterated, and converges in $\approx 20$ iterations. Calculations for the single particle response functions are performed with $10^9$ QMC steps per core and the statistics is averaged over 64 cores. The two particle Green’s functions are sampled over a larger number of cores (10000-20000) to improve the statistical error bars. We sample the local two-particle Green’s functions with CTQMC for all the correlated orbitals and compute the local polarisation bubble to solve the inverse Bethe-Salpeter equation (BSE) for the local irreducible vertex. Finally, we compute the double particle Green’s functions and combined with the local irreducible vertex $\chi^{m,d}(q, \omega)$ we obtain the full non-local spin and charge susceptibilities $\Sigma^0(k)\Sigma(k-q, \omega-\Omega)$.

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[1] A. P. Mackenzie, T. Scaffidi, C. W. Hicks, and Y. Maeno, npj Quantum Materials 2, 40 (2017).

[2] C. W. Hicks, D. O. Brodsky, E. A. Yelland, A. S. Gibbs, J. A. Bruin, M. E. Barber, S. D. Edkins, K. Nishimura, S. Yonezawa, Y. Maeno, et al., Science 344, 283 (2014).

[3] A. Steppke, L. Zhao, M. E. Barber, T. Scaffidi, F. Jerzembeck, H. Rosner, A. S. Gibbs, Y. Maeno, S. H. Simon, A. P. Mackenzie, et al., Science 355, eaaf9398 (2017).

[4] A. Pustogow, Y. Luo, A. Chronister, Y.-S. Su, D. Sokolov, F. Jerzembeck, A. Mackenzie, C. Hicks, N. Kikugawa, S. Raghu, et al., Nature 574, 72 (2019).

[5] R. Sharma, S. D. Edkins, Z. Wang, A. Kostin, C. Sow, Y. Maeno, A. P. Mackenzie, J. C. S. Davis, and Y. Madhavan, Proceedings of the National Academy of Sciences 117, 5222 (2020).

[6] V. Sunko, E. A. Morales, I. Marković, M. E. Barber, D. Milosavljević, F. Mazzola, D. A. Sokolov, N. Kikugawa, C. Cacho, P. Duden, et al., npj Quantum Materials 4, 1 (2019).

[7] S. Benhabib, C. Lupien, I. Paul, L. Berges, M. Dion, M. Nardone, A. Zitouni, Z. Mao, Y. Maeno, A. Georges, et al., arXiv preprint arXiv:2002.05916 (2020).

[8] S. Ghosh, A. Shekheter, F. Jerzembeck, N. Kikugawa, D. A. Sokolov, M. Brando, A. Mackenzie, C. W. Hicks, and B. Ramshaw, arXiv preprint arXiv:2002.06130 (2020).

[9] Y.-S. Li, N. Kikugawa, D. Sokolov, F. Jerzembeck, A. Gibbs, Y. Maeno, C. Hicks, M. Nicklas, and A. Mackenzie, arXiv preprint arXiv:1906.07597 (2019).

[10] A. T. Romer, A. Kreisel, M. A. Müller, P. J. Hirschfeld, I. M. Eremin, and B. M. Andersen, Phys. Rev. B 102, 054506 (2020).

[11] A. T. Romer, D. D. Scherer, I. M. Eremin, P. J. Hirschfeld, and B. M. Andersen, Phys. Rev. Lett. 123, 247001 (2019).

[12] Y.-C. Liu, F.-C. Zhang, T. M. Rice, and Q.-H. Wang, npj Quantum Materials 2, 1 (2017).

[13] S. A. Kivelson, A. C. Yuan, B. Ramshaw, and R. Thomale, arXiv preprint arXiv:2002.00016 (2020).

[14] Y. Maeno, H. Hashimoto, K. Yoshida, S. Nishizaki, T. Fujita, J. Bednorz, and F. Lichtenberg, Nature 372, 532 (1994).

[15] S. Acharya, D. Pashov, C. Weber, H. Park, L. Sponza, and M. van Schilfgaarde, Communications Physics 2 (2019), 10.1038/s42005-019-0254-1.

[16] D. Pashov, S. Acharya, R. L. W. Lambrecht, J. Jackson, K. Belashchenko, D. A. Chantis, F. Jamet, and M. Weber, van Schilfgaarde, Computer Physics Communications (2019).

[17] L. Sponza and et al, Phys. Rev. B 95, 041112 (2017).

[18] E. Baldini and et al, Proceedings of the National Academy of Sciences 117, 6409 (2020).

[19] S. Acharya, D. Pashov, F. Jamet, and M. van Schilfgaarde, Phys. Rev. Lett. 124, 237001 (2020).
[20] S. Acharya, D. Pashov, and M. van Schilfgaarde, arXiv preprint arXiv:2005.07729 (2020).
[21] T. Kotani, M. van Schilfgaarde, and S. V. Faleev, Phys. Rev. B 76, 165106 (2007).
[22] A. Georges, G. Kotliar, W. Krauth, and M. J. Rozenberg, Reviews of Modern Physics 68, 13 (1996).
[23] K. Haule, Physical Review B 75, 155113 (2007).
[24] E. Gull, A. J. Millis, A. I. Lichtenstein, A. N. Rubtsov, M. Troyer, and P. Werner, Reviews of Modern Physics 83, 349 (2011).
[25] S. Acharya, C. Weber, E. Plekhanov, D. Pashov, A. Taraphder, and M. van Schilfgaarde, Phys. Rev. X 8, 021038 (2018).
[26] H. Park, The study of two-particle response functions in strongly correlated electron systems within the dynamical mean field theory, Ph.D. thesis, Rutgers University-Graduate School-New Brunswick (2011).
[27] Z. Yin, K. Haule, and G. Kotliar, Nature Physics 10, 845 (2014).
[28] “Questaal website,” https://www.questaal.org.
[29] J. M. Tomczak, M. van Schilfgaarde, and G. Kotliar, Phys. Rev. Lett. 109, 237010 (2012).
[30] A. Tamai, M. Zingl, E. Rozbicki, E. Cappelli, S. Riccò, A. de la Torre, S. McKeown Walker, F. Y. Bruno, P. D. C. King, W. Meevasana, M. Shi, M. Radović, N. C. Plumb, A. S. Gibbs, A. P. Mackenzie, C. Berthod, H. U. R. Strand, M. Kim, A. Georges, and F. Baumberger, Phys. Rev. X 9, 021048 (2019).
[31] F. Aryasetiawan, M. Imada, A. Georges, G. Kotliar, S. Biermann, and A. Lichtenstein, Physical Review B 70, 195104 (2004).
[32] X. Deng, K. Haule, and G. Kotliar, Physical Review Letters 116 (2016), 10.1103/PhysRevLett.116.256401.
[33] C. N. Veenstra, Z.-H. Zhu, M. Raichle, B. M. Ludbrook, A. Nicolaou, B. Slomski, G. Landolt, S. Kittaka, Y. Maeno, J. H. Dil, I. S. Elfimov, M. W. Haverkort, and A. Damascelli, Phys. Rev. Lett. 112, 127002 (2014).
[34] S. Acharya, M. S. Laad, D. Dey, T. Maitra, and A. Taraphder, Scientific Reports 7, 43033 (2017).
[35] Q. Han, H. T. Dang, and A. Millis, Physical Review B 93, 155103 (2016).
[36] Y.-C. Liu, F.-C. Zhang, T. M. Rice, and Q.-H. Wang, npj Quantum Materials 2, 1 (2017).
[37] K. Haule and G. Kotliar, Phys. Rev. B 76, 104509 (2007).
[38] H. Park, K. Haule, and G. Kotliar, Phys Rev Lett. 107, 137007 (2011).