Electronic theory for the normal state spin dynamics in Sr$_2$RuO$_4$: anisotropy due to spin-orbit coupling

I. Eremin$^{1,2}$, D. Manske$^1$, and K.H. Bennemann$^1$

$^1$Institut für Theoretische Physik, Freie Universität Berlin, D-14195 Berlin, Germany
$^2$Physics Department, Kazan State University, 420008 Kazan, Russia

(December 27, 2021)

Using a three-band Hubbard Hamiltonian we calculate within the random-phase-approximation the spin susceptibility, $\chi(q, \omega)$, and NMR spin-lattice relaxation rate, $1/T_1$, in the normal state of the triplet superconductor Sr$_2$RuO$_4$ and obtain quantitative agreement with experimental data. Most importantly, we find that due to spin-orbit coupling the out-of-plane component of the spin susceptibility $\chi^{z\omega}$ becomes at low temperatures two times larger than the in-plane one. As a consequence strong incommensurate antiferromagnetic fluctuations of the quasi-one-dimensional $xz$- and $yz$-bands point into the $z$-direction. Our results provide further evidence for the importance of spin fluctuations for triplet superconductivity in Sr$_2$RuO$_4$.

74.20.Mn, 74.25.-q, 74.25.Ha

The spin-triplet superconductivity with $T_c=1.5$K observed in layered Sr$_2$RuO$_4$ seems to be a new example of unconventional superconductivity [1]. The non $s$-wave symmetry of the order parameter is observed in several experiments (see for example [2]). Although the structure of Sr$_2$RuO$_4$ is the same as for the high-$T_c$ superconductor La$_2$−xSr$_x$CuO$_4$, its superconducting properties resemble those of superfluid $^3$He. Most recently it was found that the superconducting order parameter is of $p$-wave type, but contains line nodes half-way between the RuO$_2$-planes [3]. These results support Cooper-pairing via spin fluctuations as one of the most probable mechanism to explain the triplet superconductivity in Sr$_2$RuO$_4$. Therefore, theoretical and experimental investigations of the spin dynamics behavior in the normal and superconducting state of Sr$_2$RuO$_4$ are needed.

Recent studies by means of inelastic neutron scattering (INS) [4] and nuclear magnetic resonance (NMR) [5] of the spin dynamics in Sr$_2$RuO$_4$ reveal the presence of strong incommensurate fluctuations in the RuO$_2$-plane at the antiferromagnetic wave vector $Q_z = (2\pi/3, 2\pi/3)$. As it was found in band structure calculations [6], they result from the nesting properties of the quasi-one-dimensional $d_{xz}$- and $d_{yz}$-bands. The two-dimensional $d_{xy}$-band contains only weak ferromagnetic fluctuations. The observation of the line nodes between the RuO$_2$-planes [7] suggests strong spin fluctuations between the RuO$_2$-planes in $z$-direction [8,9]. However, inelastic neutron scattering [10] observes that magnetic fluctuations are purely two-dimensional and originate from the RuO$_2$-plane. Both behaviors could result as a consequence of the magnetic anisotropy within the RuO$_2$-plane as indeed was observed in recent NMR experiments by Ishida et al. [11]. In particular, analyzing the temperature dependence of the nuclear spin-lattice relaxation rate on $^{17}$O in the RuO$_2$-plane at low temperatures, they have demonstrated that the out-of-plane component of the spin susceptibility can become almost three time larger than the in-plane one. This strong and unexpected anisotropy disappears with increasing temperature [12].

In this Communication we analyze the normal state spin dynamics of the Sr$_2$RuO$_4$ using the two-dimensional three-band Hubbard Hamiltonian for the three bands crossing the Fermi level. We calculate the dynamical spin susceptibility $\chi(q, \omega)$ within the random-phase-approximation and show that the observed magnetic anisotropy in the RuO$_2$-plane arises mainly due to the spin-orbit coupling. Its further enhancement with lowering temperatures is due to the vicinity to a magnetic instability. Thus, we demonstrate that as in the superconducting state [3] the spin-orbit coupling plays an important role also for the normal state spin dynamics of Sr$_2$RuO$_4$. We also discuss briefly the consequences of this magnetic anisotropy for Cooper-pairing due to the exchange of spin fluctuations.

We start from the two-dimensional three-band Hubbard Hamiltonian

$$H = H_t + H_U = \sum_{k, \sigma} \sum_{l} t_{kl} a_{k,l\sigma}^\dagger a_{k,l\sigma} + \sum_{i,j} U_i n_{i\uparrow} n_{i\downarrow},$$

(1)

where $a_{k,l\sigma}$ is the Fourier-transformed annihilation operator for the $d_{l\sigma}$ orbital electrons ($l = xy, yz, zx$) and $U_i$ is the corresponding on-site Coulomb repulsion. $t_{kl}$ denotes the energy dispersions of the tight-bindings bands calculated as follows: $t_{kl} = -\epsilon_0 - 2t_x \cos k_x - 2t_y \cos k_y + 4t' \cos k_x \cos k_y$. We choose the values for the parameter set ($\epsilon_0, t_x, t_y, t'$) as (0.5, 0.42, 0.44, 0.14), (0.24, 0.31, 0.045, 0.01), and (0.24, 0.045, 0.35, 0.01) eV for $d_{xy}$-, $d_{zx}$-, and $d_{yz}$-orbitals in accordance with band-structure calculations [6]. The electronic properties of this model in application to Sr$_2$RuO$_4$ were studied recently and as was found can explain some features of the spin excitation spectrum in Sr$_2$RuO$_4$ [13,14,15]. However, this model fails to explain the observed magnetic anisotropy at low temperatures [13] and line nodes in the superconducting
order parameter below $T_c$ which are between the RuO$_2$-planes. On the other hand, it is known that the spin-orbit coupling plays an important role in the superconducting state of in Sr$_2$RuO$_4$ [4]. This is further confirmed by the recent observation of the large spin-orbit coupling in the insulating Ca$_2$RuO$_4$ [13]. Therefore, we include in our model spin-orbit coupling:

$$H_{so} = \lambda \sum_i L_i S_i$$

where the angular momentum $L_i$ operates on the three $t_{2g}$-orbitals on the site $i$. Similar to an earlier approach [4], we restrict ourselves to the three orbitals, ignoring $e_{2g}$-orbitals and choose the coupling constant $\lambda$ such that the $t_{2g}$-states behave like an $l = 1$ angular momentum representation. Moreover, it is known that the quasi-two-dimensional $xy$-band is separated from the quasi-one-dimensional $xz$- and $yz$-bands. Then, one expects that the effect of spin-orbit coupling is small and can be excluded for simplicity. Therefore, we consider the effect of the spin-orbit coupling on $xz$- and $yz$-bands only. Then, the kinetic part of the Hamiltonian $H_i + H_{so}$ can be diagonalized and the new energy dispersions are

$$\epsilon_{k,yz}^x = (t_{k,yz} + t_{k,xz} + A_k)/2$$

$$\epsilon_{k,xz}^x = (t_{k,yz} + t_{k,xz} - A_k)/2$$

where $A_k = \sqrt{(t_{k,yz} - t_{k,xz})^2 + \lambda^2}$, and $\sigma$ refers to spin projection. One clearly sees that the spin-orbit coupling does not remove the Kramers degeneracy of the spins. Therefore, the resultant Fermi surface is consists of three sheets like observed in the experiment. Most importantly, spin-orbit coupling together with Eq. (1) leads to a new quasiparticle which we label by pseudo-spin and pseudo-orbital indices. The unitary transformation $U_k$ connecting old and new quasiparticles is defined for each wave vector and lead to the following relation between them:

$$\chi^p_{0,\|}(q, \omega)$$

FIG. 1. Calculated Fermi surface for a RuO$_2$ plane in Sr$_2$RuO$_4$ taking into account spin-orbit coupling.

$$\chi^p_{0,\perp}(q, \omega)$$

FIG. 2. Diagrammatic representation for the transverse and longitudinal components of the magnetic susceptibility. The full lines represent the electron Green’s function with pseudospin $\sigma$ and pseudo-orbital $l$-indexes. $g_+$ and $g_z$ denote the vertexes as described in the text.

$$\epsilon_{k,yz}^+ = u_{1k}a_{k,yz}^+ - iv_{1k}a_{k,xz}^+$$

$$\epsilon_{k,xz}^+ = u_{2k}a_{k,yz}^+ - iv_{2k}a_{k,xz}^+$$

$$\epsilon_{k,yz}^- = u_{1k}a_{k,yz}^- + iv_{1k}a_{k,xz}^-$$

$$\epsilon_{k,xz}^- = u_{2k}a_{k,yz}^- + iv_{2k}a_{k,xz}^-$$

where $u_{mk} = \frac{\lambda}{\sqrt{(t_{k,yz} - t_{k,xz} + A_k)^2 + \lambda^2}}$ and $v_{mk} = \frac{\lambda}{\sqrt{(t_{k,yz} + t_{k,xz} + A_k)^2 + \lambda^2}}$. The '-' and '+' signs refer to the $m = 1$ and $m = 2$, respectively.

In Fig.1 we show the resultant Fermi surfaces for each obtained band where we have chosen $\lambda = 100$meV in agreement with earlier estimations [14,17]. One immediately sees that $xz$- and $yz$-bands split around the nested parts in good agreement with experiment [13]. Thus, spin-orbit coupling acts as a hybridization between these bands. However, in contrast to hybridization spin-orbit coupling introduces also an anisotropy for the states with pseudo-spins $\uparrow$ and $\downarrow$. This will be reflected in the magnetic susceptibility. Since the spin and orbital degrees of freedom are now mixed in some spin-orbital liquid, the magnetic susceptibility involves also the orbital magnetism which is very anisotropic.

For the calculation of the transverse, $\chi^{+}_{\perp}$, and longitudinal, $\chi^{+}_{\|}$, components of the spin susceptibility of each band $l$ we use the diagrammatic representation shown in Fig. 2. Since the Kramers degeneracy is not removed by the spin-orbit coupling, the main anisotropy arises from the calculations of the anisotropic vertex $g_z = \hat{l}_z + 2s_z$ and $g_+ = \hat{l}_z + 2s_+$ calculated on the basis of the new quasiparticle states. In addition, due to the hybridization between $xz$- and $yz$-bands we also calculate the transverse and longitudinal components of the the interband susceptibility $\chi^{+\perp}$. Then, for example,

$$\chi^{+\perp}_{0,xz}(q, \omega) = -\frac{4}{N} \sum_k (u_{2k}u_{2k+q} - v_{2k}v_{2k+q})^2 \times f(\epsilon_{k,xz}^+ - \epsilon_{k+q,xz}^- + \omega + i\Omega^+)$$

and

$$\chi^{zz}_{0,xz}(q, \omega) = \chi^{\uparrow}_{xz}(q, \omega) + \chi^{\downarrow}_{xz}(q, \omega) = \frac{2}{N} \sum_k$$
transverse and longitudinal total susceptibility, \( \chi \), where \( f \), the longitudinal gets an extra term due to results from the calculated matrix elements. Moreover, between longitudinal and transverse components which calculating through the corresponding vertexes using Eq. (4). For all other orbitals the calculations are straightforward. Note, that the magnetic response of the Brillouin Zone at temperature \( T = 100K \).

\[
\sum_i \chi_{RPA,i}^{+} \uparrow \uparrow(q,\omega) = (2\pi/3, 2\pi/3) \ \text{and temperature } T=20K. \] The longitudinal component reveals a peak at approximately \( \omega_{sf} = 6meV \) in quantitative agreement with experimental data on INS [3]. The transverse component is featureless showing the absence of the incommensurate antiferromagnetic spin fluctuations. Thus, the fluctuations in the transverse susceptibility are isotropic and ferromagnetic-like. Therefore, antiferromagnetic fluctuations are present only perpendicular to the RuO\(_2\)-plane.

We also note that our results are in accordance with earlier estimations made by Ng and Sigrist [19] with one important difference. In their work it was found that the IAF are slightly enhanced in the longitudinal component of the magnetic susceptibility strongly enhances due to orbital contributions. Moreover, we show by taking into account the correlation effects within random-phase-approximation(RPA) the IAF are further enhanced in the z-direction.
In order to see the temperature dependence of the magnetic anisotropy induced by the spin-orbit coupling we display in Fig. 4 the temperature dependence of the quantity $\sum_q \frac{1}{\omega_{sf}} \tilde{\chi}_{q,\omega_{sf}}(q,\omega_{sf})$ for both components. At room temperatures both longitudinal and transverse susceptibilities are almost identical, since thermal effects wash out the influence of the spin-orbit interaction. With decreasing temperature the magnetic anisotropy arises and at low temperatures we find the important result that the out-of-plane component $\chi^{zz}$ is about two times larger than the in-plane one ($\chi^{zz} > \chi^{++}/2$).

Finally, in order to compare our results with experimental data we calculate the nuclear spin-lattice relaxation rate for $^{17}$O ion in the RuO$_2$-plane for different external magnetic field orientation ($i = a, b, c$)

$$\left[ \frac{1}{T_i/T} \right] = \frac{2 k_B \gamma_0^2}{(\gamma_c b)^2} \sum_q |A_{piq}|^2 \frac{\chi_{p}(q,\omega_{sf})}{\omega_{sf}} ,$$

where $A_{pq}$ is the $q$-dependent hyperfine-coupling constant perpendicular to the $i$-direction.

In Fig. 5 we show the calculated temperature dependence of the spin-lattice relaxation for an external magnetic field within and perpendicular to the RuO$_2$-plane together with experimental data. At $T = 250$K the spin-lattice relaxation rate is almost isotropic. Due to the anisotropy in the spin susceptibilities arising from spin-orbit coupling the relaxation rates become different with decreasing temperature. The largest anisotropy occurs close to the superconducting transition temperature in good agreement with experimental data [13].

To summarize, our results clearly demonstrate the essential significance of spin-orbit coupling for the spin-dynamics already in the normal state of the triplet superconductor Sr$_2$RuO$_4$. We find that the magnetic response becomes strongly anisotropic even within a RuO$_2$-plane: while the in-plane response is mainly ferromagnetic, the out-of-plane response is antiferromagnetic-like.

Let us also remark on the implication of our results for the triplet superconductivity in Sr$_2$RuO$_4$. In a previous study [11], neglecting spin-orbit coupling but including the hybridization between $xy$- and $xz$, $yz$-bands, we have found ferromagnetic and IAF fluctuations within the $ab$-plane. This would lead to nodes within the RuO$_2$-plane. However, due to the magnetic anisotropy induced by spin-orbit coupling, a nodeless $p$-wave pairing is possible in the RuO$_2$-plane as experimentally observed. Our results provide further evidence for the importance of spin fluctuations for triplet superconductivity in Sr$_2$RuO$_4$.

We are thankful for stimulating discussions with B.L. Gyorgy, Y. Maeno, D. Fay, M. Eremin, R. Tarento and M. Ovchinnikova for critical reading of the manuscript. We are grateful to German-French Foundation (PROCOPE) for the financial support. The work of I. E. is supported by the Alexander von Humboldt Foundation and CRDF Grant No. REC. 007.
[18] A. Damascelli \textit{et al.}, Phys. Rev. Lett. \textbf{85}, 5194 (2000).
[19] K.K. Ng, and M. Sigrist, J. Phys. Soc. Jpn. \textbf{69}, 3764 (2000).