On the Coexistence in $RuSr_2GdCu_2O_8$ of Superconductivity and Ferromagnetism

R. Weht, A. B. Shick, and W. E. Pickett

Department of Physics, University of California, Davis CA 95616

Abstract. We review the reasons that make superconductivity unlikely to arise in a ferromagnet. Then, in light of the report by Tallon and collaborators that $RuSr_2GdCu_2O_8$ becomes superconducting at $\sim$35 K which is well below the Curie temperature of 132 K, we consider whether the objections really apply to this compound. Our considerations are supported by local spin density calculations for this compound, which indeed indicate a ferromagnetic RuO_2 layer. The Ru moment resides in t_{2g} orbitals but is characteristic of itinerant magnetism (and is sensitive to choice of exchange-correlation potential and to the atomic positions). Based on the small exchange splitting that is induced in the Cu-O layers, the system seems capable of supporting singlet superconductivity an FFLO-type order parameter and possibly a π-phase alternation between layers. If instead the pairing is triplet in the RuO_2 layers, it can be distinguished by a spin-polarized supercurrent. Either type of superconductivity seems to imply a spontaneous vortex phase if the magnetization is rotated out of the plane.

INTRODUCTION

Tallon and collaborators ([1,3] report the remarkable observation of superconductivity arising up to at least $T_s = 35$ K in $RuSr_2GdCu_2O_{8-\delta}$ (Ru1212), a compound reported originally by Bauerfeind *et al.*, [2] in spite of its having become ferromagnetic (FM) already at $T_m = 132$ K. The ferromagnetism is evident from the magnetization vs. field curve and also from zero-field muon spin rotation data that indicate a uniform field in the sample below T_m. Superconductivity (SC) is evident from the loss of resistivity, reversal of the susceptibility, and from specific heat measurements that reflect a bulk transition at T_s. Although Felner *et al.* have reported related results in $R_{1.4}Ce_{0.6}Sr_2GdCu_2O_{10}$, $R =$ Eu or Gd, [4], they indicate that their system is a canted antiferromagnet, with a saturation field an order of magnitude smaller than Ru1212. Thus Ru1212 appears to be unique as a FM that becomes superconducting well within the FM phase.

Ru1212 presents scientific questions on several levels (not to mention the novel applications that a superconducting FM might have). There are questions on the phenomenological level: how can SC and FM coexist? does the magnetic field

arising from the frozen magnetization lead to supercurrent flow? is the SC pairing singlet or triplet? There are also questions on the microscopic level: what is the FM like – is it Ru, as interpreted so far? how much does FM affect the carriers in the CuO$_2$ planes? how does c-axis dispersion compare with other cuprates? In this paper we begin to address both the phenomenological and the microscopic questions.

QUESTIONS OF COEXISTENCE OF SUPERCONDUCTIVITY WITH FERROMAGNETISM

The first question, and easiest to deal with, is the possibility of paramagnetic limiting. The observed saturation magnetization is about 1 μ_B per unit cell, which translates into an internal field $B_{int} = 4\pi M = 700$ G. Spin splitting $2g\mu_B B_{int}$ of the two electrons of the pair is negligible compared to the superconducting gap $2\Delta \sim 4k_B T_c$, so paramagnetic limiting is no problem.

Since there is a strong tendency for singlet pairing in materials with CuO$_2$ layers such as Ru1212 has, and substitution of Zn for Cu strongly reduces T$_s$ as in cuprates, we examine the possibility of a singlet SC (sSC) state in FM Ru1212 by considering two fundamental problems: (1) how does the SC order parameter accommodate itself to the vector potential arising from the intrinsic magnetization, and (2) what type of pairing occurs when the exchange field splits majority and minority Fermi surfaces? We find that it is primarily the latter item (exchange splitting) that presents difficulty for singlet SC in this system, and we consider briefly whether the problem can be alleviated by development of a FFLO phase [9] or by "π phase" formation. [10] We note further that triplet pairing in the RuO$_2$ layer, which appears to occur in Sr$_2$RuO$_4$, should not be forgotten. If Ru1212 is instead a triplet superconductor (in the Ru plane), it would have several exciting characteristics, such as a polarized supercurrent.

ISSUES OF COEXISTENCE

Early on, Ginzburg [6] observed that, although constant bulk magnetization M does not induce supercurrent flow, the vanishing of M at the boundaries induces surface currents that try to shield the exterior from the internal magnetic field $B_{int} = 4\pi M$ (inverse Meissner effect). This increase in energy would suppress the SC state unless the sample cross secion is not much larger than the penetration depth. In type II superconductors, however, it was observed by Krey [7] that a spontaneous vortex phase (SVP) can be formed that avoids Ginzburg's difficulty. Since that time there have been studies of the competition between FM and SC, [8] including suggestions that a SVP may have been observed, but almost always for the case where T$_m$ is less than but comparable to T$_s$.

One exception to this regime is the recent work of Felner and collaborators, [4,5] who suggest that a SVP may occur in the canted antiferromagnet $R_{1.4}Ce_{0.6}Sr_2GdCu_2O_{10}$, R = Eu and Gd. Ru1212 is different in two ways: (1) the saturation field is itself a factor of ten larger; however, this is of limited importance because Pauli limiting is not a factor in either type of material, and (2) Ru1212, being FM, has an exchange splitting of the order of 1 eV (we show below) that will induce an exchange splitting of the paired electrons in the CuO_2 planes and thereby cause additional difficulty for SC pairing.

Tallon et al. expect the magnetization M to lie in the plane, based on the magnitude of the field at the muon site observed in zero field μSR studies. [1] If this is the case, it reduces some of the coexistence questions, because orbital kinetic energy change of the electrons (or pairs) would require the (very small) c axis hopping, and thus is expected to be negligible compared to the gap energy. Then the remaining effect of the internal field is to lift the degeneracy of the up and down spin electrons (Zeeman splitting). This effect makes the up and down Fermi surfaces inequivalent and causes singlet pairs to have a net momentum $Q \equiv k_{F\uparrow} - k_{F\downarrow} \neq 0$. The result can be a state of the type first discussed by Fulde and Farrel and by Larkin and Ovchinnikov [9] (FFLO), in which the SC order parameter becomes inhomogeneous to accommodate the non-zero momentum pairs. Another way to decrease the competition between SC and FM is to form a π phase in which the SC order parameter vanishes in the FM Ru layer. [10]

Burkhardt and Rainer [11] have made an extensive study of the two dimensional superconductor in a magnetic field, and they find that an FFLO phase is preferred over a homogeneous SC state above a lower critical field B_{c1}, but only well below T_s and at relatively high field. To some extent their analysis may be applicable to Ru1212, with the change that the "magnetic field" is frozen in and arises from electronic exchange. The exchange splitting, even if small on an electronic energy scale, may correspond to a very large field, and its magnitude is one feature we address below.

MICROSCOPIC ELECTRONIC AND MAGNETIC STRUCTURE

Electronic Structure Methods

To address the question of magnetism in Ru1212 we have applied both the local density approximation (LDA) [15] and the 'semilocal' generalized gradient approximation (GGA) [16] to describe the effects of exchange and correlation. The predictions actually differ considerably, and we report only the GGA results here. Calculations were done using the linearized augmented plane wave (LAPW) method [12–14] to find the electronic and magnetic ground state, the band structure and projected densities of states (PDOS).

Crystal Structure

The structure of Ru1212, of the triple-perovskite type, is comprised of double CuO_2 (O_{Cu} site) layers separated by a Gd layer, sandwiched in turn by SrO (O_{apical} site) layers. The unit cell in completed by a RuO_2 (O_{Ru} site) layer, making it structurally related to $YBa_2Cu_3O_7$. The difference is that the CuO chain layer is replaced by a RuO_2 square planar layer, with resulting tetragonal symmetry. The oxygen variability that occurs lies primarily in the Sr layers, and there are displacements from the ideal positions [1] for all O ions, with the displacements being largest for O_{Ru}.

Discussion of Results

Gd behaves as a magnetic trivalent ion as expected, with moment very close to 7 μ_B. In expectation that the Gd moment has little effect on the electronic and magnetic behavior in the rest of the cell, we have treated only a FM alignment of Gd ions. (They actually order antiferromagnetically around 2.6 K, but antiferromagnetic order would require doubling of the unit cell with no gain of information or insight.) We have found that the electronic and magnetic structure in the cuprate and ruthenate layers does not depend on whether the Gd moment is parallel or antiparallel to the Ru moment.

We obtain a FM Ru layer, with the band structure shown in Fig. 1 and the relevant projected densities of states presented in Fig. 2. The moment (besides that on Gd) is $\sim 2.5\mu_B$ per cell, about 1.5 μ_B of which is directly assignable to the Ru ion. The remainder is spread among the six neighboring O ions, all of which are strongly polarized (for a nominally O^{2-} ion). Our total moment is substantially more than the 1 μ_B reported by Tallon *et al.* As mentioned above, they have
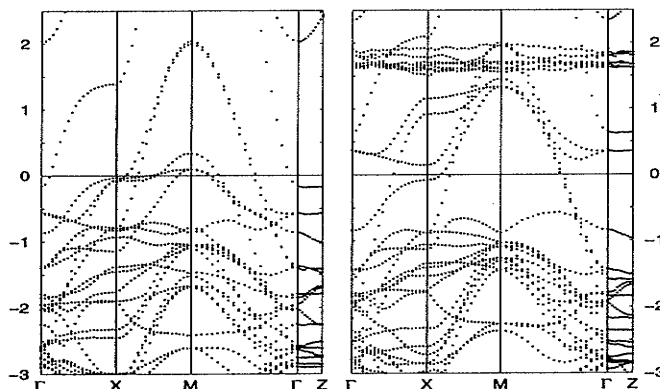

FIGURE 1. Majority (left) and minority (right) bands near E_F in tetragonal $RuSr_2GdCu_2O_8$ along high symmetry directions. Note that the distinctive Cu-O bands that reach their maximum at M lie at the same position for both spins, whereas the Ru bands are split by ~ 1 eV. The flat bands near 2 eV are the minority Gd $4f$ states.

evidence of displacements of the O ions in the Ru layer from their ideal positions by as much as 0.4 Å. We have observed in our calculations strong sensitivity of the moment to the position of the apical O and to the distance between the Ru and Cu atoms, so it is quite possible that a more realistic structure will give a moment much closer to the observed value. Stoichiometry will be important as well.

The Ru moment arises within the Ru t_{2g} subshell. The charge state is difficult to specify due to the metallic character of the Ru-O layer, but it is *not* close to Ru^{5+} and may be closer to Ru^{4+}. The exchange splitting of the Ru t_{2g} bands is 1 eV, but the induced splitting in the CuO$_2$ layers is close to two orders of magnitude smaller. The very small coupling is due to the fact that the Ru t_{2g} states couple only to the O$_{apical}$ p_x, p_y orbitals, and these to not couple either to the Cu $d_{x^2-y^2}$ or s orbitals, so the coupling path is more indirect (perhaps O$_{Ru}$-O$_{apical}$-O$_{Cu}$). The induced moment is roughly 0.01 μ_B per CuO$_2$ layer, which leads to a displacement of spin up and spin down barrel Fermi surfaces of only $\delta k_F \approx 0.01 k_F$. The work of Burkhardt and Rainer [11] suggest that a model independent feature of FFLO-like solutions is that the wavelength of modulation of the superconducting order parameter $\lambda_Q = 2\pi/\delta k_F$ must be greater than (not necessarily comparable to) the coherence length ξ. Our estimate is $\lambda_Q \sim 140a$, which is much greater than representative cuprate values $\xi \sim 5 - 15a$. Hence the exchange splitting we obtain is small enough to allow the possibility of an FFLO-like phase in Ru1212.
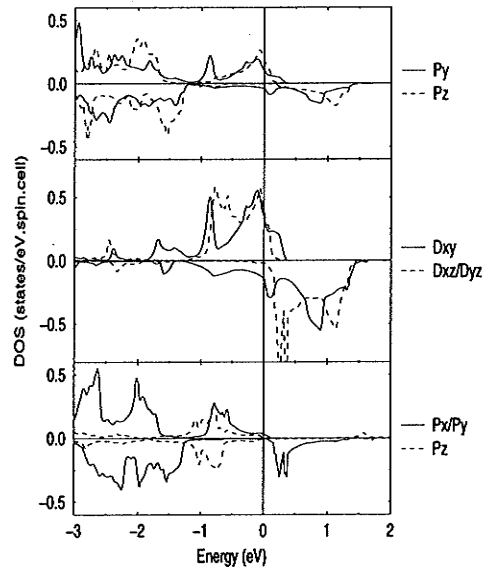

FIGURE 2. Atom projected densities of states for O$_{Ru}$ (top), Ru (middle), and O$_{apical}$ (bottom), clearly indicating spin splitting in all Ru t_{2g} states and all p orbitals of O$_{Ru}$, but in only the p_x, p_y orbitals of O$_{apical}$.

145

SUMMARY AND ACKNOWLEDGMENTS

With this paper we have begun the study of the superconducting ferromagnet Ru1212. Although magnetism in the Ru layer is strong, the exchange coupling to the Cu layers is quite small. The most likely scenario seems to be an FFLO-like inhomogeneous SC order parameter in the cuprate layers.

REFERENCES

1. J. Tallon *et al.*, unpublished, and elsewhere in this volume.
2. L. Bauernfeind, W. Widder, and H.F. Braun, J. Low Temp. Phys. **105**, 1605 (1996).
3. C. Bernhard *et al.*, cond-mat/9901084.
4. I. Felner, U. Asaf, Y. Levi, and O. Millo, Phys. Rev. B **55**, R3374 (1997).
5. E. B. Sonin and I. Felner, Phys. Rev. B **57**, R14000 (1998).
6. V. L. Ginsburg, Sov. Phys. JETP **4**, 153 (1957).
7. U. Krey, Int. J. Magnetism **4**, 153 (1973).
8. C. G. Kuper, R. Revzen, and A. Ron, Phys. Rev. Lett. **44**, 1545 (1980); O. Fischer *et al.*, Phys. Rev. Lett. **55**, 2972 (1985); T. K. Ng and C. M. Varma, Phys. Rev. Lett. **78**, 330 (1997); A. Knigavko and B. Rosenstein, Phys. Rev. B **58**, 9354 (1998).
9. P. Fulde and R. A. Ferrell, Phys. Rev. **135**, A550 (1964); A. I. Larkin and Yu. N. Ovchinnikov, Zh. Eksp. Teor. Fiz **47**, 1136 (1964) [Sov. Phys. JETP **20**, 762 (1965)].
10. V. Prokić, A. I. Budzin, and L. Dobrosavljevic-Grujic, Phys. Rev. B **59**, 587 (1999).
11. H. Burkhardt and D. Rainer, Ann. Physik **3**, 181 (1994).
12. D. J. Singh, *Planewaves, Pseudopotentials, and the LAPW Method* (Kluwer, Boston, 1994).
13. P. Blaha, K. Schwarz, and J. Luitz, WIEN97, Vienna University of Technology, 1997. Improved and updated version of the original copyrighted WIEN code, which was published by P. Blaha, K. Schwarz, P. Sorantin, and S. B. Trickey, Comput. Phys. Commun. **59**, 399 (1990).
14. The sphere radii used in fixing the LAPW basis were chosen to be 2.00 a.u for Sr and Gd, 1.9 for Ru and Cu, and 1.60 for N. Local orbitals for semicore states and for Gd $4f$ were added to the basis set for extra flexibility and to allow semicore states to be treated within the same energy window as the band states. The LAPW plane wave cutoff corresponded to energy of 14 Ryd, making the matrix dimension approximately 1400.
15. D. M. Ceperley and B. J. Alder, Phys. Rev. Lett. **45**, 566 (1980), as parametrized by J. P. Perdew and Y. Wang, Phys. Rev. B **45**, 13244 (1992) or by Vosko, Wilk and Nusair, Can. J. Phys. **58**, 1200 (1980).
16. J. P. Perdew, S. Burke, and M. Ernzerhof, Phys. Rev. Lett. **77**, 3865 (1996).

HIGH TEMPERATURE SUPERCONDUCTIVITY

Coral Gables, Florida   January 1999

EDITORS
Stewart E. Barnes
Joseph Ashkenazi
Joshua L. Cohn
Fulin Zuo
University of Miami, Coral Gables