Superconductivity in ferromagnetic $RuSr_2GdCu_2O_8$

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The phase diagram of temperature versus exchange field is obtained within a BCS model for *d*-wave superconductivity in CuO_2 layers which is coupled to ferromagnetic RuO_2 layers in $RuSr_2GdCu_2O_8$. It is found that the Fulde-Ferrell-Larkin-Ovchinnikov state is very sensitive to the band filling factor. For strong exchange field, we point out that superconductivity could only exist in the interfaces between ferromagnetic domains. The magnetization curve is calculated and its comparison with experiment is discussed. We also propose the measuring of tunneling conductance near a single unitary impurity to detect the strength of the exchange interaction.

The problem of coexistence of superconductivity (SC) and ferromagnetism (FM) has attracted keen interest since the original works of Ginzburg[1] and Matthias *et al.*[2] It was shown that singlet SC and FM are mutually exclusive and the SC can also be strongly suppressed by magnetic impurities. The competition between SC and FM were observed in the ternary compounds, $HoMo_6S_8$, $HoMo_6Se_8$, and $ErRh_4B_4$.[3] But true microscopic coexistence was found only over a narrow temperature region when FM sets in and modifies itself to a spiral or domainlike structure. The recent discovery of SC ($T_c = 16-47$ K) in the ferromagnetic ($T_M = 132$ K) ruthenate-cuprate layered compound $RuSr_2GdCu_2O_8$ (Ru-1212) (Refs. 4–10) renewed the interest in the issue of how SC and FM negotiate to coexist. Recent band-structure calculation performed by Pickett *et al.*[11] showed that the exchange splitting in the CuO_2 layer is small ($\Delta_{exc} = 25$ meV) compared to ~ 1 eV in the RuO_2 layer but is larger enough that the superconducting state in the CuO_2 layer may be of the Fulde-Ferrell-Larkin-Ovchinnikov (FFLO)-type[12,13] or finite momentum pairing state. Whether there actually exists such a pairing state even in early found magnetic superconductors remains to be controversial.[14] Indeed, until now no superconductor has been discovered to be a finite momentum pairing state. Also notice that, in earlier studies, the FFLO state was discussed by assuming a constant density of states (DOS), that is, the exchange field does not introduce structures in the electronic properties on the energy scales relevant to SC. However, this becomes not true when the DOS has singular structure. Thus a further study on the existence of this state by including the energy dependence of DOS near the Fermi surface should be interesting. Moreover, it was supposed that the SC mainly occurs in the CuO_2 layers in Ru-1212, the identification of the pairing symmetry in this material is also of fundmental importance in view of the well-established *d*-wave pairing symmetry in high-T_c cuprate superconductors, which have similar crystal structures.

The purpose of this paper is threefold: (i) By assuming a *d*-wave pairing symmetry in a two-dimensional (2D) lattice model, we present a detailed study of the temperature-exchange field phase diagram to investigate the sensitivity of the FFLO state by varying the position of the Fermi energy within the tight-binding band; (ii) arguing that the mixed state is intrinsic, we calculate the magnetization as a function of an applied magnetic field and compare it with experiment; (iii) we propose to measure the existence of the zero-energy peak (ZEP) and its splitting in the differential tunneling conductance near a single unitary impurity in the CuO_2 layer as a test of the *d*-wave pairing symmetry as well as the strength of the exchange field.

Our model system is defined on a 2D lattice with pairing interaction taking place between two electrons on the nearest-neighbor sites, which in the mean-field approximation leads to the Bogoliubov–de Gennes equations

$$\sum_j \begin{pmatrix} H_{ij,\sigma} & \Delta_{ij} \\ \Delta_{ij}^* & -H_{ij,\bar{\sigma}} \end{pmatrix} \begin{pmatrix} u_{j\sigma}^n \\ v_{j\bar{\sigma}}^n \end{pmatrix} = E_n \begin{pmatrix} u_{i\sigma}^n \\ v_{i\bar{\sigma}}^n \end{pmatrix}, \tag{1}$$

with the single particle Hamiltonian $H_{ij,\sigma} = -t\delta_{i+\gamma,j} - \mu\delta_{ij} - \sigma h_{exc}\delta_{ij} + U_i\delta_{ij}$, and the self-consistent condition $\Delta_{ij} = V/4\Sigma_{n,\sigma}(u_{i\sigma}^n v_{j\bar{\sigma}}^{n*} + u_{j\bar{\sigma}}^n v_{i\sigma}^{n*})\tanh(E_n/2k_BT)$. Here $(u_{i\sigma}, v_{i\bar{\sigma}})$ are the Bogoliubov quasiparticle amplitudes on the ith site; $\gamma = \pm\hat{x}, \pm\hat{y}$ represents the relative position of sites nearest neighboring to the ith site; t is the effective hopping integral between two nearest-neighbor sites within the CuO_2 plane; μ is the chemical potential; $h_{exc} = J(\langle S_z^a\rangle + \langle S_z^b\rangle)$ is the exchange field coming from the ordered spins in the two nearest-neighboring ferromagnetic RuO_2 layers; U_i if any accounts for the scattering from the impurities; V is the strength of the nearest-neighbor pairing interaction. Notice that the internal magnetic field on CuO_2 layers due to the magnetic moment on RuO_2 layers is only several hundred gauss and the exchange interaction should be dominant in suppressing SC. We therefore defer the effect of the internal field on the orbital motion of paired electrons to the study of the magnetization.

Temperature–exchange-field phase diagram. For a pure system in the absence of external magnetic field, the Bogoliubov quasiparticle amplitude can be generally written as

$$\begin{pmatrix} u_{i\sigma}^n \\ v_{i\bar{\sigma}}^n \end{pmatrix} = \frac{1}{\sqrt{N}} \begin{pmatrix} u_{\mathbf{k}+\mathbf{q},\sigma} e^{i(\mathbf{k}+\mathbf{q})\cdot\mathbf{R}_i} \\ v_{\mathbf{k}-\mathbf{q},\bar{\sigma}} e^{i(\mathbf{k}-\mathbf{q})\cdot\mathbf{R}_i} \end{pmatrix},$$

which yields the bond order parameter

$$\Delta_{ij}=\frac{V}{2N}e^{i\mathbf{q}\cdot(\mathbf{R}_i+\mathbf{R}_j)}\sum_{\mathbf{k}}\cos[\mathbf{k}\cdot(\mathbf{R}_j-\mathbf{R}_i)]\times[u_{\mathbf{k}+\mathbf{q},\uparrow}v^*_{\mathbf{k}-\mathbf{q},\downarrow}+u_{\mathbf{k}+\mathbf{q},\downarrow}v^*_{\mathbf{k}-\mathbf{q},\uparrow}]\tanh(E_{\mathbf{k},\mathbf{q}}/2T)=\Delta^{(0)}_{\delta}(i)e^{i\mathbf{q}\cdot(\mathbf{R}_i+\mathbf{R}_j)}. \quad (2)$$

Here $N=N_x\times N_y$ is the number of two-dimensional lattice sites. Equation (2) shows that the order parameter is not a constant in space manifesting the collective motion of paired electrons each with momentum $\mathbf{q}$. Using the definition $\Delta_d=(\Delta^{(0)}_{\hat{x}}+\Delta^{(0)}_{-\hat{x}}-\Delta^{(0)}_{\hat{y}}-\Delta^{(0)}_{\hat{y}})/4$, we find the equation determining the d-wave energy gap

$$1=\frac{V}{4N}\sum_{\mathbf{k},\sigma}\frac{(\cos k_x-\cos k_y)^2}{E^{(0)}_{\mathbf{k},\mathbf{q}}}\tanh\frac{E^{(1,\sigma)}_{\mathbf{k},\mathbf{q}}}{2T}, \quad (3)$$

where $E^{(0)}_{\mathbf{k},\mathbf{q}}=\sqrt{Z^2_{\mathbf{k},\mathbf{q},+}+|\Delta_{\mathbf{k}}|^2}$, and $E^{(1,\sigma)}_{\mathbf{k},\mathbf{q}}=-\sigma h_{\mathrm{exc}}+Z_{\mathbf{k},\mathbf{q},-}+E^{(0)}_{\mathbf{k},\mathbf{q}}$, with $\xi_{\mathbf{k}}=-2t(\cos k_x+\cos k_y)-\mu$, $Z_{\mathbf{k},\mathbf{q},\pm}=(\xi_{\mathbf{k}+\mathbf{q}}\pm\xi_{\mathbf{k}-\mathbf{q}})/2$, and $\Delta_{\mathbf{k}}=2\Delta_d(\cos k_x-\cos k_y)$. Correspondingly, the free energy per lattice site is given by

$$\mathcal{F}=\frac{1}{2N}\sum_{\mathbf{k},\sigma}\Bigg\{(\xi_{\mathbf{k}+\mathbf{q}}-\sigma h_{\mathrm{exc}})\left(1+\frac{Z_{\mathbf{k},\mathbf{q},+}}{E^{(0)}_{\mathbf{k},\mathbf{q}}}\right)f^{\sigma}_{\mathbf{k},\mathbf{q}}+(\xi_{\mathbf{k}-\mathbf{q}}+\sigma h_{\mathrm{exc}})\left(1-\frac{Z_{\mathbf{k},\mathbf{q},+}}{E^{(0)}_{\mathbf{k},\mathbf{q}}}\right)[1-f^{\sigma}_{\mathbf{k},\mathbf{q}}]+2T[(1-f^{\sigma}_{\mathbf{k},\mathbf{q}})\ln(1-f^{\sigma}_{\mathbf{k},\mathbf{q}})+f^{\sigma}_{\mathbf{k},\mathbf{q}}\ln f^{\sigma}_{\mathbf{k},\mathbf{q}}]\Bigg\}-\frac{4}{V}|\Delta_d|^2, \quad (4)$$

where $f^{\sigma}_{\mathbf{k},\mathbf{q}}=f(E^{1,\sigma}_{\mathbf{k},\mathbf{q}})$ is the Fermi distribution function.

To determine the phase boundary between the normal pairing ($\mathbf{q}=0$) state and the normal state (spin polarized), one should compare the free energies of the superconducting state and normal state, using Eq. (4). In the presence of a fairly strong exchange interaction, the system might also go into the FFLO state in which all the Cooper pairs have a single nonvanishing ($\mathbf{q}\neq 0$) center-of-mass momentum. This transition between the FFLO state and the normal state would be of the second order. To find the transition curve for the FFLO state and the normal state, we solve Eq. (3) with $\Delta_d=0$ to find the maximum value of h_{exc} by scanning through a whole set values of $\mathbf{q}$ at the same temperature T. By repeating the same calculation at a different value of T, we then obtain the phase curve h_{exc} as a function of T.

To see the sensitivity of the FFLO state to the Fermi energy position in a 2D tight-binding band, we fix the pairing interaction as $V=2t$ and consider three typical values of the chemical potential, $\mu=-t$, $-0.5t$, and $-0.14t$, corresponding to the band-filling factor $\nu\approx 0.65$, 0.82, and 0.95 ($\nu=1$ is a half filled band). For the above sets of parameters, the maximum energy gap at zero temperature and $h_{\mathrm{exc}}=0$ is found to be $\Delta_0=4\Delta_d\approx 0.65t$, $0.88t$, and $0.96t$, and correspondingly, the transition temperature is $T_{c0}\approx 0.26t$, $0.37t$, and $0.40t$, respectively. Figure 1 plots the temperature–exchange-field phase diagram. Our calculation shows that

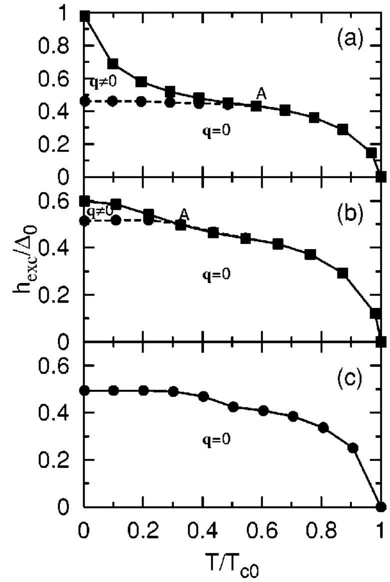

FIG. 1. The T-h_{exc} phase diagram for a d-wave superconductor with $\mu=-t$ (a), $-0.5t$ (b), and $-0.14t$ (c). The solid line represents the physical phase boundary between the superconducting state and the normal state. In panels (a) and (b), the dashed line indicates the transition between the FFLO state and the normal pairing ($\mathbf{q}=0$) state. Note that the FFLO state exists only as the temperature is below the tricritical point A. In panel (c), only the transition between the normal pairing state and the normal state is allowed.

the momentum $\mathbf{q}$ corresponding to the maximum h_{exc} is along the (10) and its equivalent direction in the whole temperature region because the energy gap reaches the maximum value along these directions so that the system can be more robust against the depairing effect from both the finite momentum and the exchange field. As shown in Fig. 1(a), when $\mu=-t$, the transition curve (solid line) is between the superconducting state (with $\mathbf{q}=0$ and $\mathbf{q}\neq 0$) and the normal state, which shows that at low temperatures, when the exchange field is increased the system initially in the normal pairing state will enter the FFLO state ($\mathbf{q}\neq 0$) through the first-order transition and then pass into the normal state by a second-order transition. Thus the FFLO state is a stable state at high exchange fields. The transition curve between the normal pairing state and the FFLO state is represented by dashed line. When the temperature is increased, two curves become closer and at $T=0.58T_{c0}$ (tricritical point) they coincide with each other, where the transition begins to be of second order and the FFLO state merges naturally to the $\mathbf{q}=0$ normal pairing state. This result is similar to that obtained within the continuum model using a constant DOS.[12,13] However, we find that the phase space for the existence of the FFLO state shrinks at $\mu=-0.5t$ as the band-filling factor shifts toward the half filling [Fig. 1(b)]. In particular, near half filling when $\mu=-0.14t$, the FFLO state will be unstable and appear as a supercooling state. Therefore as shown in Fig. 1(c), only the transition between the normal pairing state and the normal state is physically acceptable. The interesting feature comes from the influence of the exchange field on the DOS near the Fermi surface. For a 2D tight-binding band, the DOS at the energy $\mu=0$ has a singular point and it decays logarithmically as the energy goes away from zero. When $\mu=-t$, which has been far away from the zero-energy point, the DOS is flat for the energy near the Fermi surface. In this case, the splitting of the normal electron band by the exchange field has little effect on the change of the DOS and a constant DOS can be taken which corresponds to the approximation made in the continuum theory.[12,13] But as the chemical potential is close to the zero-energy point, a little splitting of the energy band will cause a strong variation of the DOS near $\mu=0$, which makes the $\mathbf{q}\neq 0$ pairing state unfavorable.

For the Ru-1212, the band-structure calculation estimated the Fermi wave vector as π/a. The filling factor should be near the half filling ($\mu=0$). Then the existence of the FFLO state in the above system becomes unlikely according to Fig. 1(c). In the following discussion, we focus on the transition as between the normal pairing state and the normal state. Consider the case of $\nu=0.95$ [Fig. 1(c)]. If the zero-field transition temperature is assumed to be $T_{c0}=90$ K, Δ_0 is about 18.7 meV. By fitting to the transition temperature $T_s \sim 36$ K as measured experimentally,[5] the exchange field should be as small as 8.8 meV. Actually, T_{c0} may be as low as 60 K when Ru is replaced by other atoms (e.g., Cu).[15] Then the allowable h_{exc} is only 5.1 meV to have $T_s \sim 36$ K. Notice that the exchange field h_{exc} from the band-structure calculation is as large as 12.5 meV,[11] and the experimental filling is very close to $\nu=1$ (or $\mu=0$); we therefore conclude that, if h_{exc} is indeed so large, SC can only exist in the CuO_2 layers between the ferromagnetic domains where the exchange interactions $h_{\text{exc}}=J(\langle S_z^a\rangle+\langle S_z^b\rangle)$ are small. It is important to emphasize that we have not ruled out the possibility of the FFLO state in the real systems because the estimated value of exchange interaction could in fact vary over a limited range depending on the method of obtaining it. In case that the real exchange interaction is somewhat smaller than that estimated in Ref. 11, the bulk FFLO state would become a reality.

Magnetization. The bulk Meissner effect has not been observed in the superconducting state of Ru-1212 (Ref. 8) until very recently.[9] Furthermore, even the absence of superconductivity has been reported in well characterized Ru-1212 samples.[16] This discrepancy indicates the delicate balance between the superconducting and ferromagnetic interactions and the experimental result appears to depend critically on the sample condition in a yet-to-be determined fashion. It is reported that the magnetic moment in each Ru atom is 1 Bohr magneton.[4] With structure parameter values, $a=3.8$ Å, and $c=11.4$ Å, the internal magnetic field $H_{int}=4\pi M_{sp}=1\mu_B/a^2c$ (M_{sp} is the spontaneous magnetization) is estimated to be 707 G in the CuO_2 layers which is larger than the first critical field $H_{c1}^{(0)}\sim 100$ G of a non-Ru layered cuprate superconductor with the comparable transition temperature, i.e., $H_{c1}^{(0)}-4\pi M_{sp}<0$. The measured H_{c1} is therefore zero and no bulk Meissner effect can be observed. Instead the superconductivity occurring in the CuO_2 layers has been driven into the mixed state. The overall magnetization in the system consists of two parts, one from the spontaneous magnetization M_{sp} of the ferromagnetic RuO_2 layers, the other M_{ob} from the diamagnetic orbital contribution of the superconducting CuO_2 layers in the mixed state, i.e., $M=M_{sp}+M_{ob}$. When an external magnetic field H_{ext} is applied, the effective magnetic field is $H=H_{ext}+H_{int}$. As an approximation, we work with the London equation for a square vortex lattice to find the magnetic induction $B\approx H-H_{c1}\ln(H_{c2}/B)/\ln\kappa$,[17] where κ is the Ginzburg-Landau parameter and $H_{c2}=2\kappa^2H_{c1}$ is the upper critical field for the CuO_2 subsystem. In Fig. 2, the total magnetization is plotted as a function of H_{ext}. As it is shown, the magnetization increases monotonically with the external magnetic field. The observed monotonic behavior of M by the experiment[8] is shown in the inset of Fig. 2. Because our study is based upon the SC in a single ferromagnetic domain, the internal magnetic field must be greater than the intrinsic first critical field H_{c1} of the CuO_2 SC. Nevertheless, when $H_{ext}=0$, our calculation gives an appreciable spontaneous magnetization which contradicts the experimentally measured zero magnetization.[8] This difference shows the existence of ferromagnetic domains in Ru-1212, where the average magnetization vanishes in the absence of an external magnetic field and the SC occurs in the interfacial regions between some of the ferromagnetic domains. Note that the magnetization has also been recently studied[18] to analyze the superconducting properties of $R_{1.5}Ce_{0.5}RuSr_2Cu_2O_{10}$, where the Meissner effect was absent at temperature region $T_d<T<T_c$ ($T_c\sim 30$ K and $T_d\sim 20$ K) but present at $T<T_d$. To interpret this phenomenon, it was proposed that the ferromagnetism appears with a domain structure but the superconductivity is a bulk phase, which is different from our explanation for Ru-1212 systems, where neither Meissner effect (down to 0.5 G) nor detectable condensation energy was observed at temperature down to 2 K.[8] Very recently, the detection of Meissner state at a field below 30 Oe was reported by Bernhard *et al.*[9] In the reported sample with a Meissner effect, the internal field was estimated to be only about 50–70 Oe (the lower critical field H_{c1} of the nonmagnetic superconductor was estimated to be of the order 80–120 Oe), in contrast to the previous reported H_{int} about 200–700 Oe by the same group and others. We argue that the experimental result of Bernhard *et al.* might make sense if the superconductivity occurs in the interferromagnetic domain region where H_{int} is much reduced and the ratio R between superconducting volume/sample volume is not too small. This would not be inconsistent with the conclusion reached for Ref. 8, where R could be rather small so that the Meissner effect was not detected.

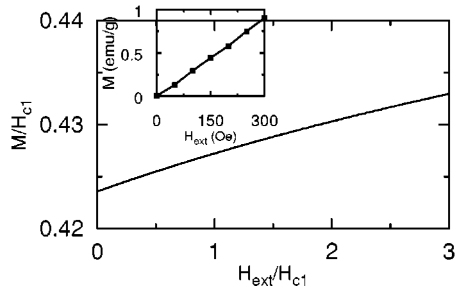

FIG. 2. Magnetization as a function of the external magnetic field. The parameter values: $H_{c1}=100$ G, $H_{int}=707$ G, and $\kappa=50$. The inset shows the measured magnetization (Ref. 8).

Quasiparticle resonant state near a single unitary impurity in CuO_2 layers. Since the ferromagnetic exchange field has pre-existed in the above system, it can affect the quasiparticle resonant states near a single impurity in the case of

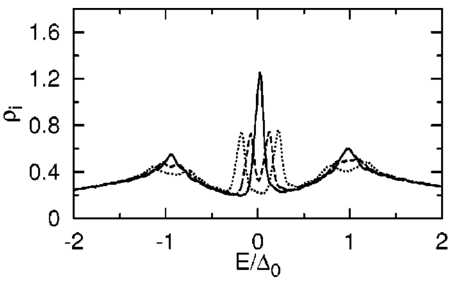

FIG. 3. Local density of states as a function of energy for $h_{\rm exc}=0$ (solid line), $0.1\Delta_0$ (dashed line), $0.2\Delta_0$ (dotted line) at the site one lattice constant away from the impurity site.

d-wave pairing symmetry. To address both issues, we solve Eq. (1) self-consistently using the exact-diagonalization method[19] and calculate the local density of states (LDOS) $\rho_i=-\Sigma_{n,\sigma}[|u_{i\sigma}|^2 f'(E-E_n)+|v_{i\bar{\sigma}}|^2 f'(E+E_n)]$. The calculation was made on 6×6 supercells each with size $35a\times 35a$ by assuming a paramagnetic pairing state. The parameters are as follows: The single-site impurity strength $U_0=100t$, $\nu=0.95$, and $T=0.02t$. From Eq. (1), we can see that the zero-field quasiparticle energy $E^{(0)}$ is shifted to $E=E^{(0)}\pm h_{\rm exc}$. Therefore the position of zero-energy states is now split to $\pm h_{\rm exc}$. Figure 3 plots the LDOS on the site nearest neighboring to the impurity site. As is shown, when $h_{\rm exc}=0$, there occurs a single ZEP in the LDOS. In the presence of exchange field, the ZEP is split into double peaks, each corresponding to one spin component. The magnitude of the splitting increases with the exchange field. Since the local differential tunneling conductance is proportional to ρ_i, the ZEP and its splitting can be detected by the scanning tunneling spectroscopy (STS) as a test of the *d*-wave symmetry as well as the pre-existing exchange field in the above system. Experimentally, the nonmagnetic impurity with strong scattering potential can be realized by substitution of Zn for Cu in the CuO_2 layers. In addition, the STS best suited for exploring the local electronic properties allows a direct examination of whether the SC in Ru-1212 appears as a bulk state or can only survive at the boundaries between ferromagnetic domains. For the former, the STS data should reveal a superconducting gap over the whole sample.

Finally, we would like to mention once again that the bulk SC in Ru-1212 prevails only when the exchange interaction is weak. For large exchange interaction, the SC could only exist at the interfacial CuO_2 layers between ferromagnetic domains, where $h_{\rm exc}$ is small. Whether the SC in this compound is bulk or interfacial like appears to depend on sample preparation and this issue needs further experimental studies.

Note added: Recently, we noticed a preprint by Shimahara and Hata[20] in which the enhancement of the possible FFLO state in a layered ferromagnetic compound such as Ru-1212 by the next-nearest-neighbor hopping was discussed.

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