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SOLIDS
Electronic Properties

Microscopic Model of the Coexistence of Superconductivity and Ferromagnetism in the Hybrid Ruthenate-Cuprate Oxide $RuSr_2GdCu_2O_8$

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Abstract—A microscopic *t–J–I* model with competing antiferromagnetic (*J*) and ferromagnetic (*I*) exchange interactions is proposed for strongly correlated electrons in $RuSr_2GdCu_2O_8$. It is assumed that $J \gg I$ for CuO_2 layers and $J \ll I$ for RuO_2 layers. A superconducting solution of $d_{x^2-y^2}$ symmetry was obtained for the CuO_2 layers while competition between ferromagnetism and *p*-type triplet superconducting pairing is obtained for RuO_2 layers. It is shown that the RuO_2 layers have a lower carrier concentration in the Hubbard subband formed by coupled $((d_{xy}–p)–\pi)$ orbitals compared with a bulk Sr_2RuO_4 crystal, which leads to stabilization of the ferromagnetic state in the RuO_2 layer. © *2000 MAIK "Nauka/Interperiodica".*

1. INTRODUCTION

In the present paper we propose a microscopic model of the electronic structure, electrical and magnetic properties of recently synthesized $RuSr_2GdCu_2O_8$ layered oxides in which the coexistence of ferromagnetism and superconductivity is observed [1, 2].

As a result of the structural similarity of ruthenate Sr_2RuO_4 and copper oxides, it has been possible to construct a hybrid compound $RuSr_2GdCu_2O_8$ in which the RuO_2 and CuO_2–Gd–CuO_2 bilayer are separated by SrO buffer layers where the RuO_2 layers are responsible for ferromagnetism below the Curie point $T_C = 133$ K and the superconductivity below $T_c = 46$ K is attributed to the CuO_2 layers. However, the bulk Sr_2RuO_4 crystal does not exhibit magnetic properties and is a superconductor with a low value of $T_c \approx 1$ K [3] and *p*-type pairing [4]. A characteristic feature of Sr_2RuO_4 is its closeness to ferromagnetic instability which is observed in the ferromagnetism of $Sr_2Ir_{1-x}Ru_xO_4$ [5] and the Ruddelsdon–Popper series $Sr_{n+1}Ru_nO_{3n+1}$ with $n \geq 2$ [6, 7].

The coexistence of ferromagnetism and superconductivity has been discussed on many occasions, beginning with [8, 9]. There are two factors which destroy the superconductivity in a ferromagnetic medium: first, exchange splitting lifts the energy degeneracy of the partners of a spin-up and spin-down Cooper pair; second, magnon exchange leads to repulsion for a singlet pair [10] (although for triplet pairs this exchange gives attraction additional to the phonon mechanism [11]). In the compound being discussed, $RuSr_2GdCu_2O_8$, coexistence occurs below T_c and the superconductivity is not destroyed in the ferromagnetic state because of the spatial separation of the CuO_2 and RuO_2 layers by SrO buffer layers which suppress the relative influence of the magnetic and superconducting layers. This means that to a first approximation the superconductivity in the CuO_2 layers and the ferromagnetism in the RuO_2 layers can be considered independently. In this approach the main question to explain the properties of $RuSr_2GdCu_2O_8$ is as follows: why does the RuO_2 layer exhibit ferromagnetic properties while the Sr_2RuO_4 is a superconductor?

Band calculations of Sr_2RuO_4 using the density functional method in the local density approximation (LDA) give a nonmagnetic ground state [12, 13] whereas GGA calculations using gradient corrections to the density functional yield a ferromagnetic Sr_2RuO_4 ground state [14]. The effective masses obtained in the band calculations differ by almost an order of magnitude from the masses observed in experiments on the de Haas–van Alfven effect [15] which suggests that we need to allow for strong electron correlations in the RuO_2 layers. For cuprates the important role of strong electron correlation effects is also widely known (see, for example, the review [16]). Thus, in the present study we shall use the *t–J–I* model which explicitly allows for strong electron correlations and was proposed in [17] to compare the superconductivity in cuprates and ruthenates. This model is generalized to the case of $RuSr_2GdCu_2O_8$ by having one set of parameters (*t*, *J*, *I*) for the CuO_2 layer and another set of the same parameters for the RuO_2 layer.

The study has the following structure: in Section 2 we describe the *t–J–I* model and its specific characteristics for $RuSr_2GdCu_2O_8$, in Section 3 we describe the superconductivity of CuO_2 layers, in Section 4 we analyze the competing ferromagnetic and superconducting states of the RuO_2 layer, and Section 5 contains a discussion of the results.

2. GENERALIZATION OF *t–J–I* MODEL FOR $RuSr_2GdCu_2O_8$

This compound, like oxides of copper and ruthenium, is characterized by a mixed type of chemical bond, with an ionic bond predominating in which all ions entering the lattice participate. The weaker metallic bond can be described using the multiband *p–d* model which allows for the e_g ($d_{x^2-y^2}$ and d_{z^2}) orbitals of copper, the t_{2g} (d_{xy}, d_{yz}, d_{zx}) orbitals of ruthenium, all the *p*-orbitals of the oxygen ions, *p–d* hopping between them, and also for the Coulomb matrix elements at copper U_{Cu} and ruthenium U_{Ru}. In this model the Cd^{3+} and SrO layers all play a passive role.

In the weak correlation regime $U_{Cu} \ll W_{Cu}$, $U_{Ru} \ll W_{Ru}$, where W_i is the band half-width in the layers i = Cu, Ru, a single-electron approach works and the band structure can be calculated from first principles. Such calculations were recently made in [18, 19] which confirmed that Gd^{3+} having the magnetic moment $\mu \approx 7\mu_B$ does not influence the band structure of the CuO_2 and RuO_2 layers. In these studies it was shown that the electronic structure of the CuO_2 layer is almost independent of the magnetic state of the RuO_2 layer; despite the strong exchange splitting (~1 eV) of the spin subbands in the ferromagnetic RuO_2 layer, the induced exchange splitting in the CuO_2 layer is small, $\Delta_{ex} \approx 25$ eV, which is attributed to characteristics of the crystal, magnetic, and electronic structures of $RuSr_2GdCu_2O_8$. In fact the magnetic moment of Ru is produced by the t_{2g} orbitals which are bound to the p_x, p_y orbitals of apical oxygen by an extremely weak $pd\pi$ bond and have a zero overlap with the p_z orbitals of apical oxygen. In turn, the $p\pi$ orbitals of apical oxygen are not bound to the copper $d_{x^2-y^2}$ and 4*s* orbitals. All these factors have the result that the electronic structures of the CuO_2 and RuO_2 layers may be considered as isolated layers to a first approximation.

The electron correlations in the band approach [18, 19] were taken into account using a generalized gradient approximation which cannot be used to describe the electronic structure of CuO_2 and RuO_2 layers over a wide range of concentrations, including the dielectric in the undoped CuO_2 layer in addition to the metallic phases. It is therefore interesting to consider a model for the case of strong intraatomic correlations $U_{Cu} \gg W_{Cu}$, $U_{Ru} \gg W_{Ru}$. Although both layers (CuO_2 and RuO_2) are metallic in this compound, the carrier concentration in these layers, as will be shown below, corresponds to weakly doped compositions in which strong electron correlation effects give substantial deviations from Fermi liquid behavior [16].

In the strong correlation regime the three-band *p–d* model at low quasiparticle energies reduces to an effective *t–J* model [20–23]. The correlations raise the energies of the two-particle polar states, eliminating these from the low-energy range. Mathematically this elimination leads to the constraint that part of the Hilbert space is inaccessible as a result of the zero occupation numbers of the polar states. As a result for the CuO_2 layer we obtain the effective *t–J* Hamiltonian (additional hopping between second neighbors *t*' and third neighbors *t*'' can also be taken into account in this approach, but from the point of view of a qualitative comparison of CuO_2 and RuO_2 layers they are unimportant in the present study):

$$H_{CuO_2} = \sum_{\mathbf{f}\sigma}(\varepsilon_{Cu} - \mu)X_{\mathbf{f}}^{\sigma\sigma} - t_{Cu}\sum_{\mathbf{f}\boldsymbol{\delta}\sigma}X_{\mathbf{f}}^{\sigma 0}X_{\mathbf{f}+\boldsymbol{\delta}}^{0\sigma} + J_{Cu}\sum_{\mathbf{f}\boldsymbol{\delta}}\left(\mathbf{S}_{\mathbf{f}}\cdot\mathbf{S}_{\mathbf{f}+\boldsymbol{\delta}} - \frac{1}{4}\hat{n}_{\mathbf{f}}n_{\mathbf{f}+\boldsymbol{\delta}}\right). \quad (1)$$

Here $\mathbf{f}$ are the lattice sites, $\boldsymbol{\delta}$ is a vector joining the nearest neighbors of the site, $X_{\mathbf{f}}^{mn} = |m\rangle\langle n|$ are the Hubbard operators, $\mathbf{S}_{\mathbf{f}}$ is the spin operator, $\hat{n}_{\mathbf{f}}$ is the electron number operator, ε_{Cu} is the energy of the single-hole state, μ is the chemical potential, t_{Cu} is the hopping integral between nearest neighbors, and $J_{Cu} = 4t_{Cu}^2/U_{Cu}$ is the indirect antiferromagnetic exchange. For hole-doped superconductors the local basis of the *t–J* model includes the states: $|0\rangle$, a Zhang–Rice singlet consisting of two holes (number of electrons $n_e = 0$) and the single-hole states $|\sigma\rangle$, $\sigma = \pm 1/2$ (number of electrons $n_e = 1$). The constraint which eliminates local two-electron states has the form

$$X_{\mathbf{f}}^{00} + \sum_{\sigma}X_{\mathbf{f}}^{\sigma\sigma} = 1. \quad (2)$$

Typical values of the model parameters for the CuO_2 layer are: $(J/t)_{Cu} = 0.4$, and the parameter t is numerically small compared with the copper–oxygen *p–d* hopping integral $t \sim 0.1t_{pd}$ [21]. For the typical value $t_{pd} = 1$ eV we obtain $t_{Cu} \sim 0.1$ eV. For these values of t_{Cu} and J_{Cu} in the exchange pairing mechanism [24, 25] we obtain $T_c \sim 100$ K.

No similar derivation of the effective Hamiltonian for the RuO_2 layer where the Ru layers have a d^4 electron configuration has been reported in the literature although it is qualitatively clear from a general approach to the derivation of the effective spin Hamil-

tonian eliminating polar states as a result of strong correlations that the hopping part of the Hamiltonian has a form similar to the term containing t_{Cu} in Eq. (1) and the exchange interaction for a d^4 configuration is determined by two contributions: ferromagnetic 180° cation–anion–cation superexchange I (Fig. 1) and antiferromagnetic cation–cation exchange $J \sim 4t_{Ru}^2/U_{Ru}$ [26]. As a result, we arrive at the Hamiltonian of the t–J–I model introduced in [17]:

$$H_{RuO_2} = \sum_{\mathbf{f}\sigma}(\varepsilon_{Ru}-\mu)X_{\mathbf{f}}^{\sigma\sigma} - t_{Ru}\sum_{\mathbf{f}\delta\sigma}X_{\mathbf{f}}^{\sigma 0}X_{\mathbf{f}+\delta}^{0\sigma} + J_{Ru}\sum_{\mathbf{f}\delta}\left(\mathbf{S}_{\mathbf{f}}\cdot\mathbf{S}_{\mathbf{f}+\delta}-\frac{1}{4}\hat{n}_{\mathbf{f}}\hat{n}_{\mathbf{f}+\delta}\right) - I_{Ru}\sum_{\mathbf{f}\delta}\left(\mathbf{S}_{\mathbf{f}}\cdot\mathbf{S}_{\mathbf{f}+\delta}+\frac{1}{4}\hat{n}_{\mathbf{f}}\hat{n}_{\mathbf{f}+\delta}\right). \quad (3)$$

For the RuO_2 layer the Hamiltonian (3) describes strongly correlated γ-band electrons formed by intraplanar hybridized (d_{xy}–p)–π states. Here $|0\rangle$ and $|\sigma\rangle$ are local (d_{xy}–p)–π states of the RuO_4 cell having the numbers of holes $n_h = 9$ and $n_h = 1$, and two-hole states of the form $|\uparrow, \downarrow\rangle$ do not fall within the low-energy range. Since the d^4 configuration contains two holes (measured from the vacuum term of the d^6 configuration), the second hole is situated in a different orbital state. According to band calculations [12, 13] for Sr_2RuO_4 this hole belongs to the half-filled β-band. For a periodic multilayer superlattice such as $RuSr_2GdCu_2O_8$ essentially is, tunneling across layers involving α- and β-band electrons formed by hybrid (d_{yz}–p) and (d_{zx}–p) orbitals is very weak and the role of strong correlation effects for these states and also for their localization is enhanced compared with a three-dimensional Sr_2RuO_4 crystal.

Typical values of the parameters for an RuO_2 layer are as follows: $t_{Ru} \sim 0.1$ eV for the same reasons as t_{Cu} (independent confirmation of this value of t_{Ru} is obtained from the value of the effective mass $m_\gamma \sim 10m_0$ in the Hubbard I approximation for the lattice parameter of the RuO_2 layer). For $U_{Ru} = 4$ eV the antiferromagnetic exchange value is $(J/t)_{Ru} = 0.1$ or $J_{Ru} = 0.01$ eV. The ferromagnetic exchange parameter can be estimated using the spin-wave formula for the Curie temperature of a quasi-two-dimensional magnetic substance:

$$T_C \sim zI/\ln(I/I_\perp), \quad (4)$$

where z is the number of nearest neighbors and for the unknown ratio of the exchanges within the RuO_2 layer (I) and between the layers ($I_\perp$) we can assume on the basis of the crystal chemistry similarity of the structures of ruthenates and cuprates that by analogy with cuprates $I/I_\perp \sim 10^4$ [quite clearly as a result of the logarithm in formula (4) the exponent in this ratio very weakly influences the result of the estimate). For $I_{Ru}/t_{Ru} \sim 0.4$ we obtain $T_C \sim 100$ K which is in good agreement with the experimental value of $T_C = 133$ K. According to the Good-enough rules [26] for indirect 180° exchange d^4–anion–d^4 ferromagnetic exchange predominates over antiferromagnetic $I_{Ru} > J_{Ru}$ as was obtained in our estimate of the parameters. The value of I_{Ru} can also be estimated independently using the temperature of the superconducting transition in Sr_2RuO_4, $T_c \sim 1$ K which is obtained for $(I/t)_{Ru} = 0.4$ and $t_{Ru} = 0.1$ eV within the limits of the triplet pairing mechanism [17].

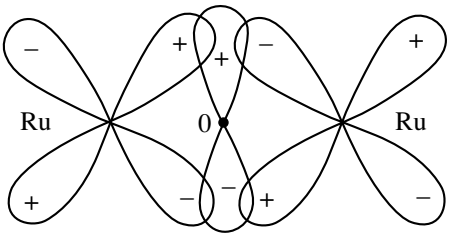

Fig. 1. Diagram of (d_{xy}–p)–π coupling in RuO_2 layer.

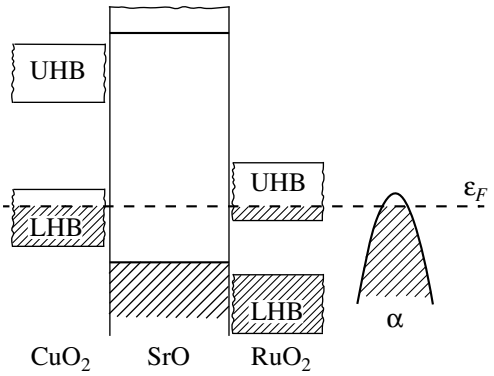

Fig. 2. Diagram of the band structure of a $CuO_2/SrO/RuO_2$ superlattice in the paramagnetic phase. Here UHB and LHB denote the upper and lower Hubbard subbands. The hole α band is also shown for RuO_2. Carriers are holes at the LHB top for CuO_2 layers and electrons at the UHB bottom for RuO_2 layers.

Interaction between the CuO_2 and RuO_2 layers not contained in the Hamiltonians (1) and (3) is small although important from the fundamental point of view since it ensures thermodynamic stability and determines the macroscopic symmetry of the superconducting and magnetic phases. We shall make a qualitative analysis of the role of these interactions in Section 5. Figure 2 shows a diagram of the electronic structure of the $CuO_2/SrO/RuO_2$ superlattice in the paramagnetic state. The carriers in the CuO_2 layer are holes in the lower Hubbard subband (LHB) and in RuO_2 they are electrons at the bottom of the upper Hubbard subband

(UHB) obtained by correlation splitting of the γ band responsible for the superconductivity in Sr_2RuO_4 [27]. The α hole band in the RuO_2 layer (and also the β band not shown in the diagram) plays the role of an electron reservoir and leads to the presence of carriers in the RuO_2 layer without doping. A general chemical potential is established as a result of the interlayer tunneling. The carrier concentration n_0 in the RuO_2 layer is a parameter of the model. For Sr_2RuO_4 we have $n_0 = 0.28$ [17]. The carriers in the CuO_2 and RuO_2 layers are spin polarons, i.e. quasiparticles surrounded by local spin fluctuations where both ferro- and antiferromagnetic correlations make a contribution in the RuO_2 layers.

The Hamiltonians (1)–(3) can be used to analyze two fundamentally different scenarios for the behavior of the system.

1. If $J > I$, which is the case for cuprates, for $n = 1$ ($n_0 = 0$, completely filled LHB) the ground state possesses long-range antiferromagnetic (AF) order. We know that the antiferromagnetic state is destroyed at fairly low hole concentrations $n_{AF} \approx 0.03$. In the range $n_0 > n_{AF}$ superconductivity may occur as a result of antiferromagnetic (J) exchange interactions.

2. If $I > J$, which is the case for ruthenates, for $n = 1$ ($n_0 = 0$ completely filled LHB), the ground dielectric state is ferromagnetic (F). In the presence of carriers, competition occurs between the saturated ferromagnetic state and the normal (nonmagnetic) N state. As a result of this competition, at concentrations $n_0 > n_F$ the system is converted to the N state and in this range superconductivity may also occur as a result of ferromagnetic (I) exchange interactions. We observe different results of this competition between the ferromagnetic and superconducting states in Sr_2RuO_4 (triplet superconductivity in RuO_4 layers) and $RuSr_2GdCu_2O_8$ (ferromagnetism in the RuO_2 layers) and thus we shall make a detailed analysis of this competition.

Omitting the index "Ru" in the Hamiltonian of the t–J–I model (3), we normalize this to the half-width of the initial electron band $W = zt$. After a Fourier transformation of the Hubbard operators

$$X_{\mathbf{k}\sigma} = \frac{1}{\sqrt{N}}\sum_{\mathbf{f}} e^{i\mathbf{k}\cdot\mathbf{f}} X_{\mathbf{f}}^{0\sigma},$$
$$X_{\mathbf{q}}^{\sigma\sigma'} = \frac{1}{\sqrt{N}}\sum_{\mathbf{f}} e^{i\mathbf{q}\cdot\mathbf{f}} X_{\mathbf{f}}^{\sigma\sigma'}, \tag{5}$$

where the vectors $\mathbf{k}$ and $\mathbf{q}$ belong to the first Brillouin band, we obtain the model Hamiltonian in the form

$$H/zt \equiv h = h_{\mathrm{kin}} + h_{\mathrm{int}}, \tag{6}$$

$$h_{\mathrm{kin}} = \sum_{\mathbf{k}\sigma} (\omega_{\mathbf{k}} - \tilde{\mu}) X_{\mathbf{k}\sigma}^{+} X_{\mathbf{k}\sigma},$$
$$\omega_{\mathbf{k}} = -\frac{1}{z}\sum_{\boldsymbol{\delta}} e^{i\mathbf{k}\cdot\boldsymbol{\delta}} = -\gamma_{\mathbf{k}}, \tag{7}$$

$$h_{\mathrm{int}} = \frac{1}{2}\sum_{\mathbf{q}\sigma} \gamma_{\mathbf{q}} \{ g(X_{\mathbf{q}}^{\sigma\bar{\sigma}} X_{-\mathbf{q}}^{\bar{\sigma}\sigma} - X_{\mathbf{q}}^{\sigma\sigma} X_{-\mathbf{q}}^{\bar{\sigma}\bar{\sigma}}) - r(X_{\mathbf{q}}^{\sigma\bar{\sigma}} X_{-\mathbf{q}}^{\bar{\sigma}\sigma} + X_{\mathbf{q}}^{\sigma\sigma} X_{-\mathbf{q}}^{\sigma\sigma}) \}, \tag{8}$$

where $g = J/t$, $r = I/t$, and $\mu/zt = \tilde{\mu}$ is the dimensionless chemical potential. The Hamiltonian h_{kin} (7) describes the electron kinetic energy and forms part of the Hubbard model for $U = \infty$.

3. SUPERCONDUCTIVITY IN CuO_2 LAYERS

In order to describe the magnetic and superconducting states of the model we use the mean-field approximation for strongly correlated systems in the formulation of the method of irreducible Green's functions [24, 28, 29] in which the higher Green's functions are projected onto the subspace of the normal $\langle\langle X_{\mathbf{k}}^{0\sigma} | X_{\mathbf{k}}^{0\sigma}\rangle\rangle$ and anomalous $\langle\langle X_{-\mathbf{k}}^{-\sigma 0} | X_{\mathbf{k}}^{\sigma 0}\rangle\rangle$ Green's functions coupled by a system of Gorkov equations [24, 25]. Fluctuations of the charge and spin states outside the limits of the mean-field theory were recently explicitly taken into account in [30] where the authors showed that the solutions for the superconducting phase obtained in the mean-field approximation are stable. The main difference between the carrier dispersion law allowing for fluctuations and the Hubbard I approximation according to [30] is that the spin correlation functions χ_1 and χ_2 between the first and second nearest neighbors are taken into account. This leads to some refinement of the Fermi surface and the appearance of satellites in the spectral density but from the qualitative point of view has little influence on the thermodynamic properties of the system and consequently in the range of parameters studied $J_{Cu}/t_{Cu} \approx 0.4$, $I_{Ru}/t_{Ru} \approx 0.4$, $J_{Ru}/t_{Ru} \approx 0.1$ we shall confine ourselves to the mean-field approximation.

In the superconducting phase the anomalous averages $B_{\mathbf{p}\sigma} = \langle X_{-\mathbf{p},-\sigma} X_{\mathbf{p}\sigma}\rangle$ determining the superconducting gap $\Delta_{\mathbf{k}\sigma}$ are nonzero:

$$\Delta_{\mathbf{k}\sigma} = \frac{1}{N}\sum_{\mathbf{p}} \{ \omega_{\mathbf{p}} (B_{\mathbf{p},-\sigma} - B_{\mathbf{p}\sigma}) + \omega_{\mathbf{k}-\mathbf{p}} [(r-g) B_{\mathbf{p},-\sigma} + g B_{\mathbf{p}\sigma}] \}. \tag{9}$$

An analysis of the symmetry of the possible solutions showed that three types of solution are possible [17].

(1) *s*-Type singlet pairings, $l = 0$,

$$\Delta_{\mathbf{k},0} = 2\Delta_0 + \lambda_s \omega_{\mathbf{k}} \Delta_0, \tag{10}$$

where the coupling constant is $\lambda_s = 2g - r$ and the dimensionless order parameter is

$$\Delta_0 = \frac{1}{2N}\sum_{\mathbf{k}} \omega_{\mathbf{k}} (B_{\mathbf{k}\uparrow} + B_{-\mathbf{k}\downarrow}). \tag{11}$$

The first term in Eq. (10) originates from the kinematic pairing mechanism [31] while the second originates from the exchange pairing mechanisms.

(2) *p*-Type triplet pairing, $l = 1$,

$$\begin{aligned} \Delta_{\mathbf{k},1} &= \lambda_p \psi_p(\mathbf{k}) \Delta_p, \\ \psi_p(\mathbf{k}) &= \frac{1}{2}(\sin k_x + i \sin k_y), \quad \lambda_p = r, \end{aligned} \tag{12}$$

the coupling constant in the *p*-pairing channel $\lambda_p = r$ is only determined by ferromagnetic exchange; this state cannot be achieved in CuO_2 layers with antiferromagnetic Cu–Cu interaction.

(3) $d_{x^2-y^2}$-Type singlet pairing, $l = 2$,

$$\begin{aligned} \Delta_{\mathbf{k},2} &= \lambda_d \psi_d(\mathbf{k}) \Delta_d, \\ \psi_d(\mathbf{k}) &= \frac{1}{2}(\cos k_x - \cos k_y), \quad \lambda_d = 2g - r. \end{aligned} \tag{13}$$

The coupling constant for the $d_{x^2-y^2}$ gap, $\lambda_d \approx 2g$ is determined by the antiferromagnetic Cu–Cu interaction (the ferromagnetic interaction $I = rt$ for the CuO_2 layers discussed in Section 2 can be neglected because of its smallness).

Expressions for the gap and the chemical potential are usually obtained from the corresponding self-consistent equations. In addition, in a strongly correlated system we also have another constraint, i.e., the constraint (2) which forbids the population of two-particle states at a single site. This condition may be written in the form [24]

$$\frac{1}{N}\sum_{\mathbf{k}} B_{\mathbf{k}\sigma} = 0, \tag{14}$$

and it is easy to see that an *s*-type solution does not satisfy this. Thus, neglecting electron–phonon interaction assuming only magnetic pairing mechanisms for the CuO_2 planes we obtain the only possible $d_{x^2-y^2}$-type state.

The gap Δ_d is determined self-consistently from the equation (for $l = 2$)

$$\frac{1}{\lambda_l} = \frac{1}{N}\sum_{\mathbf{p}} \frac{|\psi_l(\mathbf{p})|^2}{2E_{\mathbf{p}l}} \tanh\frac{E_{\mathbf{p}l}}{2\tau}, \tag{15}$$

where $\tau = k_B T/zt$ is the dimensionless temperature, $E_{\mathbf{p}l}$ is the quasiparticle energy in the superconducting phase,

$$E_{\mathbf{p}l} = \sqrt{c^2(n)(\omega_{\mathbf{p}} - m)^2 + \frac{\Delta_{\mathbf{p}l}^2}{c^2(n)}}. \tag{16}$$

Here we have $c(n) = 1 - n/2$ and m is the renormalized dimensionless chemical potential,

$$m = [\tilde{\mu} + (g + r)n/4]/c(n).$$

A similar self-consistent equation for the gap (15) was obtained earlier using a diagram technique for the *X* operators in the *t*–*J* model [32].

The solution of the equation at T_c for $d_{x^2-y^2}$ pairing shows that at moderate values of the coupling constant $\lambda_d = 0.5$ we can obtain $T_c \sim 100$ K and the high values of T_c are attributable to the closeness of the chemical potential for the optimum doped layers to the Van Hove singularity. A similar conclusion on the increase in T_c as a result of the Van Hove singularity in the superconducting state having $d_{x^2-y^2}$ symmetry caused by antiferromagnetic fluctuations was reached in an earlier study using the *t*–*J* model based on the Monte Carlo quantum method [33].

In order to estimate T_c we need to clarify the carrier concentration in the CuO_2 layers. We express the $RuSr_2GdCu_2O_8$ unit cell in the form of the successive layers:

$$\begin{aligned} &(Cu^{+2.5}O_2^{-2} - Gd^{+3} - Cu^{+2.5}O_2^{-2}) \\ &- Sr^{+2}O^{-2} - Ru^{+4}O_2^{-2} - Sr^{+2}O^{-2}, \end{aligned} \tag{17}$$

from which it follows that the number of holes per CuO_4 cell in the layer is $n_h = 1.5$. This concentration is obtained for weakly doped $YBa_2Cu_3O_{6+x}$ for $x \approx 0.5$ for we know that the $T_c(x)$ curve has a plateau at $T_c \approx 55$ K. Thus, the value of $T_c = 46$ K obtained in [1] for $RuSr_2GdCu_2O_8$ is not surprising and suggests that this compound falls within the range of weakly doped compositions. The jump in the specific heat at the transition point $\Delta\gamma(T_c) \equiv C_p/T \approx 0.35$ mJ/(g-at. K^2) is also typical of weakly doped superconductors [1].

4. COMPETITION BETWEEN FERROMAGNETIC AND SUPERCONDUCTING STATES IN THE RuO_2 LAYER

As we have noted for RuO_2 layers (and also in Sr_2RuO_4) we find $I_{Ru} \gg J_{Ru}$. The $d_{x^2-y^2}$ superconducting state with *d*-type pairing cannot be achieved in RuO_2 as a result of the antiferromagnetic correlations J_{Ru} since the coupling constant is $\lambda_d = (2J_{Ru} - I_{Ru})/t_{Ru} < 0$ [see $d_{x^2-y^2}$(13)]. Subsequently we show that $J_{Ru} = 0$

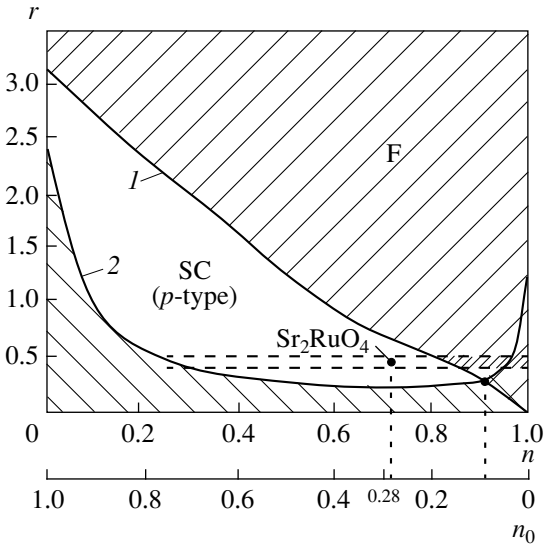

Fig. 3. *n–r* phase diagram ($T = 0$), n is the concentration, and r is the dimensionless parameter of ferromagnetic exchange interaction. Curve *1* was obtained by comparing the energies of the F and N states and the region above curve *1* corresponds to the ferromagnetic F state. Curve *2* was obtained from the equation ($T = 0$) for the superconducting gap with the minimum $\Delta_{\min} \sim 5 \times 10^{-4}$. The region above curve *2* corresponds to the *p*-type superconducting (SC) state, that below the curve is the normal N region, and the region bounded by curves *2* and *1* corresponds to the SC state.

and we retain the possibility of *p*-type superconductivity (triplet pairing) occurring as a result of ferromagnetic exchange $r = I_{\mathrm{Ru}}/t_{\mathrm{Ru}}$. The description of a system having the carrier concentration n_0 in the UHB is equivalent to its description having the hole concentration n_0 in the LHB. Using this procedure we determine the states of the system for various values of the ferromagnetic exchange parameter r and concentration $n = 1 - n_0$, i.e., we construct the *n–r* phase diagram at $T = 0$.

For fairly high values of the exchange parameter r and concentrations n in the RuO_2 layer our model gives long-range ferromagnetic order. We can establish a range of r and n values for which the ground state is ferromagnetic by comparing the energies of the normal (N) and ferromagnetic (F) states since the energy of the superconducting triplet states differs from the normal state energy by a very small relative quantity $\Delta E \sim \Delta_p^2 \rho(\varepsilon_F) \sim 10^{-2}$ eV ($\rho(\varepsilon_F)$ is the density of states at the Fermi level).

In the normal (nonmagnetic) state in the mean-field approximation [34] the distribution function at $T = 0$ has the form of a Fermi step: $f_{\mathbf{k}}^0 = \theta(m - \omega_{\mathbf{k}})$ and then the interrelation between the electron concentration n and the effective chemical potential m is written as follows:

$$n = \frac{1}{N}\sum_{\mathbf{k}\sigma} n_{\mathbf{k}\sigma} = 2c(n)\frac{1}{N}\sum_{\mathbf{k}} f_{\mathbf{k}}^0. \tag{18}$$

From this it follows that

$$\frac{n}{2-n} = \frac{1}{N}\sum_{\mathbf{k}} \theta(m - \omega_{\mathbf{k}}) = \int_{-1}^{m} \rho(\omega)\,d\omega \equiv g(m), \tag{19}$$

where $\rho(\omega) = \rho(-\omega)$ is the density of states corresponding to the $\omega_{\mathbf{k}}$ dispersion law. The energy of the N-state (at the lattice site) allowing for ferromagnetic exchange is given by

$$\epsilon_0 = \frac{1}{N}\sum_{\mathbf{k}\sigma} \omega_{\mathbf{k}}\left(1 - \frac{n}{2}\right) f_{\mathbf{k}}^0 - \frac{rn^2}{4} = (2-n)\,v(m) - \frac{rn^2}{4}, \tag{20}$$

$$v(m) \equiv \int_{-1}^{m} \omega\rho(\omega)\,d\omega,$$

and the energy of the saturated F-state is

$$\epsilon_F(n, r) = v(m_F) - \frac{rn^2}{2}, \tag{21}$$

where n is the electron concentration and m_F is the chemical potential in the F-state which is related to n by the following equation:

$$n = g(m_F) = \int_{-1}^{m_F} \rho(\omega)\,d\omega.$$

Equating the energies of the F and N states we obtain an expression for the values of the parameter r above which ferromagnetism occurs at a given concentration and below which normal or superconducting phases are possible:

$$r_{\mathrm{F}}(n) = \frac{4}{n^2}[\,v(m_F) - (2-n)\,v(m)]. \tag{22}$$

In order to establish a criterion for the occurrence of superconductivity (SC) outside the region of ferromagnetism, we set a very small fixed energy gap $D_{\min} \approx 10^{-3}$ outside the entire range of concentrations. For this gap we obtain for $\tau \longrightarrow 0$ the corresponding parameter r using formulas (15) and (16) for $l = 1$ (*p*-type solution):

$$r_{\mathrm{SC}}(n) = \left[\frac{1}{N}\sum_{\mathbf{k}} \frac{\psi_p^2(\mathbf{k})}{\sqrt{(\omega_{\mathbf{k}} - m)^2 + \psi_p^2(\mathbf{k})\dfrac{D_{\min}^2}{c^2(n)}}}\right]^{-1}. \tag{23}$$

This function $r_{\mathrm{SC}}(n)$ depends very weakly on the value of $D_{\min}$ and thus we shall assume that above this value of r, if no ferromagnetism occurs, the exchange param-

eter is sufficient for superconductivity to occur whereas below this value no superconductivity occurs.

Figure 3 gives the n–r phase diagram of the system where the curves $r_F(n)$ and $r_{SC}(n)$separate the regions of existence of the various phases. The right-hand point of intersection of the curves corresponds to the hole concentration $n_0 \approx 0.09$ and the parameter $r \approx 0.26$. The three-dimensional superconductor Sr_2RuO_4 has the hole concentration $n_0 \approx 0.28$ and the dimensionless parameter lies in the range $r \approx 0.4$–0.5 which leads to superconducting transition temperatures of ≈ 1–3 K [17]. The RuO_2 layers lie in this range of r values but since they are antiferromagnetic the hole concentrations in them are considerably lower (in Fig. 3 this range is shown by the closer shading).

5. DISCUSSION OF RESULTS

We shall compare the parameters of the model for RuO_2 in $RuSr_2GdCu_2O_8$ and for Sr_2RuO_4. The value of the exchange interaction I_{Ru} is determined by the overlap of the planar molecular $(d_{xy}$–$p)$–π orbitals, which is the same in both these cases so we shall assume that the parameter I_{Ru} remains the same. The parameter t_{Ru}, the effective Ru–Ru hopping parameter, also varies little since it is caused by overlap of the same $(d_{xy}$–$p)$–π orbitals. Thus, the dimensionless coupling constant r can be considered to be the same. The hole concentration n_0 undergoes the largest changes. As we discussed in Section 2, as a result of the narrowing of the α and β bands involving z-oriented orbitals of ruthenium and oxygen, the parameter n_0 should decrease. It can be seen from Fig. 3 that for $0.4 < r < 0.5$ the range of n_0 values where the superconducting triplet phase is replaced by a ferromagnetic one is $0.15 < n_0 < 0.20$. Consequently, a comparatively small reduction of the hole concentration in the RuO_2 layer compared with the three-dimensional Sr_2RuO_3 leads to stabilization of the ferromagnetic phase. Note that a similar conclusion on the stabilization of the ferromagnetic phase in Sr_2RuO_4 was obtained in [14] where allowance was made for gradient corrections to the local density functional whereas calculations using the density functional method in the local functional approximation give a paramagnetic Sr_2RuO_4 state [12, 13].

Direct information on the carrier concentration in the upper Hubbard subband of the RuO_2 layer could be obtained, for example, from experiments on the de Haas–van Alfven effect although such data are not available at present. In the present study the value of n_0 thus remains an undetermined parameter. At the same time, some predictions for experiments to study the influence of pressure on the thermodynamics of the system can be made from the phase diagram. For example, when pressure is applied perpendicular to the layers the Ru–Ru spacing in the layer increases and the bands become narrower whereas the ratio I/t changes little, and all these factors should lead to a reduction in n_0 and further stabilization of the ferromagnetic phase. However, pressure applied parallel to the layers reduces the Ru–Ru spacing, broadens the bands, and increases n_0 so that the ferromagnetic phase may become unstable and the RuO_2 layer will be converted to the superconducting triplet state with $T_c \sim 1$ K.

So far we have considered ferromagnetic RuO_2 layers and CuO_2 superconducting layers separately, whereas in $RuSr_2GdCu_2O_8$ they form a ferromagnetic metal/dielectric/superconductor/dielectric superlattice with atomic-scale layer thicknesses. Interlayer influences and tunneling effects were discussed in [18]. The exchange splitting t_{2g} of the Ru bands obtained in [18] is large, ~1 eV but the exchange splitting induced by them in the CuO_2 layer is two orders of magnitude smaller as a result of the absence of direct overlap of the d_{xy} states of Ru and the $d_{x^2-y^2}$ states of copper and the weak indirect interaction via the chain of oxygen ions O_{Ru}–O_{apex}–O_{Cu}. Thus, the splitting of the electron states at the Fermi surface is small and

$$Q = k_{F\uparrow} - k_{F\downarrow} \sim 0.01 k_F.$$

Under these conditions the superconductivity of the CuO_2 layer is not destroyed by exchange splitting but instead of the spatially uniform superconducting order parameter, an inhomogeneous state [35, 36] with a nonzero pair momentum Q and modulation length $\lambda = 2\pi/Q$ forms preferentially.

Coupling between the neighboring superconducting layers via the ferromagnetic layer will not destroy the superconductivity if the phase of the superconducting order parameter changes by π between the neighboring layers. Then the phase within the ferromagnetic layer will be close to zero which substantially reduces the loss of Cooper pairs in the magnetic layer [37].

To conclude we note that the coexistence of superconductivity and ferromagnetism in $RuSr_2GdCu_2O_8$ in our model is associated with a change in the ground state of the RuO_2 layer compared with Sr_2RuO_4. Both systems are close to the interface between the ferromagnetic and triplet superconducting phases and small changes in carrier concentration are responsible for a transition between these phases.

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