Effect of Antiferromagnetic Planes on the Superconducting Properties of Multilayered High-T_c Cuprates

M. Mori and S. Maekawa
Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan
(Received 26 August 2004; published 6 April 2005)

We propose a mechanism for high critical temperature (T_c) in the coexistent phase of superconducting (SC) and antiferromagnetic (AFM) CuO_2 planes in multilayered cuprates. The Josephson coupling between the SC planes separated by an AFM insulator (Mott insulator) is calculated perturbatively up to the fourth order in terms of the hopping integral between adjacent CuO_2 planes. It is shown that the AFM exchange splitting in the AFM plane suppresses the so-called *π-Josephson coupling,* and the long-ranged *0-Josephson coupling* leads to coexistence with a rather high value of T_c.

DOI: 10.1103/PhysRevLett.94.137003 PACS numbers: 74.81.–g, 74.20.De, 74.50.+r, 74.72.Jt

There is considerable interest in the superconducting critical temperature, T_c, in cuprates. In multilayered cuprates having several CuO_2 planes in a conducting block, T_c increases with the number of CuO_2 planes, n, and has a maximum at $n = 3$ [1]. Several studies have proposed that the suppression of T_c for $n > 3$ is caused by a charge imbalance among individual CuO_2 planes [2–5]; the outer-pyramidal-coordinated planes (OPs) tend to get optimal or overdoped, while the inner-square-coordinated planes (IPs) tend to get underdoped [1,2,6]. Chakravarty *et al.* have claimed that a Josephson coupling enhances the T_c up to $n = 3$, whereas a sizable charge imbalance combined with competing order parameters reduces T_c beyond $n = 3$ [4].

Recently, a coexistence of superconducting (SC) and antiferromagnetic (AFM) states has been observed in five-layered cuprates, $HgBa_2Ca_4Cu_5O_y$ and $TlBa_2Ca_4Cu_5O_y$ [7], and in a heterostructure composed of an alternating stack of $La_{1.85}Sr_{0.15}CuO_4$ and La_2CuO_4 [8]. In the five-layered cuprates, since the charge imbalance is enhanced by increasing n [2], the underdoped IPs and the optimally doped OPs show AFM and SC states, respectively [7]. It is noted that the five-layered cuprates retain rather high values of $T_c = 100$–108 K [2], despite the fact that the SC planes are separated by AFM planes in the direction perpendicular to the planes. In general, a Josephson coupling between SC planes is necessary both to stabilize the bulk SC state and to enhance T_c in layered superconductors [4,9–12]. Therefore, in the above coexistent phases, in the five-layered cuprates, the Josephson coupling is required via AFM planes not only for the stability of the superconductivity but also for such high values of T_c.

In this Letter, we study the coexistence of SC and AFM CuO_2 planes in multilayered cuprates. The AFM plane is assumed to be an insulator at half filling with no double occupancy. The Josephson coupling between SC planes separated by an AFM one is perturbatively calculated in terms of the hopping integral between adjacent CuO_2 planes. The perturbative processes comprise two parts: The first provides a positive value of Josephson coupling called *0-Josephson coupling,* while the second makes a negative contribution called *π-Josephson coupling.* Note that the sign of Josephson coupling reflects a quantum effect originating from the fermion anticommutation rules [13–15]. We find that the AFM exchange interaction suppresses the latter process, and allows the Cooper pair to tunnel through the AFM insulating (AFMI) plane. The fluctuations of the SC phase are suppressed by this long-ranged Josephson coupling, and it is this which enables the coexistence and a rather high value of T_c. The n dependence of T_c and enhancement of the Josephson coupling are discussed.

The minimal model is a three-layered system composed of two SC planes with d-wave symmetry and an AFMI plane at half filling. The SC planes are separated by the AFMI plane as shown in Fig. 1. In the five-layered cuprates, the two SC planes are separated by three AFMI planes, and the same mechanism arises in higher order. The coexistence in five-layered cuprates is explained by the Josephson coupling through the AFMI planes.

In each SC plane, the BCS mean-field Hamiltonian is adopted, and the wave functions in the two SC planes are given by

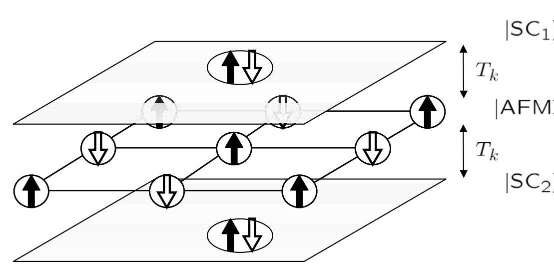

FIG. 1. Schematic figure of a conducting block in the three-layered system, SC/AFMI/SC. The thick and open arrows indicate spins of electrons. The up and down spins enclosed by the oval indicate condensed Cooper pairs. The matrix element of interlayer hopping is denoted by T_k.

$$|\mathrm{SC}_1\rangle \equiv \prod_k (u_k + v_k e^{i\phi_1} a^\dagger_{k\uparrow} a^\dagger_{-k\downarrow})|0\rangle, \tag{1}$$

$$|\mathrm{SC}_2\rangle \equiv \prod_k (u_k + v_k e^{i\phi_2} c^\dagger_{k\uparrow} c^\dagger_{-k\downarrow})|0\rangle, \tag{2}$$

where $v_k/u_k = \Delta_k/E_k$ and $E_k = \sqrt{\xi_k^2 + \Delta_k^2}$. The SC order parameter is denoted by $\Delta_k = (\Delta_0/2)[\cos(k_x) - \cos(k_y)]$ and ξ_k is the quasiparticle energy in the normal state. The true vacuum is indicated by $|0\rangle$. The operators, $a^\dagger_{k\sigma}$ and $c^\dagger_{k\sigma}$, create electrons with momentum, k, and spin, σ, in the SC_1 and the SC_2, respectively.

In the AFMI plane, the interaction between localized spins is given by $J\sum_{\langle i,j\rangle} \vec{\mathrm{S}}_i \cdot \vec{\mathrm{S}}_j$, where $\vec{\mathrm{S}}_i$ is the spin operator at the ith site, and the summation runs over the nearest neighbor sites. The Néel state is assumed for the ground state, and its wave function is given by

$$|\mathrm{AFMI}\rangle \equiv \prod_{i\in A, j\in B} b^\dagger_{i\uparrow} b^\dagger_{j\downarrow}|0\rangle, \tag{3}$$

where $b^\dagger_{i\sigma}$ is the electron creation operator at the ith site with spin σ. Up and down spins are sited on sublattices A and B, respectively. The phase convention is defined by putting the operators in order of its site index. No double occupancy is imposed on $|\mathrm{AFMI}\rangle$. The charge imbalance between the SC and the AFMI planes is induced by a site potential, W, whose value is of the order of J [5]. Because of this potential, intermediate states with a double occupancy are higher in energy than those with a single hole in the multilayered cuprates [7].

The AFM and the SC planes are connected by the tunneling Hamiltonian as

$$H_T = H_1 + H_2, \tag{4}$$

$$H_1 = \sum_{i,k,\sigma} (\phi_{i,k} a^\dagger_{k\sigma} b_{i\sigma} + \phi^*_{i,k} b^\dagger_{i\sigma} a_{k\sigma}), \tag{5}$$

$$H_2 = \sum_{i,k,\sigma} (\phi_{i,k} b^\dagger_{i\sigma} c_{k\sigma} + \phi^*_{i,k} c^\dagger_{k\sigma} b_{i\sigma}), \tag{6}$$

$$\phi_{i,k} = \frac{1}{N^{1/2}} e^{-ikr_i} T_k, \tag{7}$$

where an electron coherently hops between adjacent planes with matrix element, $T_k = (t_\perp/4)[\cos(k_x) - \cos(k_y)]^2$ [16–18], and $b_{i\sigma} = (1/N^{1/2})\sum_k e^{ikr_i} b_{k\sigma}$.

The Josephson coupling energy, $-E_J\cos\theta$, which is a function of phase difference between SC_1 and SC_2, $\theta \equiv \phi_1 - \phi_2$, is obtained by the fourth order perturbation theory in terms of Eq. (4). The wave function of the ground state is given by $|\Omega\rangle = |\mathrm{SC}_1\rangle \otimes |\mathrm{AFMI}\rangle \otimes |\mathrm{SC}_2\rangle$, where the order of $|\mathrm{SC}_1\rangle$, $|\mathrm{AFMI}\rangle$, and $|\mathrm{SC}_2\rangle$ must be maintained to define a phase convention.

The first intermediate states, $|m_1\rangle$ and $|m_1'\rangle$, are obtained by transferring an electron from the AFMI plane to the SC one as shown in Fig. 2, since the double occupancy is forbidden in $|\mathrm{AFMI}\rangle$. The energy of $|m_1\rangle$ and $|m_1'\rangle$ is given by $\Delta E_{m_1} = \Delta E_{m_1'} = E_k + J + W$, where W is the site potential in the IP [5]. Spin fluctuations and hole motions are neglected.

The second intermediate states, $|m_2\rangle$, that can provide the Josephson coupling energy, is classified into two types of spin configurations, i.e., "0-config" and "π-config." Typical processes are shown in Fig. 2. Each SC plane has one quasiparticle excitation, and the AFMI plane has no hole. The 0-config has an antiparallel spin configuration in the SC planes, while the π-config has a parallel one. The energy of $|m_2\rangle$ with 0-config is given by $\Delta E_{m_2} = 2E_k$, where the spin configuration in the AFMI plane is the same as that in the ground state. On the other hand, the

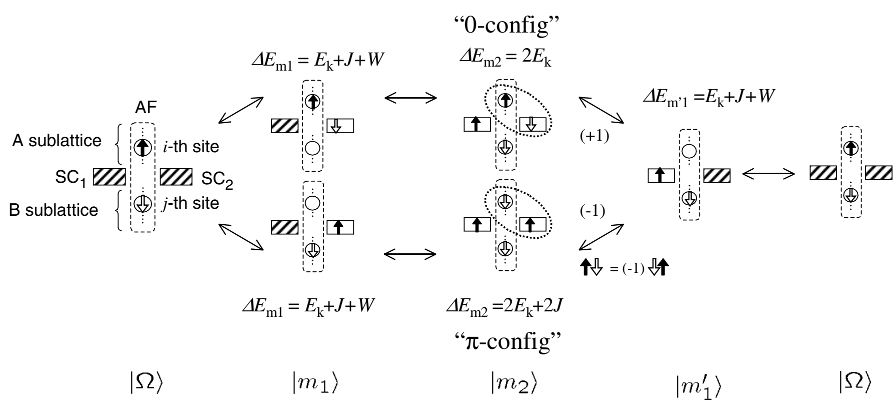

FIG. 2. Two examples of tunneling processes contributing to the Josephson coupling energy, $E_J = \sum\langle\Omega|H_T|m_1\rangle\langle m_1|H_T|m_2\rangle \times \langle m_2|H_T|m_1'\rangle\langle m_1'|H_T|\Omega\rangle/(\Delta E_{m_1}\Delta E_{m_2}\Delta E_{m_1'})$. The upper flow provides the *0-Josephson coupling,* while the lower flow leads to the *π-Josephson coupling.* The shaded rectangles imply the SC ground state. The arrows in the SC and in the AFM indicate the quasiparticle excitations and the localized spins, respectively. The open circle denotes a vacant site in the AFMI plane. The π-config has the parallel spin configuration in the SCs, while the 0-config has the antiparallel configuration. The anticommutation of fermions occurs only in the lower flow.

energy of $|m_2\rangle$ with π-config is given by $\Delta E_{m_2} = 2E_k + 2J$, since one site is filled with an opposite spin.

Finally, we find that the magnitude of Josephson coupling energy is given by

$$E_J = E_J^0 + E_J^\pi, \tag{8}$$

$$\sim\left(\frac{1}{\Delta_0} - \frac{1}{(\Delta_0 + J)}\right)\frac{t_\perp^4}{(\Delta_0 + J + W)^2}, \tag{9}$$

$$E_J^0 = 4\sum_k \frac{T_k^4}{2E_k(E_k + J + W)^2}\left(\frac{\Delta_k}{2E_k}\right)^2 \tag{10}$$

$$E_J^\pi = -4\sum_k \frac{T_k^4}{(2E_k + 2J)(E_k + J + W)^2}\left(\frac{\Delta_k}{2E_k}\right)^2. \tag{11}$$

Equations (10) and (11) are caused by 0-config and π-config, respectively. We look more carefully into the signs of E_J^0 and E_J^π. In the transitions from $|\Omega\rangle$ to $|m_2\rangle$ by way of $|m_1\rangle$, both 0-config and π-config have the same sign. On the other hand, only in π-config, the anticommutation of fermions occurs between $|m_2\rangle$ and $|m_1'\rangle$, and thus the additional minus sign is added to its transition amplitude. As a consequence, *the 0-config provides the 0-Josephson coupling, while the π-config does the π-Josephson coupling. The signs of E_J^0 and E_J^π are attributed to the quantum effect originating from the anticommutation of fermions* [13–15].

Note that, when the AFM interaction in the AFMI plane is much smaller than the SC gap, i.e., $J \ll \Delta_0$, the Cooper pair cannot go through the AFMI plane, since 0-config and π-config processes in Eq. (9) cancel out as

$$E_J \sim \left(\frac{1}{\Delta_0} - \frac{1}{(\Delta_0 + 0)}\right)\frac{t_\perp^4}{(\Delta_0 + 0 + W)^2} = 0. \tag{12}$$

To show the J dependence of Eq. (8), E_J is numerically calculated and plotted in Fig. 3 as a function of $J/t_\perp$ for $W/t_\perp = 0.2$, 0.4, and 0.8. We adopt $\xi_k = 2t[\cos(k_x) + \cos(k_y)] - \mu$ for the quasiparticle energy, and $t/t_\perp = 5$, $\Delta_0/t_\perp = 1$, and $\mu/t_\perp = -1$. One can find that *the AFM interaction generates the Josephson coupling through the AFMI plane.*

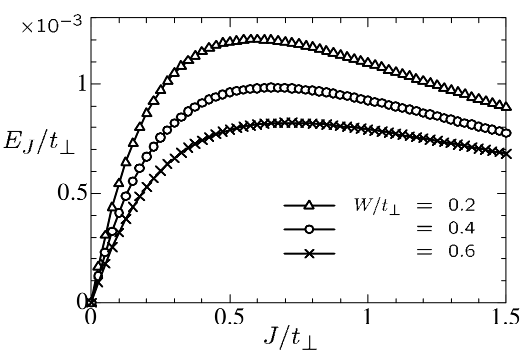

FIG. 3. J dependence of E_J. We adopted $\xi_k = 2t[\cos(k_x) + \cos(k_y)] - \mu$ for the quasiparticle energy. Parameters are scaled by $t_\perp$ as $t/t_\perp = 5$, $\Delta_0/t_\perp = 1$, $\mu/t_\perp = -1$.

We have shown that the long-ranged Josephson coupling through the AFMI, E_J, can survive due to the magnetic exchange interaction. Although the magnitude of E_J is small, it is important to provide the phase coherence, which plays an important role to determine T_c in the cuprate superconductors [19,20]. Below, we study the SC phase coherence in the multilayered systems based on a model proposed by Zaleski and Kopeć [20]. In the present study, we take account of the long-ranged Josephson coupling denoted by K that is crucial to obtain a rather high value of T_c in the coexistent phase in the five-layered cuprates.

The free energy given by a spatial variation of the SC order parameter, $\Psi(r)$, is proportional to $\int dr|\nabla\Psi(r)|^2 \sim |\Psi_0|^2 \int dr(\nabla\phi(r))^2 \sim |\Psi_0|^2 \int dr \cos(\phi_i - \phi_j)$. We assume that the amplitude, Ψ_0, is constant and the spatial variation of phase, $\phi(r)$, is slow, i.e., $\phi_i - \phi_j \sim \nabla\phi(r)$. Therefore, the phase degree of freedom in the multilayered cuprates is given by the XY model as

$$H = H_0 + H_1, \tag{13}$$

$$H_0 = -\sum_{\langle i,j\rangle,l} J_\parallel^{(\alpha)} \vec{R}_{i,l}^{(\alpha)} \cdot \vec{R}_{j,l}^{(\alpha)} - \sum_{i,l,\langle\alpha\beta\rangle} J_\perp \vec{R}_{i,l}^{(\alpha)} \cdot \vec{R}_{i,l}^{(\beta)} - \sum_{i,\langle l,m\rangle,\langle\alpha\beta\rangle} J'_\perp \vec{R}_{i,l}^{(\alpha)} \cdot \vec{R}_{i,m}^{(\beta)}, \tag{14}$$

$$H_1 = -\sum_{i,l,\langle\langle\alpha\beta\rangle\rangle} K^{(\alpha\beta)} \vec{R}_{i,l}^{(\alpha)} \cdot \vec{R}_{i,l}^{(\beta)}, \tag{15}$$

where $\vec{R}_{i,l}^{(\alpha)} = (R_{i,l}^{(\alpha),x}, R_{i,l}^{(\alpha),y})$ is the XY spin operator at the ith site on the αth plane in the lth conducting block. The single square brackets indicate sums between nearest neighboring sites, planes and blocks. The double square

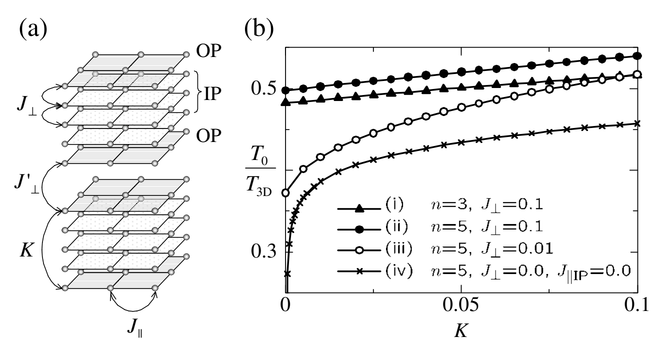

FIG. 4. (a) Schematic figure of the five-layered XY model. (b) K dependence of T_0 normalized by that of the isotropic case, T_{3D}, given by $J_{\parallel OP} = J_{\parallel IP} = J_\perp = J'_\perp = 1$ and $K = 0$. Each line is given as follows: (i) $n = 3$, $J_{\parallel OP} = J_{\parallel IP} = 1$, $J_\perp = 0.1$, and $J'_\perp = 0.01$ for triangles; (ii) $n = 5$, $J_{\parallel OP} = J_{\parallel IP} = 1$, $J_\perp = 0.1$, and $J'_\perp = 0.01$ for solid circles; (iii) $n = 5$, $J_{\parallel OP} = J_{\parallel IP} = 1$, $J_\perp = 0.01$, and $J'_\perp = 0.01$ for open circles; (iv) for cross, $n = 5$ is reduced to $n = 2$ by assuming $J_{\parallel OP} = 1$, $J_{\parallel IP} = 0$, $J_\perp = 0$, and $J'_\perp = 0.01$.

bracket indicates a sum between the OPs in one block. Schematic figures of the planes are shown in Fig. 4(a). The SC planes have a finite value of $J_{\|}^{(\alpha)}$, while $J_{\|}^{(\alpha)} = 0$ in the AFM planes. The long-ranged Josephson coupling via the AFMI plane is denoted by K. The $J'_{\perp}$ connects the SC OP in one block to that in another block. If the IP is also the SC state, $J_{\perp}$ should be included between the OP and the IP within the block. Such a case is used to discuss the n dependence of T_c.

The free energy par site for Eq. (13) is given by $f(\zeta) = -\zeta/\beta + 2/(\beta N)\sum_{k,p}^{N/2} \mathrm{Tr}\ln[\zeta - \beta\hat{M}_n]$, where we adopted an approximation that the average length of spins is restricted to 1 [20]. The matrix, $\hat{M}_n$, is the Fourier transform of the Hamiltonian, Eq. (13), in an n-layered system. The Lagrange multiplier, ζ, is determined by a saddle point equation as $\zeta_0 - \beta_c E_0^{(1)} = 0$, where $E_0^{(1)} > E_0^{(2)} > \cdots > E_0^{(n)}$ are the eigenvalues of $\hat{M}_n$ at $k = 0$.

The phase coherence develops below the critical temperature, T_0, which is determined by

$$T_0 = E_0^{(1)}\left(\frac{1}{N}\sum_k^N \frac{1}{n}\sum_{\alpha=1}^n \frac{1}{1 - E_k^{(\alpha)}/E_0^{(1)}}\right)^{-1}. \qquad (16)$$

When all interlayer couplings are zero, i.e., $J_{\perp} = J'_{\perp} = K = 0$, the k summation in Eq. (16) diverges, and then $T_0 = 0$.

The K dependence of T_0 is shown in Fig. 4(b). T_0 is normalized by T_{3D}, which denotes the critical temperature in the isotropic case on the three-dimensional cubic lattice, i.e., $J_{\|OP} = J_{\|IP} = J_{\perp} = J'_{\perp} = 1$ and $K = 0$. The ratio of T_0 to T_{3D} measures an effect of the interlayer couplings. The three- and five-layered cases with $J_{\|OP} = J_{\|IP} = 1$, $J_{\perp} = 0.1$, and $J'_{\perp} = 0.01$ are plotted by solid circles and triangles, respectively. We find that T_0 increases with n [20]. For the small value of $J_{\perp} = 0.01$ in the five-layered case with $J_{\|OP} = J_{\|IP} = 1$, and $J'_{\perp} = 0.01$, T_0 is suppressed as shown by open circles. In other words, T_0 is enhanced by the Josephson coupling, but is suppressed by the competing order, which reduces the Josephson coupling between nearest neighbor planes [4]. If all SC orders in IPs are suppressed, i.e., $J_{\|IP} = J_{\perp} = 0$, the five-layered system is reduced to the bilayer one composed of OPs. Such a case is shown in Fig. 4(b) by crosses, where $n = 5$, $J_{\|OP} = 1$, $J_{\|IP} = 0$, $J_{\perp} = 0$, and $J'_{\perp} = 0.01$. We find that T_0 is suppressed, but is strongly enhanced by small K. Therefore, even if the SC planes are separated by the AFM insulators, the SC order can coexist with the AFM order due to the Josephson coupling through the AFM plane. The high value of T_c in the coexistent phase is also retained by such a long-ranged Josephson coupling.

It is noted that, if one can eliminate all the π-config processes, E_J will be enhanced more than Eq. (8). Such a case is possible in a spin liquid state, i.e., resonating valence bond (RVB) state [21]. The RVB state does not have any transition amplitude to π-config, since the π-config processes corresponds to a triplet channel [15]. Therefore, only the 0-config process contributes to E_J, and then the Josephson coupling with the RVB state can be enhanced more than that with the AFMI state.

In conclusion, we have proposed a mechanism for high critical temperature (T_c) in the coexistent phase of superconducting (SC) and antiferromagnetic (AFM) CuO_2 planes in the multilayered cuprates. The Josephson coupling between the SC planes separated by the AFM plane is perturbatively calculated in terms of the hopping integral between adjacent CuO_2 planes. The AFM interaction provides the Josephson coupling through the AFM plane, which enables the coexistence and the high value of T_c in the multilayered cuprates. The further enhancement of Josephson coupling is expected in a resonating valence bond state.

We thank Professor G. Baskaran, Professor H. Fukuyama, Dr. W. Koshibae, and Professor S. E. Barnes for their valuable discussions. This work was supported by a Grant-in-Aid for Scientific Research on Priority Areas and the NAREGI Nanoscience Project from MEXT and CREST. One of the authors (M. M.) acknowledges support by the 21st Century COE program, Tohoku University Materials Research Center.

[1] M. Karppinen and H. Yamauchi, Mater. Sci. Eng., R **26**, 51 (1999).
[2] H. Kotegawa *et al.*, Phys. Rev. B **64**, 064515 (2001).
[3] L. Jansen and R. Block, Physica (Amsterdam) **289A**, 165 (2001).
[4] S. Chakravarty, H-Y. Kee, and K. Voelker, Nature (London) **428**, 53 (2004).
[5] M. Mori, T. Tohyama, and S. Maekawa, Phys. Rev. B **66**, 064502 (2002).
[6] A. Trokiner *et al.*, Phys. Rev. B **44**, 2426 (1991).
[7] H. Kotegawa *et al.*, Phys. Rev. B **69**, 014501 (2004).
[8] I. Bozovic *et al.*, Nature (London) **422**, 873 (2003).
[9] P. W. Anderson, Science **279**, 1196 (1998).
[10] Z. Tešanović, Phys. Rev. B **36**, 2364 (1987).
[11] A. J. Leggett, Science **274**, 587 (1996).
[12] S. Chakravarty, Eur. Phys. J. B **5**, 337 (1998).
[13] B. I. Spivak and S. A. Kivelson, Phys. Rev. B **43**, 3740 (1991).
[14] L. I. Glatzman and K. A. Matveev, JETP Lett. **49**, 659 (1989).
[15] M-S. Choi, C. Bruder, and D. Loss, Phys. Rev. B **62**, 13 569 (2000).
[16] S. Chakravarty *et al.*, Science **261**, 337 (1993).
[17] O. K. Andersen *et al.*, J. Phys. Chem. Solids **56**, 1573 (1995).
[18] A. I. Liechtenstein *et al.*, Phys. Rev. B **54**, 12 505 (1996).
[19] V. J. Emery and S. A. Kivelson, Nature (London) **374**, 434 (1995).
[20] T. A. Zaleski and T. K. Kopeć, Phys. Rev. B **71**, 014519 (2005).
[21] P. W. Anderson, Res. Mat. **8**, 53 (1973); P. Fazekas and P. W. Anderson, Philos. Mag. **30**, 432 (1974).