CuO_2 bilayer containing magnetic impurities

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The effect of magnetic impurities between the two planes of a CuO_2 bilayer on the superconducting properties is considered. To this end a previously introduced model for a CuO_2 bilayer with finite lateral size L is used. This system undergoes a bona fide transition (crossover) from two-dimensional to zero-dimensional behavior as the temperature decreases below a size-dependent value T_c^*. The model is an attempt to explain experimental findings on $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ and $Bi_2Sr_2CaCu_2O_8/Bi_2Sr_2CuO_6$ superlattices which seem to indicate that a single CuO_2 bilayer can still exhibit superconducting behavior. T_c^* is found to decrease with increasing impurity concentration. This could be a hint at the microscopic origin of the measured T_c depression in $Tl_{2-x}Sm_xBa_2Ca_2Cu_3O_y$ with increasing x. The recently discovered paramagnetic Meissner effect cannot be described by the CuO_2 bilayer model with magnetic impurities, although the Josephson term in the free-energy functional changes sign for sufficiently high impurity concentration, i.e., the bilayer behaves like a π junction.

Among the several problems concerning the theory of the high-temperature superconductors, one of the most prominent is the effect of the two dimensionality of the CuO_2 planes on the superconducting properties and the role that is played by the extension into the third dimension through a coupling mechanism between adjacent CuO_2 planes. Transport measurements on $YBa_2Cu_3O_7/PrBa_2Cu_3O_7$ (YBCO/PBCO) (Ref. 1) and $Bi_2Sr_2CaCu_2O_8/Bi_2Sr_2CuO_6$ (BSCCO/BSCO) (Ref. 2) superlattices suggest that even a one-unit-cell thick YBCO layer[1] and a half-one-unit-cell thick BSCCO layer,[2] i.e., a single CuO_2 bilayer can exhibit a (broadened) superconducting transition in the $R(T)$ curve with a T_c value of $\sim 20-30$ K.

Conventional superconductivity, characterized by off-diagonal long-range order (ODLRO), with $T_c>0$ is ruled out for two- or less-dimensional systems[3] due to destruction of ODLRO by thermal phase fluctuations of the superconducting order parameter (OP) for $T>0$.[4] One can now imagine two scenarios for the physical processes that drive the resistive transition in the CuO_2 bilayer:

(i) Spontaneous creation of vortex-antivortex pairs that undergo a Berezinskii-Kosterlitz-Thouless (BKT) phase transition[5] at a finite temperature T_{KT}. This transition is characterized, e.g., by the exponent of the nonlinear current-voltage relation $V\propto I^\alpha$ below T_{KT} and the linear resistivity above T_{KT}.[6]

(ii) Bona fide transition (crossover) from two-dimensional (2D) to zero-dimensional (0D) behavior due to finite-size effects[7] as the temperature decreases below a characteristic value T_c^*, i.e., the effective superconducting coherence length $\xi_{\text{eff}}(T)$ exceeds the lateral dimensions of the system. Since real samples are always of finite size and consist of crystallites with extensions of $\sim 10^3-10^4$ Å (Ref. 8), such 2D–0D transitions are expected to play an important role in the high-T_c materials.

In previous articles[7] a single CuO_2 bilayer has been modeled by a Hamiltonian of the form

$$H=H_0+H_P+H_T\ , \tag{1}$$

$$H_0=\sum_{i=1,2}\sum_{\alpha}\varepsilon_\alpha c_{i,\alpha}^\dagger c_{i,\alpha}\ , \tag{2}$$

$$H_P=-g\sum_{i=1,2}\sum_{Q}b_{i,Q}^\dagger b_{i,Q}\ , \tag{3}$$

$$H_T=\sum_{\alpha}(tc_{1,\alpha}^\dagger c_{2,\alpha}+\text{H.c.})\ , \tag{4}$$

with $\alpha\equiv(k,\sigma)$ and $b_Q\equiv\sum_k c_{-k\downarrow}c_{k+Q\uparrow}$. The index i refers to the plane; g and t are the intraplane pairing energy and interplane tunneling matrix element, respectively, which are taken to be constants. H_P describes a BCS-like intralayer pairing and H_T a single-particle interlayer tunneling. From Eqs. (1)–(4) a Ginzburg-Landau-type free-energy functional has been derived using a functional-integral transformation,

$$F_{\text{GL}}[\Delta^{(1)},\Delta^{(2)}]=F_0+\sum_{Qm}a_{Qm}(|\Delta_{Qm}^{(1)}|^2+|\Delta_{Qm}^{(2)}|^2)+N_0\kappa_0^2\sum_{Qm}|\Delta_{Qm}^{(1)}||\Delta_{Qm}^{(2)}|\cos(\varphi_{Qm}^{(1)}-\varphi_{Qm}^{(2)})\ , \tag{5}$$

with

$$a_{Qm}=\frac{N_0}{2}\left\{\ln\frac{T}{T_{c0}}+\frac{\pi^2}{4}|m|+\xi_0^2Q^2+\kappa_0^2\right\}+\frac{b_0}{2}\sum_{Q'}\langle|\Delta_{Q'0}|^2\rangle\delta_{m0}\ , \tag{6}$$

$\Delta_{Qm}^{(i)}=|\Delta_{Qm}^{(i)}|\exp(i\varphi_{Qm}^{(i)})$, $\kappa_0\equiv|t|/(2\omega_0)$, ω_0 is the BCS cutoff parameter, $b_0=7/(8\pi^2)\zeta(3)N_0\beta^2$, N_0 is the density of states at the Fermi surface in the normal state, and T_{c0} is the Ginzburg-Landau mean-field critical temperature. The free energy, Eq. (5), contains Gaussian fluctuations of modulus *and* phase of the superconducting OP (in contrast to Ref. 7, where it has been falsely stated that the intraplane phase fluctuations are not taken into account) and fourth-order fluctuations of the modulus of the superconducting OP in a biquadratic approximation.[9]

The interplane Coulomb interaction has been treated via inclusion of quantum fluctuations of the phase difference between the planes,[7]

$$F_C=\sum_{Qm}\frac{4\pi^2m^2}{\beta^2V_C}(\varphi^{(1)}_{Qm}-\varphi^{(2)}_{Qm})^2 , \tag{7}$$

with $V_C=e^2/(2C)$, where C is the capacitance of the bilayer. The static $(m=0)$ component of the Coulomb interaction, Eq. (7), does not contribute to the free energy. The total free-energy functional F is given by $F=F_{\rm GL}+F_C$.

Topological excitations, i.e., spontaneous creation of vortices and antivortices have been neglected within this approach, which can be justified if the vortex and antivortex densities are sufficiently low. This condition corresponds to a sufficiently high Josephson coupling strength between the planes.[10] The ansatz of Ref. 7 thus requires strong Josephson coupling between the two planes of a CuO_2 bilayer. Intrinsic Josephson effects *between* the CuO_2 bilayers in more anisotropic high-T_c materials such as BSCCO have recently been discovered.[11] Experiments concerning the nature of the interplane coupling *within* the CuO_2 bilayer are not known so far.

Using the free energy, Eq. (5), it has been shown[7] that the critical temperature T_c of the (laterally) infinitely extended bilayer, defined through the occurrence of a pole in the fluctuation propagator $\langle|\Delta_{00}|^2\rangle$ [equivalent to the divergence of the temperature-dependent superconducting coherence length $\xi_{\rm eff}(T)$], is always equal to zero, independently of the interlayer coupling strength. This result is in agreement with the works of Hohenberg[3] and Rice[4] and shows that two mutually coupled 2D systems of infinite size still form a 2D system. But in the line of thought of Hassing and Wilkins[9] it has been deduced[7] that for a bilayer with finite lateral size L there exists a fairly narrow temperature interval around a value T_c^*, defined by

$$2\pi\xi_{\rm eff}(T_c^*)=L , \tag{8}$$

where the system undergoes a bona fide 2D–0D transition (crossover) as the temperature decreases below T_c^*, i.e., the size of the locally superconducting regions with a nonvanishing local OP exceeds the system size. This transition can be characterized by the behavior of macroscopic quantities, e.g., a jump of the specific heat at T_c^*. The obtained T_c^* values depend on L and Josephson coupling strength but barely on the interlayer Coulomb interaction and are consistent with experiment.[7]

So far it cannot be decided whether the observed resistive transitions in systems which contain only one CuO_2 bilayer are caused by finite-size effects in the above sense or driven by a BKT transition. The occurrence of a BKT phase transition in high-T_c superlattices is still discussed controversely, since the functional form of the potential between vortices and antivortices is very strongly modified due to interlayer interactions.[12]

Not long ago a new set of experiments has reached remarkable interest. When they measured the field-cooling susceptibility on granular BSCCO samples[13] and very recently on YBCO single crystals,[14] several groups found a *positive* value below T_c rather than the theoretically expected negative value of $-1/(4\pi)$ at $T=0$ (the familiar Meissner-Ochsenfeld effect). This unusual behavior has been designated as "paramagnetic Meissner effect" (PME) or "Wohlleben effect." Attempts to explain the PME try to model the materials as a network of Josephson junctions[15] which are partially replaced by so-called π junctions,[16] i.e., Josephson junctions with a reverse sign of the corresponding term in the free energy (a "negative critical current"). Possible microscopic mechanisms to obtain π junctions are (i) localized magnetic moments in the junctions[16] which offer a channel for interlayer tunneling with spin flip and which contribute with a negative sign to the Josephson term in the free energy; (ii) d-wave symmetry of the superconducting OP and different preferential orientations in momentum space in the two planes.[17]

The aim of the present paper is to incorporate the effect of magnetic impurities between the two planes of a CuO_2 bilayer into the model of Ref. 7 and to examine the possibility of obtaining the PME within the framework of this approach.[18] In the presence of noninteracting, i.e., sufficiently dilute magnetic impurities the Hamiltonian, Eq. (1), contains an additional term,[16]

$$H_M=\sum_n\sum_{k\sigma\sigma'}(v_{nk}\tau_{\sigma\sigma'}\cdot\mathbf{S}_n c^\dagger_{1,k\sigma}c_{2,k\sigma'}+\text{H.c.}) , \tag{9}$$

where the n summation is over the randomly distributed impurity atoms with spin $\mathbf{S}_n$ (in units of $\hbar$), and τ is the vector of Pauli matrices. H_M describes a single-particle tunneling between the planes via magnetic impurities with spin flip. The effective tunneling matrix element v_{nk} is given by[19]

$$v_{nk}=\frac{2t_{1,kd}t_{2,dk}U}{\varepsilon_d(\varepsilon_d+U)} . \tag{10}$$

Here, $t_{i,kd}$ is the tunneling matrix element between plane i $(i=1,2)$ and impurity atom, $\varepsilon_d<0$ is the single-particle energy of an electron on the impurity atom, measured from the Fermi energy, and $\varepsilon_d+U>0$ is the energy of a second electron on the impurity atom which must have opposite sign due to Pauli's exclusion principle. Equation (9) can be derived from the Anderson Hamiltonian,[19]

$$\begin{aligned}H_{\rm Anderson}=&\sum_{k\sigma}\varepsilon_{1,k}c^\dagger_{1,k\sigma}c_{1,k\sigma}+\sum_{k\sigma}\varepsilon_{2,k}c^\dagger_{2,k\sigma}c_{2,k\sigma}\\&+\sum_\sigma\varepsilon_d n_{d\sigma}+Un_{d\uparrow}n_{d\downarrow}\\&+\sum_{k\sigma}(t_{1,kd}c^\dagger_{1,k\sigma}d_\sigma+\text{H.c.})\\&+\sum_{k\sigma}(t_{2,kd}c^\dagger_{2,k\sigma}d_\sigma+\text{H.c.}) ,\end{aligned} \tag{11}$$

via a canonical transformation.[20] Estimating[16] $|\varepsilon_d|\sim0.1$ eV, $U\sim$ several eV (i.e., $U\gg|\varepsilon_d|$), $|t_{1,kd}t_{2,kd}|\sim\varepsilon_0|t|$, $\varepsilon_0\sim1$ eV, and $|t|=50$ meV,[21] one obtains $|v_{nk}|\sim1$ eV.

After adding H_M, Eq. (9), to the Hamiltonian H, Eq.

(1), the functional-integral transformation yields a modified free energy [cf. Eqs. (5) and (6)],[22]

$$\tilde{F}_{\rm GL}[\Delta^{(1)},\Delta^{(2)}] = F_0 + \sum_{Qm} \tilde{a}_{Qm}(|\Delta^{(1)}_{Qm}|^2 + |\Delta^{(2)}_{Qm}|^2) + \frac{N_0}{4\omega_0^2}(|t|^2 - |v|^2)\sum_{Qm} |\Delta^{(1)}_{Qm}||\Delta^{(2)}_{Qm}|\cos(\varphi^{(1)}_{Qm} - \varphi^{(2)}_{Qm}) , \tag{12}$$

with

$$\tilde{a}_{Qm} = \frac{N_0}{2}\left\{\ln\frac{T}{T_{c0}} + \frac{\pi^2}{4}|m| + \xi_0^2 Q^2 + \frac{1}{4\omega_0^2}\left[|t|^2 - \frac{1}{3}|v|^2\right]\right\} + \frac{b_0}{2}\sum_{Q'} \langle |\Delta_{Q'0}|^2\rangle \delta_{m0} . \tag{13}$$

Here, $|v|^2 \equiv c\overline{|v_{nk}|^2 S(S+1)}$, c is the impurity concentration ($c \ll 1$), $S(S+1) \equiv \langle \mathbf{S}_n^2 \rangle$, and $\overline{(\cdots)}$ denotes the average over all impurity atoms and for $|v_{nk}|^2$ additionally over the Fermi surface. We assume that the density of states does not diverge at the Fermi surface so that performing a Fermi surface average makes sense. Taking, e.g., $c = 1\%$ and $S = \frac{1}{2}$, one has $|v| \sim 100$ meV so that it is realistic to obtain a sign reversal of the coefficient of the Josephson term in the free energy, Eq. (12), for $|v|^2 > |t|^2$, i.e., a π junction. The result is similar to earlier papers.[16]

To explain the PME, one has to evaluate the susceptibility $\chi_S(T)$, given by

$$\chi_S(T) = \frac{1}{\beta\Omega}\frac{\partial^2}{\partial B^2}\ln Z_B , \tag{14}$$

where Ω denotes the volume of the system, and Z_B is the partition function, calculated in the presence of a magnetic field B. But calculating Z_B means integrating out the phase difference in the Josephson term of the free energy, Eq. (12), so that Z_B and thus $\chi_S(T)$ no longer contain information about the sign of the Josephson term.[22] The physical reason for this irrelevance is that the absolute sign of the phase difference of an isolated Josephson junction makes no sense; one needs a "reference junction" to which a well-defined phase relation exists. As long as one considers Josephson networks consisting of many closed paths containing Josephson junctions, one always has these relations. Within the framework of our model, the CuO_2 bilayer is just equivalent to a single, isolated junction without any relevance of the absolute sign of the phase difference.

In Fig. 1 we plot the crossover temperature T_c^* as a function of impurity concentration [via the parameter $|v|^2$, defined below Eq. (13), which is proportional to the impurity concentration] for $|t| = 100$ meV and different values for the system size L, using $m_{\rm eff} = 6m_0$, $\xi_0 = 15$ Å, $\omega_0 = 50$ meV, and $T_{c0} = 82$ K as a set of parameters.[7] T_c^* decreases with increasing impurity concentration. For each L value there exists a maximum $|v|^2$ value (i.e., a maximum concentration of magnetic impurities) above which $T_c^* \equiv 0$. For comparison, Fig. 2 shows the measured depression of the superconducting critical temperature of $Tl_{2-x}Sm_xBa_2Ca_2Cu_3O_y$ as the concentration of magnetic Sm ions which replace the Tl ions between the CuO_2 triple layers in $Tl_2Ba_2Ca_2Cu_3O_y$ is increased.[23] Figures 1 and 2 suggest that additional spin-flip tunneling via magnetic impurities could be a possible mechanism to explain the decrease of T_c with increasing Sm concentration. However, as x exceeds ~ 0.8 the measured temperature dependence of the in-plane resistivity changes from metalliclike to insulatorlike behavior, i.e., for low temperatures the resistivity starts to increase with decreasing

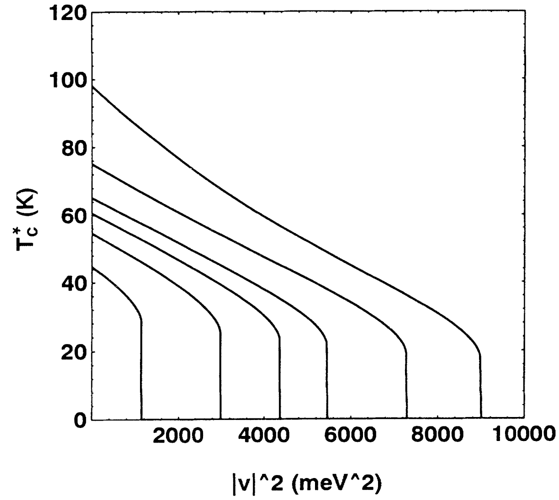

FIG. 1. T_c^* as a function of $|v|^2$ for $|t| = 100$ meV. From top to bottom: $L = 500$, 1000, 2000, 3000, 5000, and 10 000 Å.

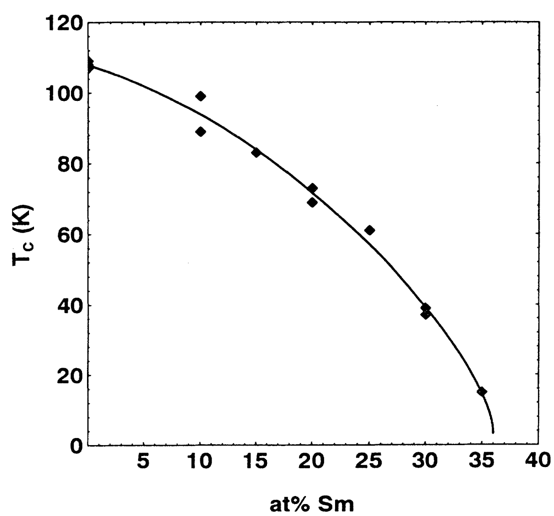

FIG. 2. T_c measured as a function of Sm concentration in $Tl_{2-x}Sm_xBa_2Ca_2Cu_3O_y$ (from Ref. 23). The solid line is a guide to the eye.

temperature before it drops to zero at T_c.[23] This influence of magnetic moments *between* the layers on the electronic properties *in* the layers is not yet understood and not included in the model described in Ref. 7 and this paper.

To conclude, we have considered the effect of magnetic impurities between the two CuO_2 planes on the 2D–0D crossover temperature T_c^*, introduced by the CuO_2 bilayer model of Ref. 7. T_c^* decreases with increasing impurity concentration, which could be a possible explanation for the observed decrease of the superconducting critical temperature of $Tl_{2-x}Sm_xBa_2Ca_2Cu_3O_y$ with increasing Sm concentration, although the change in the temperature dependence of the in-plane resistivity is not yet understood. The PME discovered in granular BSCCO and single-crystal YBCO cannot be explained by the model of a single CuO_2 bilayer in Ref. 7.

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