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EFFECT OF ASYMMETRY OF CuO_2 LAYERS IN COPPER OXIDES ON THE ANOMALOUS NEUTRON SCATTERING NEAR T_c

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Reasons for critical magnetic scattering of neutrons near T_c in copper oxides with CuO_2 layers whose nearest environment has no "up–down" symmetry are discussed. The intracrystalline electric field, which threads the CuO_2 planes on account of the asymmetry, induces coupling between the spin and momentum of the current carriers. This coupling is shown to result in a manifestation of virtual Cooper pairs in the imaginary part of the spin susceptibility. Thus spin density fluctuations as well as current fluctuations should participate in the scattering. A way of experimentally distinguishing between the two mechanisms is pointed out.

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The behavior of metals near superconducting phase transitions has been a topic of considerable interest during the last few decades. This field is attracting still more attention nowadays owing to the discovery of high-temperature superconductivity. The search for the mechanism responsible for this phenomenon has led to a thorough examination of conventional and unconventional behavior in every physical property and *inter alia* has stimulated reconsideration of many aspects of the order parameter fluctuations near T_c. In addition to ordinary magnetoresistivity measurements, new experimental approaches have been employed to study the problem, and one of them – inelastic neutron scattering – gives a positive result. An anomalous increase of the small-angle scattering near T_c in $YBa_2Cu_3O_{7-x}$ (YBCO) has been reported [1]. Critical neutron scattering usually occurs due to growing spin fluctuations near magnetic phase transitions. In that experiment [1], however, the anomaly was observed near the transition to the superconductive state. Although, to the best of our knowledge, analogous experiments with single crystals have not been conducted, there can be no doubt that fluctuations of the superconducting order parameter are capable of giving rise to critical neutron scattering. Therefore, it is important to have a complete picture of the processes through which the fluctuations can affect the scattering. In the present paper we examine the role of the crystal structure of superconducting cuprates in this problem.

The magnetic field created by the magnetic moment of a neutron interacts with both the magnetic moments of the carriers (holes) and with the electric current [2]. By virtue of this, the structure factor $S(\mathbf{q},\omega)$, to which the neutron scattering intensity is proportional, has the form

$$S(\mathbf{q},\omega) = \frac{1}{\exp(\hbar\omega/k_BT)-1}\left[\chi''(\mathbf{q},\omega) + \frac{\omega}{c^2q^2}\sigma'(\mathbf{q},\omega)\right], \qquad (1)$$

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where χ'' is the imaginary part of the spin susceptibility and σ' is the real part of the transverse conductivity.[2] Critical neutron scattering near magnetic phase transitions occurs due to critical behavior of the first term in Eq. (1). As opposed to that, in the case of a superconducting transition, it is natural to associate the anomalous scattering with the second term in the equation. Indeed, a primary source of any anomaly on approaching T_c is the formation of virtual Cooper pairs. It is known that one of two mechanisms by means of which the fluctuating Cooper pairs can affect physical quantities, the Maki – Thompson process [3], is suppressed because of strong phase breaking (which is believed to be inherent in high-T_c superconductors), while the other, the Aslamasov – Larkin process [4], is operative only in the conductivity but not in the susceptibility under usual conditions. Nevertheless, it will be shown below that owing to a feature of the crystal structure of some copper oxides, including YBCO, the superconducting fluctuations do give rise to a correction to $\chi''(\omega, \mathbf{q})$ which diverges at T_c just as $\sigma'(\omega, \mathbf{q})$.

This feature is the asymmetry of the nearest environment of the CuO_2 planes, to which the current carriers are confined. In the case of YBCO, the planes are surrounded by yttrium ions on the one side and by a Ba – O plane on the other side. The loss of "up–down" symmetry means the existence of an intracrystalline electric field threading the CuO_2 plane. For a given CuO_2 plane, the presence of the field adds the term

$$H_{so} = \frac{\alpha}{\hbar}(\mathbf{p} \times \mathbf{c}) \cdot \boldsymbol{\sigma} \tag{2}$$

to the Hamiltonian of two-dimensional (2D) holes [5], where $\mathbf{p}$, $\boldsymbol{\sigma}$, and $\mathbf{c}$ are, respectively, the 2D momentum, the Pauli matrices, and a unit vector directed along the field. Note that the coexistence of the local electric field and superconductivity is not a new fact. It was first discussed [6] in connection with A-15 compounds, the high-T_c superconductors of the sixties, some of which undergo a structural transition to a polar phase at a temperature T_M somewhat above the superconducting transition T_c. Since the vectors $\mathbf{c}$ of two adjacent CuO_2 planes are oppositely directed, YBCO may be viewed as an antipyroelectric.

The following heuristic arguments show how the mirror symmetry breaking makes it possible for the superconducting fluctuations to affect χ''. Due to the Hamiltonian H_{so}, spin–velocity correlations (SVCs) appear in the system. They would be forbidden in a centrosymmetric system because velocity and spin have different transformation properties under space inversion – the former is even and the latter is odd. One of consequences of the SVC is the magnetoelectric effect (MEE) predicted earlier [7]: the supercurrent $\mathbf{J}_s$ must be accompanied by the spin polarization $\mathbf{S}$ of the carriers in an amount proportional to $\mathbf{c} \times \mathbf{J}_s$. The naive treatment of the singlet superconductor would seem to be in definite conflict with this prediction. Indeed, the singlet Cooper pair surely is not able to possess any magnetization regardless of its motion state. However, the common belief that superconductor physics is determined solely by the properties of the order parameter (the pair wave function) is incorrect for superconductors with broken mirror symmetry. In order to understand the reason for the MEE, one should remember

[2] Note that the susceptibility and conductivity in Eq. (1) are not the same quantities that appear in the Maxwell equations. To get the material χ and σ, one should, in the Feynman-diagram language, exclude from the set of diagrams for the spin–spin and current–current correlation functions those diagrams that can be cut into two parts by cutting an internal photon line. Written in terms of the material χ and σ, the right-hand side of Eq. (1) would contain additional screening factors $Z(\mathbf{q}, \omega)_\chi$ and $Z(\mathbf{q}, \omega)_\sigma$ in front of χ and σ, respectively. However, the differences $1 - Z_{\chi,\omega}$ can be shown to be negligible under the experimental conditions [1].

that the off-diagonal correlation function (ODCF), $\langle\psi_\alpha(\mathbf{r})\psi_\beta(\mathbf{r}')\rangle$ (where $\psi_\gamma(\mathbf{r})$ is the electron field operator), which manifests a spontaneous breakdown of the $U(1)$ gauge invariance, is the basic concept of the pairing theory [8] rather than the order-parameter matrix $\Delta_{\alpha\beta}$. Usually, in centrosymmetric systems, the spinor structure of the ODCF is identical with that of $\Delta_{\alpha\beta}$. By contrast, the band spin–orbit coupling results in the presence of a triplet part in the ODCF in addition to the singlet part even for a singlet order parameter. Moreover, it turns out that the relationship between the two parts depends on the condensate motion. It is this dependence that gives rise to the MEE. Since the neighboring CuO_2 sheets are oppositely oriented, the total magnetization equals zero. However, the mean square spin polarization due to the supercurrent fluctuations above T_c need not vanish. While the band spin–orbit coupling is caused by the local electric field, the field squared will be shown to appear in the spin fluctuations. Therefore, the contributions of all CuO_2 sheets simply add up.

The model to be calculated is based on the following premises. It is assumed that the crystal under study may be considered as a system of conducting asymmetric layers with opposite orientation of the adjacent layers and that the tunneling between the layers is negligible. It is also supposed that the electron spectrum in the absence of H_{so} and of the interparticle interaction is parabolic and isotropic, and that the singlet s-type pairing takes place, i.e. the Hamiltonian of one layer has the form

$$H = \int d^2r \left\{ \frac{\hbar^2}{2m}\nabla\psi_\gamma^+(\mathbf{r})\cdot\nabla\psi_\gamma(\mathbf{r}) + \alpha\psi_\beta^+(\mathbf{r})(\frac{\nabla}{i}\times\mathbf{c})\cdot\sigma_{\beta\gamma}\psi_\gamma(\mathbf{r}) + \right.$$
$$\left. + \frac{\lambda_s}{2}[\psi_\beta^+(\mathbf{r})g_{\beta\kappa}\psi_\kappa^+(\mathbf{r})][\psi_\delta(\mathbf{r})g_{\delta\gamma}\psi_\gamma(\mathbf{r})] \right\}, \tag{3}$$

where $\hat{g} = i\sigma_2$ and λ_s is the pairing coupling constant. The SO energy αp_F is considered to be small on the Fermi energy scale, i.e., $\alpha m/p_F \ll 1$. Then, our main result, valid for the model at $T - T_c \ll T_c$, $q\xi_{ab}(0) \ll 1$, and $l \gg \xi_{ab}(0)$ (where $l = v_F\tau$ is the mean free path and $\xi_{ab}(0) = v_F/2\pi T_c$ is the in-plane coherence length at 0 K) has the form

$$\delta\chi''_{2,ij} = (\delta_{ij} - c_i c_j)\delta\chi''_2, \qquad \frac{\delta\chi''_2(\omega,\mathbf{q})}{\chi_{P,2}} = \frac{\pi\omega}{8\epsilon_F}G^2_{(2)}F_2(T,\mathbf{q}), \tag{4}$$

where $\chi_{P,2} = \mu_B^2 m/\pi$ is the 2D Pauli susceptibility, μ_B is the Bohr magneton, $\epsilon_F = (1/2m)p_F^2$ is the Fermi energy,

$$G_{(2)} = \frac{\alpha m}{2p_F} f_2\left(\frac{\alpha p_F}{\pi T_c}\right)$$

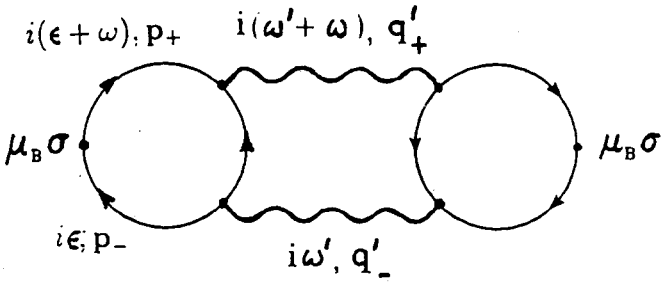
characterizes the coupling between the spin density and the supercurrent,

$$f_2(x) = (8/7\zeta(3))\sum_{n\geq 0} x^2(2n+1)^{-3}[(2n+1)^2 + x^2]^{-1},$$

$\zeta(n)$ is the Riemann zeta function, the function

$$F_2(T,\mathbf{q}) = \frac{T_c}{T - T_c}L_2\left(\frac{\kappa_2}{2}q\xi(T)\right) \tag{5}$$

plays the role of an Ornstein – Zernike factor, $L_2(x) = [x\sqrt{1+x^2}]^{-1}\sinh^{-1}x$, $\xi(T) = \xi_{ab}(0)T_c^{1/2}(T - T_c)^{-1/2}$, $\kappa_2 = [7\zeta(3)/8]^{1/2}$, and units are used in which Boltzmann's constant k_B and $\hbar$ are unity.



The diagram for the contribution of superconducting fluctuations to the spin susceptibility. The open circle represents the three-point vertex function. The wavy lines is the superconducting fluctuation propagator and the solid line is the fermion propagator. Here $\mathbf{q}'_\pm = \mathbf{q}' \pm \mathbf{q}/2$ and $\mathbf{p}_\pm = \mathbf{p} \pm \mathbf{q}/2$.

The fluctuation contribution to χ is given by the Aslamazov – Larkin type diagram shown in Figure. The fermion lines represent the one-particle Green function

$$\hat{G}(i\epsilon, \mathbf{p}) = \hat{\Pi}^{(+)}(\mathbf{p})[i\epsilon - \xi_{(+)}(p)]^{-1} + \hat{\Pi}^{(-)}(\mathbf{p})[i\epsilon - \xi_{(-)}(p)]^{-1}, \tag{6}$$

where the operator $\Pi^{(\pm)}_{\alpha\beta}(\mathbf{p}) = 1/2[\delta_{\alpha\beta} \pm (\hat{\mathbf{p}} \times \mathbf{c}) \cdot \boldsymbol{\sigma}_{\alpha\beta}]$ represents projection onto states with a definite helicity (the projection of a spin on the $\mathbf{p} \times \mathbf{c}$ direction) and $\xi_{(\pm)}(p) = (p^2/2m) \pm \alpha p - \mu$ are the energies of these states. The wavy line represents the pairing fluctuation propagator $g_{\alpha\beta} D(\mathbf{q}, i\omega') g_{\gamma\delta}$, where $\mathbf{q}$ is the momentum and $i\omega'$ is the frequency of the virtual Cooper pair, and the spinor indices $\alpha\beta$ and $\gamma\delta$ refer to the spin states of the carriers joining to form the pair and releasing after its subsequent decay, respectively. It can be shown that the fluctuation propagator has the usual form

$$D^{R(A)}(\mathbf{q}, \omega) = -\left\{ N_2(0) \left[\frac{T - T_c}{T_c} \mp \frac{i\pi\omega}{8T_c} + \frac{7\zeta(3)}{32\pi^2} \left(\frac{q v_F}{T_c} \right)^2 \right] \right\}^{-1},$$

up to corrections of the order of $(\alpha p_F/\epsilon_F)^2$. Here $N_2(0) = m/2\pi$ and the superscript $R(A)$ stands for the retarded (advanced) part of the function. The most important parts of the diagram are the left and right three-point vertex functions that describe the conversion of the supercurrent density into the spin density. The functions can be found by a method pointed out earlier [7] and under the conditions mentioned above have the form

$$\mu_B(\mathbf{c} \times \mathbf{q}')\alpha m \frac{7\zeta(3)}{16\pi^3 T_c^2} f_2\left(\frac{\alpha p_F}{\pi T_c}\right). \tag{7}$$

The imaginary part of the diagram comes from the product of the fluctuation propagators

$$Z(i\omega_n, \mathbf{q}'_\pm) = T \sum_\nu D(i\omega'_\nu, \mathbf{q}'_+) D(i\omega'_\nu + i\omega_n, \mathbf{q}'_-)$$

and has the standard form. Thus

$$\int \frac{q'^2 d^2 q'}{(2\pi)^2} Z''(\omega + i0^+, \mathbf{q}'_\pm) = 2\omega \left(\frac{T_c}{T - T_c}\right) \left(\frac{2\pi^3 T_c^2}{7\zeta(3)\epsilon_F}\right)^2 L_2\left(\frac{\kappa_2}{2} q\xi(T)\right). \tag{8}$$

Equations (4) and (5) may now be obtained immediately.

Up to this point, to accentuate the main idea, we have discussed a simple model in which the the pairing interaction was considered to be the only possible interaction between carriers. However, the cuprates are highly correlated and disordered systems;

narrow energy bands, strong Coulomb repulsion, and proximity to a metal–insulator transition are accepted as integral parts of the physics of cuprate superconductors. These factors affect the first and second term in Eq. (1) in opposite ways – they suppress the conductivity but enhance the ferromagnetic correlations (FMCs), which lead to a renormalization of the left and right σ-vertices of the diagram in Figure. (The FMCs in the hole system must not be confused with the antiferromagnetic correlations (AFMCs) of localized spins of the copper ions.) Indeed, the account of the one-site Coulomb interaction (in the random phase approximation) gives the quasi-Stoner factor $K = [1 - UN(0)]^{-1}$ to the vertices, U being the interaction. Then $\chi''(\omega, \mathbf{q})$ is enhanced by K^2. The unusually high relaxation rate of the planar ^{17}O nuclei, which is also mainly determined by the FMCs (the AFMCs are less active at those places), as well as the ferromagnetlike behavior of some relative copper oxides (e.g., the Na-doped La_2CuO_4 [9] and $La_4Ba_2Cu_2O_{10}$ [10]) also points to an essential role of the FMCs in the cuprates. This assumption is not inconsistent with the not much enhanced uniform susceptibility generally observed in high-T_c cuprates, because the latter contains contributions of both signs, being determined by all degrees of freedom. Now let us compare the fluctuation-induced corrections to the first and second term in Eq. (1). As $\delta\sigma' = (e^2/16)T_c/(T - T_c)$, one gets

$$\frac{\delta\chi''}{\delta\sigma'}\left(\frac{c^2q^2}{\omega}\right) = \left[\frac{q}{p_F}\frac{m}{m_0} f\left(\frac{\alpha p_F}{\pi T_c}\right) K \frac{\alpha p_F}{4\epsilon_F}\right]^2, \tag{9}$$

where m_0 is the electron mass in the vacuum. It is seen that, at a given α, the factors that favor the relative magnitude of the first term are the low density and the heavy mass of the carriers as well as and the enhanced value of K. The minimal q in the experiment [1] was equal to $3.5{\cdot}10^6\mathrm{cm}^{-1}$, hence $qm/p_F m_0 \sim 0.3$ at $m \sim 3m_0$ [11]. A crude estimate of α in YBCO [12] has shown $\alpha p_F \sim 10^2$K at $p_F \sim 3{\cdot}10^7\mathrm{cm}^{-1}$, i.e. one can suppose $\alpha p_F/\pi T_c \sim 1$, so that $f(\alpha p_F/\pi T_c) \sim 1$. The parameter $\alpha p_F/4\epsilon_F$ is small, but the factor K may be large. At $K \sim 10$, both terms in Eq. (1) have the same ordér of magnitude.

In order to know which fraction of the neutron scattering is due to χ'', one should compare high-T_c cuprates with asymmetric (e.g., YBCO) and symmetric (e.g., $(La,Ba)_2CuO_4$) CuO_2 layers. The difference between the paraconductivities of these crystals can be determined by resistivity measurements. In principle, the value of χ'' can also be measured in experiments on magnetic field absorption. Besides the copper oxides, all that has been said above also applies to some of the superconducting ternary silicides [13] ($CeCoSi_3$ and probably $LaRhSi_3$ and $LaIrSi_3$) which crystallize in a polar structure with space group *I4mm*. They are composed of heavy elements and exhibit strong breaking of reflectional symmetry, which is suggestive of strong spin–orbit coupling.

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