Magnetic structure of lightly doped cuprates

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The effect of impurity magnetic states on the long-range order in a layered antiferromagnet has been studied. A magnetic phase diagram has been constructed for lightly doped $La_{2-x}Sr_xCuO_4$. The layered structure and the long-range effects shrink the antiferromagnetic region and give rise to a reentrant antiferromagnetic transition.

The antiferromagnetism of high-temperature superconductors is associated with an indirect exchange between localized Cu spins. The anisotropy of these compounds leads to a pronounced difference between the exchange interaction in the CuO_2 planes, J, and that between planes, J' ($J' \ll J$) (Ref. 1). Doping creates holes in the P shells of oxygen ions from the CuO_2 planes.[2] At low temperatures T these holes are coupled with acceptors if the concentration of the latter (x) is low. Evidence for this conclusion comes from the Mott nature of the conductivity.[3] Localized holes create magnetic defects in an antiferromagnet.[4] Surprisingly, however, the antiferromagnetism of $La_{2-x}Sr_xCuO_4$ is destroyed at vanishingly low concentrations ($x_2 \approx 0.02$; Ref. 1) of such defects. In the present paper we link this fact with the long-range nature of the magnetic perturbations caused by acceptors and with the pronounced anisotropy. A

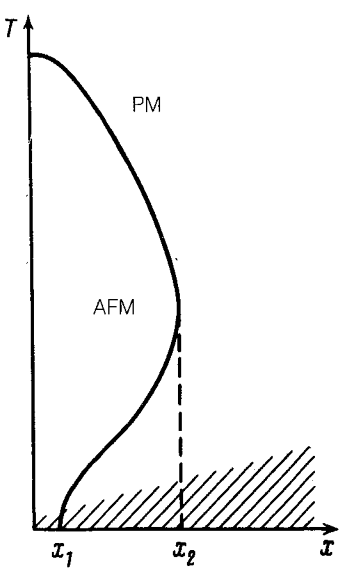

FIG. 1. Magnetic phase diagram of a lightly doped layered antiferromagnet. PM is the paramagnetic phase. The hatching shows a possible spin-glass region.

possible reason why doping has a vastly weaker effect on the antiferromagnetism in $YBa_2Cu_3O_{6+x}$ ($x_2 \approx 0.4$) is that the defects have a different symmetry, which forbids long-range effects.

A hole imposes a "defective" magnetic order, different from an antiferromagnetic order, in a region of size D near an acceptor. We studied this order in Ref. 5. The interaction of the periphery of a defective region with the antiferromagnetism leads to a slowly decreasing perturbation in the antiferromagnetic matrix (unless this decrease is forbidden by the symmetry[5]). Such perturbations were first noted by Villain[6] and, in connection with high-temperature superconductivity, by Aharony *et al.*[4] The long-range effects and the small value of the parameter J'/J make it possible for us to find the boundary of the region of three-dimensional antiferromagnetism in the T, x phase plane (Fig. 1). Impurities begin to influence the magnetic order at $x > x_1 \sim J'/J$. An antiferromagnetic order is not realized at all, at any T, if the condition $x > x_2 \sim [D^2 \ln(J/J')]^{-1}$ holds. Because of the factor $\ln^{-1}(J/J')$, the value of x_2 lies below the threshold ($x_c \sim D^{-2}$) for a percolation along defective regions; in other words, the magnetic transition is not a percolation transition. In the parametrically wide region $x_1 < x < x_2$ a reentrant transition arises; the reason for it is an intensification of the correlation between impurity spins with decreasing T.

The defective region has a finite spin M, which (like D) can be found from a calculation of the structure of the defective state at the elementary level.[5] The minimum energy of the defect-matrix interaction corresponds to the case in which the direction of the impurity spin, $\mathbf{m} = \mathbf{M}/M$, is perpendicular to the antiferromagnetism unit vector $\mathbf{n}(\mathbf{r})$ in the limit $r \to \infty$ (Ref. 6). The asymptotic form of the perturbation of $\mathbf{n}$ at large r is of dipole form:

$$\delta \mathbf{n} = (\mu / 2\pi) \mathbf{m} (\mathbf{er})/r^2 , \tag{1}$$

where the unit vector $\mathbf{e}$ runs parallel to one of the tetragonal axes ($\mathbf{a}$ or $\mathbf{b}$) in the CuO_2 plane. The four possible directions of $\mathbf{e}$ associated with the four possible positions of the defect with respect to the antiferromagnetic sublattices. The asymptotic behavior described by (1) and the random nature of $\mathbf{e}$ are universal properties, which are independent of the particular nature of the elementary Hamiltonian. The defect characteristic μ is determined by the particular structure of the defect; a calculation[5] for the simplest case yields $\mu \approx 6D$.

In a purely two-dimensional antiferromagnet, an arbitrarily low concentration of dipole defects will destroy the long-range order,[6] even at $T = 0$. In a quasi-two-dimensional antiferromagnet, the long-range order thus disappears even at a low impurity concentration (low to the extent that J'/J is low). The Néel temperature T_N for the establishment of a three-dimensional antiferromagnetic order is found[7] from the condition $T_N \sim J' \xi^2(T_N, x)$. The correlation radius ξ for a two-dimensional antiferromagnet has an exponential T dependence. Accordingly, for the purpose of calculating T_N in the leading approximation in $\ln(J/J')$, we rewrite the latter condition as

$$\xi(T_N, x) = \xi_0 \equiv (J/J')^{1/2} . \tag{2}$$

The cutoff of the weak dependence described by (1) due to layer-layer exchange occurs at $r \gtrsim \xi_0$ and does not affect $\xi(T,x)$. In the concentration region of interest here, the average distance between acceptors satisfies $x^{-1/2} \gg D$; the defective region can be replaced by a point and can be characterized by vectors **e** and **m**. As a result, the correlation radius ξ of field $\mathbf{n}(\mathbf{r})$ is determined by the thermodynamics of the $2D$ system with the Hamiltonian

$$H = (\rho/2)\int d^2\mathbf{r}(\vec{\nabla}\mathbf{n})^2 + \rho\mu\sum_i(\mathbf{e}_i\vec{\nabla})(\mathbf{n}(\mathbf{r}_i)\mathbf{m}_i), \tag{3}$$

where $\rho \sim J$ is the spin stiffness, $\mathbf{n}(\mathbf{r})$ and $\mathbf{m}_i$ are dynamic variables ($|\mathbf{n}| = 1$, $|\mathbf{m}| = 1$), and the random nature of the arrangement of impurities, $\mathbf{r}_i$, and of the vectors $\mathbf{e}_i$ determines the static disorder. The moments $\mathbf{m}_i$ form a random plane magnetic material with a characteristic dipole-dipole interaction energy $U = \rho\mu^2x/4$. A spin-glass ordering apparently does not arise in this $2D$ magnetic material,[8] and the corresponding correlation radius ζ is finite at all $T \neq 0$. In other words, there are no correlations between the spins of remote impurities. Nevertheless, fluctuations in **n** are determined by specifically the remote impurities, since perturbations (1) decay slowly.

Let us find the effective Hamiltonian of long-wave fluctuations in **n** (these are the fluctuations which determine ξ). To find it we begin with an integration over the "fast" components of $\mathbf{n}(\mathbf{r})$, with a length scale $r > L$, where $\zeta \ll L \ll \xi$. This renormalization generates an additional term in (3): $H \to H + \mathscr{H}\{\mathbf{m}\}$. This term does not depend on L, by virtue of the condition $L \gg \zeta \gtrsim x^{-1/2}$, and it is the same as the Hamiltonian of the dipole-dipole interaction:

$$\mathscr{H}\{\mathbf{m}\} = (\rho\mu^2/2)\sum_{i\neq j}(\mathbf{m}_i\mathbf{m}_j)\{(\mathbf{e}_i\mathbf{e}_j) - 2(\mathbf{e}_i\mathbf{r}_{ij})(\mathbf{e}_j\mathbf{r}_{ij})/r_{ij}^2\}\, r_{ij}^{-2}\,. \tag{4}$$

The renormalizations of the constants ρ and μ are small by virtue of the condition $\ln L \ll \ln\xi$.

The next step is to integrate over $\mathbf{m}_i$. Since $\mathbf{n}(\mathbf{r})$ now contains only long-wave components, the quantity $\vec{\nabla}\mathbf{n}$ is small, and it varies only slowly in space. In the lowest approximation in $\vec{\nabla}\mathbf{n}$, the integration over $\mathbf{m}_i$ leads to a constant which does not depend on $\vec{\nabla}\mathbf{n}$: the free energy of the system with Hamiltonian (4). The next approximation corresponds to the linear response of this system to a weak quasiuniform field $\vec{\nabla}\mathbf{n}$ and leads to a term $(-\tilde{\chi}/2)\int d^2\mathbf{r}\vec{\nabla}\mathbf{n})^2$ in the effective Hamiltonian. The generalized susceptibility is

$$\tilde{\chi} = \rho(\rho\mu^2/4TV)\sum_{i,j}\langle(\mathbf{e}_i\mathbf{e}_j)\langle(\mathbf{m}_i\mathbf{m}_j)\rangle_T\rangle_c\ , \tag{5}$$

where V is the volume of the system, $\langle...\rangle_T$ means a thermodynamic average, and $\langle...\rangle_c$ means a configurational average. At distances $L \gg \zeta$ we have ignored the spatial dispersion of $\tilde{\chi}$. The absence of any special directions in spin space and coordinate space corresponds to a scalar nature of $\tilde{\chi}$. It follows from (4) and (5) that $\tilde{\chi}$ depends on x and T only through the ratio $T/U = 4T/\rho\mu^2x$: $\tilde{\chi} = \rho f(4T/\rho\mu^2x)$, where f is a dimensionless function. The effective Hamiltonian of the long-wave part of the field $\mathbf{n}$ thus has the form

$$H_{eff} = (\rho_{eff}(4T/\rho\mu^2 x)/2)\int d^2\mathbf{r}(\vec{\nabla}\mathbf{n})^2, \qquad \rho_{eff}(z) = \rho(1 - f(z)). \tag{6}$$

For the Hamiltonian of a $2D$ $\mathbf{n}$ field the correlation radius obeys[9] $\xi \propto \exp\{2\pi\rho_{\text{eff}}/T\}$. Substituting this expression into (2), we find an equation which determines the functional dependence $T_N(x)$:

$$f(\tau/y) = 1 - \tau, \tag{7}$$

where $y = x/x_0$ and $\tau = T_N/T_N(0)$ are the reduced concentration and reduced temperature of the antiferromagnetic transition, $T_N(0) = 2\pi\rho/\ln\xi_0$, and $x_0 = 8\pi/\mu^2\ln\xi_0$. At low concentrations $x \ll x_0$ we can use a high-temperature expansion for $\tilde{\chi}$. In the leading order, only the terms with $i = j$ in (5) are important, and we have $f(z) \approx 1/z$. From (7) we find

$$T_N(x)/T_N(0) = 1 - (x/x_0) - (x/x_0)^2 \ldots \quad . \tag{8}$$

The term $\sim x^2$ in (8) does not go beyond the accuracy of this approximation, since terms $\propto z^{-2}$ vanish in the expansion of $f(z)$.

Let us discuss the shape of the magnetic phase diagram at a qualitative level. This shape is determined by the behavior of $\tilde{\chi}$ (T) at $T \sim U$. As was shown above, the system of impurity spins remains paramagnetic in this temperature range, so the ordinary susceptibility $\chi(T)$ increases monotonically with decreasing T and does not reach saturation. This susceptibility differs from $\tilde{\chi}$ [see (6)] in that it does not contain a factor $(\mathbf{e}_i\mathbf{e}_j)/2$. We can expect, however, that the functional dependences $\chi(T)$ and $\chi(T)$ will be qualitatively the same in the paramagnetic phase, and the latter will also undergo an unbounded growth. It follows from (7) that this behavior of $\tilde{\chi}(T)$ unavoidably leads to a reentrant transition from the antiferromagnetic state. It is easy to see that for a transition of this type even a weaker condition would be sufficient: The function $f(z)$, as it increases, should reach the value $f = 1$.

The physical reason for the reentrant transition is that the correlation of perturbations (1) increases with decreasing T. Such a correlation exists only under the strong inequality $x^{-1/2} \ll \xi$. This inequality is violated if $x \sim x_1$. If $x < x_1$, the impurities obviously cannot destroy the three-dimensional antiferromagnetic order at low T, and no reentrant transition will occur.

Let us estimate x_2, approximating $\tilde{\chi}(T)$ by a Curie-Weiss law $f(z) = 1/z$. As a result, we find from (7) $T_N(x)/T_N(0) = (1/2)(1 \pm \sqrt{1 - 4x/x_0})$, i.e., $x_2 = x_0/4$. Substituting in the experimental values[7] $T_N(0) \approx 300$ K and $2\pi\rho \approx 1200$ K, we find $x_2 = 0.02$ with $\mu \approx 10$. The estimate $\mu = 6D$ found above shows that the actual value of x_2 corresponds to reasonable defect dimensions: $D = 1$–2 lattice constants.

We wish to emphasize that even under the conditions prevailing in an antiferromatically ordered matrix the impurity spins remain paramagnetic. Accordingly, and despite the low concentration, the contribution (χ) of these spins to the static susceptibility is comparable to that of the matrix. How does this system behave below the line of the reentrant transition, i.e., at $T < U$? Although there is no long-range order under these conditions, the correlation length ζ increases with decreasing T. This tendency can lead to a long-term relaxation and to a strong frequency dispersion of χ. At even lower temperatures T, the three-dimensional nature of the system becomes important, as does the magnetic anisotropy. Under these conditions the magnetic impurities should go into a spin-glass state.[8]

So far, there has been no direct experimental observation of a reentrant magnetic transition in La–Sr–Cu–O. There are, on the other hand, indirect pieces of evidence in favor of such a transition (see the review by Birgeneau and Shirane[1]). We wish to thank Vik. S. Dotsenko, M. V. Feĭgel'man, and A. M. Finkel'shteĭn for discussions.

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