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Role of antiferromagnetic interlayer coupling on magnetic properties of $YBa_2Cu_3O_{6+x}$

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In this paper we have studied the role of interlayer coupling on the Néel temperature of $YBa_2Cu_3O_{6+x}$. We have used the expression for the Néel temperature derived from the three-dimensional anisotropic Heisenberg model in the random-phase approximation. With the help of numerical calculations we have obtained an analytical expression for the Néel temperature as $T_N = J_\parallel/[0.1616\ln(J_\parallel/J_\perp)+0.5055]/k_B$. This expression shows that $T_N \rightarrow 0$ as $J_\perp \rightarrow 0$. We have also compared our theoretical results with the doping-dependent Néel temperature of $YBa_2Cu_3O_{6+x}$ compounds. Here, x is the doping concentration of oxygen. Good agreement between theory and experimental results is achieved provided that the ratio of interplanar to intraplanar coupling strengths depend linearly on the doping concentration x. Further, the intraplanar coupling strength is treated as a constant independent of x in the numerical calculations.

a. Introduction. The anomalous magnetic and electronic properties of $YBa_2Cu_3O_{6+x}$ (1:2:3) and $La_{2-y}Sr_yCu_{4-x}$ (2:1:4) high-temperature superconductors (HTSC) have attracted much attention ever since their discovery. The insulating phase of these superconductors provides much information about the magnetic dynamics of these systems.[1-16] These high-temperature superconductors are antiferromagnetic in the normal state with a large anisotropy in the antiferromagnetic coupling constant within CuO_2 planes and between CuO_2 planes in the c direction. This remarkable anisotropy in the antiferromagnetic correlation coupling in these layered structures is known to be responsible for most of the unusual magnetic properties.[17]

The very structure of 1:2:3 and 2:1:4 suggests that their ratios of interplanar to intraplanar coupling would be different. It has been shown that antiferromagnetism (AFM) in these compounds is driven by weak interlayer $J_\perp$ coupling, which is a manifestation of the large amount of orthorhombic distortion present in these compounds. It has been observed that the Néel temperature depends crucially on the interplanar coupling in these materials.[18,19] It has been ascertained that as two-dimensional (2D) correlations become sufficiently long ranged in these high-temperature superconductors, 3D ordering follows due to the presence of this weak but finite interlayer coupling. In the temperature range $T < T_N$, 3D ordering sets in and the interlayer coupling plays a crucial role in determining the antiferromagnetic correlation in these materials.[20,21] Here, T_N is the Néel temperature.

A lot of experimental evidence has come up during the last couple of years suggesting the importance of interlayer coupling in the magnetic dynamics of layered superconductors.[22] It has been shown that the Néel temperature decreases as the material is doped with oxygen. At a critical value of $x \sim 0.4$ in 1:2:3 the Néel temperature is found to reduce to zero.[19] The changing concentration of oxygen in $YBa_2Cu_3O_{6+x}$ makes the system undergo an antiferromagnetic-to-superconducting transition. Addition of oxygen into these layered compounds would destroy the antiferromagnetic long-range order along with the appearance of the metallic superconducting phase. The doping is crucial, since a variety of interactions come into play because of it. These interactions are discussed extensively by Singh.[23]

The weak interlayer coupling in these layered superconductors and its role in the magnetic dynamics has recently become a highly pursued topic. Two-dimensional and quasi-two-dimensional Heisenberg models have been extensively studied to get an insight into the magnetic dynamics of these HTSC. Neutron-diffraction experiments of Tranquada *et al.*[19] revealed the structure and the variation of Néel temperature with doping in $YBa_2Cu_3O_{6+x}$. By considering the dependence of Néel temperature in layered systems as $kT_N = J_\perp^{\text{eff}}\xi_{2D}^2$ where ξ_{2D} is the correlation length in two-dimensional plane and $J_\perp^{\text{eff}}$ is the effective coupling between planes, they achieved an effective interplanar coupling of 0.002 meV in 2:1:4. 2D exchange parameters calculated from the inelastic-light-scattering experiments give an estimate of 98 meV for 1:2:3 and 128 meV for 2:1:4 compounds.[22-24]

There has been considerable theoretical effort[25] to study the role of interplayer coupling on the magnetic properties of HTSC. For instance, Chakravarty, Halperin, and Nelson[20] have calculated the correlation length at low temperatures by using the renormalization-group approach and by taking quantum nonlinear σ model. They have estimated the interlayer coupling in La_2CuO_4 (2:1:4) to be of the order of $10^{-5}J_\parallel$ and have shown that the interlayer coupling has insignificant effect on two-dimensional correlation above 3D Néel ordering temperature. Further, the zero-temperature properties calculated for an isolated CuO_2 plane are only weakly affected by such a small value of $J_\perp$. Singh and co-workers[26,27] have pursued the role of interlayer coupling to study the magnetic dynamical properties of these compounds. They have shown that due to excitation of spin waves there is a crossover from three-dimensional to quasi-two-dimensional behavior with temperature dependence of sublattice magnetization varying from $\sim T^2$ (three-dimensional case) to $\sim T\ln T$ (quasi-two-dimensional case). According to them, the Néel temperature in copper-based antiferromagnets fall logarithmically with decreasing interlayer coupling. Liu has investigated the role of interlayer coupling to study the specific heat by using the quasi-two-dimensional Heisenberg model.[28]

The three-dimensional anisotropic Heisenberg model is difficult to study and there has been very limited effort to analyze the magnetic dynamics in such a model of HTSC. Most of the above-mentioned results were obtained in a two or quasi-two-dimensional Heisenberg model. Recently we have examined the three-dimensional Heisenberg model keeping the anisotropy in the exchange coupling. By using the two-sublattice approach developed by Hewson and ter Haar,[29] and solving the equation of motion for the Green's function in random-phase approximation, we obtained an analytical expression for magnetization and Néel temperature.[30] In this paper, we study the role of interlayer coupling on the Néel temperature of 1:2:3 compounds. We have calculated the variation of Néel temperature with the doping concentration and compared with the existing experimental results. We have taken intraplanar coupling as a constant independent of the doping concentration x throughout our calculations. We discuss the behavior of Néel temperature with the variation of the ratio of interplanar to intraplanar coupling and show that in the limiting case when $J_\perp / J_\parallel \rightarrow 0$, the 3D antiferromagnetic ordering is lost. An excellent agreement between our theory and experimental results is achieved with the assumption that the ratio of interplanar to intraplanar exchange coupling strength is directly proportional to the doping concentration x.

b. Theory. In the previous paper,[30] we applied the two-sublattice approach to antiferromagnets and obtained an analytical expression for the Néel temperature by considering the 3D anisotropic Heisenberg model and using the random-phase approximation in the evaluation of Green's function. The expression is given by

$$T_N = \frac{1}{8k_B} \left\{ \frac{1}{N} \sum_{\mathbf{k}} 2J_\parallel + J_\perp \Big/ \left[4(2J_\parallel + J_\perp)^2 - \left[J_\parallel \sum_{\mathbf{j}-\mathbf{f}\parallel ab} e^{i\mathbf{k}\cdot(\mathbf{j}-\mathbf{f})} + J_\perp \sum_{\mathbf{j}-\mathbf{f}\parallel c} e^{i\mathbf{k}\cdot(\mathbf{j}-\mathbf{f})} \right]^2 \right] \right\}^{-1} , \tag{1}$$

where k_B is the Boltzmann constant and N is the total number of spin sites. $2J_\parallel$ and $2J_\perp$ are the AFM coupling constant in CuO_2 plane and perpendicular to CuO_2 plane, respectively, in high-temperature superconductors. If we assume that the summation over $\mathbf{j}$ and $\mathbf{f}$ runs over nearest neighbors, then Eq. (1) changes into the following equation:

$$T_N = J / k_B I(r) , \tag{2}$$

where $I(r)$ is given as

$$I(r) = \frac{2}{N} \sum_{\mathbf{k}} (2+r) / \{ (2+r)^2 - [\cos k_x a + \cos k_y a + r \cos k_z c]^2 \} . \tag{3}$$

Breaking the fraction in the above equation into two parts and changing the summation over $\mathbf{k}$ into integration

$$I(r) = \frac{V}{2\pi^3} \int_0^{\pi/a} \int_0^{\pi/a} \int_0^{\pi/c} \left[\frac{dk_x dk_y dk_z}{(2 - \cos k_x a - \cos k_y a) + r(1 - \cos k_z c)} + \frac{dk_x dk_y dk_z}{(2 + \cos k_x a + \cos k_y a) + r(1 + \cos k_z c)} \right] . \tag{4}$$

Here, V is the volume of the unit cell. In the above equations we have replaced the ratio of interplanar to intraplanar coupling strengths by $r(=J_\perp / J_\parallel)$. It can be proved that the last term gives the same contribution to $I(r)$ as the first term. Therefore, the above expression of $I(r)$ reduces to

$$I(r) = \frac{V}{\pi^3} \int_0^{\pi/a} \int_0^{\pi/a} \int_0^{\pi/c} dk_x dk_y dk_z / [(2 - \cos k_x a - \cos k_y a) + r(1 - \cos k_z c)] . \tag{5}$$

Making the substitution $u = k_x a$, $v = k_y a$, $w = k_z c$, and integrating over w we get

$$I(r) = \frac{1}{\pi^2} \int_0^{\pi} \int_0^{\pi} du\, dv / [\sqrt{(2 - \cos u - \cos v)} \sqrt{(2 - \cos u - \cos v) + 2r}\,] . \tag{6}$$

Now, performing the integration over v the above integral reduces to

$$I(r) = \int_0^{\pi} F(u,r) K(u,r) du , \tag{7}$$

where F and K are given as

$$F(u,r) = 2 / \sqrt{(1 - \cos u + 2r)(3 - \cos u)}] \tag{8}$$

and

$$K(u,r) = \int_0^{\pi/2} d\theta / \sqrt{1 - \alpha \sin^2 \theta} , \tag{9}$$

where

$$\alpha = 4r / (1 - \cos u + r)(3 - \cos u) . \tag{10}$$

For very small r, K becomes constant $\pi/2$ and the leading contribution to the integration comes from small u. With the help of numerical calculation, we found that $I(r)$ is proportional to $\ln(1/r)$. This point will be discussed in the next section. Incorporating this dependence of $I(r)$ in Eq. (2), we find that the Néel temperature depends inversely on $\ln(1/r)$. It might be noted here that as $r \rightarrow 0$, $T_N \rightarrow 0$. This agrees with the experimental results.

c. Numerical results and discussion. In this section we have applied our theory to calculate the Néel temperature and its variation with the doping concentration. We have compared these calculations with the experimental results of 1:2:3 compounds. It can be easily shown from expression of $I(r)$ given in Eq. (7) that when $r=0$, $I(r)=\infty$. Hence, the Néel temperature $T_N = J_{\|}/k_B I(r) = 0$ for $r=0$. This is in agreement with the Mermin and Wagner theorem that there cannot be long-range order in 2D systems at finite temperature. Note that the Mermin and Wagner theorem does not exclude long-range order for 2D systems at zero temperature. We have calculated numerically $I(r)$ as a function of r using Eq. (7). The numerical results are presented in Fig. 1 by solid lines. It is shown that the value of $I(r)$ decreases as r increases.

From our numerical calculations we find that $I(r)$ can be approximately written as

$$I(r)=0.1616\ln\left[\frac{1}{r}\right]+0.5055 \tag{11}$$

for values of r ranging from 1.0×10^{-8} to 1. By using the analytical expression of $I(r)$ given by Eq. (11), we have calculated $I(r)$ as a function of r. The results are shown in Fig. 1 by the dashed line. One can see from the figure that the above analytical expression is a good approximation for $I(r)$ given in Eq. (7). Therefore, for all practical purposes, the Néel temperature can be approximately written as

$$T_N = J_{\|}/k_B[0.1616\ln(J_{\|}/J_{\perp})+0.5055] \; . \tag{12}$$

A similar logarithmic term has been previously obtained by Singh *et al.*[26]

We have studied the dependence of the Néel temperature on the doping concentration and the role of interplanar exchange interaction on the Néel temperature. We use Eq. (2) to calculate the Néel temperature. It is clear from this equation that doping dependent behavior of T_N depends on the variation of r in $I(r)$ for a constant value of $J_{\|}$. It means there should be a direct relation between r and x. Let us for a moment assume, that the ratio of interplanar- to intraplanar-exchange-interaction strength, r is a linear function of the doping concentration. For example,

$$r(x)=c(1-x/x_0) \; , \tag{13}$$

where $c=6.0\times10^{-3}$ and $x_0=0.41$. By using this equation, we have calculated the Néel temperature as a function of doping concentration. The theoretical calculation is presented along with the experimental values in Fig. 2. In our calculation we have taken the intraplanar-exchange-coupling interaction as a constant ($2J_{\|}=98.0$ meV) throughout the entire doping range. This is consistent with the experimental results reported so far for 1:2:3 and 2:1:4 compounds.[19,22] In these experiments it was shown that as doping increases the interplanar coupling decreases and reaches zero as Néel temperature becomes zero. But, intraplanar coupling does not vanish when Néel temperature reduces to zero. We found that the agreement between the theoretical results and experimentally measured values for the Néel temperature is quite good. This means that the above approximation

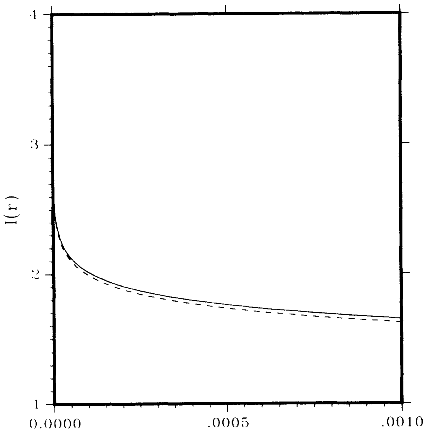

FIG. 1. The variation of $I(r)$ as a function of r. Solid curve is obtained from Eq. (7). The dashed curve is obtained from the analytical expression given in Eq. (11).

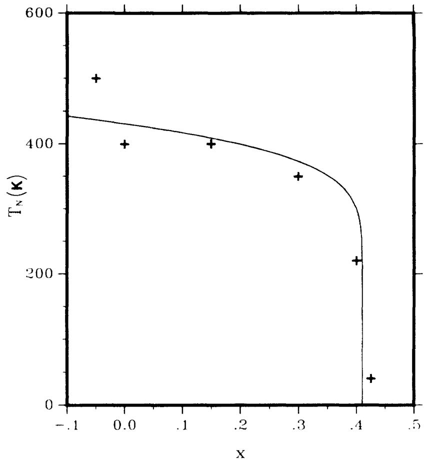

FIG. 2. The variation of Néel temperature as a function of doping concentration x in 1:2:3 compounds. The solid curve represents the theoretical results. Crosses represent the experimental points. Here, a linear relation between x and r is assumed.

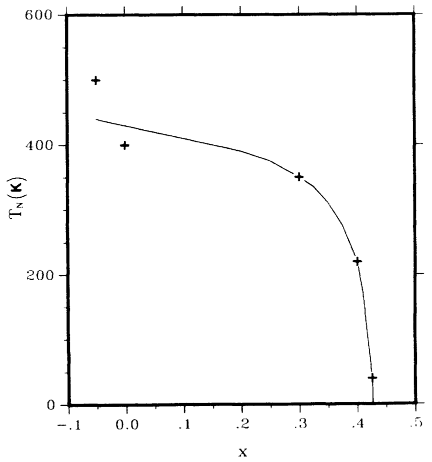

FIG. 3. The variation of Néel temperature as a function of doping concentration x in 1:2:3 compounds. The solid curve represents the theoretical results. Crosses represent the experimental points. Here, the solid curve is obtained by fitting Eq. (7) with experimental points by choosing the proper value of r.

where doping parameter is a linear function of r works pretty well, and it seems a reasonably accurate approximation.

We have also fitted the experimental results by using Eq. (2) and choosing a proper value of r at each value of doping concentration, while fixing the value of intraplanar coupling at 98.0 meV. The results are presented in Fig. 3. In Fig. 4 we plot the variation of r with x. We find the value of r by fitting our theoretical expression to the experimental points at each value of x. The dotted curve in Fig. 4 represents these values of r as a function of x. As we discussed earlier, Fig. 2 was obtained by assuming that r is a linear function of x. The solid curve in Fig. 4 represents this linear dependence as is given in Eq. (13). One can see from this figure that at $x=0$ and 0.4 there is no difference between the two curves but other than these values there is some deviation. The dashed curve seems to give a quasilinear relation between x and r for $0.05<x<0.3$. The slope of dashed curve is slightly different than that of the solid curve. For $x<0.0$ and $x>0.3$ the dashed curve shows a large nonlinearity. It can hence be concluded that the linear dependence between x and r is a fairly good approximation for most values of x.

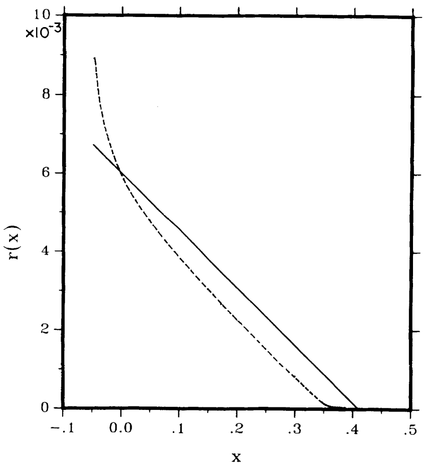

FIG. 4. The variation of r as a function of x. The solid curve represents the value of r obtained from Fig. 3 as a function of x. The dashed line represents the linear dependence of r with x.

In conclusion, we have calculated the Néel temperature as a function of doping in $YBa_2Cu_3O_{6-x}$ compounds. The comparison between theory and experiment is made. We found that if we assume a linear dependence between x and r a good agreement between theory and experiments is achieved. We also found an analytical expression for Néel temperature as a function of r, where it is shown that Néel temperature is inversely proportional to $\ln(1/r)$. This shows that as r approaches zero, Néel temperature goes to zero. This is consistent with all the experimental results.

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