

## Bilayer exchange coupling and Neel temperature of $\text{YBa}_2\text{Cu}_3\text{O}_{6.2}$

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**Abstract.** The present paper attempts to study the Neel temperature of bilayer antiferromagnetic cuprate  $\text{YBa}_2\text{Cu}_3\text{O}_{6.2}$  within anisotropic Heisenberg model. The double time Green's function formalism within random phase approximation (RPA) has been used to obtain various correlation functions. The magnetization and the Neel temperature ( $T_N$ ) are evaluated. It is observed that the ratio of intrabilayer to inplane exchange coupling ( $r = J_\perp/J_\parallel$ ) plays an important role in the magnetic dynamics of bilayer systems. The recent experimental data of bilayer system  $\text{YBa}_2\text{Cu}_3\text{O}_{6.2}$  have been used to estimate the ratio  $r$  from the expression for Neel temperature. The estimated values of spin gap and the ratio of hopping matrix elements  $t_\perp/t_\parallel$  are found to be in fairly good agreement with the existing experimental results.

**Keywords.** Bilayer antiferromagnets; high- $T_c$  superconductors; Neel temperature.

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### 1. Introduction

Among the high- $T_c$  family,  $\text{La}_{2-y}(\text{Sr,Ba})_y\text{CuO}_4$  and  $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$  are the most widely (experimentally and theoretically) studied systems both in their superconducting as well as in the normal state. In the normal state, the parent compounds  $\text{La}_2\text{CuO}_4$  (2:1:4) and  $\text{YBa}_2\text{Cu}_3\text{O}_6$  (1:2:3) are antiferromagnetic (AFM) insulators and highly anisotropic. It has been shown [1–7] that in the undoped state 3D Neel ordering occurs in these systems below 35 K and beyond this temperature, the system behaves as a quasi-2D antiferromagnet. The  $c$ -axis antiferromagnetic exchange coupling ( $J_z$ ) is essential to keep the 3D Neel ordering in these systems. Moreover, the magnetic properties are sensitive to  $J_z$  which further depends on the oxygen concentration. On increasing the oxygen concentration ( $x$ ),  $J_z$  decreases and the AFM long range order is destroyed [8]. At  $x \geq 0.41$ , the Neel temperature ( $T_N$ ) vanishes and the system undergoes a phase transition from AFM insulating state to the superconducting phase.

On the other hand, the structural difference between 2:1:4 and 1:2:3 compounds is that unlike 2:1:4, the 1:2:3 systems possess two  $\text{CuO}_2$  planes (bilayer) within the unit cell. The experimental studies [9–12] reveal that the Cu-spins of the bilayer are coupled through di-

rect intra-bilayer AFM exchange coupling ( $J_{\perp}$ ). The preliminary theoretical investigations suggest that the spin wave spectrum of the bilayer systems consist of acoustic and the optic modes [9,12]. The intra-bilayer coupling leads towards a spin gap with minimum energy  $2\sqrt{J_{\perp}J_{\parallel}}$  [13]. Most of the previous studies [3–5] on 1:2:3 compounds do not consider the role of intra-bilayer coupling in their formalism and therefore do not distinguish between the magnetic dynamics of 1:2:3 and 2:1:4 systems.

The recent observation of optical magnons in NMR experiment made on 1:2:3 system by Reznik *et al* [13] has consolidated the fact that to investigate the magnetic dynamics of these systems, it is important to take the intrabilayer coupling into account. In a recent paper [14] using linear spin wave theory, we have shown that the contribution of optical mode to the sublattice magnetization is significant in the high temperature region. In this region, the sublattice magnetization follows a  $T \ln T$  behavior and the quasi 2D picture of the system remains unaltered. Moreover, in the limit  $r(J_{\perp}/J_{\parallel}) \rightarrow 0$ , the system becomes two dimensional and the magnetization disappears.

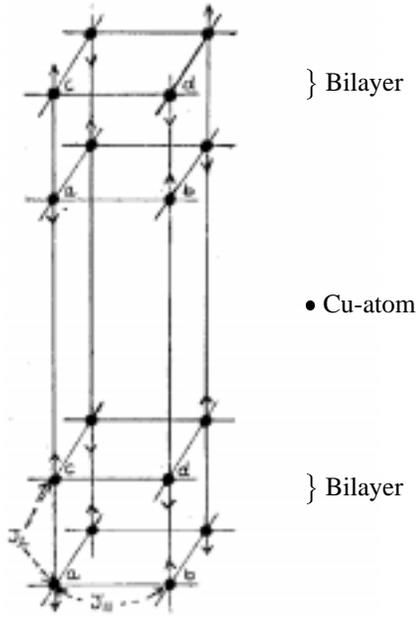
In the light of the above facts, we in the present paper evaluate the expression of Neel temperature ( $T_N$ ) for 1:2:3 systems. We adopt the four sublattice approach to treat the Heisenberg model Hamiltonian which includes the inplane and intra-bilayer coupling and therefore can describe well the bilayer antiferromagnets like  $\text{YBa}_2\text{Cu}_3\text{O}_6$ . Using Green's function formalism within random phase approximation (RPA), we obtain the equations of motion for the relevant Green's functions. The solution of the set of these equations yields the expression for sublattice magnetization. The Neel temperature ( $T_N$ ) of the system is obtained by following the standard procedure. We have shown that  $T_N$  of 1:2:3 compound depends both on  $J_z$  and  $J_{\perp}$ . Further, we have estimated the value of  $r(= J_{\perp}/J_{\parallel})$  by using the experimental data of various parameters. Our estimated value of  $r$  agrees well with the experimental as well as the other theoretical predictions.

## 2. Theoretical formulations

In the present section, we derive an expression for sublattice magnetization and Neel temperature using Green's function method in random phase approximation. For a bilayer system, the Heisenberg antiferromagnetic model Hamiltonian may be written as

$$H = J_{\parallel} \sum_{i \neq j}^{\alpha, \beta} (S_{ia\alpha} \cdot S_{jb\alpha} + S_{ja\alpha} \cdot S_{ib\alpha} + S_{ic\beta} \cdot S_{jd\beta} + S_{jc\beta} \cdot S_{id\beta}) + J_{\perp} \sum_i^{\alpha, \beta} (S_{ia\alpha} \cdot S_{ic\beta} + S_{ia\beta} \cdot S_{ic\alpha} + S_{ib\alpha} \cdot S_{id\beta} + S_{ib\beta} \cdot S_{id\alpha}), \quad (1)$$

where,  $\alpha, \beta$  are the layer indices,  $i, j$  denote the lattice sites with  $j$  being the nearest neighbour of  $i$  in the plane. The suffixes  $a, b, c, d$  denote the four basis atoms of the magnetic system.  $J_{\parallel}$  and  $J_{\perp}$  are the in-plane and intra-bilayer exchange coupling strength respectively. A figure describing various exchange coupling for a bilayer system with four basis atoms ( $a, b, c$  and  $d$ ) is shown in figure 1. We employ the Zubarev's double time Green's function formalism [15] in order to evaluate the various correlation functions. We define



**Figure 1.** A four sublattice model ( $a, b, c, d$ ) of bilayer cuprate ( $\text{YBa}_2\text{Cu}_3\text{O}_{6.2}$ ) with  $J_{||}$  (in-plane exchange coupling) and  $J_{\perp}$  (intra-bilayer exchange coupling).

$$G_{11} = \langle\langle S_{a1}^+; S_{a1}^- \rangle\rangle, \quad G_{21} = \langle\langle S_{b1}^+; S_{a1}^- \rangle\rangle,$$

$$G_{31} = \langle\langle S_{c1}^+; S_{a1}^- \rangle\rangle \quad \text{and} \quad G_{41} = \langle\langle S_{d1}^+; S_{a1}^- \rangle\rangle.$$

We write down the equation of motion of the above Green's functions and decouple the higher order Green's functions within random phase approximation (RPA). For example we have

$$\langle\langle S_{\alpha l n}^z S_{\alpha' l' n'}^{\pm}; S_{a1}^- \rangle\rangle = \langle S_{\alpha l n}^z \rangle \langle\langle S_{\alpha' l' n'}^{\pm}; S_{a1}^- \rangle\rangle \quad (2)$$

and due to the symmetry of the system, we take

$$\langle S_{a1}^z \rangle = \langle S_{d1}^z \rangle = -\langle S_{b1}^z \rangle = -\langle S_{c1}^z \rangle = \bar{S}. \quad (3)$$

Using eqs (2) and (3), the various equations of motion can be simplified. In the matrix notation these may be written as

$$\begin{bmatrix} \omega - J & -J(k) & -J'_{\perp} & 0 \\ J(k) & \omega + J & 0 & J'_{\perp} \\ J'_{\perp} & 0 & \omega + J & J(k) \\ 0 & -J'_{\perp} & -J(k) & \omega - J \end{bmatrix} = \begin{bmatrix} G_{11} \\ G_{12} \\ G_{31} \\ G_{41} \end{bmatrix} \begin{bmatrix} 2\bar{S}/2\pi \\ 0 \\ 0 \\ 0 \end{bmatrix}, \quad (4)$$

where  $J = 2\bar{S}Z_{ab}J_{\parallel} + 2\bar{S}J_{\perp}$ ;  $J(k) = 2\bar{S}Z_{ab}J_{\parallel}\gamma(k)$  and  $J'_{\perp} = 2\bar{S}J_{\perp}$  with  $\gamma(k) = (1/Z_{ab}) \sum_{\delta}^{ab} \exp(ik \cdot \delta)$  and  $Z_{ab} = 4$  being the number of nearest neighbors in the  $ab$  plane.

The solution of eq. (4) gives four roots but they are two by two equal. These may be written as

$$\omega_{1,2} = [J^2 - \{J(k) \pm J'_{\perp}\}^2]^{1/2}. \quad (5)$$

Now, the set of equations (4) may be solved easily and the simple algebra yields

$$G_{11} = (\bar{S}/2\pi) \sum_k \left[ \frac{\omega + J}{\omega^2 - \omega_1^2} + \frac{\omega + J}{\omega^2 - \omega_2^2} \right]. \quad (6)$$

Following the standard procedure [15], the correlation function  $\langle S^- S^+ \rangle$  is obtained as

$$\langle S^- S^+ \rangle = \frac{2S\bar{J}}{N} \sum_k \left[ \frac{\coth(\beta\omega_1/2)}{4\omega_1} + \frac{\coth(\beta\omega_2/2)}{4\omega_2} \right] - \bar{S}. \quad (7)$$

For spin 1/2, the sublattice magnetization may be evaluated by using the relation

$$\langle S^z \rangle = 1/2 - \langle S^- S^+ \rangle. \quad (8)$$

The expression for magnetization at  $T = 0$  and finite temperature as obtained by spin wave approximation in our previous paper [14] can be easily obtained by substituting  $\coth(\beta\omega_1/2) = 1$ , and  $\bar{S} = 1/2$  in eqs (7) and (8) respectively.

To obtain the expression of Neel temperature, we solve equation (8) under the condition that as  $T \rightarrow T_N$ ;  $\langle S^z \rangle \rightarrow 0$ . Thus, the simplified expression for  $T_N$  reads

$$K_B T_N = \frac{J_{\parallel}}{(4+r)} \frac{1}{I_{12}}, \quad (9)$$

where

$$I_{12} = (2/N) \sum_k \left( \frac{1}{\omega_1^2} + \frac{1}{\omega_2^2} \right). \quad (10)$$

Before we proceed further, we rewrite eq. (5) for  $\omega_{1,2}$  as follows:

$$\omega_{1,2} = 2\bar{S}J_{\parallel}[(4+r)^2 - \{2(\cos k_x a + \cos k_y a) \pm r\}^2]^{1/2} \quad (11)$$

with  $r = J_{\perp}/J_{\parallel}$ . It is important to note that eq. (11) gives acoustic and optic modes in the spectrum and suggests a gap with minimum energy  $\omega_{\min}^{\text{opt}} = 8\bar{S}\sqrt{J_{\perp}J_{\parallel}}$ . For  $\bar{S} = 1/2$ , this is in agreement with the predictions of the spin wave theory except that in our convention,  $J_{\parallel}$  is replaced by  $2J_{\parallel}$  [8,14].

Now, we solve eq. (10) by converting the summation over  $k$ -values into an integration. For convenience, we substitute  $k_x a = u$  and  $k_y a = v$ . Thus,  $I_{12}$  may be rewritten as

$$I_{12} = [I(\omega_1) + I(\omega_2)]/(4+r) \quad (12)$$

with

$$I(\omega_1) = (1/2\pi^2) \int_0^\pi \int_0^\pi dudv/[4 - 2(\cos u - \cos v)] \quad (13)$$

and

$$I(\omega_2) = (1/2\pi^2) \int_0^\pi \int_0^\pi dudv/[(4 + 2r) - 2(\cos u - \cos v)]. \quad (14)$$

It is important to mention that in eq. (12),  $I(\omega_1)$  is the contribution from the acoustic mode and in the absence of anisotropy, it is not possible to evaluate the integration in eq. (13). In our previous paper [14], we have discussed that there may exist several anisotropies in a layered system like 1:2:3 and the incorporation of  $c$ -axis inter-unit cell antiferromagnetic exchange coupling ( $J_z$ ) in the acoustic mode is of immense importance to keep the 3D Neel ordering at low temperature. On the other hand, the optic mode remains unaffected with such a term.

In the present calculations, though we have not considered the effects of  $J_z$  explicitly in the model Hamiltonian, we emphasize that its inclusion will modify only the acoustic mode and in such a situation, eq. (13) will be similar to that previously obtained by Singh *et al* [13] while for small values of  $J_z$ , eq. (14) remains unaltered. Therefore, corresponding to the acoustic mode, we adopt the form of  $I(\omega_1)$  as discussed in [3]. Thus,  $I(\omega_1)$  may be written as

$$I(\omega_1) \approx \{0.1616 \ln(1/R) + 0.5055\}, \quad (15)$$

where  $R = J_z/J_{||}$  is the ratio of inter-unit cell to inplane coupling strength.

On the other hand, to analyse the contribution from the optic mode, we integrate eq. (14) over  $u$  and  $v$ . This finally gives

$$I(\omega_2) \approx 1/(8\sqrt{r}). \quad (16)$$

Substituting eqs (12), (15) and (16) in eq. (9), the expression for Neel temperature reads

$$K_B T_N = \frac{J_{||}}{0.1616 \ln(1/R) + 0.5055 + (1/8\sqrt{r})}. \quad (17)$$

It is clear from (17) that Neel temperature  $T_N$  depends on inter-unit cell coupling ratio ( $R$ ), intra-bilayer coupling ratio ( $r$ ) and in-plane coupling strength ( $J_{||}$ ). The numerical estimate of  $r$  for a given  $T_N$  is presented in the next section.

### 3. Results and discussion

We have obtained the expression of  $T_N$  for a bilayer system as given by eq. (17) in the random phase approximation. From this expression we note that unlike 2:1:4 systems [1], the  $T_N$  of 1:2:3 system depends on both the inter-unit cell coupling ( $J_z$ ) and intra-bilayer coupling ( $J_{\perp}$ ). From the expression (17) we note that in the limit  $R \rightarrow 0$ ; the logarithmic term approaches to infinity and therefore  $T_N$  vanishes. This is in accordance with the previous calculations [3,5]. In addition, we also note that as soon as  $r \rightarrow 0$ , the second

term in the denominator becomes infinite and therefore  $T_N$  reduces to zero. Thus it can be pointed out that either  $r \rightarrow 0$  or  $R \rightarrow 0$ , or both tends to zero simultaneously. The Neel temperature of bilayer antiferromagnetic system vanishes. Hence, we conclude that the intra-bilayer coupling along with inter-unit cell coupling play an important role in the magnetic dynamics of bilayer systems.

Further, the expression (17) can be used to estimate the ratio  $r$ . For this, we consider the recent neutron scattering experiment of Reznik *et al* [13]. As per their estimation the Neel temperature  $T_N$  is 390 K and  $2J_{||} = 120$  meV for  $\text{YBa}_2\text{Cu}_3\text{O}_{6.2}$  system. On the other hand, it has been shown [3,5] that  $R = 6 \times 10^{-3}$  yields a good estimate of  $T_N$ . Several experiments also have confirmed that  $R$  decreases with the oxygen concentration ( $x$ ) and for  $x \geq 0.41$ ,  $R$  will be zero. Assuming a linear dependence of  $R$  on  $x$ , Singh *et al* [3] within RPA and Ajay *et al* [5] in Callen decoupling scheme have plotted  $T_N$  vs oxygen concentration ( $x$ ). Following [3,5], we assume that  $R = c(1 - x/x_0)$  with  $c = 6 \times 10^{-3}$  and  $x_0 = 0.41$ . From the results of Reznik *et al* [13] for  $\text{YBa}_2\text{Cu}_3\text{O}_{6.2}$  sample, we note that  $x = 0.2$  and therefore the above linear relationship gives  $R = 3.0 \times 10^{-3}$ .

Substituting these values of various parameters in expression (17), we estimate that  $r$  comes out to be 0.0967. Our estimated value of  $r$  is in fairly good experimental agreement with the results of Reznik *et al* [13] ( $r = 0.08$ ); Millis *et al* ( $r = 0.11$ ) [16] and the band-theory calculations due to Anderson *et al* ( $r = 0.108$ ) [17]. From the above result, it is easy to evaluate the ratio of the hopping matrix elements,  $t_{||}$  and  $t_{\perp}$ . In fact,  $t_{\perp}/t_{||} \approx \sqrt{r}$ , this gives  $t_{\perp}/t_{||} = 0.311$  and is comparable to the previous estimates of [13].

The above mentioned value of  $r$  may also be used to calculate the minimum energy gap. From eq. (11), for spin-1/2 case, we obtain  $\omega_{\min}^{\text{opt}} = 74.63$  meV which is very close to the experimentally observed value  $w = 65-70$  meV [13].

We therefore conclude that the intra-bilayer coupling and inter-unit cell coupling play an important role in the magnetic dynamics of bilayer systems. The expression for  $T_N$  exhibit significant difference from that of 2:1:4 systems and hence the dynamics of the two systems can be distinguished from each other. Further, the estimated values of  $r$  and energy gap are in agreement with those of experimental values.

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