Magnetic dynamics in La_2CuO_4 with interlayer coupling and anisotropy gaps

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The role of anisotropy gaps in La_2CuO_4—the in-plane gap of ~10 meV, mainly due to the Dzyaloshinsky-Moriya antisymmetric exchange interaction, and out-of-plane anisotropy gap of ~20 meV—together with interlayer coupling, is analyzed in the magnetic dynamics due to thermal excitation of spin waves, as revealed in temperature dependence of sublattice magnetization. By fitting with sublattice magnetization data for La_2CuO_4, obtained from a Mössbauer-spectroscopic study, and incorporating the quantum correction Z_c to spin-wave energy, we obtain the average planar exchange energy, $J_p = 1380$ K, the ratio of effective interplanar-to-planar exchange energy, $r^2 = 4.3 \times 10^{-5}$, and the characteristic energy for spin waves propagating perpendicular to the plane, $2Z_cJ_pr = 21$ K.

The orthorhombic distortion in La_2CuO_4, which causes a net interlayer exchange interaction, also allows, in the planar exchange Hamiltonian, the Dzyaloshinsky-Moriya antisymmetric exchange term, J_{bc}, which arises from spin-orbit couplings to excited-state orbitals.[1,2] This *D-M* interaction introduces an in-plane anisotropy gap, Δ_a, in the spin-wave excitation spectrum, while the anisotropy in diagonal exchange terms, $J_{cc} \approx J_{aa} > J_{bb}$, leads to the out-of-plane anisotropy gap, Δ_b. Estimates for these gaps from neutron scattering,[3] infrared spectroscopy,[4] and spin-flop transition[5,6] yield $\Delta_a \approx 10$ meV and $\Delta_b \approx 20$ meV.

The anisotropy gaps suppress quantum (spin fluctuation) corrections to sublattice magnetization, spin-wave velocity, and perpendicular susceptibility. For instance, consider the two-dimensional case with the two anisotropy gaps, Δ_a and Δ_b. Taking into account contributions from the two spin-wave polarizations, the zero-temperature reduction in sublattice magnetization, due to quantum spin fluctuations, is

$$-\delta M_0 = \frac{1}{2} \sum_{i=a,b} \sum_{\mathbf{Q}} \left(\frac{\sqrt{\Delta_i^2 + 4J_p^2}}{\sqrt{\Delta_i^2 + 4J_p^2(1-\gamma_{\mathbf{Q}}^2)}} - 1 \right) , \quad (1)$$

where J_p is the average planar exchange interaction. This vanishes in the limit of large anisotropy gaps ($\Delta_a, \Delta_b \gg J_p$), in which case spin fluctuations are almost completely suppressed. For ($\Delta_a, \Delta_b \ll J_p$), the net reduction due to spin-wave gaps in the quantum correction to sublattice magnetization is $\sim (1/2\pi)(\Delta_a+\Delta_b)/2J_p$. Interlayer coupling also reduces quantum spin fluctuations, and, in the absence of any anisotropy gaps, the zero-point reduction in sublattice magnetization as a function of strength of interlayer hopping has recently been studied.[7] The sublattice magnetization goes from 0.607 in the strictly two-dimensional (2D) case ($r = 0$) to 0.85 in the isotropic 3D case ($r = 1$), where r denotes the ratio of effective interlayer-to-planar hopping. In the limit $r \ll 1$, the reduction in quantum correction to sublattice magnetization due to interlayer hopping is $\sim (4\sqrt{2}/\pi^2)r$. In La_2CuO_4 both $(\Delta_a + \Delta_b)/2J_p$ and r are less than 1%, hence the quantum corrections are essentially unchanged relative to results of the isotropic two-dimensional case.[8]

At finite temperature, anisotropy gaps also eliminate the divergence in two dimensions in the contribution from thermal excitation of long-wavelength spin waves. This leads to a finite Néel temperature $\sim J_p/\ln(J_p/\sqrt{\Delta_a\Delta_b})$. It is essential, however, for both gaps to be nonzero. For instance, but for the Dzyaloshinski-Moriya interaction, the diagonal exchange interaction Hamiltonian, with $J_{cc} = J_{aa} > J_{bb}$, would still lead to a gapless mode due to spin deviations in the *ac* plane, corresponding to spin waves with polarization along the *b* direction.

We have recently analyzed the role of a weak interlayer exchange coupling in determining the finite-temperature magnetic dynamics, as revealed in the T dependence of sublattice magnetization, below the Néel temperature.[9] If r is the ratio of interlayer-to-planar hopping, so that J_pr^2 is the interlayer exchange coupling, it was shown that an energy scale $2Jr$, where $J \equiv J_p(1 + r^2/2)$, is introduced in the dynamics, such that the T dependence of sublattice magnetization exhibits a crossover with increasing temperature, from a 3D behavior (T^2 falloff for $k_BT \ll 2Jr$) to a quasi-2D one ($T \ln T$ falloff for $k_BT \gg 2Jr$). In this Brief Report, we examine the consequences of anisotropy gaps, together with a weak interlayer coupling, on the magnetic dynamics. We definitively show that the anisotropy gaps of ~ 10 and 20 meV in La_2CuO_4 alone are not sufficient to describe the sublattice magnetization data from the recent Mössbauer study[9,10] and that the interlayer coupling therefore must be included. With anisotropy gaps included, the difference in the nature of T dependence arises only in the low-temperature regime; however, the deviations are so small that the overall $M(T)$-versus-T curve is virtually indistinguishable from the case with only interlayer coupling.

The spin-wave energies corresponding to the in-plane and out-of-plane gaps are

$$\Omega_{\mathbf{Q}}^{i=a,b} = [\Delta_i^2 + 4J^2(1-\gamma_{\mathbf{Q}}^2)]^{1/2}, \tag{2}$$

where $J \equiv J_p(1+r^2/2)$, J_p is the average planar exchange energy $(J_{aa}+J_{bb}+J_{cc})/3$, and $\gamma_{\mathbf{Q}} = (\cos Q_x a + \cos Q_y c + r^2 \cos Q_z b)/(2+r^2)$, with the z axis oriented perpendicular to the planes. For long planar wavelengths $\theta_p = \sqrt{(Q_x a)^2 + (Q_y c)^2} < 1$, this can be written as follows, showing the two energy scales corresponding to spin waves propagating along the planes (J) and perpendicular to them ($2Jr$):

$$\Omega_{\mathbf{Q}}^{i=a,b} = [\Delta_i^2 + 4J^2(\theta_p^2/2) + 4J^2r^2(1-\cos\theta_z)]^{1/2}. \tag{3}$$

The finite-temperature correction to sublattice magnetization due to thermal excitation of spin waves is then obtained from

$$-\delta M(T) = \sum_{i=a,b}\sum_{\mathbf{Q}} \frac{\sqrt{\Delta_i^2+4J^2}}{\Omega_{\mathbf{Q}}^i}\frac{1}{e^{\beta\Omega_{\mathbf{Q}}^i}-1}. \tag{4}$$

For $k_BT \ll J$, only the long-wavelength planar modes contribute significantly; using the long-wavelength approximation for the spin-wave energy, and integrating over the planar momentum, θ_p, we obtain, after neglecting Δ_i^2 in comparison with $4J^2$ in the numerator,

$$-\delta M(T) = \sum_{i=a,b}\frac{1}{\pi^2}\left(\frac{k_BT}{J}\right)\int_0^{\pi} d\theta_z \ln\left(1-\exp\{-[\Delta_i^2+4J^2r^2(1-\cos\theta_z)]^{1/2}/k_BT\}\right)^{-1}. \tag{5}$$

In the low-temperature regime when $k_BT \ll \Delta_a, \Delta_b$, the reduction in sublattice magnetization due to thermal excitation of spin waves is exponentially suppressed by the gaps. When $r=0$, for example, $-\delta M(T) = \pi^{-1}(k_BT/J)[\exp(-\Delta_a/k_BT) + \exp(-\Delta_b/k_BT)]$. We consider the limit when $k_BT \gg \Delta_a, \Delta_b, 2Jr$, in which case we obtain, from above,

$$-\delta M(T) = \sum_{i=a,b}\frac{1}{\pi^2}\left(\frac{k_BT}{J}\right)\int_0^{\pi} d\theta_z \ln\frac{k_BT}{2Jr}[(\Delta_i/2Jr)^2 + (1-\cos\theta_z)]^{-1/2}, \tag{6}$$

which yields the $T\ln T$ behavior for the falloff, as was obtained for the case with a weak interlayer coupling but no anisotropy gaps.[9] The suppression in spin-wave reduction of sublattice magnetization due to the anisotropy gaps can be expressed by a factor in the logarithm:

$$-\delta M(T) = \frac{2}{\pi}\left(\frac{k_BT}{J}\right)\ln\frac{k_BT}{2Jr}\sqrt{2}F(\Delta_a,\Delta_b,2Jr). \tag{7}$$

The factor F equals $\sqrt{f_af_b}$, where $f_{i=a,b}$ are defined as $\pi\ln\sqrt{2}f_{i=a,b} = \int_0^{\pi} d\theta_z \ln[(\Delta_i/2Jr)^2 + (1-\cos\theta_z)]^{-1/2}$. $F \leq 1$, the equality obtained when $\Delta_a, \Delta_b = 0$.

Now we consider the temperature regime $\Delta_i \ll k_BT \ll 2Jr$, when at least one of the gaps is very small compared to k_BT. The significant contribution in this regime comes from long-wavelength modes perpendicular to the plane, and from Eq. (5), for the mode with $\Delta_i \ll k_BT$, we obtain

$$-\delta M(T) = \frac{1}{\pi^2}\left(\frac{k_BT}{J}\right)\left(\frac{k_BT}{2Jr}\right)\sqrt{2}\int_0^{\infty} dy \ln\left(1-\exp\{-[(\Delta_i/k_BT)^2+y^2]^{1/2}\}\right)^{-1}. \tag{8}$$

The integral yields $\sim(\pi^2/6 - \Delta_i/k_BT)$. In the limit $\Delta_i \ll k_BT$, this is weakly temperature dependent, and hence the sublattice magnetization falls off as T^2, the characteristic 3D behavior. Therefore, even in the presence of anisotropy gaps, it is possible, in principle, to have a crossover in the magnetic dynamics from a characteristic 3D behavior to a quasi-2D one. It should be noted that the significant role in this regime is played by the mode with the smaller gap. This particular temperature limit is, however, probably not realized in the case of La_2CuO_4, wherein the Dzyaloshinski-Moriya-interaction-induced gap is about 13 meV from the most recent study, and $2Jr = 21$ K, as we find in this study.

When quantum corrections to the spin-wave propagator are included,[8] the spin-wave energy is renormalized by a multiplicative factor, Z_c. Thus J in Eq. (2) onward can be replaced by Z_cJ. Just as for the case of sublattice magnetization, discussed earlier, Z_c is nearly unchanged from the isotropic, 2D result because the suppression in Z_c due to anisotropy gaps and interlayer coupling should be less than 1%.

In Fig. 1 we show the normalized sublattice magnetization as a function of temperature, as obtained from Eq. (5) with the best-fit parameters. Also shown is the sublattice magnetization data in La_2CuO_4, which has been inferred from Mössbauer spectroscopic studies of La_2CuO_4 doped with 0.5% ^{57}Fe.[10] We have used $\Delta_a = 13$ meV and $\Delta_b = 18$ meV from the most recent reported study.[6] Since J is determined essentially from the slope, we use the same value as before,[9] where, including the multiplicative spin-wave-energy renormalization factor, Z_c, we obtained $Z_c J M(0) = 800$ K. The best fit with these data is obtained for $2Z_c Jr = 21$ K.

We should note that the sublattice magnetization data yields the value of J essentially independently of other parameters. In the temperature regime $k_B T \gg \Delta_a, \Delta_b, 2Jr$, the leading temperature dependence in the spin-wave correction to sublattice magnetization is $k_B T/J$, the other parameters entering only in the logarithm. The slope of $M(T)$ versus T yields fairly accurately the average planar exchange energy.

Using $M(0) = 0.5$, so that $Z_c J = 1600$ K and $Z_c = 1.16$, we obtain $J_p \approx J = 1380$ K (0.12 eV), as before.[9] From $2Z_c Jr = 21$ K, we obtain $r = 0.0065$, yielding, for the ratio of effective interplanar-to-planar coupling, $J_\perp / J_\parallel = r^2 = 4.3 \times 10^{-5}$. This is in agreement with reported values of J_p in other works — $0.16/Z_c$ eV (neutron-scattering study),[11] 0.14 eV (Raman scattering),[12] 0.13 eV (by fitting the spin correlation length),[13] 1450 K (by fitting the spin correlation length within a Monte Carlo simulation of the spin-$\frac{1}{2}$ Heisenberg model),[14] and 1500 K (optical studies).[15] Finally, we should add that the importance of anisotropy gaps in the quantitative analysis of interlayer coupling has also been stressed in Ref. 16.

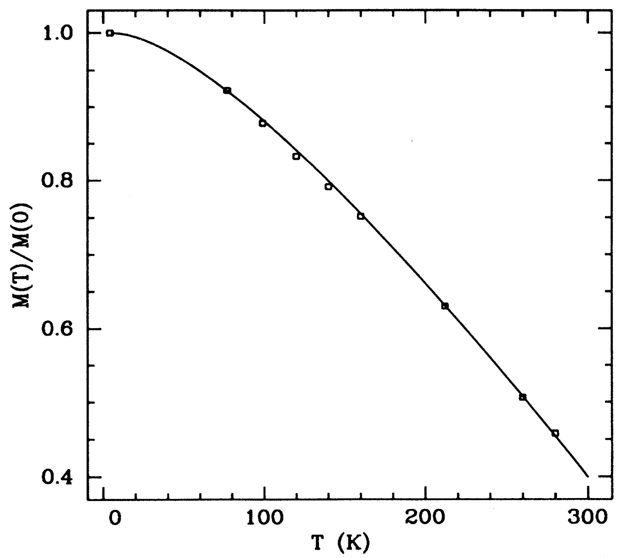

FIG. 1. The normalized sublattice magnetization data (squares) inferred from Mössbauer spectroscopic study of La_2CuO_4 doped with 0.5% ^{57}Fe, and $M(T)/M(0)$ obtained from Eq. (5) with $\Delta_a = 13$ meV, $\Delta_b = 18$ meV, and best-fit parameters of $Z_c J M(0) = 800$ K and $2Z_c Jr = 21$ K.

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