Magnetic Dynamics in Copper-Oxide-Based Antiferromagnets: The Role of Interlayer Coupling

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It is shown that thermal excitation of spin waves in a highly anisotropic antiferromagnet results in a characteristic temperature dependence of sublattice magnetization with a crossover from a 3D to a quasi-2D behavior. The magnetic dynamics in several copper-oxide-based antiferromagnets is analyzed in this context in terms of subtle details of their structural characteristics, and the temperature dependence of the Cu moment is used to determine the planar and interplanar exchange energies.

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The remarkable manifestation of the almost 2D antiferromagnetism in high-T_c cuprate superconductors has provided a great impetus in efforts to understand low-dimensional antiferromagnetism. Specifically, the discoveries of long-range antiferromagnetic (AF) order,[1] spin-wave excitations,[2] and long-range, 2D AF spin correlations above the Néel temperature[3] have contributed much to clarifying important theoretical issues. Thus, the 2D aspects of antiferromagnetism, manifested as $T > T_N$, are beginning to be understood.

However, in the temperature regime $T < T_N$, where 3D AF ordering sets in, the weak interlayer magnetic coupling becomes a most relevant piece in the physics. The very weak interlayer coupling affords us with a highly anisotropic antiferromagnet, and therefore an investigation of how it controls the magnetic dynamics is of much interest. Furthermore, the magnetic interlayer coupling in the copper-oxide systems depends, in a very subtle manner, on details of their structural characteristics. For example, if it were not for the orthorhombic distortion in the La_2CuO_4 there would be no net *exchange* coupling between two neighboring layers. Thus, as a supplement to the conductivity anisotropy, magnetic dynamics can be used as a probe to investigate magnetic aspects of the interlayer coupling, which is of importance in some theories of high-T_c superconductivity.[4]

In this Letter we report a microscopic study aimed at understanding the magnetic dynamics of several parent compounds (of the high-T_c superconductors) in terms of subtle details of their structural characteristics. We first examine, within an itinerant-electron model, the magnetic dynamics of thermally excited spin waves in a highly anisotropic antiferromagnet, as revealed in the temperature dependence of sublattice magnetization $M(T)$. We show that there are really two energy scales in the dynamics, namely, J $(=4t^2/U)$, the exchange energy, and Jr, where r is the ratio of an *effective* interplanar- to planar-hopping strength. For $k_BT < 2Jr$, the magnetization falls off as T^2, characteristic of a 3D system. However, for $k_BT > 2Jr$, we show that there is a crossover to a $T\ln T$ behavior, which is a quasi-2D behavior.

We also fit the $M(T)$ vs T behavior to experimental data for several systems and find the fits to be excellent. Moreover, the value obtained from the best fits for J and r are, respectively, in agreement with results known from neutron scattering, and the general trend expected from structural characteristics. For La_2CuO_4, we obtain $Z_cJ \approx 1600$ K, where $Z_c \sim 1.16$ is the renormalization of the spin-wave velocity.[5] This is in agreement with the reported values for J in other works: $0.16/Z_c$ eV (neutron-scattering studies),[2] 0.14 eV (Raman scattering),[6] 0.13 eV (by fitting the spin-correlation length within the nonlinear sigma model),[7] 1450 K (by fitting the spin-correlation length within a Monte Carlo simulation of the spin-$\frac{1}{2}$ Heisenberg model),[8] 1500 K (optical studies).[9] Also, we find that for the $Sr_2CuO_2Cl_2$ compound, r^2, which measures the ratio of magnetic interlayer coupling to planar coupling, is 2 orders of magnitude smaller than for the lanthanum compound. This confirms the structural viewpoint that, in the absence of any orthorhombic distortion, the effective interlayer coupling in $Sr_2CuO_2Cl_2$ is due to a much weaker (than exchange) effect, possibly a magnetic dipole coupling. Thus, the almost linear rise in magnetization with decreasing temperature, seen in this compound from neutron-scattering studies,[10] with no sign of a crossover down to 10 K, is actually the signature of an extremely weak magnetic interlayer coupling.

We now analyze the magnetic dynamics in a highly anisotropic 3D antiferromagnet. We consider a 3D simple-cubic lattice system with planar and interplanar lattice parameters of a and c, respectively. In the La_2CuO_4 compound, the effective coupling between layers is due to the orthorhombic distortion. This distortion renders unequal the two pairs of couplings by which a Cu spin in a plane is coupled to its four out-of-plane nearest neighbors. The effective coupling between planes (and the resulting AF structure) is thus governed by the larger of the two couplings. For now, we consider the couplings between planes to be due to an effective interplanar-hopping term, and later we discuss how it relates to the structural features of La_2CuO_4.

If r denotes the ratio of an effective interplanar- to planar-hopping strength, then the free-particle energy dispersion relation is $\epsilon_{\mathbf{k}} = -2t[\cos k_xa + \cos k_ya + r\cos k_zc]$. We consider the itinerant-electron description of an antiferromagnet in terms of the Hubbard model with $\epsilon_{\mathbf{k}}$ as the free-particle band energy. It has recently been shown that when quantum spin fluctuations around the Hartree-Fock (HF) state are included, the Hubbard model in the strong-coupling limit yields the same behavior as obtained from the linear spin-wave analysis of the spin-$\frac{1}{2}$ Heisenberg model.[11,12]

For the anisotropic 3D system it has been shown[12] that the spin-wave energy is given by

$$\Omega_{\mathbf{Q}} = 2J(1-\gamma_{\mathbf{Q}}^2)^{1/2}, \tag{1}$$

where J is related to the planar exchange energy $J_p = 4t^2/U$ by $J = J_p(1+r^2/2)$ and $\gamma_{\mathbf{Q}} = (\cos Q_x a + \cos Q_y a + r^2 \cos Q_z c)/(2+r^2)$. For $r=0$ one recovers the 2D result,[11,13] whereas $r=1$ yields the isotropic 3D result. The above expression for the spin-wave energy is obtained by retaining terms up to order $(t/U)^3$ in the spin susceptibility.

We first consider the zero-temperature correction to sublattice magnetization due to the zero-point, quantum spin fluctuations. Considering the contribution to self-energy arising from the spin-wave excitations,[11,12] we obtain the zero-point correction to sublattice magnetization as

$$-\delta M^0 = \frac{1}{N}\sum_{\mathbf{Q}} \left[\frac{1}{(1-\gamma_{\mathbf{Q}}^2)^{1/2}} - 1 \right]. \tag{2}$$

The sublattice magnetization in the HF state is thus lowered by the above amount due to the zero-point, spin-wave excitations. The dependence of the resulting magnetization on r is shown in Ref. 12 and goes from ~ 0.6 in the strictly 2D case ($r=0$) to ~ 0.85 in the isotropic 3D case ($r=1$), these limiting results being in exact agreement with the linear-spin-wave-analysis result for the spin-$\frac{1}{2}$ Heisenberg model.[14]

We now consider the additional reduction in the sublattice magnetization at finite temperatures arising from thermal excitation of spin waves. Extending the analysis for the self-energy correction to the finite-temperature case,[15] we obtain

$$-\delta M(T) = \frac{1}{N}\sum_{\mathbf{Q}} \left[\frac{1}{(1-\gamma_{\mathbf{Q}}^2)^{1/2}} \frac{2}{e^{\beta\Omega_{\mathbf{Q}}}-1} \right]. \tag{3}$$

For $k_B T \ll J$ only spin waves with small [mod$(\pi/a,\pi/a)$] planar wave vector ($Q_x a, Q_y a \ll 1$) will be excited and give a significant reduction in the sublattice magnetization. Therefore, retaining terms up to quadratic order in $\cos Q_x a$ and $\cos Q_y a$, and denoting $[(Q_x a)^2 + (Q_y a)^2]^{1/2}$ by θ_p and $Q_z c$ by θ_z, we obtain

$$-\delta M(T) = 2\int_{-\pi}^{\pi} \frac{d\theta_z}{2\pi} \int_0^{\infty} \frac{\theta_p\, d\theta_p}{2\pi} \left[\frac{\theta_p^2}{2} + r^2(1-\cos\theta_z) \right]^{-1/2} \frac{2}{e^{\beta\Omega_{\mathbf{Q}}}-1}, \tag{4}$$

where the spin-wave energy in the quadratic approximation for the cosines of planar momenta is $\Omega_{\mathbf{Q}} \approx 2J[\theta_p^2/2 + r^2(1-\cos\theta_z)]^{1/2}$. Integrating over θ_p yields

$$-\delta M(T) = \frac{2}{\pi^2}\left[\frac{k_B T}{J}\right] \int_0^{\pi} d\theta_z \ln\left[1 - \exp\left[-\frac{2Jr}{k_B T}(1-\cos\theta_z)^{1/2}\right]\right]^{-1}. \tag{5}$$

We now consider the above equation in the two temperature regimes. If $y \equiv (2Jr/k_B T)\sqrt{1-\cos\theta_z}$, then for $k_B T \gg 2Jr$, $y \ll 1$ and so the argument of the logarithm in Eq. (5) is $\approx (k_B T/2Jr)(1-\cos\theta_z)^{-1/2}$. The integral over θ_z then just yields a numerical factor and one gets a $T\ln T$ behavior of the sublattice magnetization with temperature. Using $-\int_0^{\pi} d\theta_z \ln(\sqrt{1-\cos\theta_z}) = \pi \ln\sqrt{2}$, we then obtain

$$-\delta M(T) = \frac{2}{\pi}\left[\frac{k_B T}{J}\right] \ln\left[\frac{k_B T}{2Jr}\sqrt{2}\right] \quad (k_B T \gg 2Jr). \tag{6}$$

In the limit $k_B T \ll 2Jr$, y can become very large if θ_z is not very small compared to 1; using $\ln(1-e^{-y})^{-1} \approx e^{-y}$ for large y, we notice that the contribution is exponentially small when θ_z is not small compared to 1 and hence conclude that the only significant contribution comes from spin-wave modes with long wavelength in the z direction. Using $y \approx (2Jr/k_B T)(\theta_z/\sqrt{2})$, valid for $\theta_z \ll 1$, the integral over θ_z can be converted into one over y, and we obtain a T^2 behavior of the sublattice magnetization with temperature. Using $\int_0^{\infty} dy \ln(1-e^{-y})^{-1} = \pi^2/6$, we obtain

$$-\delta M(T) = \frac{\sqrt{2}}{3}\left[\frac{k_B T}{J}\right]\left[\frac{k_B T}{2Jr}\right] \quad (k_B T \ll 2Jr). \tag{7}$$

In this low-temperature regime ($k_B T \ll 2Jr$), when the significant contribution comes only from long-wavelength modes in *all* directions, the temperature dependence is therefore that of a 3D system. However, the temperature dependence is still over two energy scales, namely, J from long-wavelength, planar spin-wave modes and $2Jr$ from long-wavelength, spin-wave modes along the z direction.

A recent experimental study of the temperature dependence of the sublattice magnetization in La_2CuO_4 has revealed an initial weak dependence at very low temperatures, changing over to a faster, approximately linear falloff with temperature. The sublattice magnetization in La_2CuO_4 has been inferred from Mössbauer-spectroscopic studies of La_2CuO_4 doped with about half a percent of ^{57}Fe.[16] Previous studies have established that Fe, as a dopant, exclusively goes into the Cu sites and that the Fe spin is antiferromagnetically coupled to the Cu spins.[17,18] The Fe nuclear ground state with spin $\frac{1}{2}$ and first excited state with spin $\frac{3}{2}$ are split by the hyperfine field. Transitions between these states with a dipole selection rule result in the completely split-out sextet seen in the Mössbauer spectra below the Néel temperature. This allows a determination of the magnetic hyperfine field, which is a measure of the sublattice magnetization. The low level of doping ensures that the magnetic properties of the Fe-doped sample are barely altered, if at all, from those of its parent compound. The temperature dependence of the hyperfine field at the Fe nucleus is thus expected to reflect the genuine temperature dependence of the sublattice magnetization of the antiferromagnetic Cu-spin system.

Figure 1 shows the reduction in the normalized sublattice magnetization due to thermal excitation of spin waves, as obtained by numerically evaluating Eq. (5). The parameters J and r are chosen to obtain a best fit with the normalized Mössbauer hyperfine-field-strength data,[16] for which the data points are also shown on the same plot. The excellent fit with theory clearly confirms that the magnetic dynamics in La_2CuO_4 is characteristic of thermal excitation of spin waves in a highly anisotropic antiferromagnet. The best fit yields $J=800/M(0)$ K and $r/M(0)=0.022$. Using $M(0)=0.5$ we then obtain $J=1600$ K and $r=0.011$. This value of J, it should be realized, includes the 16% correction to the spin-wave velocity.

We now discuss how the effective interlayer hopping, used in our analysis of an anisotropic antiferromagnet, is related to structural characteristics of La_2CuO_4. In this regard, the most important feature is the orthorhombic distortion, because of which the two pairs of exchange terms by which a Cu spin is coupled to its out-of-plane nearest-neighbor spins (on each side) are not equal. If J_1 and J_2, respectively, denote out-of-plane nearest-neighbor couplings inclined along the crystallographic a and c directions in the plane,[1] then the La_2CuO_4 structure results when $J_1 > J_2$, whereas for $J_1 < J_2$ the La_2NiO_4 antiferromagnetic structure results with AF ordering of out-of-plane nearest neighbors along the c direction.[19] The effective interplanar exchange coupling is therefore $2(J_1 - J_2)$. If we express the various exchange couplings in terms of respective hopping strengths $(J=4t^2/U)$, then the effective interplanar-hopping strength $t'_{\rm eff}$ is related to the average (t') and the difference $(\Delta t')$ of the out-of-plane nearest-neighbor hoppings by $(t'_{\rm eff})^2 \approx 4t'\Delta t'$. Dividing by t^2, the planar nearest-neighbor hopping, we obtain

$$r^2 \equiv \frac{(t'_{\rm eff})^2}{t^2} \approx 4\left(\frac{t'^2}{t^2}\right)\left(\frac{\Delta t'}{t'}\right). \quad (8)$$

The ratio $\sigma_\perp/\sigma_\parallel$ of the conductivities perpendicular to the copper-oxide plane and along the plane is expected to be proportional to the ratio of the squares of the relevant hopping terms, t'^2/t^2. Therefore, r^2 contains not only the anisotropy in the conductivities, but also the fractional anisotropy in the out-of-plane nearest-neighbor hoppings.

The $Sr_2CuO_2Cl_2$ compound differs from La_2CuO_4 in that it stays in the tetragonal phase down to the lowest temperatures; there are no octahedral rotations and hence no anisotropy in the out-of-plane nearest-neighbor (nn) exchange terms. Therefore the exchange interaction energy due to the coupling between a spin and its eight out-of-plane nn spins, ordered antiferromagnetically, vanishes by symmetry, leading to a frustration between planes. Most likely, magnetic dipole interactions break this frustration and introduce a very weak coupling between layers. The magnetic behavior in $Sr_2CuO_2Cl_2$ should therefore be expected to be even more 2D in nature.

We have also obtained J and r for the $Sr_2CuO_2Cl_2$ system by fitting with the magnetic superlattice reflection data of Vaknin *et al.*[10] which are shown in Fig. 2. In this case also, the theoretical curve fits the data very well, and the best fit with $JM(0)=800$ K (as for the lanthanum compound) yields $r/M(0)=0.004$. Using $M(0)=0.34$ (Ref. 10) we obtain $r=0.0014$. Thus r^2 is about 2 orders of magnitude smaller than that in La_2CuO_4. This lends strong support to the structural viewpoint that it is a much weaker (than exchange type) interaction, possibly a magnetic dipole interaction, which is responsible for coupling between layers in the absence of any orthorhombic distortion.

Recently, neutron-scattering studies have been made to investigate the magnetic structure of the L_2CuO_4 (L = Pr, Nd, Sm) family of systems. For the praseodymium compound, the temperature dependence of the sublattice magnetization, as inferred from the intensity of the magnetic $(\frac{1}{2}, \frac{1}{2}, 1)$ peak, again shows an approximately linear falloff with temperature over a fairly large temperature range from 45 K to just below the Néel tem-

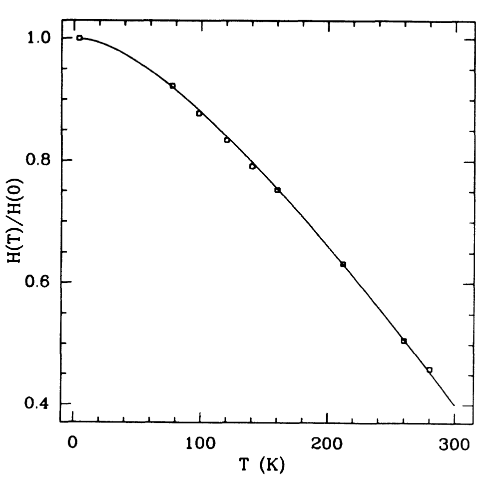

FIG. 1. The normalized hyperfine field strength at different temperatures obtained from the Mössbauer study (open squares) and the normalized sublattice magnetization $M(T)/M(0)$ obtained from Eq. (5) with best-fit parameters of $J=800/M(0)$ K and $r/M(0)=0.022$.

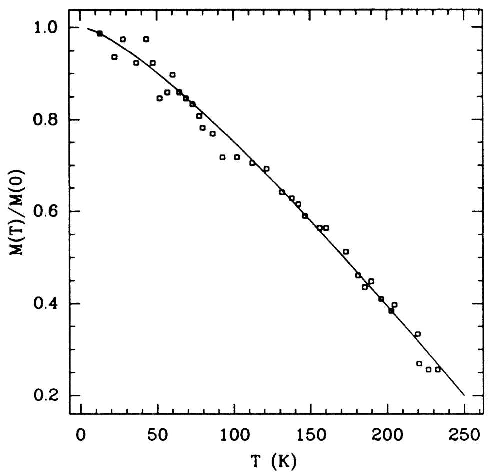

FIG. 2. Normalized intensity of the $[\frac{1}{2}\ \frac{1}{2}\ 0]$ magnetic superlattice reflection at different temperatures in $Sr_2CuO_2Cl_2$ (from Ref. 6) and $M(T)/M(0)$ obtained from our theory with same $JM(0)=800$ K and $r/M(0)=0.004$.

perature.[20,21] The linear falloff is a signature of very weak interlayer magnetic coupling and is again consistent with its tetragonal structure (characteristic of all three compounds). The sublattice magnetization, however, remains almost unchanged below 45 K, which indicates the presence of an energy gap in the spin-wave spectrum. The extremely anisotropic susceptibility due to these L ions[22] is the likely source of this anisotropy gap. Studies of the neodymium compound[23,24] have indicated many complicated reorderings at intermediate temperatures and L^{3+} ions are believed to participate in the ordering at low temperatures.[23]

In conclusion, we have shown that the magnetic dynamics in several copper-oxide-based antiferromagnets, as manifested macroscopically in the temperature dependence of the sublattice magnetization, can be understood in terms of subtle details of their structural characteristics. Thermal excitation of spin waves in a highly anisotropic antiferromagnet, with a ratio r of an effective interplanar- to planar-hopping strength, results in a characteristic magnetization versus temperature behavior with a crossover (at $T=2Jr/k_B$) from a 3D ($\sim T^2$) behavior to a quasi-2D ($\sim T\ln T$) behavior. An estimate of the Néel temperature from this $T\ln T$ falloff leads to $k_BT_N \sim J/\ln(r^{-1})$.[25] Hence the Néel temperature decreases logarithmically with decreasing interlayer coupling. For the orthorhombic, La_2CuO_4 system, the best fit with the magnetization data yields $Z_cJ=1600$ K and $r=0.011$, leading to a crossover temperature of 35 K. An estimate of T_N then yields about 350 K. The magnetization behavior with temperature thus clearly correlates very well with the experimental Néel temperature. In the tetragonal $Sr_2CuO_2Cl_2$ system, however, the net exchange coupling between planes vanishes due to symmetry. The effective coupling in this case is most likely due to a much weaker magnetic dipole interaction. Indeed, for this system we find that r^2, which measures the effective interlayer magnetic coupling, is almost 2 orders of magnitude smaller than in the lanthanum system. The best fit yields $r=0.0014$, implying that the interlayer coupling is $\sim 2\times 10^{-6}$ times the planar coupling, which is about one-fortieth of the ratio of interlayer coupling to planar coupling in La_2CuO_4. Thus we have shown that the magnetic dynamics can be used as a probe to investigate subtle, magnetic aspects of the interlayer coupling.

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25This is in agreement with the estimate obtained by comparing the exchange energy due to interplanar coupling of spins within a domain of size ξ_{2D} and the thermal energy (k_BT_N).