Theory of nuclear relaxation in La_2CuO_4

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We calculate the nuclear spin-lattice relaxation rates $(1/T_1)$ in the quasi-two-dimensional antiferromagnet La_2CuO_4, both above and below the Néel temperature T_N, paying particular attention to the form factors associated with different nuclear sites. The smallness of the interplanar coupling J' compared with the intraplanar coupling J and the absence of on-site Ising anisotropy result in some interesting behaviors of the relaxation rates. For $J \gg T > T_N$, and to leading order, (i) $1/T_1^{Cu} \sim (aT/\hbar c)^{3/2}\xi/a$, where ξ is the two-dimensional correlation length which diverges exponentially at low T, c is the $T=0$ two-dimensional spin-wave velocity (proportional to J) and a is the lattice spacing; (ii) $1/T_1^{O} \sim (aT/\hbar c)^3$; and (iii) $1/T_1^{La} \sim A(aT/\hbar c)^3 + B(aT/\hbar c)^{3/2}\xi/a$, where we expect $B \ll A$ although a precise estimate is unavailable. If anisotropies of the couplings (in spin space) are neglected, the only other relevant temperature scale is set by $\Delta = 2\sqrt{JJ'}$, which defines the crossover between two- and three-dimensional behavior; in La_2CuO_4, $\Delta \approx 20$ K. In the two-dimensional regime $T_N \gg T \gg \Delta$, (i) $1/T_1^{Cu} \sim (aT/\hbar c)T/\Delta$, (ii) $1/T_1^{O} \sim (aT/\hbar c)^4 \ln(T/\Delta)$; and (iii) $1/T_1^{La} \sim A'(aT/\hbar c)^4 \ln(T/\Delta) + B'(aT/\hbar c)^2 T/\Delta$, with $B'/A' \sim B/A$. In the three-dimensional regime $T < \Delta$, one has (i) $1/T_1^{Cu} \sim (aT/\hbar c)(T/\Delta)^2$; (ii) $1/T_1^{O} \sim (aT/\hbar c)^4 (T/\Delta)^3$; and (iii) $1/T_1^{La} \sim A''(aT/\hbar c)^4 (T/\Delta)^3 + B''(aT/\hbar c)^2 (T/\Delta)^3$, with $B''/A'' \sim B/A$. These results for $T < T_N$ are sensitive to the hyperfine interactions assumed; we take the coupling to the Cu nuclei to be highly anisotropic and the couplings to La and O to be isotropic. However, gaps in the spin-wave spectrum, of, say, magnitude E, give rise to rates which vanish as $e^{-E/T}$ at temperatures $T < E$. In La_2CuO_4, Dzyaloshinskii-Moriya interaction gives rise to an E not too different from Δ, and so a clean two- to three-dimensional crossover should not be present even though the quasi-two-dimensional behavior should hold over a wide range of temperature between T_N and Δ, and the relaxation rate for Cu in that temperature regime should be considerably larger than what one might expect based on the rates in cubic cuprate materials.

I. INTRODUCTION

There appears to be considerable interest in the subject of nuclear magnetic resonance (NMR) in high-temperature superconductors and particularly in the spin-lattice relaxation rates $1/T_1$.[1] In particular, differing behaviors of $1/T_1$ for Cu and O nuclei are being used as a diagnostic tool to understand the low-frequency spin dynamics in these materials.[2] Because a number of interpretations[3] of $1/T_1$ rely on strong short-range antiferromagnetic correlations, it is useful to study the insulating, antiferromagnetic parent compounds. Possibly, the nature of *short-range* spin correlations is not drastically modified as the parent compounds are doped to form superconductors; at the very least, most theories of NMR in high-temperature superconductors assume this to be the case.[4] Of course, magnetic resonance in quasi-two-dimensional, $S = \frac{1}{2}$, quantum antiferromagnets is interesting in its own right and can be used to explore the low-frequency spin dynamics of such systems. In the present paper we develop a theory for the relaxation rates both above and below the Néel temperature T_N. For the sake of concreteness, we focus our attention specifically on La_2CuO_4, though similar considerations should apply to the parent compounds of the other cuprate superconductors.

The organization of this paper is as follows. In Sec. II the important magnetic properties of La_2CuO_4 are summarized, and the hyperfine interactions are discussed in Sec. III. The key ideas underlying the calculations are described in Sec. IV, followed by the actual calculations in Sec. V, which is supplemented by an Appendix. Section VI contains a summary, a discussion of the relevance of this work to experiments, and comments on possible extensions of the calculations.

II. MAGNETIC PROPERTIES OF La_2CuO_4

It is by now well established, on both experimental and theoretical grounds, that La_2CuO_4 is a quasi-two-dimensional antiferromagnet which is well described by an $S = \frac{1}{2}$ Heisenberg model.[5] The intraplanar exchange constant J is about 1500 K, while the effective[6] interplanar exchange J' is antiferromagnetic, but much smaller: $J' \approx 0.035$ K $\approx 2 \times 10^{-5}$ J. One of the most prominent features which distinguishes the cuprate perovskites from previously studied antiferromagnets such as K_2NiF_4 is its complete lack of on-site Ising anisotropy, because $S = \frac{1}{2}$ for Cu^{2+}.[7] The three-dimensional Néel transition seen in La_2CuO_4 at 200–300 K (depending on the precise concentration of oxygen) is due to the weak interplanar coupling and takes place well below J. In contrast, the Néel transition in K_2NiF_4 is primarily a two-dimensional Ising transition occurring at $T_N \approx J$ (more precisely,

$T_N = 97.23$ K and $J = 104 \pm 1$ K), with a crossover from two-dimensional Heisenberg to two-dimensional Ising behavior taking place at a temperature readily distinguishable from T_N, at about 102 K.[7,8] (Of course, for temperatures sufficiently close to T_N the critical behavior should ultimately become $d = 3$ Ising-like, but experimentally this asymptotic critical region has been unobservably narrow in K_2NiF_4.) It is therefore not unreasonable to expect that the spin dynamics in La_2CuO_4 must be very different from that in K_2NiF_4.

The dominant anisotropy in the intraplanar couplings in La_2CuO_4 turns out to be an antisymmetric Dzyaloshinskii-Moriya (DM) interaction and corresponds to an energy scale E of roughly 10–20 K (as will be discussed below in Secs. V and VI).[9] The strength of this anisotropy is proportional to the orthorhombic distortion of the crystal structure, and it vanishes for tetragonal crystals. Other anisotropies, such as the dipolar anisotropy, are much weaker. Hence it is sensible for the purpose of discussing the cuprate perovskites generally to consider the behavior of the relaxation rates for a wide range of values of E.

We now summarize the remaining relevant facts concerning La_2CuO_4. From various experiments it is now known that the in-plane spin-wave velocity in this material is $\hbar c \approx 0.85$ eV Å.[10–12] Moreover, inelastic neutron-scattering experiments have demonstrated that the dynamics of spin fluctuations above T_N is, to an excellent approximation, independent of the momentum component perpendicular to the CuO planes.[13] Thus a two-dimensional model is reasonable for the disordered phase. At present our understanding of the neutron-scattering experiments in the thermally disordered phase appears to be quite satisfactory, as there is good agreement between the data and theory developed by Chakravarty, Halperin, and Nelson[14] (CHN) and Tyc, Halperin, and Chakravarty[15] (THC). With respect to static properties, the correlation length ξ is found to diverge as $\xi/a \approx C_\xi \exp(2\pi\rho_s/T)$ for $T > T_N$, where C_ξ is a pure number of order unity and ρ_s ($=0.16J$ in spin-wave theory at $S = \frac{1}{2}$) is the spin stiffness of a two-dimensional square-lattice antiferromagnet at $T = 0$.[11] With respect to dynamics the agreement between theory and experiment is equally good.[11] In the theory, relatively long-lived spin waves must be present for length scales smaller than ξ, while for scales larger than ξ the spin dynamics must be diffusive, corresponding to a dynamic structure factor which exhibits a peak at zero frequency known as the quasielastic peak. (This *intrinsic* quasielastic peak is distinct from the *extrinsic* central peak phenomenon, unfortunately also known as the quasielastic peak, which may arise from magnetic defects such as two-level systems.)

As mentioned earlier, the three-dimensional transition in this material is due to interplanar coupling. Even though the interplanar coupling is extremely small, the system orders antiferromagnetically well above J', when ξ becomes sufficiently large. In mean-field theory the Néel transition is determined by the criterion $(N/N_0)^2(\xi/a)^2 J' = T_N$,[14] where N/N_0 ($=0.606$, in spin-wave theory) is the ratio of the sublattice magnetization at $T = 0$ in the two-dimensional antiferromagnet to its maximum possible value.

III. HYPERFINE INTERACTIONS

In La_2CuO_4 the orbital angular momentum is quenched.[16] Therefore, the spin-lattice relaxation is due to the coupling of the nuclear moments to the electronic spin degrees of freedom, described by a suitable hyperfine Hamiltonian. It is now generally believed that the relevant spin degrees of freedom are described by Wannier functions of $d_{x^2-y^2}$ symmetry centered at Cu sites. This orbital has vanishing amplitude at the nucleus, and hence the Fermi contact interaction is not relevant for the Cu nuclei. Instead, the hyperfine interaction for Cu must be due to electron-nuclear dipolar interaction or core polarization effects. This leads to an anisotropic Hamiltonian for Cu, given by

$$H_{\mathrm{Cu}} = {}^{\mathrm{Cu}}\mathbf{I}_{\mathbf{n}} \cdot \mathbf{A}_{\mathrm{Cu}} \cdot \mathbf{S}_{\mathbf{n}} , \tag{3.1}$$

where ${}^{\mathrm{Cu}}\mathbf{I}_{\mathbf{n}}$ is the nuclear spin at site $\mathbf{n}$, $\mathbf{S}_{\mathbf{n}}$ is the electronic spin, and $\mathbf{A}_{\mathrm{Cu}}$ is the hyperfine tensor with two independent components $A_{\mathrm{Cu},xx}$ ($= A_{\mathrm{Cu},yy}$) and $A_{\mathrm{Cu},zz}$. Here we have ignored the orthorhombic distortion (present below about 520 K) which may give rise to further anisotropy, but which should be negligible for our purposes. However, Mila and Rice[17] have pointed out that it is necessary to incorporate an additional term in H_{Cu}. Because the Wannier function centered on a given site has a nonvanishing projection on neighboring Cu $3s$ or $4s$ orbitals, it is possible to have a sizable additional term due to the large magnitude of the Fermi contact interaction. Thus we may write

$$H_{\mathrm{Cu}} = {}^{\mathrm{Cu}}\mathbf{I}_{\mathbf{n}} \cdot \mathbf{A}_{\mathrm{Cu}} \cdot \mathbf{S}_{\mathbf{n}} + B_{\mathrm{Cu}} \sum_{\boldsymbol{\delta}} {}^{\mathrm{Cu}}\mathbf{I}_{\mathbf{n}} \cdot \mathbf{S}_{\mathbf{n}+\boldsymbol{\delta}} , \tag{3.2}$$

where the vectors $\boldsymbol{\delta}$ connect the site $\mathbf{n}$ to its neighboring Cu sites $\mathbf{n}+\boldsymbol{\delta}$. Because the source of the second term is the contact interaction, B_{Cu} can be assumed to be isotropic. As will become evident below, the additional term does not contribute to the leading temperature dependence in the Cu relaxation rate below T_N (as the two-magnon relaxation mechanism associated with the first term dominates the three-magnon relaxation coming from the second), while above T_N it only modifies the magnitude, but not the temperature dependence, of the rate. Hence, although the full interaction (3.2) is necessary to understand some aspects of the problem[18] (particularly the dependence of the Knight shift and relaxation rate on the orientation of the magnetic field), (3.1) will be adequate for our purposes.

The Hamiltonian for the O nucleus can be obtained in a similar manner, as shown by Shastry,[19] for example, giving

$$H_{\mathrm{O}} = A_{\mathrm{O}} \sum_{\boldsymbol{\delta}'} {}^{\mathrm{O}}\mathbf{I}_{\mathbf{n}} \cdot \mathbf{S}_{\mathbf{n}+\boldsymbol{\delta}'} , \tag{3.3}$$

where the vectors $\boldsymbol{\delta}'$ connect the O site to the two neighboring Cu sites. It is commonly assumed that A_{O} is isotropic, as displayed here. Strictly speaking, this is not

correct. The observed anisotropy in A_O has been interpreted as being due to the fact that it is the O p_σ orbital which hybridizes with the Cu $d_{x^2-y^2}$ orbital.[20] However, because the anisotropy is small, it is probably not important for our purposes.

Similarly, one would expect the dominant hyperfine interaction for a La nucleus to be

$$H_{\rm La} = A_{\rm La} \sum_{\delta''} {}^{\rm La}\mathbf{I}_{\mathbf{n}} \cdot \mathbf{S}_{\mathbf{n}+\delta''} , \tag{3.4}$$

where the vectors δ'' connect the La site to the four Cu sites in the plane adjacent to it. As above, the coupling will be assumed to be isotropic. In writing (3.4) we have neglected any coupling to Cu spins on the next-nearest CuO plane; while this may seem eminently reasonable (because the next-nearest plane is roughly twice as far as the nearest plane), it is not fully adequate.[21] In that next-nearest plane, a *single* Cu spin is coupled most strongly to the La nuclear spin, rather than a set of two or four Cu spins. Thus $H_{\rm La}$ has an additional term which we write as

$$B_{\rm La} {}^{\rm La}\mathbf{I}_{\mathbf{n}} \cdot \mathbf{S}'_{\mathbf{n}+\xi} , \tag{3.5}$$

where the prime indicates that the Cu spin lies in a different CuO plane than the spins **S** in (3.4). It seems best to assume that $B_{\rm La}$ is an isotropic coupling as well. We expect $B_{\rm La} \ll A_{\rm La}$, but, as will become evident, the contribution of (3.5) to the La relaxation may be important, nonetheless. Let us also point out here that the hyperfine interaction for Y in Y-Ba-Cu-O is much like that for La in La_2CuO_4, but there is no contribution of the form (3.5), because of its particular crystal structure.

As we shall see, the anisotropy (or lack thereof) of the hyperfine interaction plays an important role in determining the temperature dependence of the relaxation rates below the Néel transition. Above the transition, when the electronic spin correlations are isotropic, anisotropy in the hyperfine couplings does not affect the temperature dependence of the rates, though it does affect the magnitudes and field dependences of the rates.

IV. OVERVIEW OF THE CALCULATIONS

In the present section we summarize some of the key ideas employed in the calculations which are given in the following section. Consider first the regime above T_N. The relaxation rate for Cu has been calculated by Chakravarty and Orbach[22] using the dynamic structure factor of CHN (Ref. 14) and THC.[15] That part of the La relaxation associated with the hyperfine interaction (3.5) may be calculated in the same way, since the coupling is the same as that for Cu. However, this structure factor is valid only in the regime $k \ll T/\hbar c$, that is, for wavelengths larger than the thermal de Broglie wavelength of the spin waves. [Here and below k is measured with respect to the antiferromagnetic ordering wave vector $Q \equiv (\pi/a, \pi/a)$]. Although the Cu rate can be accurately calculated, the O rate cannot be calculated using this structure factor because its associated form factor [see (5.29) below] vanishes as $k \to 0$. The integral over the Brillouin zone which determines $1/T_1^O$ is not dominated by the critical fluctuations, and *a priori* the entire Brillouin zone appears to contribute. To the extent that the La rate is due to the interaction (3.4), similar considerations apply in that case as well.

A calculation of the O rate above T_N has been given by Bulut *et al.*,[23] who use the dynamic structure factor due to Arovas and Auerbach.[24] However, the correctness of that form for the structure factor has been questioned,[25] and it is not clear whether the resulting expression for $1/T_1^O$ is correct.

Our method is a spin-wave expansion, which, at least in principle, can be systematically improved. Let us first discuss the situation for $T > T_N$. Even though the system is disordered, the correlation length is quite large for temperatures small compared to J, and thus magnons with wave vector k satisfying $k\xi \gg 1$ should be well defined. Indeed, explicit calculations have confirmed that the damping is small compared to the frequency for such magnons.[26] The recent neutron-scattering experiments of Aeppli *et al.* also support such a picture.[10]

As noted above, the expressions for the O and (part of) the La relaxation rates involve Brillouin-zone integrals which contain little weight at small wave vectors because of form factors associated with the position of those nuclei relative to the lattice of Cu spins. Thus, at first sight, spin-wave calculations for the O and La rates appear to be meaningful even above T_N; indeed, one can set the long-wavelength cutoff on the magnon wave vectors to zero with impunity. However, this can lead to completely erroneous results as the spin-wave picture explicitly breaks rotational invariance, because magnons are, by definition, transverse excitations in the broken-symmetry phase. But the symmetry is unbroken in the disordered phase, and therefore a naive spin-wave expansion is not meaningful at any wavelength even if the integrals determining the relaxation rates are convergent. In order to circumvent this problem, one may make a local spin-wave expansion in which different domains, of typical size ξ, have their order parameter (the staggered magnetization) oriented in different directions.[27] In this picture rotational invariance is restored. Equivalently, and what is done here, is to use the magnon picture to calculate *only* rotationally invariant quantities, so that any "memory" of a particular broken-symmetry direction that one may have chosen is lost. Since the relaxation rates in the thermally disordered phase can indeed be written in terms of a rotationally invariant dynamic structure factor, this approach permits us to calculate them.

We now discuss the regime below T_N. Of course, in this regime a spin-wave expansion is sensible, and such calculations of relaxation rates have a long history.[28,29] However, an interesting feature that should be kept in mind is the highly (spatially) anisotropic nature of the spin-wave dispersion in La_2CuO_4, due to the exceedingly small interplanar coupling. The spin waves are nearly dispersionless for momentum components perpendicular to the CuO layers. Above an energy scale Δ given by $\Delta = 4S\sqrt{JJ'}$, the dynamics of the system is effectively two dimensional, with a low-energy cutoff Δ. In La_2CuO_4 we

estimate Δ to be between 10 and 20 K, and so there is a large temperature range even below T_N in which the spin dynamics is essentially two dimensional.

Finally, it might be argued that our treatment leaves out an important bit of physics associated with spin diffusion, which in principle may contribute significantly to the low-frequency spin dynamics in low-dimensional materials. However, it has already been pointed out that spin diffusion is actually irrelevant at the temperatures of concern to us, which are small compared to J.[30]

V. CALCULATIONS AND RESULTS

A. General considerations

Consider the Heisenberg Hamiltonian for spins situated on a simple cubic lattice:

$$H=\tfrac{1}{2}J\sum_{\mathbf{j},\delta}\mathbf{S}_\mathbf{j}\cdot\mathbf{S}_{\mathbf{j}+\delta}+\tfrac{1}{2}J'\sum_{\mathbf{j},\delta'}\mathbf{S}_\mathbf{j}\cdot\mathbf{S}_{\mathbf{j}+\delta'}\ , \tag{5.1}$$

where the vectors δ connect the site $\mathbf{j}$ with its nearest neighbors on a square lattice (representing the CuO planes), and δ' connects $\mathbf{j}$ with its neighbors on the adjacent planes. Since the Cu^{2+} in La_2CuO_4 do not form a simple cubic lattice, this must be considered to be an effective Hamiltonian (recall Ref. 6); but as will be seen, the only effect of the interplanar coupling is to generate a small energy scale, and we expect the precise lattice structure to be unimportant.

Let us now use the Holstein-Primakoff transformation[31] to rewrite the spin operators in terms of boson operators. Divide the lattice into two sublattices A and B, and define on the A sublattice

$$S_i^+=\sqrt{2S}\left[1-\frac{1}{2S}a_i^\dagger a_i\right]^{1/2}a_i\ , \tag{5.2}$$

$$S_i^-=\sqrt{2S}\,a_i^\dagger\left[1-\frac{1}{2S}a_i^\dagger a_i\right]^{1/2}\ , \tag{5.3}$$

$$S_i^z=S-a_i^\dagger a_i\ , \tag{5.4}$$

and on the B sublattice

$$S_j^+=\sqrt{2S}\,b_j^\dagger\left[1-\frac{1}{2S}b_j^\dagger b_j\right]^{1/2}\ , \tag{5.5}$$

$$S_j^-=\sqrt{2S}\left[1-\frac{1}{2S}b_j^\dagger b_j\right]^{1/2}b_j\ , \tag{5.6}$$

$$S_j^z=-S+b_j^\dagger b_j\ . \tag{5.7}$$

The Fourier transforms of the boson operators are defined to be

$$a_\mathbf{k}=N^{-1/2}\sum_j e^{i\mathbf{k}\cdot\mathbf{x}_j}a_j\ , \tag{5.8}$$

$$b_\mathbf{k}=N^{-1/2}\sum_j e^{-i\mathbf{k}\cdot\mathbf{x}_j}b_j\ , \tag{5.9}$$

where N is the number of sites on each of the sublattices, so that the total number of lattice sites is $2N$. These transformations, when applied to the spin Hamiltonian (5.1), lead to a boson representation of that Hamiltonian which is nonlinear in the boson operators. Consider for the moment the linearized Hamiltonian which describes noninteracting spin waves, namely,

$$H_L=-NS^2\gamma_0+S\sum_k\gamma_0(a_\mathbf{k}^\dagger a_\mathbf{k}+b_\mathbf{k}^\dagger b_\mathbf{k})+S\sum_k\gamma_\mathbf{k}(a_\mathbf{k}^\dagger b_\mathbf{k}^\dagger+a_\mathbf{k}b_\mathbf{k})\ , \tag{5.10}$$

where

$$\gamma_0=zJ+z'J'\ , \tag{5.11}$$

$$\gamma_\mathbf{k}=\frac{zJ}{d}[\cos(k_xa)+\cos(k_xa)]+z'J'\cos(k_za_\perp)\ , \tag{5.12}$$

z (the intraplanar coordination number) is 4, z' (the interplanar coordination number) is 2, d (the planar dimensionality) is 2, a is the intraplanar lattice constant, and $a_\perp$ is the interplanar lattice constant.

A Bogoliubov transformation diagonalizes H_L. The new boson operators are defined by

$$\alpha_\mathbf{k}=u_\mathbf{k}a_\mathbf{k}-v_\mathbf{k}b_\mathbf{k}^\dagger\ , \tag{5.13}$$

$$\beta_\mathbf{k}=u_\mathbf{k}b_\mathbf{k}-v_\mathbf{k}a_\mathbf{k}^\dagger\ , \tag{5.14}$$

where

$$u_\mathbf{k}^2-v_\mathbf{k}^2=1\ , \tag{5.15}$$

and

$$2u_\mathbf{k}v_\mathbf{k}=-\frac{\gamma_\mathbf{k}/\gamma_0}{[1-(\gamma_\mathbf{k}/\gamma_0)^2]^{1/2}}\ . \tag{5.16}$$

In the long-wavelength limit,

$$-u_\mathbf{k}v_\mathbf{k}\approx u_\mathbf{k}^2\approx v_\mathbf{k}^2\approx\frac{\gamma_0}{2(\gamma_0^2-\gamma_\mathbf{k}^2)^{1/2}}\ . \tag{5.17}$$

The diagonalized form of H_L can be written as

$$H_L=-NS(S+1)\gamma_0+\sum_\mathbf{k}\varepsilon_\mathbf{k}+\sum_\mathbf{k}\varepsilon_\mathbf{k}(\alpha_\mathbf{k}^\dagger\alpha_\mathbf{k}+\beta_\mathbf{k}^\dagger\beta_\mathbf{k})\ , \tag{5.18}$$

with $\varepsilon_\mathbf{k}$ the spin-wave dispersion, namely,

$$\varepsilon_\mathbf{k}=S(\gamma_0^2-\gamma_\mathbf{k}^2)^{1/2}=4JS(1+\tfrac{1}{2}r)(1-\lambda_\mathbf{k}^2)^{1/2}\ , \tag{5.19}$$

where

$$r=J'/J\ , \tag{5.20}$$

and

$$\lambda_\mathbf{k}=\frac{1}{2+r}[\cos(k_xa)+\cos(k_ya)+r\cos(k_za_\perp)]\ . \tag{5.21}$$

Since we are concerned with temperatures small compared to J, it will be sufficient to linearize the spectrum in $k_\parallel=(k_x^2+k_y^2)^{1/2}$, and thus

$$\varepsilon_\mathbf{k}\approx2\sqrt{2}JS\{(k_\parallel a)^2+2r[1-\cos(k_za_\perp)]\}^{1/2}\ . \tag{5.22}$$

Given the spectrum $\varepsilon_\mathbf{k}$, one can readily calculate the density of states $\rho(\varepsilon)$, obtaining (for $S=\frac{1}{2}$)

$$\rho(\varepsilon)\approx\frac{1}{2\pi\sqrt{2r}}\left[\frac{a}{\hbar c}\right]^3\varepsilon^2,\quad \varepsilon\ll\frac{\hbar c}{a}\sqrt{2r}\ , \tag{5.23}$$

$$\rho(\varepsilon)\approx\frac{1}{2\pi}\left[\frac{a}{\hbar c}\right]^2\varepsilon,\quad J\gg\varepsilon\gg\frac{\hbar c}{a}\sqrt{2r}\ . \tag{5.24}$$

If we define the energy scale Δ by

$$\Delta=\frac{\hbar c}{a}\sqrt{2r}\approx 2\sqrt{JJ'}\ , \tag{5.25}$$

then for energies smaller than Δ the density of states has the form appropriate to a three-dimensional system, while for energies greater than Δ the system appears to be two dimensional. Here the spin-wave velocity c in the plane was taken to be $\hbar c=\sqrt{2}Ja$, which is its value in the linear spin-wave approximation for $S=\frac{1}{2}$.

The nuclear relaxation rate can be obtained from Fermi's golden rule:

$$1/T_1=2W\ , \tag{5.26}$$

where W is the transition rate given by

$$W=\sum_{n,m}\frac{\exp(-\beta E_n)}{Z}|\langle m|H_i|n\rangle|^2\delta(E_n-E_m)\ . \tag{5.27}$$

This is an exact formula within linear response theory. Here E_n are the exact energy eigenvalues of the electronic spin system, and the hyperfine interaction Hamiltonian H_i, which was already discussed in Sec. III, causes transitions between the exact initial and final states of the spin system. For simplicity, we shall take the nuclear matrix elements equal to unity when we evaluate Eq. (5.27) explicitly. This means that we have, in effect, calculated $1/T_1$ for the $\pm\frac{1}{2}$ transition for an $I=\frac{1}{2}$ nucleus. The extension to a transition rate between given nuclear spin states is straightforward. The factor Z is the partition function of the electronic spin system.

In our calculation we shall take E_n to be the energy spectrum of the noninteracting spin-wave states. Thus there should be two sources of corrections to our results: (i) $T=0$ renormalization of the magnon spectrum and (ii) thermal renormalization. It is now well established that the renormalization of the $T=0$ spectrum is not substantial. The magnon-magnon interaction simply renormalizes the spectrum, for all $\mathbf{k}$, by a multiplicative factor, which amounts to roughly a 15% correction for $S=\frac{1}{2}$ in two dimensions (and is a smaller correction for larger S).[32] The spectrum remains concave throughout the entire Brillouin zone, which implies that a magnon cannot spontaneously decay, into, say, three magnons, because of the restrictions of energy and momentum conservation.[33] Moreover, the correction as calculated by the spin-wave expansion (which is an asymptotic expansion) agrees remarkably well with more refined numerical calculations.[34] Thus it appears that the magnon spectral function at $T=0$ should have a well-defined quasiparticle peak and a broad multimagnon background above that peak. The broad background is due to the decay of magnons off resonance; the situation is similar to that of quasiparticles in Fermi liquid theory.[33] At $T>0$ the system should behave as though it were at $T=0$ on length scales smaller than the thermal de Broglie wavelength proportional to $\hbar c/T$; hence, on these scales, the above considerations imply that one should have only a harmless $T=0$ renormalization of the physical parameters.

The effects of thermal renormalization are more serious. In particular, in a strictly two-dimensional system such renormalization would become increasingly important on longer length scales, and magnons would not exist as well-defined quasiparticles at scales larger than the finite-temperature correlation length ξ. Of course, the situation is better for the three-dimensional system in the Néel-ordered phase. However, a correct and consistent calculation of finite-temperature renormalization effects (due to thermal vertex and self-energy corrections) appears to be formidable (see the Appendix.) Even if one were to perform the calculation, it would not change the leading temperature dependences of the relaxation rates, although the absolute magnitudes would change since the magnon-magnon interaction is expected to renormalize the spin stiffness, which would become scale dependent. We are assuming that for short length scales such renormalizations are small, and that the quantities we calculate are dominated by short-wavelength excitations.

B. $T>T_N$

A rather detailed and unified description of the calculations of the relaxation rates in the disordered phase, but at temperatures small compared to J, is given in the Appendix. Here we will present brief discussions of the relevant calculations and the results.

1. *Cu relaxation*

The hyperfine Hamiltonian associated with Cu nuclei was given in (3.1). As shown by Chakravarty and Orbach,[22] $1/T_1^{\mathrm{Cu}}$ is dominated by critical fluctuations at the incipient antiferromagnetic ordering wave vector and, hence, can be calculated using the dynamic structure factor of THC. We shall not repeat this calculation, except to note the principal temperature dependence:

$$1/T_1^{\mathrm{Cu}}\propto T^{3/2}\xi\propto T^{3/2}\exp(\kappa J/T)\ , \tag{5.28}$$

where the constant κ is close to unity.

2. *O relaxation*

As noted earlier, there appears to be no justification in using the dynamical structure factor due to THC to calculate $1/T_1^{\mathrm{O}}$ as it is not dominated by critical fluctuations. Instead, we shall use spin-wave theory, since the short-wavelength magnons are well defined, and as will become evident, there is little weight at long wavelengths. However, as stressed above, we are restricted to calculating only rotationally invariant quantities.

In the disordered phase, the relaxation rate for the O nuclei can be expressed as

$$\frac{1}{T_1^{\mathrm{O}}}=\frac{A_{\mathrm{O}}^2}{6N\hbar^2}\sum_{\mathbf{q}}f^{(\mathrm{O})}(\mathbf{q})S(\mathbf{q},\omega\approx 0)\ , \tag{5.29}$$

where the sum covers the *full* two-dimensional Brillouin zone, not the antiferromagnetic Brillouin zone, and $S(\mathbf{q},\omega)$ is the dynamic structure factor of the electronic spins:

$$S(\mathbf{q},\omega)=\sum_j \int_{-\infty}^{\infty} dt\, e^{i(\omega t-\mathbf{q}\cdot\mathbf{r}_j)}\langle \mathbf{S}_j(t)\cdot\mathbf{S}_0(0)\rangle \ . \quad (5.30)$$

The form factor $f^{(O)}$ is given by

$$f^{(O)}(\mathbf{q})=1+\cos(q_x a) \ . \quad (5.31)$$

This arises from the observation that the relaxation takes place through the transferred hyperfine coupling to two adjacent Cu spins, assumed here to lie along the x direction. The calculation of the rate is described in detail in the Appendix. Here we quote the final answer for $S=\frac{1}{2}$ systems:

$$\frac{1}{T_1^O}\approx\frac{\pi}{3}\frac{(A_O/\hbar)^2}{(c/a)}\left[\frac{aT}{\hbar c}\right]^3 . \quad (5.32)$$

The result for the low-temperature O relaxation rate in an isolated CuO plane quoted by Bulut *et al.*[23] is identical to this, except that theirs is smaller by a factor of $6/\pi^2$.

3. *La relaxation*

Let us first set $B_{La}=0$; that is, we consider only the hyperfine interaction with the four nearest Cu spins. In this case the calculation of the relaxation rate for La is similar to that for O. The form factor is different, but like that for O, it vanishes at the antiferromagnetic ordering wave vector so that critical fluctuations do not provide the dominant relaxation channels. The spin-wave calculation yields

$$\frac{1}{T_1^{La}}\approx\frac{4\pi}{9}\frac{(A_{La}/\hbar)^2}{(c/a)}\left[\frac{aT}{\hbar c}\right]^3 . \quad (5.33)$$

A nonzero B_{La} can change the results; the correlations one needs to determine [cf. Eqs. (5.29) and (5.30)] are $\langle \mathbf{T}(t)\cdot\mathbf{T}(0)\rangle$, with

$$\mathbf{T}=B_{La}\mathbf{S}'_{n+\zeta}+A_{La}\sum_{\delta''}\mathbf{S}_{n+\delta''} \ , \quad (5.34)$$

but $\langle \mathbf{S}'_{n+\zeta}\cdot\mathbf{S}_{n+\delta''}\rangle$ is entirely negligible because J' is so small. Thence, dropping the cross terms in the correlation function, one is left with the result that, in addition to the contribution to $1/T_1^{La}$ described by (5.33), one also has a term proportional to B_{La}^2, which is of the same form as $1/T_1^{Cu}$ [see (5.28)]. (Note that in the disordered phase of the electronic spins, $T>T_N$, the anisotropy of the hyperfine couplings does not affect the temperature dependence of the relaxation rate.) It is possible that this additional contribution to $1/T_1^{La}$ may be significant, at least for small enough T/J, even though the associated hyperfine coupling may be small. The experimental signature would be unmistakable: The relaxation rate would increase, rather than decrease, with decreasing temperature.

C. $\Delta \ll T < T_N$

We have already noted that in the disordered phase the temperature dependences of the relaxation rates do not depend on the degree of anisotropy in the hyperfine interactions, because the electronic spin correlations are isotropic. Below T_N, however, this is no longer true. The calculations that follow are similar to those of Beeman and Pincus,[29] except that the system considered here is highly spatially anisotropic. We should note, however, that we do not explicitly take into account the exchange enhancement considered by Beeman and Pincus; this is precisely the $T=0$ renormalization effect which, as argued above, should cause only a multiplicative renormalization of the rates without changing their temperature dependences. We also do not take into account the thermal renormalization effects noted before. Such effects are presumably small for temperatures well below the Néel transition, but may become important close to the transition. However, one advantage of the present problem is that $T_N \ll J$, unlike the case considered by Beeman and Pincus. At least we may expect our theory to be correct to leading order in T, and the leading behavior may be the dominant contribution until quite close to T_N.

1. *Cu relaxation*

For Cu, with its highly anisotropic hyperfine interaction, the term in the interaction Hamiltonian that dominates the relaxation process is of the form

$$H_i = AI^+S^z \ , \quad (5.35)$$

which gives rise to two-magnon relaxation processes. In contrast, the term in the interaction of the form I^+S^- gives rise to three and higher magnon processes, which we shall show below to yield contributions to the relaxation rate which vary to higher powers of T than that of the two-magnon processes.

For the interaction (5.35), one finds[35]

$$\begin{aligned}\frac{1}{T_1^{Cu}}&=\frac{8\pi A^2}{\hbar N^2}\sum_{k_1,k_2}|u_1u_2|^2(1+n_1)n_2\delta(\varepsilon_1-\varepsilon_2)\\ &=\frac{8\pi A^2}{\hbar}\int_0^\infty d\varepsilon\, u^2(\varepsilon)\rho^2(\varepsilon)n(\varepsilon)[1+n(\varepsilon)]\\ &\approx\frac{A^2S^2\gamma_0^2}{2\pi\hbar}\left[\frac{a}{\hbar c}\right]^4\int_\Delta^\infty d\varepsilon\, n(\varepsilon)[1+n(\varepsilon)]\\ &\approx\frac{1}{\pi}\frac{(A/\hbar)^2}{(c/a)}\frac{aT}{\hbar c}\frac{T}{\Delta} \ . \end{aligned} \quad (5.36)$$

To see the effect of higher-order processes, consider an additional interaction term of the form

$$H_i = A'I^+S^- \ . \quad (5.37)$$

Note that this term does not interfere with the two-magnon relaxation calculated above. The spin operator can now be expanded using the Holstein-Primakoff transformation. The one-magnon term does not contribute to nuclear relaxation, while the three-magnon contribution is

$$\frac{1}{T_1^{\mathrm{Cu}}(3)}=\frac{\pi A'^2}{2S\hbar N^3}\sum_{k_1,k_2,k_3}(u_1^2u_2^2u_3^2+4u_1^2u_2^2v_3^2+4v_1^2u_2^2v_3^2+v_1^2v_2^2v_3^2)(1+n_1)n_2n_3\delta(\varepsilon_1-\varepsilon_2-\varepsilon_3)\ . \tag{5.38}$$

This contribution can be evaluated like the previous term, and one finds

$$\frac{1}{T_1^{\mathrm{Cu}}(3)}\approx\frac{5}{16\sqrt{2}\pi^2S}\frac{(A'/\hbar)^2}{(c/a)}\left[\frac{aT}{\hbar c}\right]^2\int_{\Delta/T}^{\infty}dx\int_{\Delta/T}^{\infty}dy\frac{e^{x+y}}{(e^x-1)(e^y-1)(e^{x+y}-1)}$$
$$\approx\frac{5\ln 2}{16\sqrt{2}\pi^2S}\frac{(A'/\hbar)^2}{(c/a)}\left[\frac{aT}{\hbar c}\right]^2\frac{T}{\Delta}\ . \tag{5.39}$$

2. O relaxation

We have noted earlier that the hyperfine interaction for O nuclei is isotropic, and so below T_N there are no two-magnon relaxation processes and the leading contribution to the relaxation comes from three-magnon processes. In this case the interaction Hamiltonian is given by

$$H_i=\tfrac{1}{2}A_{\mathrm{O}}{}^{\mathrm{O}}I^-(S_1^+ +S_2^+)\ , \tag{5.40}$$

where the O nucleus is coupled to its nearest-neighbor electronic spins. As above, we rewrite this in terms of magnon operators, truncate to lowest order, and apply Fermi's golden rule. The result is

$$\frac{1}{T_1^{\mathrm{O}}}=\frac{\pi A_{\mathrm{O}}^2}{2S\hbar N^3}\sum_{k_1,k_2,k_3}(1+n_1)n_2n_3\delta(\varepsilon_1-\varepsilon_2-\varepsilon_3)$$
$$\times\{[u_1u_2u_3+v_1v_2v_3g(1,2,3)]^2+4[u_1u_2v_3+v_1v_2u_3g(2,1,3)]^2$$
$$+4[v_1u_2v_3+u_1v_2u_3g(3,1,2)]^2+[v_1v_2v_3+u_1u_2u_3g(1,2,3)]^2\}\ , \tag{5.41}$$

where

$$g(l,m,n)=e^{ia(k_{lx}+k_{mx}+k_{nx})}\ . \tag{5.42}$$

Introducing the magnon density of states, this can be rewritten as

$$\frac{1}{T_1^{\mathrm{O}}}\approx\frac{5}{16\sqrt{2}\pi^2S}\frac{(A_{\mathrm{O}}/\hbar)^2}{(c/a)}\left[\frac{aT}{\hbar c}\right]^4\left[\int_{\Delta/T}^{\infty}dx\int_{\Delta/T}^{\infty}dy\frac{(2x^2+xy)e^{x+y}}{(e^x-1)(e^y-1)(e^{x+y}-1)}\right]$$
$$\approx\frac{5}{24\sqrt{2}S}\frac{(A_{\mathrm{O}}/\hbar)^2}{(c/a)}\left[\frac{aT}{\hbar c}\right]^4\ln\left[\frac{1.1T}{\Delta}\right]\ . \tag{5.43}$$

3. La relaxation

As in Sec. V A, let us first consider the case $B_{\mathrm{La}}^2=0$. Since the hyperfine interaction for La is isotropic, just as in the case of O, the leading contribution to the relaxation is due to three-magnon processes. Recall that each La nucleus is coupled to four electronic spins, and thus the interaction Hamiltonian is given by

$$H_i=\tfrac{1}{2}A_{\mathrm{La}}{}^{\mathrm{La}}I^-(S_1^+ +S_2^+ +S_3^+ +S_4^+)\ . \tag{5.44}$$

Consequently,

$$\frac{1}{T_1^{\mathrm{La}}}=\frac{\pi A_{\mathrm{La}}^2}{2S\hbar N^3}\sum_{k_1,k_2,k_3}(1+n_1)n_2n_3\delta(\varepsilon_1-\varepsilon_2-\varepsilon_3)$$
$$\times\{[u_1u_2u_3f_a(1,2,3)+v_1v_2v_3f_b(3,2,1)]^2+4[u_1u_2v_3f_1(2,1,3)+v_1v_2u_3f_b(3,1,2)]^2$$
$$+4[v_1u_2v_3f_1(3,2,1)+u_1v_2u_3f_b(1,2,3)]^2+[v_1v_2v_3f_a(1,2,3)+u_1u_2u_3f_b(3,2,1)]^2\}\ , \tag{5.45}$$

where

$$f_a(l,m,n)=1+e^{ia(k_{lx}+k_{ly}-k_{mx}-k_{my}-k_{nx}-k_{ny})}, \quad (5.46)$$

(the a in the exponent signifying the lattice spacing), and

$$f_b(l,m,n)=e^{-ia(k_{lx}+k_{mx}-k_{nx})}+e^{-ia(k_{ly}-k_{my}+k_{ny})}. \quad (5.47)$$

A calculation similar to the one for O yields

$$\frac{1}{T_1^{\rm La}}\approx\frac{1}{4\sqrt{2}S}\frac{(A_{\rm La}/\hbar)^2}{(c/a)}\left[\frac{aT}{\hbar c}\right]^4\ln\left[\frac{T}{1.1\Delta}\right]. \quad (5.48)$$

What is the effect of the $B_{\rm La}$ term in the hyperfine interaction? Although $\mathbf{S}'_{n+\zeta}$ and the $\mathbf{S}_{n+\delta''}$ are correlated below T_N, their fluctuations are uncorrelated, just as in the disordered phase, because the spin waves have little dispersion perpendicular to the CuO plane. We may simply add to $1/T_1^{\rm La}$ a contribution of the form (5.39) (i.e., three-magnon relaxation), but with A' replaced by $B_{\rm La}$.

D. $T\ll\Delta$

The calculations for the temperature regime $T\ll\Delta$ proceed in the same manner as those for $T_N>T\gg\Delta$. The only difference is that the relevant momentum integrals must be done using the three-dimensional density of states (5.23), rather than the two-dimensional form (5.24) used above, because the integrals in every case are dominated by wave vectors corresponding to energies of order T. We now just quote the final results for the leading contributions to the relaxation rates:

$$\frac{1}{T_1^{\rm Cu}}\approx\frac{\pi}{3S}\frac{(A_{\rm Cu}/\hbar)^2}{(c/a)}\frac{aT}{\hbar c}\left[\frac{T}{\Delta}\right]^2, \quad (5.49)$$

$$\frac{1}{T_1^{\rm O}}\approx\frac{P^{\rm O}}{S}\frac{(A_{\rm O}/\hbar)^2}{(c/a)}\left[\frac{aT}{\hbar c}\right]^4\left[\frac{T}{\Delta}\right]^3, \quad (5.50)$$

$$\frac{1}{T_1^{\rm La}}\approx\frac{P_A^{\rm La}}{S}\frac{(A_{\rm La}/\hbar)^2}{(c/a)}\left[\frac{aT}{\hbar c}\right]^4\left[\frac{T}{\Delta}\right]^3+\frac{P_B^{\rm La}}{S}\frac{(B_{\rm La}/\hbar)^2}{(c/a)}\left[\frac{aT}{\hbar c}\right]^2\left[\frac{T}{\Delta}\right]^3, \quad (5.51)$$

where $P^{\rm O}$, $P_A^{\rm La}$, and $P_B^{\rm La}$ are linear combinations of various lattice sums (Riemann ζ functions and two-dimensional generalizations thereof) for which numerical evaluations yield

$$P^{\rm O}=3.415,\quad P_A^{\rm La}=3.652,\quad P_B^{\rm La}=0.194. \quad (5.52)$$

E. Anisotropy in the spin Hamiltonian

So far, all the calculations of relaxation rates have assumed that the Hamiltonian governing the electronic spins is given by (5.1), which is fully isotropic. However, as noted in Sec. II, in La_2CuO_4 the spin Hamiltonian is not isotropic. More precisely, the DM interaction splits the magnon spectrum into two branches with distinct energy gaps $E_0^{(a,b)}$ at the zone center. These gaps have been measured both directly, by neutron scattering,[9(a)] and indirectly, by determinations of the magnetic field required to induce various spin-flop transitions at low temperature;[9(b)] roughly speaking, the gaps are 10 and 25 K. The generalization of Eq. (5.22), the long-wavelength magnon spectrum, in the presence of anisotropy is

$$\varepsilon_{\mathbf{k}}^{(a,b)}\approx 2\sqrt{2}JS\{(k_\parallel a)^2+2r[1-\cos(k_z a_\perp)]+E_0^{(a,b)}/(2\sqrt{2}JS)\}^{1/2}. \quad (5.53)$$

It should be clear that if the larger of the anisotropy gaps is sufficiently large (i.e., of order or larger than Δ), then there is no temperature regime in which the formulas of Sec. V C above are applicable, since (5.23) is never an accurate approximation for the density of states. What about temperatures small compared to the anisotropy gaps? Magnon populations are then exponentially small in $1/T$, and one expects all relaxation rates to vanish exponentially as well as $T\to 0$. Let us consider explicitly the case of Cu and suppose, for the sake of simplicity, that $E_0^a=E_0^b\equiv E_0\ll\Delta$. Then one has

$$\frac{1}{T_1^{\rm Cu}}\approx\frac{2}{\pi}\frac{(A/\hbar)^2}{(c/a)}\frac{aT}{\hbar c}\left[\frac{T}{\Delta}\right]^2\times\int_{E_0/T}^{\infty}dx\left[x^2-\left[\frac{E_0}{T}\right]^2\right]\frac{e^x}{(e^x-1)^2}. \quad (5.54)$$

Observe that in the limit $E_0\to 0$ the integral here yields $\frac{1}{3}\pi^2$ and the result (5.49) is recovered. However, for $E_0\gg T$ the integral is approximately $2(E_0/T)e^{-E_0/T}$ and thus

$$\frac{1}{T_1^{\rm Cu}}\approx\frac{4}{\pi}\frac{(A/\hbar)^2}{(c/a)}\frac{aE_0}{\hbar c}\left[\frac{T}{\Delta}\right]^2 e^{-E_0/T}. \quad (5.55)$$

VI. CONCLUSIONS

We have presented a rather detailed account of the temperature dependences of nuclear spin-lattice relaxation rates in La_2CuO_4 which should also be largely applicable to the other antiferromagnetic, insulating cuprate perovskites. Experimental investigations of such materials would shed considerable light on the properties of low-energy spin fluctuations, an interesting topic in the area of low-dimensional magnetic systems involving $S=\frac{1}{2}$ spins, small anisotropy in spin space, and small interlayer coupling. The functional forms of the temperature dependences of the relaxation rates for Cu and O in various regimes (disordered phase, ordered phase with two- and three-"dimensional" spin waves) is fully determined by the analysis; in contrast, for La the behavior of the rate depends on the ratio of the hyperfine interaction $A_{\rm La}$ with the nearest Cu spins to the interaction $B_{\rm La}$ with the nearest spin on the next-nearest CuO plane. Even though one expects $A_{\rm La}\gg B_{\rm La}$, the farther-neighbor interaction may play an important role: It allows the La nuclei to couple to critical fluctuations in the disordered phase.

We are not aware of any experimental investigations of the temperature dependence of relaxation rates to which our theory should clearly pertain. Indeed, there is only a single published work describing results on which we can comment at all. Alloul, Ohno, and Mendels[36] studied the relaxation rate for Y in $YBa_2Cu_3O_{6.41}$, which they presumed to be an antiferromagnetic insulator, and found that the rate was proportional to T. The temperature dependence of the Y rate in antiferromagnetic Y-Ba-Cu-O should be the same as that of the La rate in La_2CuO_4 [but with $B_{La}=0$; recall the discussion following Eq. (3.5)]. The latter is in turn proportional to T^3—or to an even higher power of T in the ordered phase—so we must conclude that the observed relaxation cannot be ascribed to short-wavelength spin fluctuations. If correct, this indicates the presence of other relaxation channels besides those present in the Heisenberg model. It is an amusing coincidence that a T^1 law for the Y relaxation rate follows if the spin dynamics are assumed to be that of the classical spherical model,[19] but there is no reason to suspect that such dynamics is appropriate for the real systems. Further experimental investigations are clearly called for.

One result of the calculations that is important to note is that the Cu relaxation rates in the layered cuprate perovskites should be considerably larger than those in cubic cuprate perovskites; for example, in the ordered phase (but above the crossover to anisotropy-dominated or quasi-three-dimensional behavior), the relaxation rates are larger by a factor of order $(J/T)^2(J/J')^{1/2}$.

It is also appropriate to comment briefly on an experimental and theoretical study of the temperature dependence of the sublattice magnetization in La_2CuO_4 by Singh *et al.*[37] Although we have not discussed the sublattice magnetization explicitly in our work, it may be calculated by the spin-wave approach straightforwardly (indeed, that is the theoretical approach of Singh *et al.*), and similar considerations regarding the relevant energy scales apply. For an isotropic spin Hamiltonian, the same energy Δ separates quasi-two-and quasi-three-dimensional behaviors $[(T/J)\ln(T/\Delta)$ and $(T/J)(T/\Delta)$, respectively, in the deviation from the $T=0$ magnetization]. Singh *et al.* did more, and calculated the relevant integrals numerically so that the results could be applied to fit the experimental data over the entire temperature range studied; they claimed to be able to estimate Δ in this way, and their result was consistent with other estimates. However, it appears to us that one cannot be completely confident of their analysis. In particular, the DM anisotropy should also affect the temperature dependence of the sublattice magnetization, and as the experimental data at temperatures comparable to or smaller than the observed crossover temperature is quite sparse (indeed, it appears to consist of a single data point!), it seems impossible to ascertain whether there is ever a crossover to isotropic, quasi-three-dimensional behavior.

We should conclude by noting that it may be difficult to observe any of the low-temperature crossovers in the temperature dependences of the relaxation rates. If the samples contain magnetic impurities or defects such as magnetic two-level systems, these will give rise to extrinsic relaxation, which may overwhelm the intrinsic relaxation channels at low temperatures, where the intrinsic rates are vanishing rapidly as $T\rightarrow 0$.

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APPENDIX: RELAXATION RATES FOR $T > T_N$

The relaxation rates for all the nuclei can be written in terms of the dynamic structure factor $S(\mathbf{q},\omega\approx 0)$ [defined in Eq. (5.30)] in the form

$$\frac{1}{T_1^{(X)}}=\frac{A_X^2}{6N\hbar^2}\sum_{\mathbf{q}} f^{(X)}(\mathbf{q})S(\mathbf{q},\omega\approx 0)\ , \tag{A1}$$

where A_X sets the magnitude of the hyperfine coupling for a nucleus of type X, and the sum is over the full Brillouin zone (BZ) of the square lattice. Recall that the total number of lattice sites is $2N$. The form factors are given by

$$f^{(\mathrm{Cu})}(\mathbf{q})=1\ , \tag{A2}$$

$$f^{(\mathrm{O})}(\mathbf{q})=1+\cos(q_x a)\ , \tag{A3}$$

$$f^{(\mathrm{La})}(\mathbf{q})=1+\cos(q_x a)+\cos(q_y a)+\cos(q_x a)\cos(q_y a)\ . \tag{A4}$$

(We have neglected the B_{La} term in the hyperfine interaction for La with the knowledge that its contribution to $1/T_1^{La}$ has the same form as $1/T_1^{Cu}$.) Although the relaxation rates involve the correlation functions $\langle S_j^+(t)S_0^-(0)\rangle$, they can be written in terms of the rotationally invariant correlation function because of the rotational symmetry of the disordered phase.

The relaxation rate for Cu has been calculated using the structure factor of CHN (Ref. 14) for the square-lattice antiferromagnet. However, that structure factor is valid only for $k\lambda_{th}\ll 1$, with the thermal wavelength $\lambda_{th}\equiv\hbar c/T$ and where $\mathbf{k}\equiv\mathbf{q}-\mathbf{Q}$ is measured with respect to the antiferromagnetic ordering wave vector $\mathbf{Q}\equiv(\pi/a,\pi/a)$. Note that for wave vectors close to $\mathbf{Q}$,

the form factors $f^{(X)}$ for O and La vanish as $(ka)^2$ and $(ka)^4$, respectively. Hence, although the Cu rate can be accurately calculated using the structure factor of CHN, the latter cannot be naively taken over to calculate the rates for O and La, since the form factors kill substantially the weight at $k\approx 0$. On the other hand, for $k\xi \gg 1$ spin-wave calculations should be reliable.

Because we prefer to work in the reduced BZ of the antiferromagnet, we fold the outer part of the full BZ into the reduced BZ and write

$$\frac{1}{T_1^{(X)}}=\frac{A_X^2}{6N\hbar^2}\sum_{\mathbf{k}}[\,f^{(X)}(\mathbf{k})S(\mathbf{k},\omega\approx 0)+f_{\rm st}^{(X)}(\mathbf{k})S_{\rm st}(\mathbf{k},\omega\approx 0)]\,, \tag{A5}$$

where $\mathbf{k}$ runs over the reduced BZ, and we have defined the staggered form factor and structure factor via $f_{\rm st}^{(X)}(\mathbf{k})\equiv f^{(X)}(\mathbf{k}+\mathbf{Q})$ and $S_{\rm st}(\mathbf{k},\omega)\equiv S(\mathbf{k}+\mathbf{Q},\omega)$. When $\mathbf{k}$ is restricted to the reduced BZ, we shall from now on refer to $S(\mathbf{k},\omega)$ as the total-spin structure factor. The staggered form factors for O and La are, explicitly,

$$f_{\rm st}^{(\rm O)}(\mathbf{k})=1-\cos(k_x a)\,, \tag{A6}$$

$$f_{\rm st}^{(\rm La)}(\mathbf{q})=1-\cos(k_x a)-\cos(k_y a)+\cos(k_x a)\cos(k_y a)\,. \tag{A7}$$

At low temperatures, that is, $a/\lambda_{\rm th}\sim T/J\ll 1$, the calculation of the relaxation rate greatly simplifies, because the structure factors depend only on $|k|$ and are exponentially small if $k\lambda_{\rm th}\gg 1$. This justifies the use of small wave-vector expansions for the form factors. Hence we write

$$\frac{1}{T_1^{\rm O}}=\frac{A_{\rm O}^2}{6N\hbar^2}\sum_{\mathbf{k}}[2S(\mathbf{k},0)+\tfrac{1}{2}k_x^2a^2S_{\rm st}(\mathbf{k},0)]\,, \tag{A8}$$

$$\frac{1}{T_1^{\rm La}}=\frac{A_{\rm La}^2}{6N\hbar^2}\sum_{\mathbf{k}}4S(\mathbf{k},0)\,. \tag{A9}$$

The leading-order contribution from $S_{\rm st}$ to the La rate vanishes, because the staggered form factor for La vanishes as k^4. Higher-order momentum dependence will introduce a correction factor of the form $[1+O((a/\lambda_{\rm th})^2)]$. In contrast, the two contributions to the O rate have qualitatively the same properties [see Eqs. (A11) and (A12) below]. The fact that the La rate is less sensitive to the staggered-spin fluctuations than the O rate is clear: the La nuclei are coupled to four spins with equal weight, while O nuclei are only coupled to two spins. Because the average of four spins is a better approximation to the total spin than the average of two spins, the total-spin structure factor is comparatively more important for the La rate than the O rate.

Because $S_{\rm st}(\mathbf{k},0)$ diverges exponentially in $2\pi\rho_s/T$ for $k\approx 0$, it cannot be calculated perturbatively for small momenta. However, the factor k_x^2 in (A8) will reduce the weight of the region $k\approx 0$ in the sum. To estimate the contribution of this long-wavelength regime to $1/T_1^{\rm O}$ quantitatively, we write

$$\sum_{\mathbf{k}}k_x^2S_{\rm st}(\mathbf{k},0)=\sum_{|\mathbf{k}|<k_0}k_x^2S_{\rm st}(\mathbf{k},0)+\sum_{|\mathbf{k}|>k_0}k_x^2S_{\rm st}(\mathbf{k},0) \tag{A10}$$

and choose k_0 so that for $|\mathbf{k}|<k_0$ the expression for the structure factor given by THC (Ref. 15) may be used, and for $|\mathbf{k}|>k_0$ the rotationally invariant structure factor may be calculated by spin-wave theory. A reasonable choice for k_0 is $k_0=\eta/\lambda_{\rm th}$ with $\eta\ll 1$. The fact that k_0 should be chosen proportional to $1/\lambda_{\rm th}$, and not any other power of $\lambda_{\rm th}$, is a consequence of the fact that the Bose occupation factor $n(k\lambda_{\rm th})$ cuts off the momentum integration at $k\approx 1/\lambda_{\rm th}$. Using then the scaling form for $S_{\rm st}$ given by THC,[15] we find that the contribution of the first term in (A10) is proportional to $\eta(T/2\pi\rho_s)^2(a/\lambda_{\rm th})\sim T^4$, and it will become evident that this term is negligible compared to the second term of (A10).

It can be shown[39] that within perturbation theory the total- and staggered-spin structure factors at $\omega=0$ and $\xi^{-1}\ll k\ll a^{-1}$ for $Ta/\hbar c\ll 1$ are of the form

$$S(\mathbf{k},0)=\frac{Ta^2}{\hbar c^2}\frac{T}{\hbar ck}\left[R^{(1)}\left(\frac{T}{\hbar ck}\right)+\frac{T}{2\pi\rho_s}R^{(2)}\left(\frac{T}{\hbar ck}\right)+O(T^2)\right], \tag{A11}$$

$$S_{\rm st}(\mathbf{k},0)=4\left(\frac{1}{ka}\right)^2\frac{Ta^2}{\hbar c^2}\frac{T}{\hbar ck}\left[R_{\rm st}^{(1)}\left(\frac{T}{\hbar ck}\right)+\frac{T}{2\pi\rho_s}R_{\rm st}^{(2)}\left(\frac{T}{\hbar ck}\right)+O(T^2)\right], \tag{A12}$$

where the functions $R^{(j)}(x)$ and $R_{\rm st}^{(j)}(x)$ are exponentially small at large x and have at worst integrable singularities at the origin. The functions $R^{(1)}$ and $R_{\rm st}^{(1)}$ are given explicitly by[39]

$$R^{(1)}(x)=\frac{x^2}{2\pi}\int_0^\infty d\tau\, g_1(\tau)g_2(\tau,x)\,,\qquad R_{\rm st}^{(1)}(x)=\frac{x^2}{2\pi}\int_0^\infty d\tau\frac{g_2(\tau,x)}{g_1(\tau)}\,, \tag{A13}$$

with

$$g_1(\tau)=\sqrt{1+\tau^2}\,,\qquad g_2(\tau,x)=e^{xg_1(\tau)/2}/(e^{xg_1(\tau)/2}-1)^2\,. \tag{A14}$$

It follows that the rates at low temperatures ($Ta/\hbar c\ll 1$) are given by

$$\frac{1}{T_1^{\rm O}}=C_{\rm O}^{(1)}\frac{A_{\rm O}^2a}{3\hbar^2c}\left(\frac{Ta}{\hbar c}\right)^3\left[1+C_{\rm O}^{(2)}\frac{T}{2\pi\rho_s}+O(T^2)\right], \tag{A15}$$

$$\frac{1}{T_1^{\rm La}}=C_{\rm La}^{(1)}\frac{A_{\rm La}^2a}{3\hbar^2c}\left(\frac{Ta}{\hbar c}\right)^3\left[1+C_{\rm La}^{(2)}\frac{T}{2\pi\rho_s}+O(T^2)\right]. \tag{A16}$$

The prefactors $C_O^{(1)}$ and $C_{La}^{(1)}$ can be determined from a one-loop calculation. To this order only the S^zS^z part of the rotationally invariant correlation function survives the $\omega \to 0$ limit. It is easily shown that

$$\int_0^\infty dx\, R^{(1)}(x) = \int_0^\infty dx\, R_{st}^{(1)}(x) = \frac{2\pi^2}{3} , \tag{A17}$$

and it follows that the prefactors in (A15) and (A16) are

$$C_O^{(1)} = \frac{1}{\pi}\int_0^\infty dx \left[R^{(1)}(x) + \frac{1}{2} R_{st}^{(1)}(x) \right] = \pi , \tag{A18}$$

$$C_{La}^{(1)} = \frac{1}{\pi}\int_0^\infty dx\, 2R^{(1)}(x) = \frac{4}{3}\pi . \tag{A19}$$

Let us now describe the calculation of the two-loop coefficients $C_O^{(2)}$ and $C_{La}^{(2)}$. In terms of the functions $R^{(2)}(x)$ and $R_{st}^{(2)}(x)$, which appear in (A11) and (A12), the coefficients are given by

$$C_O^{(2)} = \frac{1}{\pi^2}\int_0^\infty dx \left[R^{(2)}(x) + \frac{1}{2} R_{st}^{(2)}(x) \right] , \tag{A20}$$

$$C_{La}^{(2)} = \frac{3}{2\pi^2}\int_0^\infty dx\, R^{(2)}(x) . \tag{A21}$$

using the Dyson-Maleev formalism,[33,38,39] we have explicitly calculated the two-loop functions $R^{(2)}(x)$ and $R_{st}^{(2)}(x)$. The coefficients $C_O^{(2)}$ and $C_{La}^{(2)}$ can then be written as four-dimensional integrals. At this order both the transverse and longitudinal parts of the correlation functions contain logarithmic divergences, which are only removed when the two parts are added to form the desired rotationally invariant quantity. Motivated by (A15) and (A16), one might imagine using precise magnetic resonance data to determine ρ_s without having to know the strength of the hyperfine interactions—all one needs to know, in principle, are the two-loop coefficients and the ratio of the T^4 to T^3 terms in the relaxation rate. Unfortunately, even the leading T^3 behavior has not yet been seen experimentally, and so it does not seem worthwhile to evaluate the two-loop coefficients numerically.

Nonetheless, let us give here some details of the calculation of $R^{(2)}(x)$, to show how the logarithmic divergences cancel when one forms the rotationally invariant quantity. We separate $R^{(2)}(x)$ into contributions from the transverse and longitudinal parts of the correlation function, that is,

$$R^{(2)}(x) = R_{+-}^{(2)}(x) + R_{zz}^{(2)}(x) . \tag{A22}$$

The explicit expressions for each part, obtained within the Dyson-Maleev formalism, are

$$R_{+-}^{(2)}(x) = \frac{2x^3}{(2\pi)^2}\int d\mathbf{u}\int d\mathbf{v}\, n(xu)n(xv) \left[\left[\frac{u+v}{2uv} \right] \delta(|\hat{\mathbf{x}}-\mathbf{u}-\mathbf{v}|-u-v)[1+n(xu+xv)] + \left[\frac{u-v}{uv} \right] \delta(|\hat{\mathbf{x}}-\mathbf{u}-\mathbf{v}|-u+v)[1+n(xu-xv)] \right] , \tag{A23}$$

$$R_{zz}^{(2)}(x) = -\frac{2x^3}{(2\pi)^2}\int d\mathbf{u}\int d\mathbf{v}\, n(xu)[1+n(xu)]\delta(|\hat{\mathbf{x}}-\mathbf{u}|-u)\frac{1}{v} \left[\left[\frac{|\hat{\mathbf{x}}-\mathbf{u}|+v}{|\hat{\mathbf{x}}-\mathbf{u}|-v} \right] [n(xv)-n(x|\hat{\mathbf{x}}-\mathbf{v}|)] + \left[\frac{|\hat{\mathbf{x}}-\mathbf{u}|-v}{|\hat{\mathbf{x}}-\mathbf{u}|+v} \right] [n(xv)+n(x|\hat{\mathbf{x}}-\mathbf{v}|)+1] \right] . \tag{A24}$$

Here $\mathbf{u}$ and $\mathbf{v}$ are two-dimensional vectors, and $\hat{\mathbf{x}}$ is an arbitrary fixed unit vector. (The integrals are independent of the choice of $\hat{\mathbf{x}}$.) Both integrals are logarithmically divergent. To isolate the singular terms in the integral of $R^{(2)}$, let us first define

$$I_{+-}(\delta) = \int_0^\infty dx\, \tilde{R}_{+-}^{(2)}(x) , \tag{A25}$$

$$I_{zz}(\delta) = \int_0^\infty dx\, \tilde{R}_{zz}^{(2)}(x) , \tag{A26}$$

where the functions $\tilde{R}^{(2)}$ are obtained from the functions $R^{(2)}$ by removing the logarithmic divergences via the introduction of a cutoff $\delta \ll 1$ in the relevant lower limits of integration. It turns out that the integral $I_{+-}(\delta)$ can then be done (almost) exactly. We obtain

$$I_{+-}(\delta) = \frac{4\pi^2}{3}\ln\left[\frac{1}{\delta}\right] + 2(K_1 - 2K_2) + O(\delta) , \tag{A27}$$

with

$$K_1 = \int_0^\infty dx \int_0^\infty dy \frac{xye^{x+y}}{(e^x-1)(e^y-1)(e^{x+y}-1)} \approx 3.60 , \tag{A28}$$

and

$$K_2 = -\int_0^\infty dx \frac{x^2 e^x}{(e^x-1)^2} \ln(1-e^x) \approx 1.51 \ . \tag{A29}$$

The integral $I_{zz}(\delta)$ can be written as

$$I_{zz}(\delta) = -\frac{4\pi^2}{3} \ln\left[\frac{1}{\delta}\right] - 2[K_3 + K_4 + 3\zeta(3)] + O(\delta) \ . \tag{A30}$$

Here ζ is the Riemann ζ function, and K_3 and K_4 are given by the following integrals (in which the auxiliary vector $\mathbf{r}$ is defined as $u\hat{\mathbf{x}} - \mathbf{u}$):

$$K_3 = \frac{1}{(2\pi)^2} \int d\mathbf{u} \int_{|\mathbf{v}|<1} d\mathbf{v} \frac{u}{v} n(u)[1+n(u)] \left[\left[\frac{|\mathbf{v}-\mathbf{r}|+v}{|\mathbf{v}-\mathbf{r}|-v}\right] \left[n(v) - n(|\mathbf{v}-\mathbf{r}|) + \frac{1}{v} - \frac{1}{|\mathbf{v}-\mathbf{r}|} \right] + \left[\frac{|\mathbf{v}-\mathbf{r}|-v}{|\mathbf{v}-\mathbf{r}|+v}\right] \left[n(v) + n(|\mathbf{v}-\mathbf{r}|) - \frac{1}{v} - \frac{1}{|\mathbf{v}-\mathbf{r}|} \right] \right] , \tag{A31}$$

$$K_4 = \frac{1}{(2\pi)^2} \int d\mathbf{u} \int_{|v|>1} d\mathbf{v} \frac{u}{v} n(u)[1+n(u)] \left[\left[\frac{|\mathbf{v}-\mathbf{r}|+v}{|\mathbf{v}-\mathbf{r}|-v}\right] [n(v) - n(|\mathbf{v}-\mathbf{r}|)] + \left[\frac{|\mathbf{v}-\mathbf{r}|-v}{|\mathbf{v}-\mathbf{r}|+v}\right] [n(v) + n(|\mathbf{v}-\mathbf{r}|)] \right] . \tag{A32}$$

Note that $n(x) - x^{-1} \sim -\frac{1}{2} + x/12 + O(x^2)$ for small x, and hence the above integrals are finite and of order unity. Combining these results, we obtain

$$\begin{aligned} \int_0^\infty dx \, R^{(2)}(x) &= \lim_{\delta \to 0} \left[\frac{4\pi^2}{3} \ln\left[\frac{1}{\delta}\right] + 2(K_1 - 2K_2) - \frac{4\pi^2}{3} \ln\left[\frac{1}{\delta}\right] - 2[K_3 + K_4 + 3\zeta(3)] + O(\delta) \right] \\ &= 2[K_1 - 2K_2 - K_3 - K_4 - 3\zeta(3)] \ . \end{aligned} \tag{A33}$$

The coefficient $C_{\mathrm{La}}^{(2)}$ is then obtained from Eq. (A21).

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