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Spin-wave theory for anisotropic Heisenberg antiferromagnets

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The anisotropic Heisenberg antiferromagnet (AF), which is defined as a three-dimensional simple-cubic lattice with in-plane antiferromagnetic interaction $J_\parallel$ and interplane coupling $J_\perp=\gamma J_\parallel$ and is believed to describe the magnetic properties of the cupric oxide materials, is studied using low-temperature spin-wave theory. The dependence of the $T=0$ staggered magnetization, ground-state energy, transverse susceptibility, spin-wave velocity, and the Néel temperature T_N on the anisotropy parameter γ ($0\leq\gamma\leq 1$) are obtained. These results are found to be in satisfactory agreement with existing experiments on cupric oxide materials. The apparent difference between the muon-spin resonance and neutron-scattering results for the ordered moment in the AF state is well explained.

The discovery of the copper-oxide superconductors[1] has produced great interest in the Heisenberg antiferromagnetic (AF) model. This is because the antiferromagnetic order of Cu^{2+} spins has been observed[2] in La_2CuO_4 and $YBa_2Cu_3O_6$, which become superconducting when doped. While it has been proposed that magnetism may play an important role in high-temperature superconductivity, the study of magnetic ordering in these families of materials is interesting in its own right. These systems are highly anisotropic with a very strong coupling between Cu spins in the plane and very weak coupling between planes. Thus, these systems have interesting magnetic properties which may be representative of a wider class of quasi-two-dimensional antiferromagnets. For this reason, we decided to carry out a detailed study of the magnetism of a classical Heisenberg model with interactions in the range appropriate to cupric oxides.

It is now well established from numerical work (quantum Monte Carlo,[3] Green-function Monte Carlo,[4] series analysis[5]) and different variational methods[4] that the ground state of the two-dimensional (2D) $S=\frac{1}{2}$ Heisenberg AF does indeed have long-range Néel order. All of its ground-state properties as well as the long-wavelength excitations are well described by straightforward spin-wave theory.[6,7] Spin-wave theory is based upon a large-S expansion, starting from a ground state with Néel order. Why spin-wave theory is so accurate even for $S=\frac{1}{2}$ is still an open and interesting question. Due to these successes, we decided to use the regular spin-wave theory to study the ground-state properties of the anisotropic Heisenberg antiferromagnet on a simple cubic lattice. In order to investigate the dependence of the Néel temperature, T_N, on the anisotropy parameter γ we generalized the Green's function formalism[8-10] with its random-phase decoupling to the anisotropic Heisenberg antiferromagnet. This random-phase approximation gives a better estimate of T_N compared with Monte Carlo-simulation results[11] than mean-field-theory and low-T spin-wave theory results. One of the most interesting results of this study is that it is possible to have large changes in T_N as the anisotropy γ changes, while the staggered magnetization, i.e., the ordered moment of the AF state, remains constant. This is consistent with muon-spin rotation experiments[12] on La_2CuO_{4-y} and not with the neutron-scattering experiments.[13] Details of the calculation and discussion of the results will be presented in the remainder of the paper.

We have studied the Heisenberg antiferromagnetic model on a simple cubic lattice with in-plane antiferromagnetic interaction $J_\parallel$ and interplane antiferromagnetic interaction $J_\perp$, which is much weaker than $J_\parallel$. The Hamiltonian for this model is

$$H=-J_\parallel\sum_{i,j}\mathbf{S}_i\cdot\mathbf{S}_j-J_\perp\sum_{i,j}\mathbf{S}_i\cdot\mathbf{S}_j\,, \tag{1}$$

where the first sum is over four nearest neighbors in the plane and the second sum is over the nearest-neighbor interplane interactions. The ratio of the two interactions, $\gamma=J_\perp/J_\parallel$, is inversely proportional to the strength of the anisotropy of the system and should determine the transition temperature T_N of the model. When $\gamma=0$, we have a 2D Heisenberg AF with no interplane interactions and $T_N=0$. When $\gamma=1$, we recover a simple cubic Heisenberg AF with equal interactions for which $T_N=1.35$ for classical spins[11] S_i with magnitude equal to one. We will use the Boltzmann constant $k_B=1$ and $|J_\parallel|=1$.

To obtain, within the spin-wave theory, the ground-state properties, as well as the spin-wave excitation energy, for the anisotropic Heisenberg AF model given by Eq. (1), we follow the standard two-sublattice approximation.[6,7] Using the Holstein-Primakoff transformation in the two sublattices, the Fourier transform of Eq. (1) can be easily diagonalized. Following the notation in Callaway's book,[7] we have the following results. The ground-state energy is given by

$$E_0=-\tfrac{1}{2}N|J_\parallel|zS^2(1+\lambda/S)\,. \tag{2}$$

The staggered magnetization is

$$\langle S_0^z\rangle=(S-\lambda'/S)\,, \tag{3}$$

the perpendicular susceptibility at $T=0$ is

$$x_\perp(0)=\frac{Ng^2\mu_B^2}{2z|J_\parallel|}[1-(1/2S)(\lambda+\lambda')]\,, \tag{4}$$

and the spin-wave excitation energy is

$$\varepsilon(k)=\sqrt{2z}\,|J_\parallel|S(1+\lambda/2S)ka\,, \tag{5}$$

where

$$\lambda = 1 - \frac{2}{N}\sum_k (1-\gamma_k^2)^{1/2} a ,$$
$$\lambda' = \frac{2}{N}\sum_k \frac{1}{(1-\gamma_k^2)^{1/2}} - 1b , \quad (6)$$
$$\gamma_k = (2\cos k_x a + 2\cos k_y a + 2\gamma \cos k_z a)/zc$$

and $z = 4+2\gamma$. The values of λ and λ' have been calculated numerically and our results for $\gamma = 0$ (square lattice) and $\gamma = 1$ (simple cubic lattice) agree with those[6] of Anderson and Kubo.

To understand the dependence of the Néel temperature T_N on the anisotropy parameter γ, one must develop a theory that will give $T_N = 0$, when the interplane coupling $J_\perp = 0$, i.e., when the model given by Eq. (1) becomes a 2D Heisenberg antiferromagnet. The simplest of the mean-field approximations is that in which the field spin feels is just determined from the sum of its interactions with its neighbors. For the classical Heisenberg model with spins of unit magnitude, $T_N = \frac{1}{3}(4|J_\parallel| + 2|J_\perp|) = \frac{1}{3}|J_\parallel|(4+2\gamma)$, which obviously is wrong because for $\gamma = 0$, it gives a finite T_N. This simple mean-field approach is not good, since it does not correctly take into account the strong fluctuations which destroy long-range order in the limit of $J_\perp \to 0$. We have, therefore, calculated the Néel temperature T_N from the theory of ferromagnetism which is based on the Green function method.[8-10] Bogolyubov and Tyablikov[8] were the first to apply the Green function method to ferromagnetism for the spin-$\frac{1}{2}$ case and they have evoked the random-phase decoupling scheme to reduce the Green functions to the lowest order.[9] We have followed[9,10] the random-phase decoupling method to calculate the T_N for the general spin S for the anisotropic Heisenberg model. Within this approximation, the result for T_N can be written as

$$T_N = \frac{|J_\parallel|}{3G(0)} , \quad (7)$$

where $G(0) = \sum_k 1/z(1-\gamma_k)$, $z = 4+2\gamma$, and γ_k is given by Eq. (6). $G(0)$ is the Green function[14] of the periodic anisotropic tight-binding system at the bottom of the band. $G(0)$ can be calculated analytically for $\gamma = 0$ (square lattice) and numerically for all the other values of the anisotropy γ. A simpler expression for $G(0)$ can be obtained[14] by integrating over the two variables k_x and k_y and yielding $G(0) = (1/2\pi^2)\int_0^\pi dk_z\, tK(t)$, where $t = 2/(2+\gamma-\gamma\cos k_z a)$ and K is the complete elliptic integral of the first kind. In the limit of small anisotropy parameter $\gamma \to 0$, we obtain $G(0) \simeq (1/4\pi)\ln(32/\gamma)$, which gives an analytic expression for the dependence of T_N versus γ. In this weak limit, we have

$$T_N \simeq \frac{4\pi}{3}|J_\parallel| / \ln\left(\frac{32}{\gamma}\right) . \quad (8)$$

Similar results were obtained[15] for the dependence of the mobility edge on anisotropy parameter for a disordered anisotropic tight-binding model. Equation (8) can also be written as $(\gamma/32)\exp(4\pi|J_\parallel|/3T_N) = 1$, which is similar to a renormalized mean-field theory[11] that treats the spin correlations in the planes exactly, and the interplane interactions within the mean-field approximation. The relation between T_N vs γ is very important and in Fig. 1 we compare the random-phase approximation results [Eq. (7)] for T_N with the Monte Carlo simulations results on the classical anisotropic Heisenberg system. Notice that the agreement is extremely good for all the anisotropies γ. T_N increases very rapidly as γ increases for small γ, but for $\gamma > 0.1$, T_N is nearly linear with γ. In particular, the small γ dependence of T_N is described very well by the expression given in Eq. (8). However, our Monte Carlo techniques cannot handle very small γ, since at these smaller interplane interactions the magnetic correlation length is much larger than the sample size [most of the simulation runs[11] were on sizes $N = 18000$ $(30\times30\times20)$]. This is a very interesting result. It shows that the transition from a paramagnetic $T_N = 0$ for $\gamma = 0$ to an antiferromagnetic $T_N \neq 0$ takes place abruptly. Notice that even for very small anisotropic coupling, $\gamma \simeq 0.001$, $T_N \simeq 0.404$, of the same order of magnitude as the $\gamma = 1$ value of T_N, which is roughly equal to 1.36. This is simply due to the fact that the two-dimensional character ($T_N = 0$) is very unstable, being approached (as $\gamma \to 0$) very slowly. In other words, even a very small out-of-plane coupling is enough to destroy the two-dimensional character of the transition. In Fig. 1, we also plot the results for T_N versus γ which we have obtained[11] using the modified spin-wave theory of Takahashi.[16] While the agreement with the Monte Carlo results is good close to $\gamma = 1$, for all the other cases the spin wave T_N are always lower than our Monte Carlo simulations. In Takahashi's approach, T_N is like a Bose-Einstein condensation temperature below which long-range order results. The random-phase approxima-

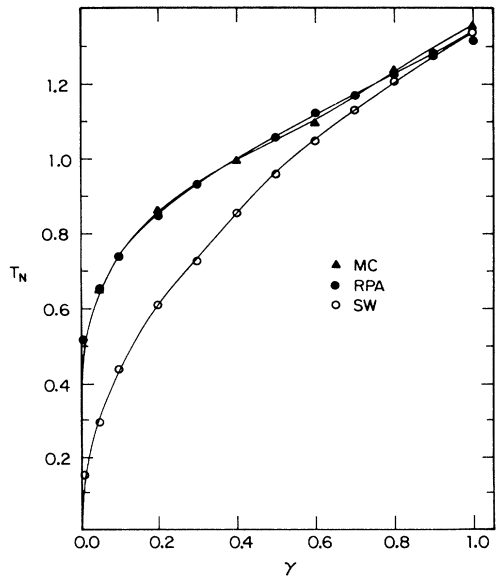

FIG. 1. Néel temperature T_N as a function of the anisotropy parameter $\gamma = J_\perp/J_\parallel$. Results from the Monte Carlo (MC) simulations (solid triangles), from the random-phase approximation (RPA) (solid circles), and the low-temperature spin-wave theory (open circles) are presented.

tion[8-10] scheme to reduce the Green's functions to the lowest order takes into account the hierarchy of coupled equations. As expected, the naive mean-field theory results (not shown in Fig. 1) overestimate T_N for all the values of γ (e.g., $T_N=1.33$ for $\gamma=0$ and $T_N=2.00$ for $\gamma=1$). We have also compared[11] the results of the renormalized mean-field theory with the Monte Carlo simulations, not shown in Fig. 1 and have found[11] that T_N obtained within this renormalized mean-field theory is much higher than the Monte Carlo[11] for $\gamma \geq 10^{-3}$.

In Fig. 2, we plot the spin-wave results for the staggered magnetization $\langle S_0^z \rangle$ and the $T=0$ perpendicular susceptibility $x_\perp(0)$ versus the Néel temperature T_N for the case of $S=\frac{1}{2}$, in order to be able to compare our results with experiments on the cupric oxide materials. Notice that the ordered moment $\langle S_0^z \rangle$ in the AF state remains the same, close to its $\gamma=0$ value of 0.303, as T_N increases from its zero value, all the way to $T_N \simeq 0.3$. As T_N increases further, the system is more three-dimensional and its $\langle S_0^z \rangle$ increases almost linearly from $T_N \simeq 0.4$ to $T_N \simeq 0.9$ and then $\langle S_0^z \rangle$ saturates to its three-dimensional value of 0.4216. The behavior of $x_\perp(0)$ vs T_N follows that of $\langle S_0^z \rangle$. It has been known[17] that the magnetic properties of the AF compound La_2CuO_{4-y} depend sensitively on small differences in the oxygen content $y=0$ to 0.03. The input of O^{2-}, equivalent to the substitution of Sr^{2+} for La^{3+}, removes electrons and creates holes in the system, and therefore, suppresses the three-dimensional AF ordering. Neutron scattering experiments[13] on La_2CuO_{4-y} have revealed that the $T=0$ ordering moment of the antiferromagnet decreases by as much as a factor of 3 as the Néel temperature T_N decreases. However, muon-spin rotation experiments[12] on the same samples show that the ordered Cu moment remains unchanged close to $\simeq 0.6$ μ_B/Cu despite the large difference in the ordering temperature T_N (from 300 to 15 K). A similar behavior has been observed in $YBa_2Cu_3O_{6+x}$ (Ref. 18) and $Sr_2CuO_2Cl_2$ (Ref. 19) samples. In this respect, muon-spin rotation and neutron measurements appear inconsistent. Our theoretical results from Fig. 2 show that the $T=0$ ordered moment ($g\mu_B\langle S_0^z \rangle$) remains constant, closed to 0.60 μ_B/Cu, while T_N can change a lot. These results are consistent with the muon-spin rotation experiments. Within our spin-wave theory, the introduction of oxygen defects does not change the $T=0$ moment but changes mostly $J_\parallel$ and possibly $J_\perp$ which then drastically changes T_N, since T_N is a very sensitive function of the anisotropy γ (see Fig. 1). To reconcile with the neutron-scattering results, one has to argue that either the long-range order is not over the whole sample, and that is the reason for the reduction of the $T=0$ moment, or that the magnetic ordering becomes more and more short ranged with increasing oxygen content (i.e., with increasing number of holes) resulting[12] in the decrease of T_N. Alternatively, if the $T=0$ magnetic moment of 0.34 μ_B/Cu seen in the neutron-scattering experiments for low values of T_N is uniform throughout the volume of the crystal, then its reduction to such low values, much lower than the spin-wave value, is due to as yet an unidentified mechanism. From Fig. 2, we have that $x_\perp(0)$ remains constant equal to its two-dimensional value of $0.449\, Ng^2\mu_B^2/2z|J_\parallel|$, while T_N drastically changes. Some preliminary results[13] support this picture, i.e., that T_N can change drastically while $x_\perp(0)$ remains constant. However, more careful experiments on single crystals have to measure $x_\perp(T)$, $\langle S_0^z \rangle$, the spin-wave excitation energy, and T_N to check the predictions of our anisotropic spin-wave theory. In Fig. 3, we plot the ground-state energy E_0 and the spin-wave velocity v_s versus the anisotropy γ for the $S=\frac{1}{2}$ case. We have normalized E_0 by the Néel energy $E_{\text{Neel}}=-N|J_\parallel|zS^2/2$ and the v_s by $\sqrt{2z}\,|J_\parallel|Sa$. Notice that the behavior of E_0

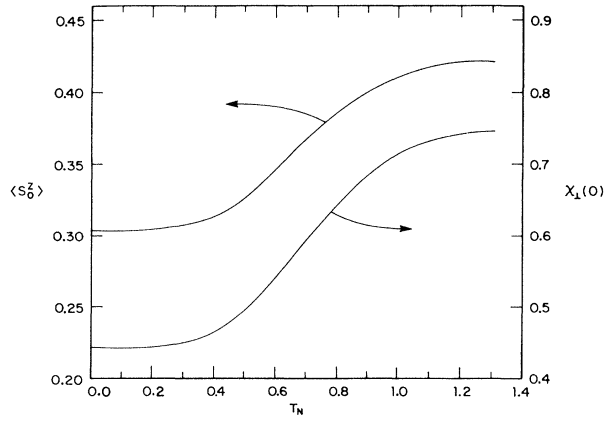

FIG. 2. The $T=0$ staggered spin $\langle S_0^z \rangle$ and perpendicular magnetic susceptibility $x_\perp(0)$ vs the Néel temperature T_N for an anisotropic Heisenberg antiferromagnet with $S=\frac{1}{2}$. The $x_\perp(0)$ is normalized by $Ng^2\mu_B^2/2z|J_\parallel|$.

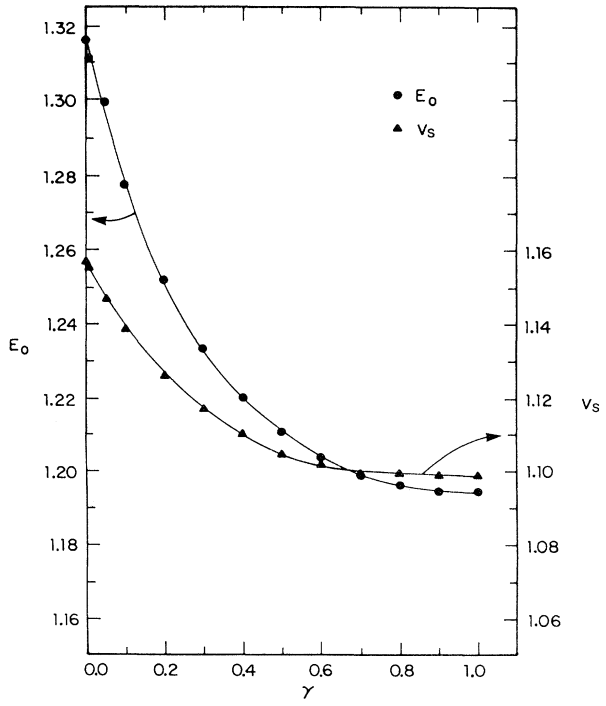

FIG. 3. The ground-state energy E_0 and the spin-wave velocity v_s vs the anisotropy parameter γ. E_0 is normalized by $E_{\text{Neel}}=-\frac{1}{2}Nz|J_\parallel|S^2$ and v_s by $\sqrt{2z}\,|J_\parallel|Sa$.

and v_s vs γ is quite similar. For $\gamma=0$, we have the 2D results of $E_0=+1.3159\ E_{\text{Neel}}$ and $v_s=1.158\sqrt{2}|J_\parallel|a$. As γ increases, E_0 and v_s decreases and monotically reach their 3D results for the simple cubic lattice with $\gamma=1$.

Within the anisotropic Heisenberg AF model, the only parameter that enters the theory is the anisotropy parameter γ. For La_2CuO_4, we have that $T_N/J_\parallel\simeq 300/1000 \simeq 0.300$, which gives a value of $\gamma\simeq 3\times 10^{-5}$. The $T=0$ staggered moment is $\langle S_0^z\rangle=0.304$, while $x_\perp(0)=0.45\ Ng^2\mu_B^2/2z|J_\parallel|=2.11\times 10^{-5}$ cm^3/mole using $g=2$ and the spin-wave velocity $\hbar v_s=d\varepsilon(k)/dk=1.158\sqrt{2}|J_\parallel|a\simeq 0.54$ eV Å using $a=3.8$ Å and $J_\parallel=1000$ K. For $YBa_2Cu_3O_6$, we have that $T_N/J_\parallel\simeq 450/1200\simeq 0.375$, which gives that $\gamma\simeq 4\times 10^{-4}$, and therefore, $\langle S_0^z\rangle=0.31$, $x_\perp(0)=1.80\times 10^{-5}$ cm^3/mole, and $\hbar v_s=0.65$ eV Å using $a=3.85$ Å. For $Sr_2CuO_2Cl_2$, we have that $T_N/J_\parallel\simeq 250/900\simeq 0.278$, which gives $\gamma\simeq 1\times 10^{-5}$, and therefore, $\langle S_0^z\rangle\simeq 0.303$, while $x_\perp(0)\simeq 2.35\times 10^{-5}$ cm^3/mole, and $\hbar v_s=0.50$ Å using $a=3.8$ Å and $J_\parallel=900$ K. Notice that the most anisotropic system is the $Sr_2CuO_2Cl_2$ since the distance between the CuO_2 planes is the largest, while the coupling between the planes in $YBa_2Cu_3O_6$ is stronger than in La_2CuO_4. The above predictions for $x_\perp(0)$ and $\hbar v_s$ are in qualitative agreement with the experiment.[17–19] However, more experiments are needed to check the predictions of this theory.

In conclusion, we have calculated within the traditional spin-wave theory the ground-state properties of the anisotropic Heisenberg antiferromagnet on a simple cubic lattice, this model is believed to describe the magnetic properties of the cupric oxide materials. We have also calculated the Néel temperature T_N versus the anisotropy parameter γ. While the dependence of the $T=0$ ordered moment versus T_N is in agreement with muon experiments, more detailed experiments on single crystals have to measure $x_\perp(T)$, $\langle S_0^z\rangle$, v_s, and T_N in order to be able to check the predictions of the anisotropic spin-wave theory.

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[1] J. G. Bednorz and K. A. Müller, Z. Phys. B **64**, 188 (1986); C. W. Chu *et al.*, Phys. Rev. Lett. **58**, 405 (1987).

[2] D. Vaknin *et al.*, Phys. Rev. Lett. **58**, 2802 (1987); G. Shirane *et al.*, *ibid.* **59**, 1613 (1987).

[3] J. D. Reger, J. A. Riera, and A. P. Young, J. Phys. Condens. Matter **1**, 1855 (1989).

[4] N. Trivedi and D. M. Ceperly, Phys. Rev. B **41**, 4552 (1990), and references therein.

[5] R. R. P. Singh, Phys. Rev. B **39**, 9760 (1990); **41**, 4873 (1990).

[6] P. W. Anderson, Phys. Rev. **86**, 694 (1952); R. Kubo, *ibid.* **87**, 568 (1952); T. Oguchi, *ibid.* **117**, 117 (1960).

[7] J. Callaway, *Quantum Theory of the Solid State* (Academic, New York, 1976), Chap. 2.

[8] N. N. Bogolyubov and S. V. Tyablikov, Dokl. Akad. Nauk. SSSR **126**, 53 (1957) [Sov. Phys. Dokl. **4**, 604 (1959)].

[9] A. S. Chakravarty, *Introduction to the Magnetic Properties of Solids* (Wiley, New York, 1980), Chaps. 17 and 18.

[10] R. A. Tahir-Kheli and D. ter Haar, Phys. Rev. **127**, 88 (1962).

[11] W. Xue, G. S. Grest, M. H. Cohen, S. K. Sinha, and C. M. Soukoulis, Phys. Rev. B **38**, 6868 (1988).

[12] Y. J. Uemura *et al.*, Physica C **153–155**, 769 (1988).

[13] K. Yamada *et al.*, Solid State Commun. **64**, 753 (1987).

[14] E. N. Economou, *Green's Functions in Quantum Physics,* 2nd ed. (Springer, Heidelberg, 1983), Chap. 5.

[15] Qiming Li, C. M. Soukoulis, E. N. Economou, and G. S. Grest, Phys. Rev. B **40**, 2825 (1989).

[16] M. Takahashi, Phys. Rev. B **36**, 3791 (1987); **40**, 2494 (1989).

[17] D. C. Johnston *et al.*, Phys. Rev. B **36**, 4007 (1987).

[18] J. Rossat-Mignod *et al.*, J. Phys. (Paris) Colloq. **49** (Suppl. 12), C8-2119 (1988), and references therein.

[19] D. Vaknin, S. K. Sinha, C. Stassis, L. L. Miller, and D. C. Johnston, Phys. Rev. B **41**, 1926 (1990).