# $J_1$-$J_2$ model revisited: Phenomenology of $CuGeO_3$

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We present a mean-field solution of the antiferromagnetic Heisenberg chain with nearest ($J_1$) and next-to-nearest neighbor ($J_2$) interactions. This solution provides a way to estimate the effects of frustration. We calculate the temperature-dependent spin-wave velocity, $v_s(T)$ and discuss the possibility to determine the magnitude of frustration $J_2/J_1$ present in quasi-one-dimensional compounds from measurements of $v_s(T)$. We compute the thermodynamic susceptibility at finite temperatures and compare it with the observed susceptibility of the spin-Peierls compound $CuGeO_3$. We also use the method to study the two-magnon Raman continuum observed in $CuGeO_3$ above the spin-Peierls transition. [S0163-1829(97)01009-6]

## I. INTRODUCTION

The discovery of the spin-Peierls (SP) transition in the inorganic material $CuGeO_3$ (Ref. 1) has led to an intense investigation of the magnetic properties of this system. It is becoming increasingly clear that this material may not be a prototype SP system such as the quasi-one-dimensional organic material TTFCuBDT.[2] For example, the temperature dependence of the SP gap[3] is unlike what is expected for conventional SP systems.[2] A recent inelastic-neutron-scattering experiment[4] reports observing a spin gap at temperatures above the SP transition temperature $T_{SP}$. So far, there has been no evidence for the presence of a phonon soft mode.[5] X-ray-scattering studies of the incommensurate phase of $CuGeO_3$ show the existence of a soliton lattice with the width of the soliton being much larger than that predicted by calculations.[6] These results, taken together, call for a better understanding of the homogeneous state of $CuGeO_3$ above $T_{SP}$ which should eventually shed light on the nature of the SP transition itself.

The basic structure of $CuGeO_3$ consists of edge-sharing $CuO_6$ octahedra forming $CuO_4$ chains along the crystallographic $\hat{c}$ axis. The dimerization of the $S=\frac{1}{2}$Cu ions below 14 K has been determined by neutron-diffraction measurements.[7] From inelastic-neutron-scattering (INS) measurements, Nishi *et al.*[8] estimated the intrachain and interchain exchange parameters $J_c \approx 120$ K, $J_b \approx 0.1 J_c$, suggesting that interchain effects may not be negligible in this compound. On the other hand, INS measurements above $T_{SP}$[9] clearly show the two-spinon continuum which is characteristic of a one-dimensional $S=\frac{1}{2}$ antiferromagnetic chain.[10] The susceptibility of $CuGeO_3$ above $T_{SP}$[1] shows a broad maximum as expected for Heisenberg chains. From the temperature at which this maximum is observed (56 K), one can estimate the value of $J_c$ using the results of Bonner and Fisher,[11] yielding $J_c = 88$ K. This discrepancy between the value of $J_c$ estimated from static susceptibility measurements and INS led to the proposal,[12,13] that a minimal model to describe the magnetic properties of this system above $T_{SP}$ is the so-called ''$J_1$-$J_2$'' model. This model describes an antiferromagnetic Heisenberg chain with nearest neighbor (NN) and next-to-nearest neighbor (NNN) exchange interactions. The model Hamiltonian is written as

$$H = J \sum_i (\mathbf{S}_i \cdot \mathbf{S}_{i+1} + \alpha \mathbf{S}_i \cdot \mathbf{S}_{i+2}), \tag{1}$$

where $J \equiv J_c$ is the intrachain superexchange between neighboring Cu ions along the $\hat{c}$ direction. The second term in the Hamiltonian (1) is the exchange interaction between next to nearest neighbor Cu ions. In $CuGeO_3$, the NNN superexchange path is through Cu-O-O-Cu and is identical to that in the cuprate superconductors. A detailed analysis of the structure of $CuGeO_3$ and its relation to the magnetic interaction can be found in Refs. 14 and 15.

The model Hamiltonian (1) has been studied by several authors.[16] Though the Hamiltonian is not exactly solvable for all values of $\alpha$, the phase diagram is well understood qualitatively. For $0 < \alpha < \alpha_{cr}$, the ground state remains gapless (as is the case when $\alpha = 0$). The effect of $\alpha$ is to renormalize the spin wave velocity in this regime. The value of $\alpha_{cr}$ has been estimated to be 0.2411.[17] When $\alpha > \alpha_{cr}$, the spectrum becomes gapped and for $\alpha = \frac{1}{2}$, the Hamiltonian (1) is exactly solvable.[18] As mentioned earlier, there has been a renewed interest in this model in the context of $CuGeO_3$. Riera and Dobry[12] as well as Castilla, Chakravarty, and Emery[13] computed the thermodynamic susceptibility of the Hamiltonian (1) numerically and compared it with the experimental values. Both groups found that the presence of a nonvanishing $\alpha$ is needed to provide a consistent description of both the INS and susceptibility results.

If indeed the $J_1$-$J_2$ model is an appropriate starting point to embark on a study of $CuGeO_3$, it would be desirable to have some way of calculating physical quantities, especially considering the wealth of experimental data now available on $CuGeO_3$. This is the primary objective of this study where we present some results obtained from a solitonic mean-field theory of the $J_1$-$J_2$ model. This method, which is meaningful only for $\alpha<\alpha_{cr}$, provides a simple self-consistent way of evaluating the effects of frustration. We use this solution to obtain the spinon dispersion relation as a function of $\alpha$. We also calculate the ground-state energy and bulk susceptibility at finite temperatures as a function of $\alpha$. The results for the bulk susceptibility are compared with the experimentally observed values in $CuGeO_3$. We then examine the Raman continuum seen experimentally above $T_{SP}$ and show how this arises as a natural consequence of competing magnetic interactions. We compute the Raman intensity using the mean-field solution and compare with experimental results. The paper is organized as follows. In Sec. II, we present the mean-field solution of the Hamiltonian (1) and use it to compute the static susceptibility. In Sec. III, we compute the two-magnon Raman scattering intensity arising from competing magnetic interactions. Section IV contains a brief summary of our results.

## II. A MEAN-FIELD SOLUTION OF THE $J_1$-$J_2$ MODEL

In this section, we propose a mean-field solution of the Hamiltonian (1). This solution is based on a mapping introduced by Gómez Santos[19] between the spin-$\frac{1}{2}$ Heisenberg chain with NN interactions and a Hamiltonian describing the dynamics of antiferromagnetic domain walls. It has also been used by Weng and collaborators[20] for the one-dimensional $t$-$J$ model. We show below how the mapping can be generalized to the spin Hamiltonian (1) with NNN interactions as well. In this mapping, the local degrees of freedom are given by the nature of the bond (ferromagnetic or antiferromagnetic) between two interacting spins. We work using periodic boundary conditions and choose the following convention: Néel ordering is characterized by an ‘‘up ’’ spin at the first site. Since, by definition, the Néel ordered state does not have a ‘‘kink’’ (henceforth, we shall use the words ‘‘kink,’’ ‘‘soliton,’’ and ‘‘domain wall’’ interchangeably), this state is the vacuum state $|0\rangle$ of the solitons, which we write symbolically as

$$|0\rangle\equiv|\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow\cdots\rangle.$$

A state with a kink between sites $i$ and $i+1$ is defined to be a one-soliton state $d_i^\dagger|0\rangle$. For example, $d_4^\dagger|0\rangle$ defines the spin configuration

$$d_4^\dagger|0\rangle\equiv|\downarrow\uparrow\downarrow\uparrow\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow\uparrow\downarrow\cdots\rangle.$$

With these definitions, it is easy to verify that the NN term in Eq. (1) is mapped into

$$\frac{J}{2}\sum_i\left\{(d_{i-1}^\dagger d_{i+1}+d_{i-1}^\dagger d_{i+1}^\dagger+\text{H.c.})(1-d_i^\dagger d_i)+\left(d_i^\dagger d_i-\frac{1}{2}\right)\right\}.$$

Here, we have avoided the sign problem by assuming the solitons to be hard-core bosons. On performing the Jordan-Wigner transformation

$$d_i\rightarrow\exp\left(i\pi\sum_{m<i}d_m^\dagger d_m\right)d_i,$$

one sees that the NN term written above preserves its form. By looking at the action of the NNN term in Eq. (1) on a pair of interacting spins (or equivalently, a given bond), one can write down the NNN term in terms of the soliton operators in the same manner as above. Doing this and performing a Jordan-Wigner transformation, we obtain the $J_1$-$J_2$ model in terms of fermionic soliton operators as

$$\begin{aligned}H=&\frac{J}{2}\sum_i\left\{(d_{i+1}^\dagger+d_{i+1})(1-d_i^\dagger d_i)(d_{i-1}^\dagger-d_{i-1})+\left(d_i^\dagger d_i-\frac{1}{2}\right)\right\}\\&+\frac{\alpha J}{2}\sum_i(d_{i+2}^\dagger+d_{i+2})(d_{i+1}^\dagger d_i+d_i^\dagger d_{i+1})(d_{i-1}^\dagger-d_{i-1})\\&+\frac{\alpha J}{4}\sum_i(2d_i^\dagger d_i-1)(2d_{i+1}^\dagger d_{i+1}-1).\end{aligned}\quad(2)$$

The above mapping may also be verified by using the definition of the original spin operators in terms of the solitonic operators, viz.,

$$S_i^+=\frac{1}{2}(d_{i-1}^\dagger-d_{i-1})(d_i^\dagger+d_i)(1-2S_i^z),$$

$$S_i^-=\frac{1}{2}(d_{i-1}^\dagger-d_{i-1})(d_i^\dagger+d_i)(1+2S_i^z),$$

$$S_i^z=\frac{1}{2}(-)^{i+1}\exp\left(i\pi\sum_{j<i}d_j^\dagger d_j\right).$$

Substituting the above expressions for the spin operators in Eq. (1), we recover Eq. (2). It should be noted that the maximum number of fermion operators occurring in Eq. (2) is four, both for the NN term and for the NNN term. Longer ranged interactions such as $\mathbf{S}_i\cdot\mathbf{S}_{i+3}$ would, on the other hand, lead to terms containing products of six or more fermion operators.

The Hamiltonian (2) is solved by treating the quartic terms in mean-field theory. We define the following averages that are determined self-consistently: $\overline{n}=\langle d_i^\dagger d_i\rangle$, $\Delta_1=\langle d_{i-1}^\dagger d_{i+1}\rangle$, and $\Delta_2=\langle d_{i-1}^\dagger d_{i+1}^\dagger\rangle$. In terms of these averages, the mean-field Hamiltonian is given by

$$\begin{aligned}H_{\text{MF}}=&\sum_i d_i^\dagger d_i\left\{\frac{J}{2}[1-2(\Delta_1+\Delta_2)]+\alpha J(2\overline{n}-1)\right\}\\&+\sum_i(d_{i-1}^\dagger d_{i+1}+d_{i-1}^\dagger d_{i+1}^\dagger+\text{H.c.})\\&\times\left\{\frac{J}{2}(1-\overline{n})+\alpha J(\Delta_1+\Delta_2)\right\}+E_0,\end{aligned}$$

where $N$ is the number of spins in the chain and $E_0$ is defined by

$$E_0 = NJ(\Delta_1+\Delta_2) - \frac{NJ}{4} - NJ(1-\overline{n})(\Delta_1+\Delta_2) - N\alpha J(\Delta_1+\Delta_2)^2 - N\alpha J\left(\overline{n^2} - \frac{1}{4}\right).$$

The mean-field Hamiltonian can be simplified further by defining the following two quantities:

$$J_B \equiv \frac{J}{2}[1-2(\Delta_1+\Delta_2)] + \alpha J(2\overline{n}-1),$$

$$J_A \equiv \frac{J}{2}(1-\overline{n}) + \alpha J(\Delta_1+\Delta_2).$$

Using these definitions and Fourier transforming, the mean-field $J_1$-$J_2$ model is written as

$$H_{\rm MF} = \sum_k (J_B + 2J_A\cos 2k) d_k^\dagger d_k + i\sum_k J_A \sin 2k (d_k^\dagger d_{-k}^\dagger - d_{-k}d_k) + E_0, \qquad (3)$$

where the lattice constant has been set to be unity. The mean-field Hamiltonian (3) can now be solved by introducing the Bogoliubov transformation

$$d_k = u_k\alpha_k - i v_k \alpha_{-k}^\dagger,$$

where

$$u_k = \frac{1}{\sqrt{2}}\left(1+\frac{\epsilon_k}{E_k}\right)^{1/2},$$

$$v_k = \frac{1}{\sqrt{2}}\left(1-\frac{\epsilon_k}{E_k}\right)^{1/2} {\rm sgn} k,$$

with $\epsilon_k = J_B + 2J_A\cos 2k$, $\Delta_k = 2J_A \sin 2k$, and $E_k = \sqrt{\epsilon_k^2 + \Delta_k^2}$. It is then easy to see that the Hamiltonian (3) reduces to

$$H_{\rm MF} = \sum_k E_k \alpha_k^\dagger \alpha_k + \frac{1}{2}\sum_k (\epsilon_k - E_k) + E_0.$$

On evaluating the mean-field quantities and substituting them in the definitions for $J_A$ and $J_B$ we get two mean-field equations that have to be solved for self consistency,

$$J_B = \frac{J}{2}\left[1+\frac{1}{N}\sum_k \frac{J_B\cos 2k + 2J_A}{E_k}(1-2n_k)\right] - \frac{\alpha J}{N}\sum_k \frac{2J_A\cos 2k + J_B}{E_k}(1-2n_k), \qquad (4)$$

$$2J_A = \frac{J}{2}\left[1+\frac{1}{N}\sum_k \frac{2J_A\cos 2k + J_B}{E_k}(1-2n_k)\right] - \frac{\alpha J}{N}\sum_k \frac{J_B\cos 2k + 2J_A}{E_k}(1-2n_k),$$

where $n_k$ is the usual Fermi distribution function. When $\alpha=0$, the above equations reduce to the mean-field equations written down in Refs. 19 and 20. The solution in that case was obtained as $J_B = 2J_A$. By inspection, we see that the solution of the mean-field equations is still given by $J_B = 2J_A \equiv \overline{J}$, where now, $\overline{J}$ is modified by $\alpha$. The dispersion relation is as before, $E_k = 2\overline{J}|\cos k|$. On substituting the solution in either of the above two equations, we determine $\overline{J}$ by the equation

$$\overline{J} = \frac{J}{2}\left[1+\frac{1-2\alpha}{N}\sum_k |\cos k| \tanh(\beta\overline{J}|\cos k|)\right].$$

At zero temperature, the above expression gives us the dispersion relation,

$$E_k = J\left[1+\frac{2(1-2\alpha)}{\pi}\right]|\cos k|. \qquad (5)$$

When $\alpha=0$, (5) describes the dispersion relation for spinons obtained by Faddeev and Takhtajan.[21] As pointed out in Ref. 20, the spinon velocity obtained from the mean-field theory of antiferromagnetic domain walls, namely $(1+2/\pi)J \approx 1.64J$ is quite close to the exact value $(\pi/2J \approx 1.57J)$. When $\alpha$ is nonzero, our result shows that the spinon velocity is reduced because of frustration arising from the NNN interaction. This is to be expected from physical grounds. On comparing the spinon velocity in Eq. (5) with the spinon velocity for $\alpha=0$, we see that the effect of the NNN term is to reduce the spinon velocity as $v_s(\alpha) = v_s(0)[1-4\alpha/(\pi+2)]$. The ground-state energy at $T=0$ is given by $E_0 = E_0(\alpha=0) + 2\alpha NJ/\pi^2$, where $E_0(\alpha=0)/NJ = -(1/\pi + 1/\pi^2) \approx -0.420$ is the ground-state energy of the NN Heisenberg chain within this approach, which compares well with the exact result $1/4 - \ln 2 \approx -0.443$.[22]

The mean-field solution described above provides a simple way to calculate physical quantities in the $J_1$-$J_2$ model. In particular, one can see how the presence of the NNN term in the Hamiltonian (1) alters the susceptibility at finite temperatures. Before we proceed to show this, we discuss the limitations of the mean-field theory. First, we point out that though the mean-field *solution* is valid for any value of $\alpha$, the theory itself is meaningful only in the gapless regime, i.e, when $\alpha < \alpha_{\rm cr}$ where the spinons are deconfined. This mean-field theory is not suited for studying the transition between the gapless and gapped phases of the $J_1$-$J_2$ model. We reemphasize that our objective is not to undertake such a study (indeed, far more powerful techniques are available and have been used to study this problem) but to provide a simple self-consistent prescription to calculate physical quantities in the gapless phase of the $J_1$-$J_2$ model. Next, we point out that the mean-field solution is plagued by its reluctance to move away from the universality class of the $XY$ model. This can be seen by writing down the (mean-field) ground-state wave function in terms of the soliton operators $d_k$ as $|\Psi\rangle = \Pi_k(u_k + v_k d_k^\dagger d_{-k}^\dagger)|0\rangle$. Whereas in the $XY$ model, the Hamiltonian can be solved exactly to determine $u_k$ and $v_k$, the mean-field solution of the Heisenberg as well as the $J_1$-$J_2$ model give the same $u_k$ and $v_k$. In this sense, this theory has the same difficulties as the mean-field

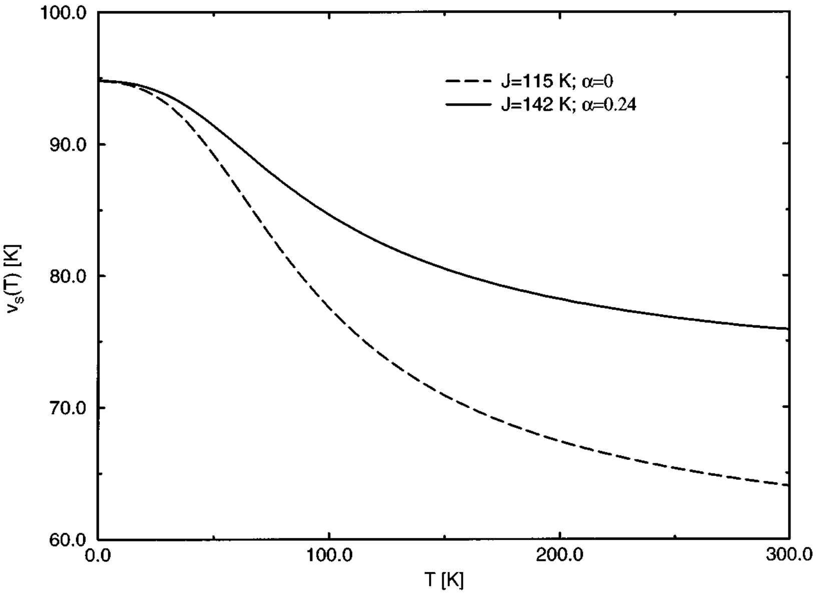


FIG. 1. Spinon velocity as a function of temperature $T$, obtained by the solitonic mean-field theory. The long-dashed line shows results for $J=115$ K and $\alpha=0$. The solid line is obtained with $J=142$ K and $\alpha=0.24$. The values of $J$ are chosen such that the velocities at zero temperature are the same in both cases.

theory of Bulaevskiĭ[23] though the nature of the quasiparticles in the two theories are entirely different. Finally, let us consider the reduction in the spinon velocity as a function of the NNN coupling $\alpha$. From Eq. (5), we see that the velocity $v_s(\alpha)\approx v_s(0)(1-0.8\alpha)$. This result can be checked numerically. In a recent numerical study, Fledderjohann and Gros[24] found that the spinon velocity in the $J_1$-$J_2$ model can be fit to the relation $v_s(\alpha)=v_s(0)(1-1.12\alpha)$. Thus we see that the results from mean-field theory *underestimate* the effects of frustration.

We are now ready to consider the static susceptibility at finite temperatures. In particular, we are interested in seeing how the presence of a NNN interaction alters the susceptibility of an antiferromagnetic Heisenberg chain. Now, it is well known that the susceptibility of the Heisenberg chain is constant at zero temperature and increases to a broad maximum at $T=0.641J$.[11] The increase in susceptibility is due to the gapless spinon excitations. As we shall see, the NNN term causes a suppression in the maximum value of this susceptibility. The reason for this is that the spinon velocity now has a different temperature dependence with a nonzero $\alpha$. To illustrate this, we show in Fig. 1, the spinon velocity as a function of temperature. The long-dashed line shows the results for the case $\alpha=0$. The solid line shows the spinon velocity as a function of temperature for $\alpha=0.24$. For the latter case, we have, for purposes of illustration, chosen $J$ such that the velocity at $T=0$, given by Eq. (5) is the same as that for $\alpha=0$. It should be noted that while the low temperature velocities for different $\alpha$ are identical, the results begin to differ as temperature increases. In particular, they are perceptibly different at $T\approx 0.6J$. Therefore, we should expect a difference in the finite temperature susceptibilities as a function of $\alpha$ at these temperatures.

We have calculated the static susceptibility at finite temperatures from the mean-field solution. At this point, it is crucial to realize that the results from the mean-field theory violate spin-rotational invariance of the underlying Hamiltonian, and that the physically relevant susceptibility would be given by the average

$$\chi=\frac{1}{3}(\chi_{xx}+\chi_{yy}+\chi_{zz}),$$

where $\chi_{xx}$, $\chi_{yy}$, $\chi_{zz}$ are the susceptibilities for the applied magnetic fields in $x$, $y$, and $z$ directions, respectively. Next we note that our mean-field theory becomes exact for the $XY$ model for which the susceptibilities $\chi_{zz}^{XY}=1/\pi\approx 0.318$ and $\chi_{xx}^{XY}=\chi_{yy}^{XY}\approx 0.075\approx\chi_{zz}^{XY}/4$,[25] in dimensionless units and at zero temperature. We therefore have for the $XY$ model, the relation

$$\chi\approx\frac{1}{3}\left(\frac{1}{4}+\frac{1}{4}+1\right)\chi_{zz}^{XY}=\frac{1}{2}\chi_{zz}^{XY}. \tag{6}$$

We have applied this relation to our mean-field solution of the Heisenberg model.

A straightforward calculation shows that the susceptibility at zero temperature is given by

$$\frac{\chi}{g^2\mu_B^2 N}=\frac{1}{2\pi v_s},$$

where $g$ is the gyromagnetic ratio, $\mu_B$ is the electron Bohr magneton and $v_s$, as before, is the spinon velocity. The factor 2 in the denominator comes from the average over the directions of the applied magnetic field as given by Eq. (6).

As a quick check of our results, let us consider the case $\alpha=0$ and compare with the results of Bonner and Fisher.[11] At $T=0$, from Eq. (5) and the above expression for susceptibility, we get $\chi(T=0)J/(g^2\mu_B^2 N)\approx 0.106$, which compares very well with the Bethe-Ansatz results $1/\pi^2\approx 0.101$.[11] At finite temperatures, we solved the mean-field Eqs. (4) numerically and used it to determine the susceptibility. We found that the temperature at which the susceptibility attains its maximum value is given by $k_B T_{\max}/J=0.61$, while the corresponding value quoted in Ref. 11 is 0.641. These comparisons encourage us to proceed to the case $\alpha\neq 0$.

Our results for the case $\alpha\neq 0$ are summarized in Fig. 2 where they are compared with experimentally obtained values from a polycrystalline sample of $CuGeO_3$.[26] We have chosen $g=2.0$ in all our calculations. The long-dashed line

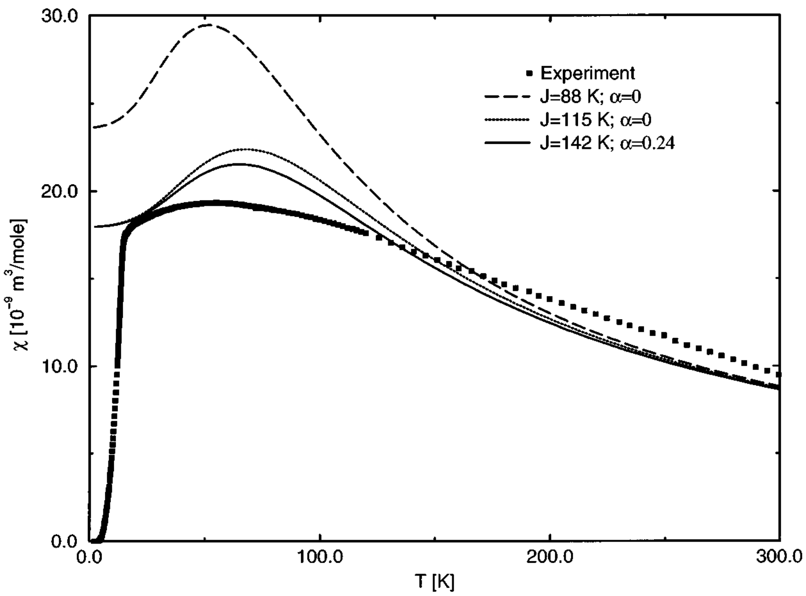


FIG. 2. Experimental susceptibility of $CuGeO_3$ [filled squares, (Ref. 26)] as a function of temperature compared with results from the mean-field theory of the $J_1$-$J_2$ model. The long-dashed line shows results with $J=88$ K and $\alpha=0$. The dotted line shows results with $J=115$ K and $\alpha=0$. The solid line is obtained with $J=142$ K and $\alpha=0.24$.

shows the results for $J=88$ K, $\alpha=0$ that are clearly in disagreement with experiment. The choice of $J=88$ K (Ref. 1) stems from the observed temperature (56 K) at which the susceptibility attains its maximal value and its relation to $J$ from the results of Ref. 11 or equivalently, our results. The dotted line shows the results we obtained from choosing $J=115$ K, $\alpha=0$. This choice of $J$ yields the observed value of spinon velocity from INS (Ref. 8) without including the effects of frustration. It is clear that there still remains a discrepancy between the observed and calculated values at intermediate temperatures. Our results for the susceptibility with a nonvanishing $\alpha$ are given by the solid line. Here, we have chosen $J=142$ K and $\alpha=0.24$. This choice of parameters when substituted in Eq. (5) gives the value of the spinon velocity observed in INS. (Note that the corresponding value of $J$ determined numerically in Ref. 13 is $J=150$ K.) At low temperatures, results with and without $\alpha$ are identical (see Fig. 1 and our discussion of those results). However, we see that as temperature increases, a nonvanishing $\alpha$ does tend to suppress the susceptibility. We see that there is still a discrepancy between theory and experiment which is of the order of 10%. We believe this discrepancy is because the mean-field theory underestimates the effects of frustration. We also considered the possibility that the discrepancy between theory and experiment may well be due to the effect of interchain coupling. As mentioned earlier, the interchain superexchange $J_b \approx 0.1 J_c$ and this could play an important role. A qualitative argument against this possibility is that at temperatures where the discrepancy is maximal ($T > 0.5 T_{SP}$), the magnetic correlation lengths should be small. To substantiate this argument, we did a simple calculation using renormalized spin wave theory with the appropriate values of $J_b$ and $J_c$. Were it to order antiferromagnetically, the observed values of the superexchange interactions suggest that the ''Néel temperature'' of $CuGeO_3$ would be $\sim 10$ K which is lower than $T_{SP}$. At temperatures above $T_{SP}$, the effect of (short-range) antiferromagnetic ordering can be easily estimated by spin wave theory. A standard calculation using the ''decoupling approximation''[27] gives us the susceptibility $\chi$ in terms of the integral equation

$$\frac{1}{\pi^2}\int_0^{\pi}\int_0^{\pi} dz\ dy \frac{1}{2(\gamma_o-\gamma_k)-\chi^{-1}} = \frac{1}{4k_BT},$$

where $\gamma_o \equiv J_c + J_b$ and $\gamma_k \equiv J_c \cos z + J_b \cos y$. We have ignored $J_a \approx -0.01 J_c$ and set $g\mu_B$ to be unity for convenience. One integration in the left-hand side of the above expression can be done trivially and we obtain

$$\int_0^{\pi} dz \frac{1}{\sqrt{(\overline{\chi}+\cos z)^2-0.01}} = \frac{\pi}{2}\left(\frac{J}{k_BT}\right),$$

where $\overline{\chi} \equiv (2J\chi)^{-1} - 1.1$ and $J_c \equiv J = 10 J_b$. Solving the above equation numerically with $J=115$ K, we obtain the susceptibility as shown in Fig. 3. It should be remembered that the theoretical results are valid only at temperatures well above $T_{SP}$ where the effects of phonons as well as effects due to competing antiferromagnetic and SP instabilities can be ignored. The results in Fig. 2 and Fig. 3 show that a one-dimensional model with frustration is indeed a better starting point to study $CuGeO_3$ rather than an anisotropic two-dimensional model. This is in agreement with a number of experimental observations such as (i) a nonzero nuclear spin-lattice relaxation rate seen in a Cu NQR study,[28] (ii) the two-spinon continuum observed in INS above $T_{SP}$,[4,9] and (iii) a broad continuum of magnetic excitations seen in two-magnon Raman scattering above $T_{SP}$,[29] which will be discussed in the next section. All these experiments show characteristic features of a one-dimensional Heisenberg antiferromagnetic chain.

Since our mean-field solution underestimates the effects of frustration, we are unable to determine the exact ratio of $J_2/J_1$ in $CuGeO_3$. As we mentioned in the Introduction, the value of $J_2/J_1$ was estimated to be 0.36 (Ref. 12) and 0.24 (Ref. 13) by two independent studies. Since these two values lead to two different fixed points (gapped and ungapped spin

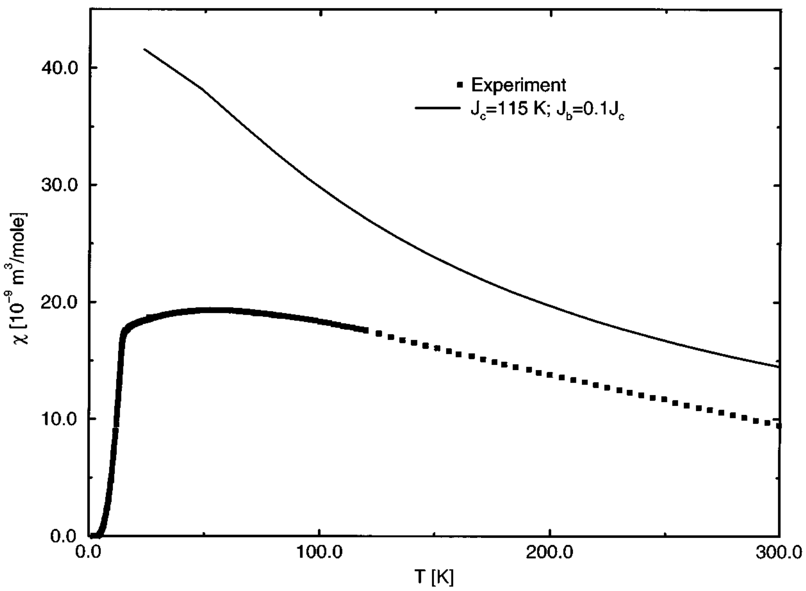


FIG. 3. A comparison between the experimental susceptibility of $CuGeO_3$ [filled squares, (Ref. 26)] and results obtained from a 2D spin wave theory with $J_c = 115$ K and $J_b = 0.1 J_c$.

excitation spectra, respectively), careful experimental investigation of spin dynamics above $T_{SP}$ may provide further clues. Recently, Kuroe *et al.*[30] have claimed that $J_2/J_1 = 0.35 \pm 0.05$ leads to a good fit of the magnetic specific heat as determined by quasielastic Raman scattering. It should be noted, however, that this choice does not fit the observed magnetic susceptibility.

Before we conclude this section, we discuss how the temperature dependence of the spinon velocity may be used to determine the presence of competing magnetic interactions in quasi-one-dimensional (1D) materials. As shown in Fig. 1, a nonzero value of $J_2/J_1$ leads to a difference in the temperature dependence of $v_s(T)$. This feature may be detected experimentally. As a possibility, we suggest INS experiments in the presence of a magnetic field. In the presence of a magnetic field, it is known that the spinon dispersion $E_k$ goes to zero at $k$ given by $\cos k = -2H/v_s(T)$.[31] This is a feature of low-dimensional spin excitations and is seen in the $XY$ model as well. This behavior should be contrasted with spin wave (magnon) excitations, where no such node appears in the magnon dispersion as a function of the magnetic field. An examination of the temperature dependence of this node with moderately high fields $H/J \sim 0.2$ should indicate the presence of frustration as shown in Fig. 1.

## III. TWO-MAGNON RAMAN CONTINUUM IN $CuGeO_3$

In this section, we consider the Raman spectra in the homogeneous phase of $CuGeO_3$ observed in two-magnon scattering experiments.[29] A partial account of the results in this section has been published elsewhere.[32] For completeness, we first give a brief summary of the pertinent experimental results.

Scattering of light from magnetic (as opposed to phonon) excitations in $CuGeO_3$ is strongest in the $(zz)$ geometry, viz., when both incident and scattered light are polarized along the crystallographic $\hat{c}$ direction. This is consistent with the fact that exchange interactions are strongest along this direction. The Raman spectra above $T_{SP}$ are rather featureless and the spectral weight is distributed over a wide range of energies (100–300 cm$^{-1}$). A broad maximum is observed around 250 cm$^{-1}$. This should be contrasted with the Raman spectra observed in antiferromagnetically ordered compounds such as the rutile or perovskite halides. In these compounds, one typically observes a well defined peak at characteristic magnon energies which is broadened by magnon-magnon interactions.[33] This comparison immediately suggests light scattering in $CuGeO_3$ from a continuum of excitations rather than single-particle excitations (magnons).

The magnetic Raman scattering in $CuGeO_3$ can be studied using the scheme of Fleury and Loudon.[34] The basic idea is that in an exchange-coupled system, an incident photon can create particle-hole excitations accompanied by a pair of spin flips that propagate as magnons. In this scheme the Raman operator $H_R$ has essentially the same form as the Heisenberg exchange interaction, i.e., $H_R \sim \Sigma_i \mathbf{S}_i \cdot \mathbf{S}_{i+1}$. In one dimensional chains with NN exchange interaction, we are faced with an intriguing situation where the Raman operator for scattering in the $(zz)$ geometry is proportional to the Hamiltonian itself. Thus, there should be no Raman scattering from a NN Heisenberg spin chain, a fact which has not been appreciated previously.[29] However, there are two ways in which Raman scattering can occur: (i) interchain couplings and (ii) exchange interactions that are extended in space, i.e., NNN interactions. Let us first consider the former possibility. The role of the interchain couplings is to establish short-range antiferromagnetic order. In such a case, incident light is scattered off paramagnon excitations. However, as we mentioned earlier, the featureless spectra observed in $CuGeO_3$ seem to indicate scattering from a continuum of excitations. This is basically a one-dimensional feature. The second possibility is therefore a more viable explanation of the experimental results. In view of the results discussed in the previous section, it is also a more natural choice. Thus, we work with a Hamiltonian which has both NN and NNN interactions. This implies that the Raman operator will also have both NN and NNN terms. For conve-

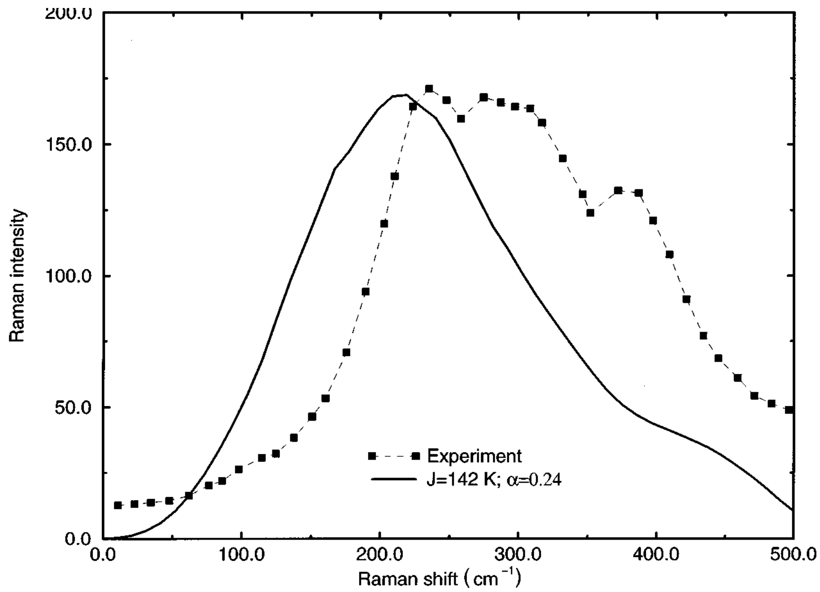


FIG. 4. The two-magnon Raman continuum seen in experiment [filled squares, (Ref. 32) see also (Ref. 29)] at $T=20$ K compared with results obtained from mean-field theory (solid line). The theoretical results were obtained with $J=142$ K and $\alpha=0.24$.

nience, we subtract from the Raman operator, a part which commutes with the basic Hamiltonian of $CuGeO_3$ (1), we can write down a Raman operator of the form

$$H_R=A\sum_i \mathbf{S}_i\cdot\mathbf{S}_{i+2}.$$

The Raman intensity is then evaluated by considering the appropriate spectral function.

We now use the mean-field theory described in the previous section to compute the Raman intensity. The Raman operator $R$ can be written in terms of the soliton operators and transformed into quasiparticle or spinon operators just as we did in the previous section. We find that there are three basic physical processes caused by $R$: (i) spinon-spinon scattering, (ii) two-spinon creation/annihilation, and (iii) four-spinon creation/annihilation. Of these, (ii) corresponds to one-magnon excitations. On using the mean-field solution, one can show that the contribution to light scattering from these excitations vanishes, which is just the statement that one-magnon excitations with $k=0$ have zero spectral weight. Two-magnon scattering is given by (iii). We find that the matrix element for two-magnon (four-spinon) creation (Stokes component of the scattering intensity) is given by

$$\begin{aligned}\frac{1}{N}\sum_{kk'qq'}&\delta(k+k'+q+q')\,\alpha_k^\dagger\alpha_{k'}^\dagger\alpha_q^\dagger\alpha_{q'}^\dagger\\&\times[\tfrac{1}{2}\exp\{-i(2k+k'-q')\}\\&\times\{(u_ku_{k'}+iv_ku_{k'})(v_qv_{q'}+iv_qu_{q'})\\&-(u_kv_{k'}+iv_kv_{k'})(u_qv_{q'}+iu_qu_{q'})\}\\&-\exp\{-i(q+q')\}u_kv_{k'}u_qv_{q'}],\end{aligned}$$

where the summation is over the range of spinon momenta and the quantities $u_k$ and $v_k$ are as defined in the previous section. The matrix element can be understood as arising from a decomposition of two magnon excitations into four spinon excitations with total momentum zero. Given this matrix element, we can calculate the Raman intensity. The mean-field solution thus provides a way of incorporating matrix element effects arising from the Raman operator instead of postulating *ad hoc* filtering functions that mimic matrix element effects. As noted elsewhere,[32] such effects could be very important and ignoring them often leads to erroneous conclusions.

The results for the Raman intensity at $T=20$ K with $J=142$ K and $\alpha=0.24$ are shown in Fig. 4 (solid line) where they are compared with experimental results (squares) obtained from a single-crystal sample. Phonon lines at 184 and 330 cm$^{-1}$ have been subtracted from the experimental data. Since we do not know the value of $A$ in our definition of the Raman operator $R$, we have normalized the maximal theoretical value of the intensity to the maximal experimental value. A uniform background of 50 counts has also been subtracted from the experimental data. We see that there is a reasonable agreement between theory and experiment as far as the observed continuum is concerned. The shoulder observed experimentally at $\sim$ 390 cm$^{-1}$ is not understood and our calculations show no indications of this feature. We also see that the maximum of the experimental data is shifted slightly toward larger energies than the theoretical results. This could either be due to interchain couplings and phonon effects that have been neglected or a mismatch in the parameters we have chosen. Our calculations are in good agreement with results from exact diagonalization[32] as well as those of Singh and collaborators[35] who have studied the same problem recently using a finite temperature Lanczos method. We have also calculated the Raman intensities at higher temperatures. We find that the continuum broadens out as temperature is increased and there is no accumulation of spectral weight at lower energies as seen in experiment.[29]

From the results shown in Fig. 4, we conclude that the Hamiltonian (1) provides a good description of the observed Raman continuum owing solely to the presence of the NNN interaction. These results as well as the reasoning behind them also lead us to conclude that there would be no inelastic

Raman intensity in one-dimensional chains such as $KCuF_3$ where NNN interactions are negligible. In such materials, while INS can observe the *one-magnon* continuum, the *two-magnon* continuum cannot be observed in Raman scattering. These conclusions are indeed borne out by experiments.[36]

## IV. SUMMARY

To summarize the results of this study, we have proposed a mean-field solution to the antiferromagnetic Heisenberg chain with NN and NNN interactions. This was motivated by recent interest evinced in the spin-Peierls compound $CuGeO_3$ where it is suspected that NNN interactions play an important role. Our mean-field solution provides a self-consistent way to include the effects of frustration and calculate physical quantities of interest as a function of temperature. Though we have limited our attention to experimental results in $CuGeO_3$, the results in this paper can be used to study the effects of frustration in generic quasi-1D systems.

As a first application, we have evaluated the temperature dependent spin-wave velocity, $v_s(T)$ and found that (i) $v_s(T)$ decreases rapidly for $\alpha=0$ with increasing $T$, $\alpha$ being the ratio between the NNN and NN interactions and (ii) this decrease is much less pronounced for $\alpha\neq 0$. We propose that this fact could be exploited to determine the frustration parameter $\alpha$ directly from measurements of $v_s(T)$, e.g., by inelastic-neutron-scattering studies in other quasi-1D systems.

We then used the mean-field theory to evaluate the static susceptibility $\chi$. A comparison with experimental results shows that the inclusion of frustration does reproduce the suppression of susceptibility at finite temperatures seen experimentally. We have been able to identify the physical reason for the suppression of $\chi$ as a function of $\alpha$ with the differing behavior of the respective $v_s(T)$. We have also presented the results of a spin wave calculation for an anisotropic two-dimensional Heisenberg model which do not, on the other hand, compare well with the experimental results for the susceptibility. This suggests that a one-dimensional model with frustration is a more appropriate starting point to model the homogeneous phase of $CuGeO_3$.

We also used the mean-field solution to calculate the two-magnon Raman intensity from a one-dimensional antiferromagnetic chain and showed that frustration is necessary for obtaining a nonvanishing spectral weight. Considering the fact that NNN interactions are needed to produce a good fit to the static susceptibility of $CuGeO_3$, using the same idea to explain the Raman continuum provides a consistent picture as far as modeling goes. Quite apart from the relevance to $CuGeO_3$, this method of computing Raman intensity can be used to study experimental results now available in $Sr_2CuO_3$ and $SrCuO_2$.[37] Clearly, a complete understanding of $CuGeO_3$ calls for the inclusion of interchain and phonon effects. The mean-field solution presented in this paper provides a way to incorporate these effects systematically. Work is in progress on these issues and results will be reported in a forthcoming publication.

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