Raman Scattering from Frustrated Quantum Spin Chains

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Abstract

Magnetic Raman scattering from a frustrated spin–1/2 Heisenberg–chain is considered with a focus on the uniform phase of the spin–Peierls compound CuGeO₃. The Raman intensity is analyzed in terms of a Loudon–Fleury scattering process using a spinless–fermion mean-field theory developed for the frustrated spin–chain. A comparison to experimental data is presented and the frustration and temperature dependence is studied. In good agreement with observed spectra a broad inelastic four–spinon continuum is found at low temperatures above the spin–Peierls transition. At high temperatures the intensity develops a quasi–elastic line analogous to experiment.

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Elementary excitations of one–dimensional (1D) quantum spin–chains exhibit a number of remarkable subtleties, such as the magnon continuum of the isotropic 1D spin–1/2 Heisenberg model or the excitation–gap for integer–spin systems as conjectured by Hal–dane. Recent and extensive studies of ‘spin–ladder’ compounds as well as of the germanate CuGeO$_3$ [1] have lead to renewed interest in quantum spin–chains. In this context, magnetic Raman scattering, if symmetry–allowed, has developed into an important tool to investigate the local spin dynamics [2–4]. Here I will establish a simple framework to interpret the magnetic Raman scattering from a frustrated quantum spin–chain with a particular focus on the uniform phase of CuGeO$_3$.

CuGeO$_3$ is a quasi–1D anorganic spin–Peierls compound with a dimerization transition at a temperature $T_{SP} \simeq 14K$ [1, 5, 6]. Its structure comprises of weakly coupled CuO$_2$ chains, with copper in a spin–1/2 state [7]. Since the nearest–neighbor (n.n.) exchange–coupling between copper spins along the CuO$_2$ chains is strongly reduced by almost orthogonal intermediate oxygen states [8], the next–nearest–neighbor (n.n.n.) exchange is relevant. Both, n.n. and n.n.n. exchange, are antiferromagnetic [9] implying intra–chain frustration. A minimal model of CuGeO$_3$ is the $J_1$–$J_2$–$\delta$ model

$$H = J_1 \sum_l [(1 + (-)^l \delta) \mathbf{S}_l \cdot \mathbf{S}_{l+1} + \alpha \mathbf{S}_l \cdot \mathbf{S}_{l+2}] .$$

Here, $\mathbf{S}_l$ is a spin–1/2 operator, $J_1 \approx 160K$ [10, 12] is the n.n. exchange–coupling constant and $\delta$ resembles the lattice dimerization which is finite for $T < T_{SP}$ only. $\alpha$ is the intra–chain frustration–ratio $\alpha = J_2/J_1$ where $J_2$ is the n.n.n. exchange–coupling constant. A final consensus on the precise magnitude of $\alpha$ is still lacking. Studies of the magnetic susceptibility have resulted in $\alpha \approx 0.24$ [10] as well as in $\alpha \approx 0.35$ [11]. The latter is consistent with a comparative investigation of magnetic susceptibility and thermal expansion [12]. Therefore, very likely, CuGeO$_3$ displays a frustration induced contribution to the spin–gap at zero temperature irrespective of the actual lattice dimerization [13]. Depending on
the magnitude of the frustration the zero–temperature dimerization $\delta(T = 0)$ has to be on the order of $\delta(0) = 0.01...0.03$ such as to enforce the size of the spin–gap observed in inelastic neutron scattering (INS) \cite{5}.

Magnetic excitations in CuGeO$_3$ are clearly distinct among the uniform, i.e. $T > T_{SP}$, and the dimerized, i.e. $T < T_{SP}$, phase. While the dynamic structure factor exhibits a gapless two–spinon continuum similar to that of the 1D Heisenberg chain above $T_{SP}$ \cite{14}, well defined magnon–like excitations have been observed below $T_{SP}$ \cite{15}. These magnons reside within the spin–gap and are split off from the two–spinon continuum. They have been interpreted as two–spinon bound states \cite{16,17}.

Magnetic Raman scattering from CuGeO$_3$ has been observed both, in the dimerized as well as in the uniform phase \cite{2–4}. In the low temperature uniform phase, for $T_{SP} < T \ll J_1$, the Raman spectrum displays a broad continuum centered at $\hbar \omega \approx 2.4J_1$. Early on this continuum has been related to four–spinon excitations \cite{2–4,18}. In the high temperature uniform phase, and in addition to the four–spinon continuum, the spectra show a pronounced quasi–elastic line. In the dimerized phase the Raman intensity develops a gap at approximately 1.5–1.8$\Delta_{ST}$ \cite{20} where $\Delta_{ST}$ is the singlet–triplet gap observed in INS \cite{5}. Moreover, four characteristic peaks appear in the spectrum the lowest one of which at 30cm$^{-1}$ 'coincides’ with the Raman gap. At present the interpretations of these peaks are controversial. Tentatively the 30cm$^{-1}$ line has been attributed to a continuum of two–magnon bound states \cite{19,20}. A possible dimensional crossover effect has been invoked for the 225cm$^{-1}$ line \cite{3,20}.

In this respect the influence of inter–chain coupling in CuGeO$_3$ is an open problem \cite{21}.

Here I will focus on Raman scattering from the uniform phase of the $J_1$–$J_2$–$\delta$ model, i.e. at $\delta = 0$. First I will describe a spinless–fermion mean–field theory to treat the $J_1$–$J_2$ Hamiltonian. Next the Raman intensity is expressed in terms of a four–fermion correlation function. Finally results for the Raman spectra are compared with experimental findings and are contrasted against other theoretical approaches.
II. MEAN–FIELD THEORY OF THE $J_1$–$J_2$ MODEL

The spinless–fermion mean–field (MF) theory for the $J_1$–$J_2$ model is based on the Jordan–Wigner (JW) representation \cite{22} of the spin algebra, i.e. $S^z_l = (c^\dagger_lc_l - \frac{1}{2})$ and $S^+_l = (-)^l \prod_{j<l}(1 - c^\dagger_jc_j)c^\dagger_l$, where $c^{(t)}_l$ are fermion operators. Inserting this into (1) at $\delta = 0$ one obtains

$$H = \sum_l \left\{ -\frac{1}{2}c^\dagger_{l+1}c_{l+1} - \frac{1}{2}c^\dagger_{l+1}c_l - c^\dagger_{l+1}c^\dagger_{l+1}c_{l+1} + \frac{1}{4} + \frac{\alpha}{2}c^\dagger_{l+2}c_{l+2} + \frac{\alpha}{2}c^\dagger_l c_l + \alpha \left(c^\dagger_{l+1}c^\dagger_{l+2}c_l - c^\dagger_{l+1}c^\dagger_{l+2}c_{l+1} - c^\dagger_{l+1}c_l + c^\dagger_{l+2}c^\dagger_l c^\dagger_{l+1}c_{l+2} + \frac{1}{4}\right) \right\} ,$$

(2)

where $J_1$ is set to unity in section II and III. $l$ runs over the lattice sites. The terms proportional to $\alpha$ are absent in the JW representation of the isotropic $n.n.$ Heisenberg model. In contrast to the $n.n.$ model, both, the transverse as well as the longitudinal $n.n.n.$ exchange–interaction lead to four–fermion vertices. Longer range spin–exchange leads to even higher order couplings, as is obvious from the JW representation which implies $2l$–fermion vertices for $l$–th.–nearest–neighbor spin–exchange. To proceed I treat Hamiltonian (4) in MF–approximation. Allowing for all contractions of type $\langle c^\dagger_l c_m \rangle$ one gets

$$H_{MF} = \sum_k \left\{ -[1 + (A + B)(1 - 2\alpha)] \cos(k) c^\dagger_k c_k + \frac{i}{2}(A - B)(1 + 2\alpha) \sin(k) (c^\dagger_k c_{k+\pi} - c^\dagger_{k+\pi} c_k) \right\} + \text{const.} ,$$

(3)

where $k$ is the momentum, $A = A^* = \langle c^\dagger_{2l}c_{2l+1} \rangle$, and $B = B^* = \langle c^\dagger_{2l+1}c_{2l} \rangle$. In principle contractions of type $D = D^* = \langle c^\dagger_{2l+2}c_{2l} \rangle$ do occur, however, their selfconsistent value can be shown to vanish identically. Note that (3) allows for both, a uniform, if $A - B = 0$, and a gaped phase, if $A - B \neq 0$. Here no attempt will be made to describe the spin–dimer state using $A \neq B$ and the relevance of the gaped solution of the MF–theory will be discussed elsewhere. In the uniform case (3) resembles a single–band spinon gas with a hopping amplitude $t(T, \alpha)$ to be determined selfconsistently

$$H_{MF} = \sum_k \epsilon_k c^\dagger_k c_k$$
\[ \epsilon_k = -t(T, \alpha) \cos(k) = -[1 + 2A(1 - 2\alpha)] \cos(k) \]  
\[ A = \frac{2}{N} \sum_{0 \leq k \leq \pi} \cos(k) f(\epsilon_k) \],

where \( f(\epsilon) = [\exp(\epsilon/T) + 1]^{-1} \) is the Fermi function. At zero temperature the spinon dispersion simplifies to

\[ \epsilon_k(T = 0) = -[1 + 2(1 - 2\alpha)/\pi] \cos(k) \]  

For vanishing frustration this is identical to Bulaevskii’s result \(- (1 + 2/\pi) \cos(k)\) \[23\]. The latter is known to compare reasonably well with the exact spinon dispersion \( \epsilon_k = -\pi/2 \cos(k) \) by des Cloizeaux and Pearson \[24\]. For finite \( \alpha \) the MF–theory results in a frustration induced softening of the spinon stiffness which, for \( T = 0 \), can be expressed as

\[ \frac{v_s(\alpha)}{v_s(0)} = 1 - \frac{4}{2 + \pi} \alpha \approx 1 - 0.778\alpha \],

where \( v_s(\alpha) = \partial \epsilon_k / \partial k \| k = \pi/2 \) is the spinon velocity. Eqn. \(3\) is qualitatively consistent with a numerical study where \( v_s(\alpha)/v_s(0) \approx 1 - 1.12\alpha \) has been found \[17\]. In fig. 1 \( t(T, \alpha) \) is depicted as a function of temperature for various values of frustration. As is obvious the MF spinon–stiffness is decreased, both, as a function of increasing frustration and temperature. At zero temperature the MF ground state energy is given by \( E_0(\alpha) = N(-1/\pi - 1/\pi^2 + 2\alpha/\pi^2) \) which, for \( \alpha = 0 \), leads to \( E_0(0) \approx -0.420 \) \[23\]. This agrees reasonably well with the Bethe–Ansatz result \( E_0 = 1/4 - \ln(2) \approx -0.443 \). Moreover, the linear frustration dependence of the MF ground state energy, \( (E_0(\alpha) - E_0(0))/N \approx 0.203\alpha \) is close to that found in finite–chain diagonalization \[25\], where \( (E_0(\alpha) - E_0(0))/N \approx 0.177\alpha \) for \( \alpha \leq 0.3 \).

III. RAMAN SCATTERING

The MF–theory is a convenient tool to study Raman scattering from the \( J_1–J_2 \) model. The Raman vertex is given by Loudon–Fleury’s photon–induced super–exchange operator \[26\]
\[ R = \sum_{lm} T_{lm} \langle \mathbf{E}_{in} \cdot \mathbf{n}_{lm} \rangle \langle \mathbf{E}_{out} \cdot \mathbf{n}_{lm} \rangle \mathbf{S}_l \cdot \mathbf{S}_m. \quad (7) \]

Here \( T_{lm} \) sets the coupling strength, \( \mathbf{E}_{in} \) (\( \mathbf{E}_{out} \)) refers to the field of the in(out)going light, and \( \mathbf{n}_{lm} \) labels a unit vector connecting the sites \( l \) and \( m \). By symmetry, in a strictly 1D situation, (7) leads to scattering only for parallel polarization along the \( c \)-directed chains. Since the real-space decay of \( T_{lm} \) is comparable to that of the exchange integrals in (1) it is sufficient to consider a Raman operator with at most \( n.n.n. \) spin-exchange

\[ R = C \sum_l (\mathbf{S}_l \cdot \mathbf{S}_{l+1} + \beta \mathbf{S}_l \cdot \mathbf{S}_{l+2}) \quad . \quad (8) \]

The scattering intensity \( I(\omega) \) is obtained via the fluctuation dissipation theorem \( I(\omega) = \chi''(\omega)/(1 - e^{-\omega/T}) \) from the dynamical susceptibility of the Raman operator

\[ \chi''(\omega) = Im[\chi(\omega + i\eta)] = Re \int_0^{+\infty} dt \, e^{i(\omega + i\eta)t} \langle [R(t), R] \rangle , \quad (9) \]

where \( \langle (...) \rangle \) denotes the thermal average.

Frustration of unequal magnitude regarding the \( J_1 - J_2 \) model and the Raman operator is mandatory for non-vanishing inelastic scattering. If \( \alpha = \beta \) the Hamiltonian for \( \delta = 0 \) and the Raman operator commute which leads to elastic scattering only. The inelastic intensity which results from (8) is identical to that of a Raman operator \( R' = R - \gamma H \). Setting \( \gamma = C\beta/\alpha \) or \( \gamma = C \) leads to scattering by a renormalized \( n.n. \) or \( n.n.n. \) Raman operator only

\[ R_1 = C(1 - \frac{\beta}{\alpha}) \sum_l \mathbf{S}_l \cdot \mathbf{S}_{l+1} \quad \text{or} \quad R_2 = C(\beta - \alpha) \sum_l \mathbf{S}_l \cdot \mathbf{S}_{l+2} . \quad (10) \]

At present, an exact treatment of (9) is not feasible and the Raman intensity of any approximate evaluation will depend on \( \gamma \). In this respect \( R_2 \) is the proper choice for \( \alpha, \beta \ll 1 \) since it guarantees that \( I(\omega) \propto (\beta - \alpha)^2 \). This is not obvious for \( R_1 \) or other values of \( \gamma \). In those cases, even for \( \alpha, \beta \to 0 \), \( R' \) may contain a component of order unity, proportional to the Hamiltonian, which has to be projected out. To avoid this complication \( R_2 \) will be considered hereafter.
Using the JW–fermions the Raman operator $R_2$ can be expressed in terms of a four–fermion operator

$$R_2 = C(\beta - \alpha) \sum_{k,k',q} h(k,k',q) c_k^\dagger c_{k+q} c_{k'-q}^\dagger + \Lambda_{1ph} \quad (11)$$

$$h(k,k',q) = \cos(2q) - \cos(2k + q) - \cos(2k' - q) \ ,$$

where $\Lambda_{1ph}$ labels all one–particle–hole excitations of the JW–transform of $R_2$. Since these occur at zero total momentum they do not contribute to inelastic Raman scattering within MF–theory [27]. Using (11), the Raman intensity (9) is written in terms of the four–fermion propagator

$$\chi(\tau) = C^2(\alpha - \beta)^2 \sum_{k,k',q,p,p',r} h(k,k',q) h(p,p',r) \langle T_{\tau}(c_k^\dagger(\tau) c_{k+q}(\tau) c_{k'-q}(\tau) c_{p'-r} c_{p+r} c_p) \rangle \ . \quad (12)$$

Here $\tau$ is the imaginary time and $\chi''(\omega)$ results from the usual analytic continuation $\chi''(\omega) = -Im[\chi(i\omega_n \rightarrow \omega + i0^+)]$ where $\omega_n = 2n\pi T$ is a Bose Matsubara-frequency. On the level of MF–theory the four–fermion propagator (12) is evaluated neglecting all vertex corrections and using the MF one–particle Green’s functions corresponding to (4). After a number of standard manipulations I obtain

$$\chi_{MF}'(\omega) = -\frac{1}{2\pi^2} \int_{-\pi}^{\pi} dq \int_{-\pi}^{\pi} dk \sum_{k'} \left\{ \frac{g^2(k,k'+q,q)}{\sqrt{[2t \sin(q/2)]^2 - (\epsilon_{k+q} - \epsilon_k - \omega)^2}} \right\} \ 
\left[ f(\epsilon_k) - f(\epsilon_{k+q}) \right] \left[ f(\epsilon_{k'}) - f(\epsilon_{k'+q}) \right] [n(\epsilon_{k'+q} - \epsilon_{k'} + \omega) - n(\epsilon_{k'+q} - \epsilon_{k'})] \right\} \ , \quad (13)$$

where $g(k,k',q) = \frac{1}{2}[h(k,k',q) + h(k,k',k' - k - q)]$ and the discrete sum on $k'$ runs over the set of solutions of

$$\sin(k' + q/2) = \frac{\epsilon_{k+q} - \epsilon_k - \omega}{2t \sin(q/2)} ; \quad k' \in [\pi,\pi] \ . \quad (14)$$

This concludes the MF–theory of Raman scattering.
IV. DISCUSSION

In fig. 2 I compare the MF–intensity obtained from a numerical integration of (13) (solid and dashed lines) at \( T = 20K \) and \( 12.8K \), for \( J_1 = 160K \) and two values of frustration \( \alpha = 0.24 \) and \( \alpha = 0.35 \) with an experimental Raman spectrum observed at \( T = 20K \) in the uniform phase of CuGeO\(_3\) [4,18] (dashed–dotted line with markers). Phonon lines at 184cm\(^{-1}\) and 330cm\(^{-1}\) have been removed. This Raman data is consistent with other published results [2,3]. The absolute magnitude of the observed Raman intensity as well as the coupling strength \( C \) of the scattering operator (8) are unknown quantities. Therefore the two sets of MF–intensities for distinct \( \alpha \) and the experimental spectrum have been normalized on a scale of arbitrary units. The theoretical parameters in fig. 2 represent no attempt at a ‘best fit’ to the data, rather they have been chosen among those obtained independently from studies of the magnetic susceptibility of CuGeO\(_3\) [10–12]. Figure 2 displays reasonable agreement between theory and experiment, although details of the experimental spectrum, i.e. the structure at 375 wave numbers and the initial curvature are not reproduced by the MF–theory. Evidently a value of \( \alpha = 0.24 \) [10] is slightly more favored by this comparison at \( J_1 = 160K \) than \( \alpha = 0.35 \) [11,12]. Similar findings have been made in numerical studies [20]. However, since the maximum of the MF–intensity roughly scales with the maximum of the spinon–dispersion \( \propto J_1[1 + 2(1 - 2\alpha)/\pi] \), a better agreement with \( \alpha = 0.35 \) instead of \( \alpha = 0.24 \) can also be reached simply by increasing \( J_1 \) by \( \sim 10\% \).

The comparison in fig. 2 strongly corroborates a study [18] of Raman scattering from the uniform phase of CuGeO\(_3\) which is based the ‘solitonic’ mean–field description of the XXZ Heisenberg chain by Gómez–Santos [28]. This approach employs a domain–wall representation of the spin chain which leads to formal developments very different from the straightforward application of the Jordan–Wigner type of MF–theory presented here.

In fig. 3 the temperature dependence of the MF–spectrum is shown, both, in terms of the intensity \( I(\omega) \) – which is observed in experiment – and the Raman–operator susceptibility \( \chi''(\omega) \). The latter exhibits a left–shift of its maximum upon increase of the temperature.
This leads to enhanced low-frequency spectral weight which, by virtue of the Bose–prefactor, turns into a quasi–elastic line in $I(\omega)$ for $T \gtrsim J_1$. A corresponding tendency has been detected in finite–temperature Lanczos studies \[32\]. In MF–theory the high–temperature enhancement of the low–frequency spectral weight in $\chi''(\omega)$ is due to the reduction of the spinon–stiffness as a function of increasing temperature, see fig. \[1\], and due to the 'smearing' induced by the Fermi– and Bose–functions contained in (13). It is tempting to relate the behavior depicted in fig. \[3\] to the high–temperature quasi–elastic line observed in the uniform phase of CuGeO$_3$ \[3–4\]. However, in CuGeO$_3$ the four–spinon continuum is only weakly shifted by temperature and remains more clearly separated from the quasi–elastic line for $T_{SP} \lesssim T \lesssim J_1$. This discrepancy may be due to an overestimation of the temperature dependence of the spinon–stiffness in MF–theory \[29,30\].

Finally, I emphasize that the MF–theory gives only a limited description of spinon interaction effects. In the case of the two–spinon propagator this is known to result in an incorrect description of the spectral weight distribution \[31\]. This caveat of MF–theory has stimulated a study of approximate vertex corrections to the four–spinon–propagator \[32\]. However, the resulting Raman spectra show no agreement with experiment. At present the impact of spinon interaction effects beyond MF–theory on the Raman spectra remain unclear.

In conclusion I have described a finite temperature MF–theory for frustrated spin–chains and consequently detailed its application to magnetic Raman scattering. At low–temperatures, in the gapless phase, I find a scattering continuum which is due to frustration induced four–spinon excitations and is compatible with observed Raman spectra of CuGeO$_3$. In the high–temperature regime the MF Raman–intensity is dominated by a quasi–elastic line which results from the temperature dependence of the spinon spectrum.
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REFERENCES

[1] M. Hase et al., Phys. Rev. Lett. 70, 3651 (1993).

[2] H. Kuroe et al., Phys. Rev. B 50, 16468 (1994).

[3] P.H.M. van Loosdrecht et al., Phys. Rev. Lett. 76, 311 (1996).

[4] P. Lemmens et al., Physica B 223&224, 535 (1996).

[5] M. Nishi et al., Phys. Rev. B 50, 6508 (1994); M.C. Martin et al., Phys. Rev. B 53, R14713 (1996); L.P. Regnault et al., Physica B 213 & 214, 278 (1995);

[6] J.P. Pouget et al., Phys. Rev. Lett. 72, 4037 (1994); O. Kamimura et al., J. Phys. Soc. Jpn. 63, 2467 (1994); S.B. Oseroff et al., Phys. Rev. Lett. 74, 1450 (1995).

[7] H. Völlkenke et al., Monatsh. Chem. 98, 1352 (1967); G.A. Petakovskii et al., Zh. Eksp. Teor. Fiz. 98, 1382 (1990) [Sov. Phys. JETP 71, 772 (1990)]; K. Hirota et al., Phys. Rev. Lett. 73, 736 (1994); J.E. Lorenzo et al., Phys. Rev. B 50, 1278 (1994); Q.J. Harris et al., Phys. Rev. B 50, 12606 (1994); M. Arai et al., J. Phys. Soc. Jpn. 63, 1661 (1994).

[8] M. Braden et al., Phys. Rev. B 54, 1105 (1996).

[9] W. Geertsma and D. Khomskii, Phys. Rev. B 54, 3011 (1996).

[10] G. Castillia et al., Phys. Rev. Lett. 75, 1823 (1995).

[11] J. Riera et al., Phys. Rev. B 51, 16098 (1995).

[12] B. Büchner, unpublished.

[13] R. Chitra et al., Phys. Rev. B 52, 6581 (1995).

[14] M. Arai et al., Phys. Rev. Lett. 77, 3649 (1996).

[15] M. Áin et al., preprint.

[16] G.S. Uhrig and H.J. Schulz, Phys. Rev. B 54, R9624 (1996).
[17] A. Fledderjohann and C. Gros, preprint cond-mat/9612013

[18] V.N. Muthukumar et al., Phys. Rev. B 54, R9635 (1996).

[19] P.H.M. van Loosdrecht et al., preprint cond-mat/9612167.

[20] C. Gros et al., preprint cond-mat/9612101.

[21] G.S. Uhrig, preprint, (1996).

[22] P. Jordan and E. Wigner, Z. Phys. 47, 631 (1928).

[23] L.N. Bulaevskii et al., Zh. Eksp. Teor. Fiz. 43, 968 (1962), [Sov. Phys. JETP 16, 685 (1963)]

[24] J. des Cloizeaux and J.J. Pearson, Phys. Rev. 128, 2131 (1962).

[25] T. Tonegawa and I. Harada, J. Phys. Soc. Jpn. 56, 2153 (1987).

[26] P.A. Fleury and R. Loudon, Phys. Rev. 166, 514 (1968).

[27] In Raman scattering photon–momentum transfer to the electronic system can be neglected which is implicit in the definition of $R$. Therefore spinon one–particle–hole excitations will contribute to the inelastic intensity only in case of a finite quasi–particle relaxation rate, which is absent in MF–theory.

[28] G. Gómez–Santos, Phys. Rev. B 41, 6788 (1990).

[29] K. Frabricius et al., preprint cond-mat/9611077

[30] O. A. Starykh et al., preprint cond-mat/9701052.

[31] G. Müller, H. Beck, and J.C. Bonner, Phys. Rev. Lett. 43, 75 (1979).

[32] R.P.R. Singh et al., preprint (1996).
FIGURES

FIG. 1. Temperature dependence of the mean–field hopping amplitude for various values of frustration.

FIG. 2. Mean–field Raman intensity (solid and dashed lines) at two temperatures and for two values of frustration as compared to the experimental Raman spectrum of CuGeO$_3$ [18] (dashed–dotted line & markers).

FIG. 3. Mean–field Raman intensity and susceptibility for various temperatures.
Fig. 1, Brenig
$J_1 = 160 \text{K}$

$\approx 111.2 \text{cm}^{-1}$

$\alpha = 0.24$

$\alpha = 0.35$

$\text{exp. } T = 20 \text{K}$

$\text{MF-theor. } T = 20 \text{K}$

$\text{MF-theor. } T = 12.8 \text{K}$

Fig. 2, Brenig
Fig. 3, Brenig