Minimal model to describe the magnetism of CuGeO$_3$

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We show that to describe properly the low energy excitations of CuGeO$_3$ one must include the effects of a transverse antiferromagnetic coupling, which is estimated to be $J_{\perp} = 0.15J$. Owing to this coupling the frustration in the chains is significantly lower than recent 1D estimates based on purely one-dimensional arguments, we find $J_2 = 0.2J$. Furthermore we have found a strong modulation of the nearest neighbour coupling due to the static distortion $\delta = 0.065$, which is 5 times higher than that previously deduced from a 1D chain approach. Our set of parameters gives, i) a value of the distortion which agrees well with some recent estimate for a lower bound, we are able to perfectly reproduce ii) the dispersions, iii) the experimental susceptibility at both high and low temperatures. By performing DMRG calculations for 2 coupled chains we have analysed the effect of the transverse coupling on the ratio of singlet to triplet gaps. The ratio is very sensitive to the parameters and the universality reported in the strict one dimensional case is lost. As an additional point, we provide a simple picture to explain the interesting new feature observed in recent inelastic neutron scattering experiments: the existence of a second branch of excitations.

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I. INTRODUCTION

The discovery of the non-organic compound CuGeO$_3$ has attracted considerable attention as a laboratory for low dimensional many-body quantum mechanics. This compound is believed to exhibit a spin Peierls transition at $T = T_{Sp}$. Below this temperature the ground-state is dimerized, and simultaneously a gap opens in the excitation spectrum. Most of the attempts to extract the appropriate magnetic couplings have been in the framework of purely one dimensional system. This restriction to a one-dimensional picture was mainly justified by the features of Inelastic Neutron Scattering, the dispersion is the largest in the chain direction (c-direction) \( c \). The model which has been widely studied that with dimerization and frustration:

\[
H_{1D} = J_c \sum_i \left[ (1 + \delta(-1)^i) \mathbf{S}_i \cdot \mathbf{S}_{i+1} + \alpha \mathbf{S}_i \cdot \mathbf{S}_{i+2} \right] \quad (1)
\]

\( \delta \) measure the distortion of the lattice at zero temperature and \( \alpha \) is the measure of frustration, the ratio of second-nearest to nearest neighbour antiferromagnetic exchange. Attempts to fit the susceptibility data and the singlet-triplet gap have provided two different set of parameters \( (J_c, \alpha, \delta) \):

a) \( (J_c = 150K, \alpha = 0.24, \delta = 0.030) \) in ref. \cite{1}

b) \( (J_c = 160K, \alpha = 0.36, \delta = 0.016) \) in ref. \cite{2}

The discrepancy in these two sets of parameters came from differing emphasis in the fitting procedures: In reference \cite{2} the value \( \alpha \) was first constrained to be at most 0.24 by the observation that the triplet gap scaled with temperature as a power of the lattice distortion and should thus be less than the (one-dimensional) values giving a spontaneous dimerization. On the other hand it was made as large as possible to give a reasonable fit to the susceptibility. The distortion \( \delta \) was fit from the dispersion at low temperatures. In reference \cite{2} the value of \( \delta \) was simply chosen to make the best fit possible to the susceptibility and \( \delta \) was taken to fit the triplet gap at zero wave vector.

Recently, Fabricius et al. \cite{3} carefully reexamined the fit of the susceptibility to a strictly one-dimensional model. They deduced a value of \( \alpha = 0.354 \), which is almost identical to the estimate of Riera and Dobry. Their fit reproduced perfectly the susceptibility from 40\( K \) up to 1000\( K \). For this reason the value \( \alpha = 0.36 \) has been preferred to \( \alpha = 0.24 \) and has been considered as the value from susceptibility measurements.

In fact the issue is not settled, as apart from the observed relation of the gap to the lattice distortion, Raman scattering experiments lead to a value of \( \alpha \) which is very much smaller, as we shall see: a value of \( \alpha \) close to 0.2 was suggested. The argument for this is as follows: It is known that the a system described by (1) can exhibit a singlet bound-state excitation below the continuum \cite{5}. A recent detailed analysis have shown that it appears at \( q = 0 \) momentum when the frustration is strictly non zero, for any given distorsion of the lattice \cite{6}. The most interesting feature is that the ratio \( R = \Delta_s/\Delta_t \) of the gap to singlet excitations measured in Raman scattering and the gap to triplet excitations as visible in inelastic neutron scattering was shown to be
a universal function of the frustration parameter, i.e. is independent of the dimerization parameter. This then, provides a direct tool to measure the amplitude of the frustration. Singlet excitations are directly observable in Raman scattering experiments. Such a bound state was first observed by Kuroe et al.\[6\]. The excitation energy is $\Delta_\perp \approx 1.78\Delta_\parallel \[3\]$, it could be directly concluded from fig.1 in ref.\[3\] that this leads to $\alpha \approx 0.20$. Note that this value is very close to that proposed by Castilla et al\[1\]. In contrast a value of $\alpha \approx 0.35$ would give $R = 1.50$, inconsistent with the experimental value.

In conclusion, a description purely in terms of uncoupled chains lead to a conflict:

i) Susceptibility data $\rightarrow \alpha = 0.36$.

ii) Raman scattering data $\rightarrow \alpha \approx 0.2$.

The two values proposed suggest very different picture of the spin-Peierls transition: for a single chain there is a critical $\alpha_c = 0.24$ below which dimerization requires a coupling to phonons, and above which even without coupling to the lattice the magnetic chain spontaneously develops dimer order. While this may seem an academic point, given that a lattice dimerization is observed, but it is not if we wish to give a quantitative account of the physics: the estimate of that exchange dimerization depends on the frustration. It should be also stressed that Braden et al. have recently given an estimate of the lower bound value of the distortion parameter which has clearly excluded the value $\delta = 0.016$ of scenario b)\[9\]. Indeed they have found that $\delta > \delta_{\text{min}} = 0.03$. To solve this issue some have argued that an adiabatic approach was not appropriate and that the phonons degrees of freedom should be treated exactly (‘dynamical phonon picture’)\[10\]. All recent attempts to include dynamical phonons were made in the framework of strictly 1D system. Unfortunately, because of the large size of the Hilbert space, the exact calculations are restricted to very small cluster\[11\].

In the following we will show that if we take into account properly the transverse coupling between chains, these fundamental issues are solved. Within our approach, described in the next section, we will be able to reproduce perfectly many experimental features and provide a more realistic value of the distortion parameter $\delta$.

II. MINIMAL 2D MODEL

As was already mentioned in the first experimental papers, CuGeO$_3$ is not a strictly one-dimensional compound: a naive spin wave approach provide an estimate of the antiferromagnetic coupling in the perpendicular direction to the chain (b-direction), $J_b \approx 0.1J_c$, which is not so small\[12\]. Unfortunately, the problem with a two-dimensional system is that much less is known both analytically and numerically. Numerical methods such as Exact Diagonalization and Density Matrix Renormalization Group can resolve only small clusters far from the thermodynamic limit.

Most attempts to study CuGeO$_3$ as a quasi 2D compound were essentially within the framework of mean field theory\[3\]. An alternative, and powerful, approach for weakly coupled low-dimensional systems is to maintain a precise treatment of the one dimension and treat only the interchain coupling perturbatively. We shall follow this method and supplement it with exact treatment of two chains for certain parameters as a check on the numerical parameters found. The low temperature phase of CuGeO$_3$ actually has a checker-board structure\[3\]: the dimerization alternates from chain to chain. The minimal magnetic Hamiltonian is then:

$$H_{2D} = H_{1D} + J_\perp \sum_{i,r} S_{i,r} \cdot S_{i,r+1}$$

(2)

with,

$$H_{1D} = J_c \sum_{i,r} ([1 + \delta(-1)^{i+r}]S_{i,r} \cdot S_{i+1,r} + \alpha S_{i,r} \cdot S_{i+2,r})$$

(3)

The variable $r$ counts the different chains. The factor $(-1)^r$ explicitly takes into account the fact that the structure of the real system is checker-board.

Starting from the limit of strong dimerization (in which triplet excitations are on nearest neighbour sites), we can treat the coupling between the chains perturbatively. It is easy to find, at the lowest order, that the dispersion is,

$$\omega(k_\parallel, k_\perp) = \omega_{1D}(k_\parallel) - J_\perp \cos k_\perp$$

(4)

where $k_\parallel$ is the wave-vector along the strongly coupled direction and $k_\perp$ is transverse. Eq.\[4\] implies the simple relation:

$$\Delta = \Delta_{1D} - J_\perp$$

(5)

$\Delta$ is the real gap (including $J_\perp$), and $\Delta_{1D}$ denotes the gap in absence of coupling between the chains.

Note that if the structure were not checker-board, but simply repeated in the b direction the dispersion would be different: $\omega(k_\parallel, k_\perp) = \omega_{1D}(k_\parallel) - J_\perp \cos k_\perp$.

While equation (4) is strictly true for very strong dimerization, ie $J_\perp < < \omega(k_\parallel)$ it is a very good approximation even when this is not satisfied, as we have verified by diagonalisation of two chains using Density Matrix Renormalization Group (DMRG). We then exploit this equation and use the exact one-dimensional dispersion from exact diagonalisation $\omega_{1D}(k_\parallel)$. From exact diagonalisation, in the parameter region relevant for our discussion, a good fit for the dispersion is\[13\],

$$\omega_{1D}(k) = J_c \sqrt{a - b \cos(2k)}$$

(6)

where $a = \frac{1}{2} + \frac{\Delta_{1D}}{J_c} + (\frac{\Delta_{1D}}{J_c})^2$ and $b = \frac{1}{2} + \frac{\Delta_{1D}}{J_c}$.
III. FIXING THE PARAMETERS.

In order to fix the parameters of the model, let us first start with the transverse coupling \( J_{\perp} \). For this purpose, we will use the experimental data for the dispersion as measured by inelastic neutron scattering [4]. From equation (4) the dispersion in the \( b \)-direction gives the transverse coupling from the difference between the spin wave energy at \( q_0 \) the band edge and the centre, one has directly

i) \( J_{\perp} = (5.6 - 2.0)/2 = 1.8 \text{meV} \)

From equation (4) the width of the 1D dispersion, \( \omega(\pi/2,k_{\perp}) - \omega(0,k_{\perp}) = \omega_{1D}(\pi/2) - \omega_{1D}(0) = \Delta \omega \). From eq. (4) the width of the 1D dispersion is \( \Delta \omega \approx J_c \). Thus using the given experimental data, we find,

ii) \( J_c \approx 12.2 \text{meV} = 146 \text{K} \)

Hence, from i) and ii) we obtain \( J_{\perp} \approx 0.15 J_c \), this value of the transverse coupling confirms the poor 1D character of \( \text{CuGeO}_3 \). Note that the ratio of couplings is somewhat larger than estimated by the theorists who used an expression appropriate for a gap induced by anisotropy. As the measured anisotropy in spin is small \( \text{this is not appropriate.} \)

Now our task is to fix the value of the two remaining parameters, \( \alpha \) and \( \delta \). As said before, this was easy in the 1D picture, since the ratio singlet-triplet gap was shown to be a universal function of the frustration parameter only. Unfortunately, this is not the case anymore in the 2D model \( (J_{\perp} \neq 0) \). This will be illustrated later on. An alternative is to make use the high-temperature susceptibility data. As a reference for the high-T data, we have recalculated for the one-dimensional system the susceptibility with \( \alpha = 0.35 \) and \( J_c = 156 \text{K} \) and \( g = 2.25 \), since with this set of parameters, the experimental data have been perfectly reproduced from 40K up to 1000K \( \text{[3]} \). It is easy to get the first terms in the high T expansion for the susceptibility, in this limit one gets straightforwardly,

\[
\chi(T) = \frac{N_A}{k_B T} (g \mu_B)^2 (a_0 - \frac{\alpha_1}{T})
\]

where \( a_0 = \frac{1}{2} \) and \( \alpha_1 = \frac{J_{\perp}}{J_c} (1 + \alpha + J_{\perp}/J_c) \)

For the purely 1D case, i.e \( J_{\perp} = 0 \), one requires a value of \( \alpha_1 D = 0.35 \). Thus the condition to reproduce the high T part of the susceptibility is,

\[
\alpha_{\text{reed}} + \frac{J_{\perp}}{J_c} \approx \alpha_1 D \approx 0.35 \tag{8}
\]

Since \( J_{\perp} = 0.15 J_c \), the previous equation fixes unambiguously the amplitude of the frustration:

iii) \( \alpha_{\text{reed}} \approx 0.20 \)

From this relation, it can be concluded that a purely 1D approach overestimates the real value of the frustration parameter. In other words, to reproduce the High T behavior of the susceptibility in the one-dimensional limit, one requires a larger value of the frustration parameter.

As a check, we have performed the exact calculation of the susceptibility for a two chains system. This was done for a \( 2 \times 8 \) system, using periodic boundary conditions in both directions, this means for the two chain problem changing \( J_{\perp} \) to \( 2J_{\perp} \). For this size we can fully diagonalize the Hamiltonian in each subspace and calculate the thermodynamic functions. Since we are interested in the high temperature phase we have set \( \delta = 0 \). Note that, at high temperature the number of chains does not limit the accuracy of the calculation. In fig.3 we see that the agreement between the experimental data and the high-T expansion is very good for sufficiently large temperatures. We also see that the agreement with the experimental data is very good down to 100 K. One should not pay too much attention to the fact that the pure one-dimensional approach was very good down to 45K.

In our case, the finite size effect are definitely stronger: the chains are shorter and at low temperature the number of chains is important in the present case we have considered only two chains. Nevertheless the agreement goes beyond the strict applicability of the leading term.

The last step consists in fixing the single remaining parameter, i.e the static distortion amplitude. For this purpose, we had to perform a calculation of the gap in the one-dimensional case, with the frustration parameter set to \( \alpha = 0.2 \). Using eq. (5) one immediately deduces \( \Delta_{1D} = 0.322 J_c \). Following the method of ref. \( \text{[1]} \) to extrapolate the data in the thermodynamic limit, we have obtained straightforwardly,

iv) \( \delta \approx 0.065 \)

We remark that this value is significantly larger that previously reported: it is almost 5 times larger than the value obtained in the purely one-dimensional approach \( \text{[3]} \) and closer to that estimated from the structure \( \text{[9]} \). At this point all the parameters of the model have been fixed from the experimental data. In order to test the validity of our set of parameters, we must confront it to different tests. This will be done in the following section.

IV. PARAMETER TESTS.

The realistic value of \( \delta \) we have found is already a first check of the validity of our set of parameters. We now proceed to a second test; let us check that we can accurately reproduce the dispersion data in both directions. Using eq. (6) and (7) we have performed the direct calculation of the dispersion in the chain and transverse directions. The plot of fig.3 is rather convincing, the agreement between experimental and theoretical data is excellent.

Now, as a third check of the validity of our approach, let us perform the calculation of the low temperature susceptibility \( (T < T_{SP}) \) and compare it to available experimental data. For this purpose we assume that \( \delta(T) = \delta \), which is reasonable except very close to the transition point \( \text{[1]} \). Following the method of ref. \( \text{[7]} \) the suscepti-
bility is accurately given by:

\[ \chi(T) = \frac{N_A}{k_B T} (g \mu_B)^2 \frac{z(T)}{1 + 3z(T)} \]  

whith,

\[ z(T) = \frac{1}{(\pi^2)^2} \int dk \|dk \| \text{exp}(-\frac{\omega(k)}{T}) \]  

The expression for the dispersion is given by equation (9).

We have plotted in fig. 4 both the calculated susceptibility at low T and the experimental data [18]. We observe that the agreement is very good up to \( T \approx 0.85 T_{SP} \). Note that between 0.85T_{SP} and T_{SP}, one naturally expects a deviation from the experimental data due to the sudden drop of \( \delta(T) \) when approaching the transition temperature. As a remark one has to keep in mind that we did not use any fitting parameter.

At this point, it will be also an interesting point to estimate with our set of parameters the energy of the singlet bound state. As a preliminary step, let us first analyse the effect of the transverse coupling on the singlet-triplet ratio. Indeed, as said in the introduction part, the one dimensional calculation have shown that this ratio does only depend on the frustration only, leading in this case to \( \alpha \approx 0.2 \), to get the experimental value \( R_{exp} \approx 1.8 \). As we are now considering a weak, but non-zero transverse coupling we must know whether this ratio will change substantially with interchain exchange. To test this we have carried out Density Matrix Renormalization Group calculations on the system of two coupled chains.

V. EFFECT OF J_⊥ ON THE SINGLET-TRIPLET RATIO R.

We have performed numerical calculations by applying the DMRG method [14] on the model defined by the Hamiltonian in Eq. 1. Since this method is more accurate for systems with free ends, we will consider our two coupled chain model with open boundary conditions in the long direction. An unfavorable consequence of the DMRG is that the total momentum is not a good quantum number. As our aim is to determine the singlet and triplet energy gaps at \( k = 0 \) momentum we have used the spin reflection symmetry to rule out excited levels belonging to other \( k \) values [20]. The singlet gap was calculated from the energy difference of the two lowest lying energy levels in the \( S^z = 0 \) spin sector with odd parity under spin reflection while the triplet gap was obtained from the lowest levels of the \( S^z = 0 \) and \( S^z = 1 \) subspaces. Another unfavorable consequence of open boundary conditions, as in the nearest-neighbor valence-bound configuration of the bilinear biquadratic model [21], is that free \( S = 1/2 \) spins remain at the ends of one of the chains, giving rise to a fourfold degenerate ground state. This is exactly the case for any finite chain length for the extreme dimerization limit (\( \delta = 1 \)) and is true asymptotically for \( \delta < 1 \). Since the extra degeneracies make the analysis of the spectrum more difficult we have removed the two outmost spins on one chain and set the end-coupling to \( J = 1 + \delta \) on the other chain. It worth mentioning that the total symmetry of all states becomes opposite under the spin reflection symmetry as it had been for the original problem. In most of the calculations we have used the more accurate version of DMRG, the so-called finite-lattice method to determine the energies more precisely.

To further improve the efficiency of the calculations, we have also included the left-right reflection symmetry and all the investigated states were targeted independently. Since for finite dimerization the system is gapped, it was adequate to keep 100 – 200 block states to have the truncation error below \( 10^{-5} \) for chain up to 100 sites. The absolute error of our calculation was estimated by comparing the ratio of the singlet and triplet energies (\( R \)) obtained for the ladder model using the DMRG procedure at \( \alpha_c = 0.24, J_\perp = 0 \) to the exact value \( R = \sqrt{3} \) given by the sine-gordon model. For stronger dimerization the deviation was \( 10^{-3} \) while for weaker dimerization it was found to be \( 10^{-2} \).

The results for the ratio of the singlet and triplet energy gaps (\( R \)) as a function of \( J_\perp \) in the strong dimerization regime (\( \delta = 0.2 \)) and \( 0.2 < \alpha < 0.26 \) values are plotted in fig. 4. It is seen from the figure that there exists a finite region of \( J_\perp \) where the value of \( R \) is below 2, thus we have confirmed the the bound state is stable even in the 2D case for not too large interchain couplings. As a second result, we have found that the triplet gap is a linear function of \( J_\perp \) in agreement with eq. 1. In general the coefficient of this linear function should be different from unity and indeed this is apparent from the two chain results. In order to estimate parameters, however, we take the linear coefficient as unity as the two chain estimate is too heavily biased by finite (transverse) size: doubling the transverse coupling to take into account the periodic boundary condition clearly overestimates the correlations in the two transverse directions. The nonlinear behavior of \( R \) follows from the nonlinear dependence of the singlet gap on \( J_\perp \). Taking the parameters for the frustration and \( R \) obtained from the 1D chain calculations, namely \( \alpha = 0.2, R = 1.80 \), one finds the same value of \( R \) for a different parameter set: \( \alpha \approx 0.26 \) \( J_\perp \approx 0.12 \).

In order to check the effect of dimerization in the case of 2D system we have performed the similar calculations, but for a weaker dimerization parameter \( \delta = 0.065 \). The result for \( R \) as a function of \( J_\perp \) is also shown on the figure. It is apparent from the curves that the \( R \) depends on \( \delta \), thus the ratio is no longer a universal function of the frustration parameter for any finite interchain coupling. On the other hand, we have found the triplet gap is linear function of the interchain coupling and the slope does not depend on the strength of the static distortion.

It is clear that for weaker dimerization the ratio gets very sensitive to the interchain coupling. For a fixed frus-
tration, increasing interchain coupling leads to the disappearance of the bound state into the continuum. The fact that the ratio exceeds 2 means only that this state is no longer well defined. For instance, we find that when $\alpha = 0.2$ the singlet bound-state disappear into the continuum for $J_{\text{perp}} > 0.1$. While the value of the ratio of singlet to continuum can no longer be used to extract directly a value of the frustration, the very existence of the state gives a lower bound on the frustration. To conclude this section, one can say that it is extremely difficult from this two chain analyse to reproduce the experimental value of the singlet-triplet ratio: one requires very precise values of the parameters of the model.

VI. EXISTENCE OF A SECOND BRANCH IN INS

Up to now we have assumed that the magnetic structure is of equivalent but coupled chains at high temperature and below the spin-Peierls transition an identical dimerization along each chain in a checker-board structure, ie the dimerization out of phase from chain to chain. There are two modifications to this view: one which we take to be unconfirmed, the other which we take to be reliable.

The first is that X-ray analysis of the room temperature structure have suggested a larger unit cell $[22,23]$ even above the spin-Peierls transition, in contrast to that found by neutrons $[2]$. Recent examination by neutron diffraction $[24]$ confirmed the earlier neutron results that these distortions are absent in the crystals studied by neutron diffraction, so while the differences in the experimental structures remain to be fully elucidated, we shall assume that the structure at high temperatures is as taken.

The second modification we take as necessary to take into account. Recent inelastic neutron results $[21]$ on the dynamics have shown a new magnetic mode, that the experimentalists call an optic mode. We shall now show that the dispersion and amplitudes of this second mode are well explained by a slight refinement of the Hamiltonian considered so far in supposing two inequivalent structures at high temperatures. As the numerical differences are small, this alters very little the dispersion and the susceptibility at low temperatures. We now give a simplified version based on the limit of strong dimerization.

It is enough for our purpose to consider a system which consists of two inequivalent chains $A$ and $B$ coupled antiferromagnetically through $J_{\perp}$. We first define the following set of parameters $(J_A = J_c + dJ, \delta_A = \delta + d\delta, \alpha_A = \alpha + da)$ and $(J_B = J_c - dJ, \delta_B = \delta - d\delta, \alpha_B = \alpha - da)$ respectively for the chain $A$ and $B$ where it is assumed that $dJ$, $d\delta$ and $da$ are small. We also define $\omega_A(k_{\parallel})$ and $\omega_B(k_{\parallel})$ as the excitation energy for each chain, and $\Phi_A(k_{\parallel})$, $\Phi_B(k_{\parallel})$ the associated eigenvectors. In the strong dimerization limit, it is relatively easy to perform the calculation of the eigemodes. For a given $k_{\parallel}$, there are two possible modes $\omega_+$ and $\omega_-$ corresponding to $k_{\perp} = 0$, or $\pi$.

\[
\omega_{\pm}(k_{\parallel}) = \omega(k_{\parallel}) + \sqrt{\delta\omega(k_{\parallel})^2 + [J_{\perp}\cos(k_{\parallel})]^2},
\]

Their corresponding eigenstates are,

\[
\Psi_{\pm}(k_{\parallel}) = \Psi_A(k_{\parallel}) + (1 - \frac{\delta\omega(k_{\parallel})}{J_{\perp}\cos(k_{\parallel})})\Psi_B(k_{\parallel}),
\]

where $\omega = (\omega_A + \omega_B)/2$ and $\delta\omega = (\omega_A - \omega_B)/2$. For simplicity we have assumed that we are only working in the vicinity of $k_{\parallel} = 0$ or $\pi$ and that $\delta\omega \ll J_{\perp}\cos(k_{\parallel})$, which is reasonable since the chains are only weakly inequivalent. Now the crucial question is: Will we effectively observe both branches in INS experiment?

To answer this question we have to calculate the matrix elements, $F_+ = \langle \Psi_+ | S(k) | \Psi_GS \rangle$ and $F_- = \langle \Psi_- | S(k) | \Psi_GS \rangle$, where $| \Psi_GS \rangle$ is the Ground-state wave function and $S(k) = S_A^2(k_{\parallel}) + e^{ik_{\perp}b}S_B^2(k_{\parallel})$. When performing the calculation at the lowest order one gets,

\[
F_+ = f_0(1 + e^{ik_{\perp}b} + \frac{\delta\omega(k_{\parallel})}{J_{\perp}\cos(k_{\parallel})}) - df_0(1 - e^{ik_{\perp}b})
\]

\[
F_- = f_0(1 - e^{ik_{\perp}b} + \frac{\delta\omega(k_{\parallel})}{J_{\perp}\cos(k_{\parallel})}) - df_0(1 + e^{ik_{\perp}b})
\]

where $f_0 = \langle \Psi(k_{\parallel}) | S^2(k_{\parallel}) | \Psi_GS \rangle$ for an isolated chain, considering the ‘average’ parameters $(J_c, \delta, \alpha)$, $f_0$ corresponds to its variation. If we now fix for instance $k_{\perp} = 0$ then the spectral weight in both branches is,

\[
A_+ = ||f_0||^2(1 + \frac{\delta\omega(k_{\parallel})}{2J_{\perp}\cos(k_{\parallel})})^2
\]

\[
A_- = ||f_0||^2(\frac{\delta\omega(k_{\parallel})}{2J_{\perp}\cos(k_{\parallel})})^2 + ||df_0||^2 + \frac{\delta\omega(k_{\parallel})}{2J_{\perp}\cos(k_{\parallel})} df_0^* f_0^*
\]

It is clear from these expressions that one will effectively observe two excitation branches, one with a significant spectral weight (the main branch) and another one with a relatively small one (secondary), in agreement with the experimental observation. Typically the order of magnitude of the ratio of the spectral weight is,

\[
\frac{A_-}{A_+} \approx \frac{\delta\omega(k_{\parallel})}{2J_{\perp}\cos(k_{\parallel})}
\]
This very simple calculation can be refined to make more quantitative comparison with the available experimental data. Let us repeat again that the previous simplified expressions are only valid in the vicinity of \( k_y = 0 \) or \( \pi \) and we are considering only two chains: ie \( k_\perp \) is by definition 0 or \( \pi \).

VII. CONCLUSION

In this paper we have proposed a minimal model to describe the magnetism of \( \text{CuGeO}_3 \). It is shown unambiguously that the compound is two-dimensional (the coupling in the third direction is very small). We clearly identify the origin of the conflicts in the one-dimensional approach: i) small value of \( \delta \) and ii) large value of \( \alpha \) estimated from the susceptibility data not consistent with the Raman experiments. The susceptibility previously calculated in a pure one-dimensional picture has in fact strongly overestimated the amplitude of the frustration. The smallness of \( \delta \) previously quoted, is simply due to underestimating the 1D gap (see eq. 5) As a test of our set of parameters, we now provide a value of the distortion which is more realistic, we are also able to reproduce perfectly the high temperature (uniform phase) and low temperature (dimerized phase) behavior of the susceptibility without any additional fitting parameter. We could also perfectly reproduce the dispersion in both directions. As mentioned above, it is difficult to reproduce the singlet-triplet ratio from a study of two coupled chains, because of the extreme sensitivity of the ratio to the parameters. However, we believe that it would be interesting to perform the calculation of the Raman intensity, this will provide a measurable effect of the transverse coupling. We believe that this model with the parameters b) \( (J_c = 146 K, \alpha = 0.2, \delta = 0.065, J_b/J_c = 0.15) \) should be the starting point for further studies on \( \text{CuGeO}_3 \). For instance, the need to consider two inequivalent chains, as explained from observation of the extra branch in the inelastic scattering, can be included by taking two slightly different values of the parameter \( \delta \).

As we have remarked, a value of \( \alpha \) well below the critical value for a chain implies that understanding of the dimerization inevitably involves the coupling to the lattice: the observed lattice distortion is not a secondary effect. The change of estimate implies that calculation of dilute phase diagram is quantitatively very different. While we consider the model with these parameters as the correct minimal model consistent with what is currently known.

Copper Germanate is more correctly considered as a (spatially) anisotropic spin system rather than as a quasi-one-dimensional spin chain. The one-dimensional analyses that have been used until now therefore needed to be quantitatively modified. The apparent conflict between parameter sets previously obtained from different experiments reflects the simplification. While we can give a satisfactory account to the experimental results to date, their remain aspects that require precision:

- A more precise value of the interchain coupling requires a quantitative calculation in the anisotropic two dimensional spin system. Our simple estimate based on the strong dimer limit \( J_b/J_c = 0.15 \) may be modified by this. This also implies that \( \alpha \) and \( \delta \) will be also affected.
- The method proposed \( \delta \) to fix the frustration from the ratio of the singlet excitation seen in Raman to the triplet in neutron scattering is correct for a strictly one-dimensional model but must be modified in the two dimensional case. We have verified the linearity of the triple gap predicted by the extreme dimer limit by means of a numerically exact two chain result. The existence and the position of a bound state in the Raman spectrum certainly constrain the parameters, but in a way which defies precise calculation at present. That is why, it may be useful to extend calculations of the Raman scattering to include the form of the continuum.
- As yet there is no detailed understanding of anisotropic terms in the Hamiltonian. The bulk susceptibilities differ \( \alpha \) but this has been interpreted in terms of anisotropy in the tensor of g factors. This may change with better understanding of the polarized neutron scattering cross-sections and/or more quantitative theories of the ESR \( \delta \).
- The observation of the second inelastic branch in the neutron scattering indicates that the magnetic unit cell is doubled by a small inequivalence of adjacent chains in the low temperature phase. This is also seen in recent ESR experiments \( \delta \). It would be interesting to explore what physical parameter is inequivalent.

VIII. ACKNOWLEDGEMENTS

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FIG. 1. Dispersion in both directions. The symbols are experimental data from taken from ref. [1].

FIG. 2. Data for the effect of $J_\perp$ on the Singlet-Triplet ratio R. The distorsion \( \delta \) is fixed to 0.2 (a) and to 0.065 (b). We have considered several values of the frustration parameter $\alpha$.

FIG. 3. Susceptibility at High Temperature. Indirect Comparison between high-T expansion and experimental data."Reference" was recalculated in the 1D picture for a $L = 16$ sites system using the parameters of ref. [4].

FIG. 4. Susceptibility at Low Temperature. We compare the experimental data [18] with the analytical calculations.
\text{delta}=0.2, \text{alpha}=0.2...0.26
$\delta = 0.065$, $\alpha = 0.20, ..., 0.26$
$\chi \text{(emu/mol)}$

$T \text{(K)}$

$J_c = 146 \text{ K}$
$J_b = 0.15 J_c$
$\alpha = 0.2$

-- reference
- - - coupled chains
-- HT expansion
