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Does a Spin-Peierls System Have One Gap or Two?

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We investigated the collective excitations of the spin-Peierls phase of $CuGeO_3$ by inelastic neutron scattering. We measured the dispersion curve of these excitations, with and without magnetic field. The main result is to show that there exist a second gap feature which separate the spin singlet-triplet excitation from a 'continuum' of excitation extending to relatively high energies. Moreover magnetic field produces a loss of intensity in the energy scan.

KEYWORDS: spin-Peierls, spin gap, strongly correlated electrons, inelastic neutron scattering, low-dimensional magnetism

§1. Introduction

The one-dimensionnal $s = 1/2$ Heisenberg antiferromagnetic chain has a disordered state at $T \neq 0$K. Its excitation spectrum is gapless with a well defined dispersive mode, in addition it is surmounted by a region of continuous excitations called the continuum (see for a review ref. 1). When magnetoelastically coupled, to three-dimensionnal phonons, such a highly fluctuating chain is unstable to a dimerization induced by 3d phonons. It undergoes at some $T_{sP} \neq 0$, a spin-Peierls structural transition to a phase where the ground state is a singlet and hence, where magnetism has disapeared. The exchange interaction J along the chain is no longer uniform but alternated (dimerized), with values J and J'. As a consequence the excitation spectrum becomes gapped throughout the whole Brillouin zone.[2)]

This uniform (undimerized) $s = 1/2$ Heisenberg AF chain with nearest neighbor exchange, has a magnetic excitations spectrum (MES) sketched on figure 1(a). Within the frame of a classical approach[2,3)] the dimerized spin-Peierls chain has a gap and its MES is expected to resemble to figure 1(b); yet in a recent letter[4,5)] we presented experimental evidence proving that there are in fact two gaps in this system and not only one as predicted by the classical approach. A simple way of understanding this feature amounts to say that a dimerized system has an obvious excitation which consists of breaking a dimer bond into a triplet at a cost of a certain amount of magnetoelastic energy. The triplet delocalizes along the chain generating eigenstates of definite momentum. However there is another possible excitation in this system because the triplet can absorb a second amount of energy, which corresponds to the second gap, and thus dissociate into two $s = 1/2$ traveling solitons that generate the continuum as shown on figure 1(c). This point of view was discussed by Khomskii *et al.* [6)] that took into account inter-chain elastic coupling.

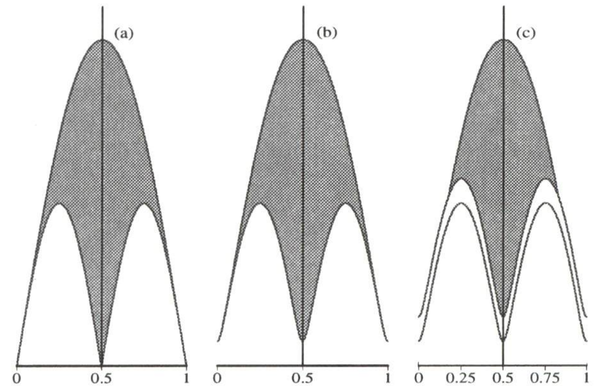

Fig. 1. Sketches of magnetic excitations as function of wavevector in (a) Heisenberg, (b) classical spin-Peierls (c) actual spin-Peierls chains. The shadowed areas are the continuums.

§2. Experiment

Inelastic neutron scattering measurements have been performed on two cold source triple axis spectrometers, 4F1 and 4F2, at Orphée (LLB Saclay). These apparatus have an incident beam that is fixed in direction, it is extracted by a pair of graphite monochromators (the second one being vertically focussing). They were operated at constant scattered wave-vector of k_f=1.55 Å^{-1} or 1.3 Å^{-1} with a horizontally focussing graphite analyser and a berylium filter to cut out higher-order components of the diffracted beam. On 4F2 we installed a vertical magnetic field ranging from 0 to 6 Teslas.

Hase[7)] has proved that $CuGeO_3$ was a spin-Peierls compound. In $CuGeO_3$, Magnetic chains of Cu^{++}, $s = 1/2$ ions are parallel to the c axis.[8)] Copper ions are coordinated by four oxygens. The spin-Peierls gap is observable at $k_{AF} = (0, 0, 0.5) \neq k_{sP}$ or equivalent points, also noteworthy is that no magnetic Bragg peaks could be observed since the ground state is a singlet. Estimates for intrachain nearest neighbor exchange (NNE) along c gave $J \approx J' \approx 120$ K. Estimates for interchain NNE, yielded smaller values of respectively one and two orders of magnitude, along directions b and a.[9)]

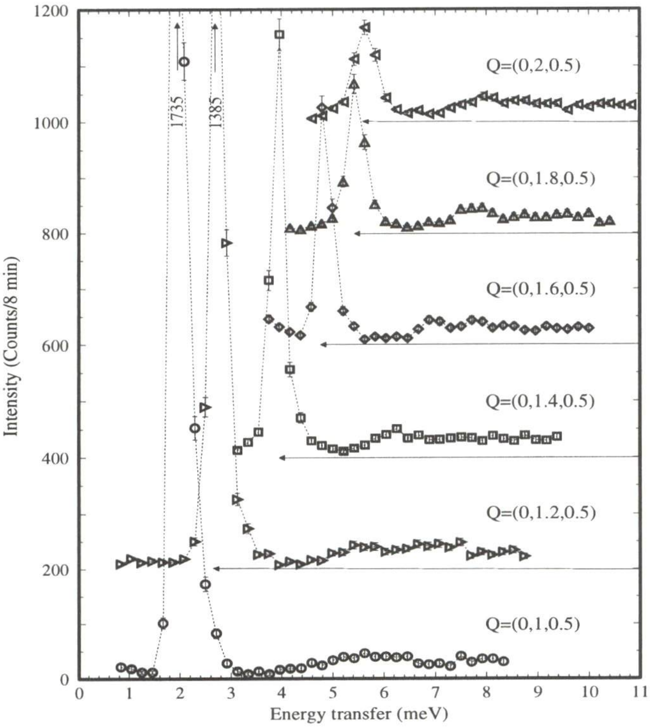

Fig. 2. Six energy scans at $Q = (0, Q_b, 0.5)$ for $Q_b = \{1, 1.2, 1.4, 1.6, 1.8, 2\}$ at $T = 1.7$ K. The sharp peak of the magnon-like mode, the 'solitonic gap' and the continuum are clearly visible in all scans. The peak drifts towards higher energy due to interchain coupling.

§3. Results

Figure (2) shows a series of energy scans performed at several points in reciprocal space, regularly spaced along Q_b between $Q = (0, 1, 0.5)$ and $Q = (0, 2, 0.5)$, they were made at $T = 1.7$ K. If we examine the scan at $Q = (0, 1, 0.5)$ we see from left to right 1- Firstly the gap that is expected by the classical theory, called hereafter the 'triplet gap' it has a value of $\Delta = 1.93$ meV at $Q = (0, 1, 0.5)$. 2- A very sharp magnon-like peak first observed by Nishi *et al.*[10] This peak correspond in fact to a spin-triplet mode as shown by measurements under magnetic field[11,12] where it has been observed that it splits in three. Its asymmetric shape is due to the steep curvature of this dispersive mode in the vicinity of $Q = (0, 1, 0.5)$. 3- The scattering between the middle peak (the triplet mode) and the plateau, falls to the background level. This is clearly a new gap in energy that we call hereafter the 'solitonic gap'. At $Q = (0, 1, 0.5)$, this 'solitonic gap' is slightly higher than 2 meV. 4- Finally, brought out by the 'solitonic gap' we find some signal (low but clearly present) which constitute the expected continuum that extends at least up to the maximum energy transfer permitted by the configuration of the instrument that we used, i.e. 11.5 meV.

The dispersive mode followed by the 'solitonic gap' and the continuum are visible on this series of scans. Owing to coupling between chains there is a small dispersion along Q_b and the positions of the peaks are not constant in energy as it should be expected for a pure one-dimensional system. Here, some technical remarks are due concerning the nature of the scattering above the "solitonic gap". The examination of this scattering suggests that it is magnetic in origin, but what is the evidence for this. The Q dependance can be used to check if the scattering follows a magnetic form factor dependance. Such an expression for the magnetic from factor of two dimerized spins 1/2 is presently not available. Nevertheless the intensity associated with the singlet-triplet excitation varies very strongly with Q_b as Q_b is varied from 1 to 2 in the scans of the form $Q = (0, Q_b, 0.5)$, although this may be due to its dispersion in energy. Ideally polarized neutrons scattering would answer this question appropriately. The argument of the Q-dependance would not point conclusively towards a purely magnetic origin as the continuum (above the solitonic gap) seems to vary little with Q_b in contrast to the singlet triplet excitation peak. Therefore it is possible that the continuum of scattering is due to some spin charge hybridization. This raises a question: what is the relationship between the continuum of scattering above the "solitonic gap" at low temperatures and the continuum of scattering known to exist in the uniform state of the $s = 1/2$ Heisenberg chain? Presumably this is the situation one has in $CuGeO_3$ for temperatures above $T_{sP} = 14.2$ K.

We felt necessary to investigate the whole dispersion curve of $CuGeO_3$ under magnetic field, and doing this we came across a new anomalous behavior. Figure 3 shows two energy scans on a point equivalent to the one at $Q_c = 0.25$ on figure 1(c)); left H=0 T, right H=5.5 T. On each scan we see two groups, in the first one at low energy, we identified a spurion ($k_i = 2k_f$) and two phonons, while above 3.25 tHz we see the MES group. We have tentatively fitted the latter group into 4 resolution limited peaks, three for the dispersive mode plus one at the highest energy, for the continuum; note that the program we used for fitting the data takes into account the slope of the dispersion curve. The new information brought by these measurments is that 1- the dispersive mode is already splitted in absence of magnetic field as soon as we depart from k_{sP} and 2- the intensity of MES decreases with increasing field while the positions of the different four peaks seem not to be affected. Note also that the spurion and the phonons on the low energy side are not affected by the field as we could expect. We can only rule out the attribution of this spontaneous splitting to anisotropy since it would uniformely split the dispersion curve even at $Q_c = 0.5$, obviously not the case,[4] we incline to believe that this effect has more to do with spin-phonon coupling.

§4. Discussion

We wish to draw attention on some aspects of the excitations in $CuGeO_3$ that behave in a non conventional manner, wondering to what extent these are representative of the ideal one-dimensional spin-Peierls system.

Much of the theoretical work on spin-Peierls systems incorporate a frustrating exchange coupling between next nearest neighbors, partly ignoring phonon dynamics; the lattice distortion being considered as static and simply producing an alternating exchange between nearest neighbors. Khomskii *et al.* [6] have proposed a

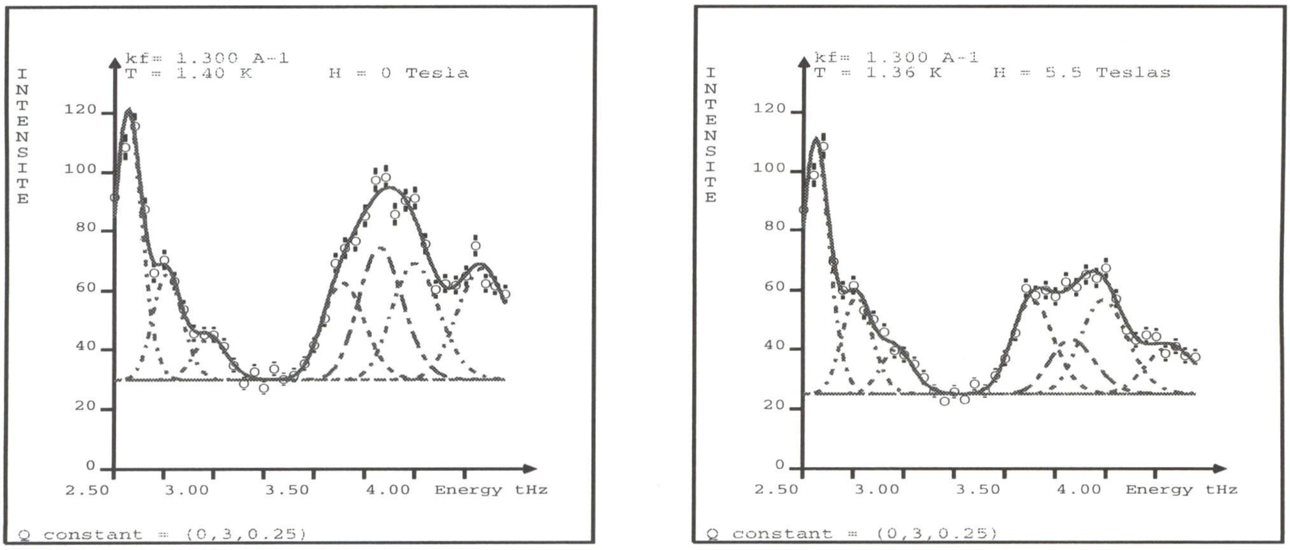

Fig.3. Energy scans at $Q = (0, 3, 0.25)$ (the horizontal axis is in tHz; 1 tHz = 4.13 meV), left figure $H = 0$ T and $T = 1.4$ K; right figure $H = 5.5$ T and $T = 1.36$ K.

simple theory of the spin-Peierls transition based on solitons; they take into account the one-dimensional phonon dynamics exactly, and introduce the interchain couplings through mean field theory. In their approach the basic excitations of the system are solitons.

Let us define a set of elementary diagrams(altough inacurate but quite handy) , to be used as visual aids. Labelling a dimer in its singlet state by: $\bullet - \bullet$, the triplet state by $\Uparrow$ and a spin 1/2 on a copper site by $\uparrow$ (soliton s) or $\downarrow$ (antisoliton $\bar{s}$), then we can illustrate the states of the spin-Peierls chain in diagrammatic language. The travelling triplet which generates the sharp peaks shown on figure (2) can be schematically represented by $\bullet - \bullet \quad \bullet - \bullet \quad \Uparrow \quad \bullet - \bullet \quad \bullet - \bullet$ (again, please note that strictly speaking this is not an eigenstate) and then above the 'solitonic gap' the continuum would correspond to delocalized spins 1/2 such as

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It is easy to see on the above diagram that the pairing of dimers in between a couple $s\bar{s}$ (on the third line), is opposite to the pairing on the neighboring lines, this costs an energy proportional to the $s\bar{s}$ separation x. In such a model the soliton-antisoliton are confined in a potential V and their effective Hamiltonian is given by (see Affleck[13]):

$$H_{eff} = -\frac{1}{2m}\frac{d^2}{dx^2} + V|x|$$

This model has discrete boundstates and explains quite naturally the existence of at least two gaps. Note that if the separation between s and $\bar{s}$ stretches too far, then it will eventually become more favorable to nucleate a new triplet excited dimer in between them as sketched below:

$\bullet - \bullet \quad \uparrow \quad \bullet - \bullet \quad \bullet - \bullet \quad \Uparrow \quad \bullet - \bullet \quad \bullet - \bullet \quad \downarrow \quad \bullet - \bullet$

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