Excitations of the Field-Induced Quantum Soliton Lattice in $CuGeO_3$

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(Received 2 August 2000; published 4 October 2001)

The incommensurate magnetic soliton lattice in the high-field phase of a spin-Peierls system results from quantum fluctuations. We have used neutron scattering techniques to study $CuGeO_3$, allowing us to obtain the first complete characterization of the excitations of the soliton lattice. Three distinct excitation branches are observed, all of which are gapped. The two highest energy modes have minimum gaps at the commensurate wave vector and correspond to the creation or annihilation of soliton pairs. The third mode is incommensurate and is discussed in relation to theoretical predictions.

DOI: 10.1103/PhysRevLett.87.177203 PACS numbers: 75.10.Jm, 75.45.+j, 75.50.Ee, 75.50.Gg

The Heisenberg uncertainty principle implies that a single particle with one well-defined physical variable experiences quantum fluctuations of its conjugate variable. In many-particle systems, coupling produces collective quantum fluctuations which reduce, and can even completely quench, the classical order parameter. What then emerges is a new macroscopic quantum ground state without a classical analog. Often, the wave functions of these quantum ground states cannot be accessed directly by experiments. However, each of them has a characteristic excitation spectrum, which determines the physical properties of the system. Perhaps the simplest example of such a macroscopic quantum ground state is the one-dimensional spin-$\frac{1}{2}$ Heisenberg antiferromagnet (AF), which is disordered even at $T = 0$ due to quantum fluctuations. Coupled to a three-dimensional (3D) phonon field, it can undergo a spin-Peierls phase transition at a finite temperature into a dimerized quantum ground state (D phase). Although both have zero total spin, the ground states of the spin-Peierls and the unperturbed system are qualitatively different, as revealed by their excitation spectra.

In the D phase of a spin-Peierls material, the lowest magnetic excitation is an isolated triplet of bound domain wall pairs with respect to the dimer order parameter. These domain walls, or solitons, have magnetic and distortive components. The triplet has finite energy over the entire Brillouin zone, with minima at wave vectors 0 and $\frac{\pi}{c}$ (the AF zone center, c being the average distance between two spins in the chain direction). The triplet ($S_{\rm tot} = 1$) becomes Zeeman split in an applied magnetic field. Approximately at the field where the lowest mode softens to zero energy, the domain walls condense into the ground state. They repel each other and are frozen into a static incommensurate (IC) pattern by the 3D elastic interactions. The amplitude of the antiferromagnetic modulation reaches about 10% of the full spin value only, and is oriented entirely parallel to the magnetic field. For the classical Heisenberg antiferromagnet, the staggered magnetization flops perpendicular to the field.

In contrast to other IC magnetic structures, the IC modulation of the spin-Peierls high-field phase is caused entirely by quantum fluctuations, which makes it very interesting to investigate its excitation spectrum. Of the great variety of quantum ground states that can be constructed theoretically, it represents one of the few for which physical realizations are known. Prior to this paper, the excitation spectrum was addressed only in a limited number of theoretical studies [1–5], with very little supporting experimental work [6,7].

$CuGeO_3$ is the first spin-Peierls compound where large single crystals are available [8], thus allowing the complete phonon and magnetic excitation spectra to be measured using neutron scattering techniques. The IC phase is also accessible, since magnetic fields up to 14.5 T have become available for neutron experiments, well in excess of the critical field in $CuGeO_3$ of $\mu_0 H_c \approx 12.5$ T. Our recent neutron scattering study of the soliton lattice in the IC phase [9] reveals a static magnetic modulation with the same period as the distortive modulation, and in particular the superlattice Bragg peaks at $(\frac{1}{2}, 1, \frac{1}{2} \pm \delta k_{\rm sp})$ were found to be almost entirely magnetic in origin. More importantly, the staggered magnetization was indeed found to be collinear with the magnetic field, as expected for a quantum IC phase, and quantities, such as the soliton width and amplitude, were found to be in good agreement with theoretical predictions.

In this Letter we report on the magnetic excitation spectrum in the IC phase, measured close to the superlattice peak $(\frac{1}{2}, 1, \frac{1}{2} \pm \delta k_{\rm sp})$ and also at the AF zone center $(0, \frac{1}{2}, 1)$, where the dispersion of the magnetic excitations

 0031-9007/01/87(17)/177203(4)$15.00

in the D phase has its minimum. The experiments were performed at the cold triple axis spectrometer FLEX at BENSC, Berlin, using the vertical cryomagnet VM-1. Single crystals of $CuGeO_3$ of 0.34 and 0.49 cm^3 were aligned in the $(0, k, \ell)$ and $(h, 2h, \ell)$ scattering planes, respectively. The collimation was guide-60′-60′-60′ with either $k_f = 1.3$ Å fixed and a Be filter in k_i, or $k_f = 1.54$ Å and the Be filter in k_f. The critical fields at $(0, 1, \frac{1}{2})$ and $(\frac{1}{2}, 1, \frac{1}{2})$ differ (12.58 and 12.31 T) because of the different field directions and hence g factors (2.152 and 2.199) in the respective scattering geometry.

Figure 1 shows the magnetic field dependence of the excitations at the AF zone center $(0, 1, \frac{1}{2})$. With increasing field, the lowest triplet branch decreases to 0.43 ± 0.02 meV but never becomes completely soft. At the critical field, two new sharp excitations appear at 0.61 ± 0.02 meV and 1.01 ± 0.03 meV. The 1 meV mode shows barely any field dependence between 12.5 and 14.5 T, while the energy of the lower mode increases to 0.76 ± 0.02 meV. The excitations at $(\frac{1}{2}, 1, \frac{1}{2})$, close to the magnetic Bragg peaks at $(\frac{1}{2}, 1, \frac{1}{2} \pm \delta k_{sp})$, display the same field dependence as those at $(0, 1, \frac{1}{2})$, apart from an overall shift towards higher energies (1.33 ± 0.02 meV and 1.61 ± 0.03 meV at 14.5 T) which is entirely accounted for by the ferromagnetic interchain interaction along a^*.

In Fig. 2 we show high-resolution scans at a wave vector of $(\frac{1}{2}, 1, \frac{1}{2})$ for selected magnetic fields. At 12 T, the widths of the middle and the lowest triplet excitation are resolution limited. Both broaden significantly at the critical field (12.31 T) and then disappear. In a small field region, the new sharp IC-phase excitations coexist with the broadened D-phase modes (see scan at 12.5 T), confirming the first-order character of the transition which has also been observed with other methods.

The spectral weight of the two higher excitations in the IC phase is concentrated in a very small region of reciprocal space, extending from the commensurate position $\ell = \frac{1}{2}$ to the incommensurate one at $\ell = \frac{1}{2} \pm \delta k_{sp}$. The upper part of Fig. 3 shows the dispersion from $(\frac{1}{2}, 1, \frac{1}{2})$ along the chain direction c^*, at $T = 2$ K and $B = 14.5$ T, where $\delta k_{sp} = 0.015$. The dispersion (solid lines) appears significantly flatter than the dispersion calculated from the zero field spin-wave velocity (dashed lines). Neither at $(0, 1, \frac{1}{2})$ nor at $(\frac{1}{2}, 1, \frac{1}{2})$ do the dispersions of the two sharp modes exhibit minima at the IC positions $\ell = \frac{1}{2} \pm \delta k_{sp}$.

A search was also made for excitations emanating from the magnetic IC Bragg peaks at $(\frac{1}{2}, 1, \frac{1}{2} \pm \delta k_{sp})$. This revealed a mode at ~0.29 meV (Fig. 4), at $T = 2$ K and $B = 14.5$ T. Fits with Lorentzian line shapes including the Bose factor at 2 K show that the energy of this mode (at the respective IC wave vector) changes little, if at all, between 13 T ($\delta k_{sp} = 0.011$) and 14.5 T ($\delta k_{sp} = 0.015$). The dispersion was studied at $B = 14.5$ T and is plotted in the bottom part of Fig. 3. Its spectral weight is concentrated at the IC wave vector $(\frac{1}{2}, 1, \frac{1}{2} \pm \delta k_{sp})$, the minimum of its dispersion.

In interpreting the data the first thing to establish is whether the three modes shown in Fig. 3 are magnetic or nuclear in origin. It is known from our earlier study that the satellites at $(\frac{1}{2}, 1, \frac{1}{2} \pm \delta k_{sp})$ are predominantly magnetic (by a factor of ~40) [9], and it follows that the low-lying mode emanating from the satellites must also have a predominantly magnetic character. For the higher-lying modes it was observed that their intensity decreased at equivalent ℓ, but larger total wave vectors transfer as expected from the magnetic form factor. They, too, can therefore be ascribed to be mainly magnetic in origin. It should be noted, however, that a structural contribution cannot be excluded, and indeed a mixing of magnetic and elastic degrees of freedom is expected in a spin-Peierls material.

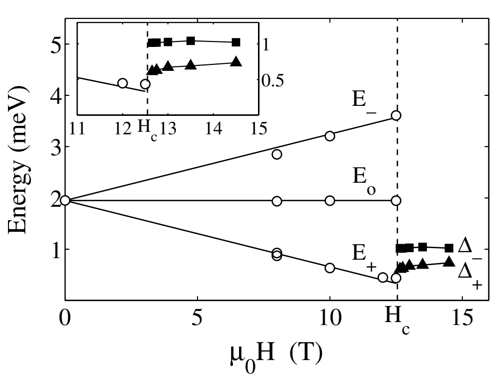

FIG. 1. Field dependence of the excitations in $CuGeO_3$ at $(0, 1, \frac{1}{2})$, 2 K. Open circles and solid lines: triplet splitting $g_a \mu_B H S^a_{tot}$, $g_a = 2.152$. Closed symbols: new excitations in the IC phase, enlarged in the inset. The critical field is $\mu_0 H_c = 12.58$ T, as indicated by the vertical dashed line.

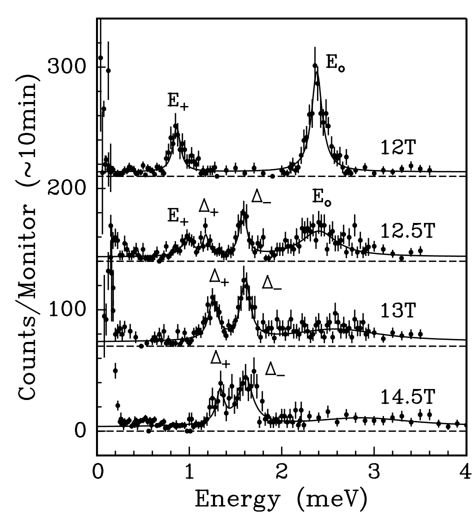

FIG. 2. Energy scans at $(\frac{1}{2}, 1, \frac{1}{2})$, $T = 2$ K. Solid lines: fits to Lorentzian-type profiles including the Bose factor. Close to the critical field, $\mu_0 H_c = 12.31$ T, the D-phase modes $E_{+,0}$ coexist with the two new modes $\Delta_{\pm}$.

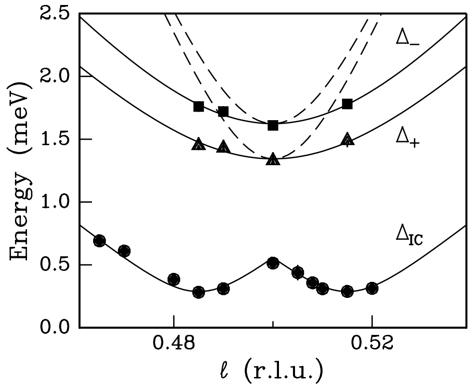

FIG. 3. $(\frac{1}{2}, 1, \ell)$ dispersion of the magnetic excitations in $CuGeO_3$ at 14.5 T, 2 K. Solid lines: dispersion fit to $\sqrt{\Delta_{\min}^2 + A^2(\ell - \ell_{\min})^2}$, where $\ell_{\min} = \frac{1}{2}$ for $\Delta_\pm$ and $\frac{1}{2} \pm \delta k_{sp}$ for the incommensurate excitation Δ_{IC}, and $A_{\Delta_+,\Delta_-,\Delta_{IC}} = 41(5)$, 48(5), and 32(2) meV/r.l.u. (reciprocal lattice units). The magnetic Bragg peaks are at $\frac{1}{2} \pm \delta k_{sp}$ with $\delta k_{sp} = 0.015$. Dashed lines: dispersion assuming the $H = 0$ spin-wave velocity, $A = 96.3$ meV/r.l.u.

In the following we relate our findings to the few theoretical results that are presently available. The fact that we observe three gapped excitations is in contrast to the soft modes in classical IC structures [10,11], but confirms recent arguments for the spin—Peierls IC phase [1,4]. The magnetic excitation spectrum is believed to have finite excitation gaps, since at each given magnetic field the lattice, and hence the intrachain exchange, adapts to the magnetization and the number of solitons. Three excitation branches would be expected, two with polarization perpendicular to H and one longitudinal [1]. The transverse branches, $\Delta_\pm$, corresponding to adding or removing a soliton pair with a resulting increase or decrease of the total spin by 1, should have minima at the commensurate wave vector $\ell = \frac{1}{2}$. The longitudinal branch Δ_0 should have minimum energy $\Delta_0^{\min} \gtrsim \frac{1}{2}(\Delta_+^{\min} + \Delta_-^{\min})$ at $\ell = \frac{1}{2} \pm \delta k_{sp}$, and an energy of $g\mu_B H$ at $\ell = \frac{1}{2}$.

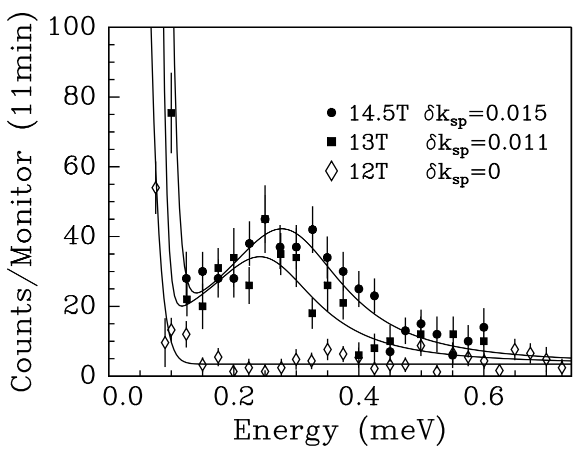

FIG. 4. Field dependence of the minimum energy of Δ_{IC}, energy scans at 12, 13, and 14.5 T at the respective modulation vector $(\frac{1}{2}, 1, \frac{1}{2} + \delta k_{sp})$. Solid lines are fits with Lorenzian line shapes including the Bose factor at 2 K, giving finite energy gaps of 0.25 ± 0.02 meV and 0.29 ± 0.01 meV at 13 T and 14.5 T, respectively.

This picture has been supported by numerical calculations for different choices of the parameters (ϵ, α), respectively the dimerization parameter $\epsilon = \frac{J_{i+1} - J_i}{J_{i+1} + J_i}$ and the next-nearest-neighbor intrachain exchange interaction $\alpha = \frac{2J^{\text{nnn}}}{J_i + J_{i+1}}$, where J_i is the nearest-neighbor coupling between sites i and $i + 1$. To date, three parameter sets have been investigated: (0.014, 0.24) [12], (0.14, 0.36) [4], and (0.4, 0) [2]. The spectral weight is predicted to be concentrated around the minima $\ell = \frac{1}{2}$ for $\Delta_\pm$ and around $\ell = \frac{1}{2} \pm \delta k_{sp}$ for Δ_0 [12]. The wave-vector dependence of the intensity and dispersion of the upper two modes found in our experiment identifies them as $\Delta_\pm$. The predicted energies depend on the choice of ϵ and α, and a quantitative comparison with our data could potentially resolve the present uncertainty in the correct values for $CuGeO_3$. However, the existing calculations refer to larger magnetic fields and predict far too high energies. It is, however, perhaps worth noting that the calculation with a finite and large value of α [4] predicts that Δ_+ should increase with increasing field, while Δ_- should be field independent, in qualitative agreement with the data shown in Fig. 1. In this sense our data are consistent with a large value of α [13].

We now address the origin of the observed IC excitation Δ_{IC} at 0.29 meV. Δ_{IC} could be the longitudinal mode Δ_0, for which minima are expected at the IC wave vectors. However, the predicted parameter-free relation $\Delta_0^{\min} \gtrsim \frac{1}{2}(\Delta_+^{\min} + \Delta_-^{\min})$ would be violated, and moreover the observed energy at $\ell = \frac{1}{2}$ is much lower than $g\mu_B H = 1.85$ meV. We therefore have to explore other possible explanations.

A low-energy excitation with incommensurate dispersion could result from phase oscillations of the magnetic and distortive soliton lattice, a so-called phason. The IC long-range order in the high-field phase breaks the quasi-continuous translation symmetry of the soliton lattice with respect to simultaneous shifts of the soliton centers along the chain axis. The Goldstone modes corresponding to this symmetry breaking should have zero energy at the IC superlattice Bragg peaks. In spite of their magnetostructural character, phasons are believed to disperse like magnetic excitations [14]. Indeed the experimentally observed dispersion along ℓ is comparable to that of the transverse magnetic excitations (Fig. 3).

The interpretation of the IC mode as a gapped phason is also consistent with other known experimental facts. A phason, gapped or otherwise, is expected to have a magnetostructural character. Raman experiments probe mainly the structural component, and have found a mode at the same energy (2.3 ± 0.2 cm^{-1}, i.e., 0.29 meV), in support of the phason interpretation [6]. If the phason is gapless, then this should lead to a reduction in the staggered magnetization by a factor 4–6 compared to field theory [3]. This is in contrast to our neutron diffraction data [9] which agree with field theory within 20%. One potential problem with this interpretation is the increase of the T^3 contribution to the specific heat capacity in the IC phase [15], which has been attributed to gapless phasons [14]. However, the specific heat data do not exclude a small finite phason energy, and might instead be dominated by a change in elastic constants.

If Δ_{IC} is indeed a phason, it remains to be explained why it has a gap. Of the various candidate mechanisms, we can probably exclude pinning to impurities, as this would disorder the soliton lattice and produce a broadening of the satellite peaks not observed in our earlier diffraction study [9]. The bare pinning energy of the phason to the discrete lattice is of the order 0.003 meV [3], and is too small to account for the gap. However, the bare pinning energy may be enhanced by different mechanisms [10], including spin-phonon interaction [16], interchain coupling, etc., but further theoretical studies are required to explore these and other possibilities.

In summary, we have measured the excitation spectrum in the quantum IC soliton phase of the spin-Peierls material $CuGeO_3$, and discovered three different magnetic excitation branches. Two are identified as the transverse magnetic excitations $\Delta_\pm$ which correspond to the creation or destruction of a soliton pair. The third, Δ_{IC} has energy minima and maximum spectral weight at the position of the IC modulation wave vector. It seems incompatible with the expected longitudinal mode Δ_0, and may instead be identified as a phason. Since the phason is also a longitudinal excitation, one may perhaps question whether there is a physical difference between these two theoretically predicted modes. We hope that our data will stimulate and guide the development of a microscopic model of $CuGeO_3$ that is capable of providing a consistent description over the entire temperature-field phase diagram of this unconventional spin-Peierls system, and so ultimately lead to a full understanding of its macroscopic quantum ground states.

We thank G. Uhrig, R. A. Cowley, W. J. L. Buyers, and K. Knorr for helpful discussions. The work was supported by the German Bundesministerium für Bildung und Forschung, Grant No. 03-KN5SAA (M. E.), and the European Community through Marie Curie Contracts No. HPRICT-1999-00020 (BENSC) and No. HPMFCT-2000-00488 (H. M. R.).

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