Journal of Physics: Conference Series

Low-energy excitations in DTN below T_c: ESR studies

To cite this article: Zvyagin *et al* 2009 *J. Phys.: Conf. Ser.* **150** 042244

Low-energy excitations in DTN below T_c: ESR studies

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Abstract. $NiCl_2$-$4SC(NH_2)_2$ (known as DTN) is an $S = 1$ system with an easy-plane anisotropy dominating over the exchange interaction and exhibiting a field-induced antiferromagnetic ordering with critical fields B_{c1}=2.1 T, B_{c2}=12.6 T and temperature $T_c^{max} \sim$ 1.2 K. A systematic study of the low-energy excitation spectrum of DTN in the ordered phase at temperatures down to 0.45 K is presented. It is showed that two observed gapped modes can be consistently interpreted within a four-sublattice antiferromagnet model with a weak isotropic corner-center interaction of magnetic ions in the body-centered tetragonal lattice.

Field-induced phase transitions in magnets with gapped excitation spectra have recently received a considerable amount of attention [1, 2, 3, 4], particularly in the context of the so-called Bose-Einstein condensation (BEC) of spin degrees of freedom. In accordance with the BEC scenario, for a uniform gas of identical particles, at some finite temperature T_c when the de Broglie wavelength becomes comparable to the average distance between the particles, a macroscopic fraction of the gas can be "condensed" into a single coherent quantum state and BEC occurs. An important property of BEC is the presence of U(1) symmetry, which corresponds to the global rotational symmetry of the bosonic field phase. It is worth mentioning that the field-induced BEC of spin degrees of freedom is not a full analogue of BEC as initially defined by Bose and Einstein [5, 6], since in magnets (i) there is a finite spin-spin interaction and (ii) the number of quasi-particles participating in the field-induced magnetic ordering is not conserved, resulting as the matter of fact in zero chemical potential [7]. Nevertheless, it still appears to be interesting to test the availability of applying the BEC formalism to the field-induced ordering in quantum magnets. Investigation of the low-energy excitation spectrum in such materials is of particular importance, since it can give important information on the magnetic structure and the symmetry of magnetic interactions across different regions of their phase diagrams. For instance, the re-opening of the energy gap in the excitation spectrum of $TlCuCl_3$ (which, based on the analysis of critical exponents, was regarded as the best realization of BEC of spin degrees of freedom in magnets [1]) in the field-induced ordered state [8, 9] with the same order of magnitude as the exchange coupling is a clear evidence for a broken uniaxial symmetry. Such observation rules out the description of the magnetic ordering in this compound in terms of BEC (at least on the energy scale reported).

$NiCl_2$-4SC$(NH_2)_2$ (known as DTN) is a gapped $S = 1$ system with a single-ion anisotropy D dominating over the intrachain exchange coupling J, exhibiting a field-induced antiferromagnetic (AF) ordering below $T_c \leq 1.2$ K, with critical fields $B_{c1} = 2.1$ T and $B_{c2} = 12.6$ T ($B \parallel c$) and critical exponents corresponding to the BEC scenario [3, 4]. In the present work, the magnetic excitation spectrum of DTN is probed in the field-induced AFM phase by means of electron spin resonance (ESR) measurements.

The ESR measurements were performed at the Kapitza Institute using a transmission-type ESR spectrometer equipped with a cylindrical multimode resonator and a ^{3}He-cryostat. High-quality single-crystalline DTN samples from the same batch as in Ref. [10] were used. The magnetic field was applied along the tetragonal c axis.

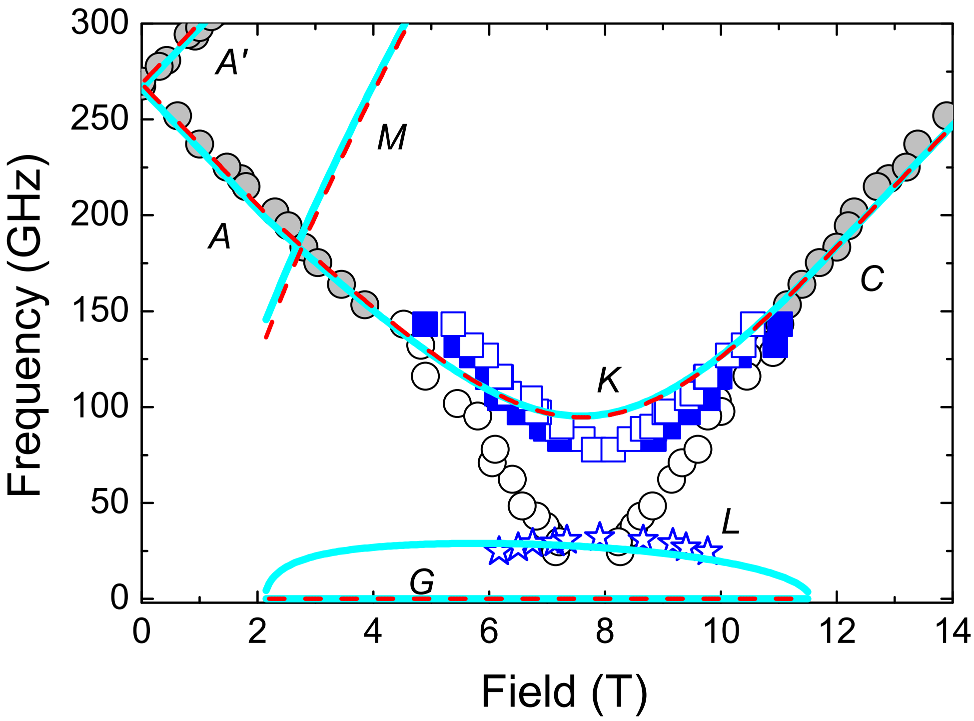

Figure 1. (Color online) The frequency-field dependence of ESR modes in DTN measured at $T = 1.4$ K (open circles) and 0.45 K (squares and stars). Filled grey circles denote the high frequency data ($T = 1.6$ K) taken from Ref. [10]. The dashed (red) lines correspond to calculation results for the simplest axially symmetric two-sublattice AF model with parameters $g_c = 2.26$, $D = 9.4$ K, $J = 2.0$ K. The solid (cyan) lines were obtained by model calculations assuming $d = 0.02$ K (see text for details).

DTN is characterized by the $I4$ space group [11] with a body-centered tetragonal lattice that may be viewed as two interpenetrating tetragonal subsystems. At $B \parallel c$ the spin dynamics can be described by the spin-Hamiltonian

$$\mathcal{H}_0 = \frac{1}{2}\sum_{\vec{n},\vec{\delta}} J_{\vec{\delta}} \vec{S}_{\vec{n}} \cdot \vec{S}_{\vec{n}+\vec{\delta}} + D\sum_{\vec{n}} (S^z_{\vec{n}})^2 - h\sum_{\vec{n}} S^z_{\vec{n}} + \mathcal{H}_{\text{int}}, \quad (1)$$

where $\vec{S}_{\vec{n}}$ are spin-1 operators at site $\vec{n}$, the vectors $\vec{\delta}$ connect the site $\vec{n}$ to its nearest neighbors within the same subsystem, $h = g_c \mu_B B$ is the Zeeman term, and $\mathcal{H}_{\rm int}$ describes (yet unspecified) additional interactions. Assuming that the latter are much weaker than the interchain interaction within tetragonal subsystems, the Hamiltonian parameters were estimated as $D = 8.9$ K, $J_c = 2.2$ K, $J_{a,b} = 0.18$ K, and $g_c = 2.26$ [10].

The frequency-field dependences of magnetic excitations in DTN at temperatures 1.4 K and 0.45 K are presented in Fig. 1. Two ESR lines (denoted as A and C) have been observed at 1.4 K. Comparison with the data of Ref. [10] (filled grey circles, Fig. 1) reveals that the mode A continues smoothly from the ESR line in the low-field disordered phase, while the mode C corresponds to single-magnon excitations in the high-field phase. Several important features have been observed at lower temperatures. Upon cooling, the modes A and C shift towards each other. While at a temperature of 1.4 K they seem to cross at zero frequency in the vicinity of $B \sim 8$ T, in the low-temperature AF ordered phase they are converted into a new gapped mode K. This mode exhibits a slight but distinct splitting, the origin of which will be discussed below (the corresponding frequency-field dependence of the resonances is denoted in Fig. 1 by pairs of open and closed squares). The low-temperature spectrum demonstrates one more set of resonances appearing at low frequencies (mode L, denoted by stars in Fig. 1). Unlike mode K, the integrated intensity of line L is $50 - 100$ times smaller than that of the modes outside the AF phase, e.g. modes A and C. The observation of the gapped mode L and splitting of mode K in a field range of $6 - 10$ T clearly indicates the presence of additional interactions, that are not accounted for in the simplest axially symmetric two-sublattice AF model (which would have the Goldstone mode as its lowest energy excitation and a single mode K as the next excitation branch as indicated by the dashed (red) lines in Fig. 1). In order to explain the observed excitation behavior, several scenarios have been examined.

First we have considered the possibility of axial-symmetry breaking, caused either by rhombic in-plane anisotropy or by the Dzyaloshinskii-Moriya (DM) interaction inside a tetragonal subsystem, with the DM vector deviating from the anisotropy c axis (note that DM is generally allowed in DTN due to the absence of inversion symmetry). It is worth mentioning that $\Delta M_s = 2$ ESR transitions from the ground state to two-magnon bound states observed in the high-field phase [10] indicate a small nonconservation of the quantum number S^z, which might be a signature of the broken axial symmetry in DTN. On the other hand, one should keep in mind that even a slight (a few degrees) misorientation of the sample with respect to the applied field might allow such transitions. In case of in-plane anisotropy or DM interaction inside tetragonal subsystems, the axial symmetry break-down would open a gap in the excitation spectrum, 'lifting' the Goldstone mode. Even so, this mode corresponds to a coherent collective excitation of the magnon condensate below T_c, and thus would be expected to be particularly strong in comparison with ESR excitations in the disordered phase. As mentioned, the gapped mode L observed in our experiments is much less intense than ESR absorptions above T_c, raising questions about the above scenarios. In addition, the rhombic in-plane anisotropy would necessarily cause splitting of the doublet at $B = 0$ (roughly of the same strength as the maximum energy of the L mode), which was not observed in the experiment. Further, close to the fully spin-polarized state, the DM contribution provides an energy gain linear in the AF order parameter, while the corresponding Zeeman energy loss is quadratic, so AF order would be favored for arbitrarily large B. Consequently (and this is of particular importance for our analysis), such a symmetry-breaking DM term would lead to the absence of the second critical field B_{c2}, which is incompatible with the results of [3, 4].

Contrary to the scenarios mentioned above, a weak isotropic corner-center interaction of magnetic ions in the body-centered tetragonal lattice (i.e., an interaction between tetragonal subsystems that preserves the axial symmetry) can explain the observed features. However, the theoretical analysis of such a model is very difficult: if, as indicated by previous studies [10], the

Journal of Physics: Conference Series **150** (2009) 042244 doi:10.1088/1742-6596/150/4/042244

exchange interactions within each tetragonal sublattice are AF, the system is highly frustrated and its mean-field ground state at $H > H_{c1}$ is infinitely degenerate. For that reason, we would like to illustrate the effect of a sublattice coupling by assuming a finite DM interaction d between two tetragonal sublattices. In case of such interactions in addition to conventional (relativistic) modes, modes with antiphase oscillations of interacting AF sublattices (exchange modes) should be present. Having much weaker coupling to the microwave field [12], exchange modes should be less intensive than relativistic modes. In accordance to our observations, the gapped mode L would then be an exchange mode, while the mode K is relativistic.

The results of model calculations [13] using $g_c = 2.26$, $D = 9.4$ K, $J = 2.0$ K, $d = 0.02$ K are shown in Fig. 1 by solid (cyan) lines. Due to a finite interaction between the two tetragonal subsystems, the low-energy mode is split into a doublet with a gapped upper component and a zero frequency lower component. One can see that the model qualitatively describes the frequency-field dependences of all observed ESR modes assuming the existence of a gapless Goldstone mode G, which can not be detected experimentally. It is important to mention that it was not possible to fit the frequency-field dependence of magnetic excitations in DTN in the AF phase using the set of parameters obtained in Ref. [10], although they are very close to those used in our calculations. This discrepancy mainly stems from neglecting quantum fluctuations in the mean-field-theory approach used in the present paper. In the above calculation the mode K (determined by $\varepsilon_{\vec{k}=0}$) comes out doubly degenerate, which is an artefact of the model assumption of purely DM inter-tetragonal subsystem interaction, making the interaction matrix U vanish at $\vec{k} = 0$. Any finite symmetric exchange interaction between the tetragonal subsystems will be sufficient to lift this degeneracy and thus will explain the observed slight splitting of the K mode. The theory also predicts the existence of a third ESR mode M in the AF phase (Fig. 1), which corresponds to the second magnon branch at $\vec{k} = \vec{Q}_B$.

In summary, high-field ESR studies of low-energy magnetic excitations in the field-induced ordered phase of DTN have been performed. Two gapped modes were observed. Based on our analysis, we showed that the above excitation spectrum can be consistently interpreted within a four-sublattice AF model with an intact axial symmetry (at least on the energy scale down to 1.2 K, which corresponds to the lowest frequency used in our experiments, 25 GHz). The latter is of particular importance, being a necessary prerequisite for the interpreting of the AF ordering in DTN in terms of the BEC scenario.

We thank C. D. Batista, M. Kenzelmann, S. Zherlitsyn, and A. A. Zvyagin for discussions. This work was partly supported by the DFG, and by the RFBR. AK is supported by the DFG Heisenberg Program. APF is grateful for support from CNPq and FAPESP (Brazil).

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