

Coexisting 1D and 3D magnetic interactions in the insulating copper–oxygen compound CuB_2O_4

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Abstract

We performed inelastic neutron scattering measurements on CuB_2O_4 with a magnetic field applied in the tetragonal basal plane. The spin dynamics in this cuprate is determined by the interplay of two magnetic Cu^{2+} subsystems, the magnetic cage with predominant 3d Heisenberg exchange and quasi 1d zig-zag chains. The comparison of the dispersion spectra along the chain direction in zero field and under applied field suggests a decoupling of the two magnetic sublattices under field.

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CuB_2O_4 is an insulating cuprate with interesting magnetic properties as evidenced by the existence of a stable magnetic soliton lattice that is not induced by an external magnetic field but is the result of interplay between exchange, Dzyaloshinskii–Moriya and anisotropic interactions [1,2]. The soliton lattice forms at the magnetic phase transition temperature between an antiferromagnetic commensurate phase (CP) and an incommensurate helical phase (ICP). We point out that the ICP is the stable phase at low temperatures, which is unusual compared to other helimagnets. In CuB_2O_4 , the CuO_4 units have no direct bond between each other and exchange interaction takes place through Cu–O–B–O–Cu paths. The magnetic unit cell of CuB_2O_4 is best described by separating the Cu^{2+} ions into two distinct magnetic sublattices, further on referred to as ‘cage’ and spin-one-half zig-zag chains (see inset of Fig. 1). The magnetic properties of both sublattices are different. The magnetic moments in the cage order below the Néel temperature $T_N = 20$ K and their magnitude increases to almost saturation $\mu_{\text{Cage}} = 0.9\mu_B$ at $T = 10$ K. Through analysis of the spin-wave spectrum it

was found that the magnetic interactions between spins in the cage are well described by an easy-plane Heisenberg hamiltonian [3]. The zig-zag chains are embedded inside the cage and propagate along the tetragonal c -axis. The magnitude of the Cu^{2+} magnetic moments in the chains are strongly reduced above the CP–ICP phase transition with about $\mu_{\text{Chain}} = 0.2\mu_B$ at $T = 12$ K. Below $T = 10$ K, the ordered magnetic moments gradually increase with decreasing temperature and reach $\mu_{\text{Chain}} = 0.5\mu_B$ at $T = 1.5$ K. The spin dynamics of this Cu^{2+} sublattice was recently explained by spin-wave calculations on the basis of a 1D antiferromagnetic zig-zag chain model, that reproduces the observed low energetic dispersion branch along the c^* -axis [4]. However, the calculations were based on linear spin-wave theory and cannot account for the line-shape of the inelastic spectra having magnetic excitations much broader than expected for spin-wave excitations.

The picture of two separated sub-systems, which can correctly reproduce the measured excitation spectra in the CP, is inadequate below the CP–ICP transition. From neutron diffraction it is found that the magnetic structure in the ICP phase is described by a single propagation vector k , which continuously changes from $k = (0, 0, 0)$ r.l.u at $T = 10$ K to $k = (0, 0, 0.15)$ r.l.u. at $T = 200$ mK. As both

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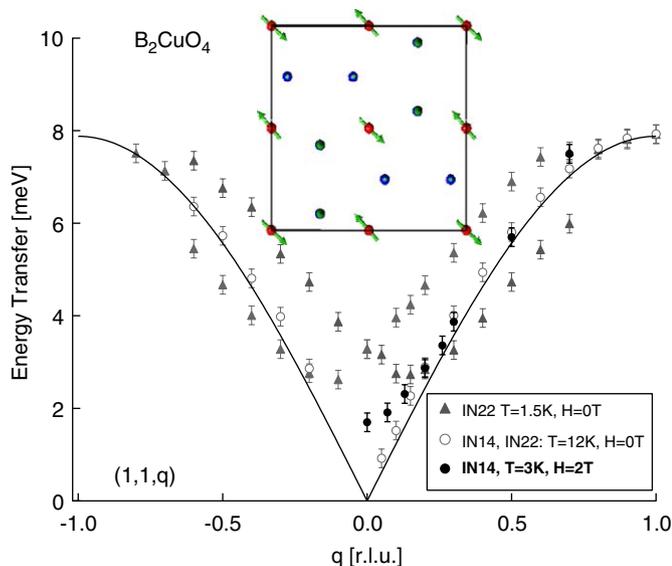


Fig. 1. Magnetic excitation spectra along the c^* -axis around the magnetic zone center $(1, 1, 0)$ at different temperatures and fields. The inset shows the magnetic unit cell of CuB_2O_4 in the commensurate phase at $T = 12$ K. The structure is projected into the ab plane. Chains are propagating along c with a Cu–Cu distance of 0.5 r.l.u.

magnetic sublattices have the same periodicity in the ICP, the exchange interactions between both sublattices cannot be neglected.

In a recent theory based on mean-field calculations [5], the antiferromagnetic inter-lattice interactions are found to be frustrated resulting in a quasi-uncoupled behavior of the two subsystems. The change of the magnetic structure in the ICP leads to a breaking of the frustration and an increasing influence of inter-lattice interactions on the dynamical properties. However, the static mean field turned out to be not sufficient to account for the large observed energy gap of $E = 2.7$ meV at $T = 2$ K in the spin-wave spectrum and a temperature-dependent anisotropy term induced by the fluctuating magnetic moments was added [5,6]. In a different approach [7,8] magnetic exchange between the sub-systems is already assumed below T_N . It has been shown that in this case that the magnetic structure is incommensurate above $T = 10$ K, with, however, an almost commensurate propagation vector above the CP–ICP phase transition as a result of competing exchange interactions.

In order to obtain additional information about the nature of the inter-sublattice interaction, we decided to measure the magnetic excitations in an external magnetic

field. The measurements were done at the cold three-axes spectrometer IN14 (ILL) with vertically focusing PG002 monochromator and horizontally focusing PG002 analyser with fixed final energy $E_f = 3.5$ meV. A Be-filter was installed after the sample position to avoid higher-order contamination. A 5 T cryomagnet was mounted with the field vertical to the scattering plane that is defined by the $[00c^*]$ and $[a^*a^*0]$ directions of the sample. We performed const.- Q -scans at $T = 3$ K and $H = 2$ T along the c^* -axis up to the zone boundary around the magnetic zone center $(1, 1, 0)$. The obtained dispersion curve is shown in Fig. 1 and compared with the magnetic excitation spectrum measured in the absence of magnetic field at $T = 12$ K (open circles) and $T = 1.5$ K (triangles), respectively. In zero field, the spectrum consists of two branches of excitations with minima at $\pm k_0$, as expected for a helicoidal magnetic ground-state. In the field-induced CP, the spin-wave spectrum contains one branch only, as previously observed above the CP–ICP phase transition in zero field. The minimum coincides with the magnetic zone center of the zero field CP structure, but in contrast to the latter an energy gap persists, which is lower in energy compared to the ICP. Surprisingly, the periodicity of the lower excitation (not shown) is not affected by the application of an external field. The minimum of this branch remains at $|q_0| = 0.15$ r.l.u., which corresponds to the magnetic propagation vector of the ICP in zero field. Under the assumption that the upper and lower branch are tightly linked to the cage and the chains, respectively, we suggest that the application of an external field separates the two sublattices into a commensurate (cage subsystem) and an incommensurate (spin chains) magnetic state along the c -axis. A detailed analysis of the experimental results is in progress and will be published elsewhere.

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