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TEMPERATURE DEPENDENT DISORDERING IN RANDOM ANTIFERROMAGNETIC Rb$_2$Mn$_{0.70}$Cr$_{0.30}$Cl$_4$

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Abstract. - A random antiferromagnetic sample of the mixed crystal system Rb$_2$Mn$_x$Cr$_{1-x}$Cl$_4$ ($x = 0.70$) with competing anisotropies and exchange interactions has been studied by quasi-elastic neutron scattering in the temperature range of 4.2-37 K. A linear temperature dependence of the observed scattering intensities as well as considerably broadened peak shapes have been found, indicating finite correlation lengths and a dynamic disorder of the antiferromagnetic phase. Below $T = 11$ K the onset of an additional freezing behaviour has been found.

1. Introduction

The random quasi-two-dimensional magnetic system Rb$_2$Mn$_x$Cr$_{1-x}$Cl$_4$ has attracted considerable attention due to its random disorder in the magnetic properties. In fact, it has become a model representing quasi-two-dimensional magnetic systems with competing exchange interactions in addition to competing anisotropies [1, 2]. By varying the concentration parameter $x$ several magnetic phases with antiferromagnetic (afm), ferromagnetic (fm) ordering and spin glass behaviour close to $x \approx 0.5$ have been found. In these randomly disordered phases the magnetic excitation spectra exhibit a coexistence of both wave-like excitation and dispersionless magnetic clusters within the same band of frequencies [1, 2]. Rb$_2$Mn$_{0.70}$Cr$_{0.30}$Cl$_4$ has a random antiferromagnetic order below $T_a = 42$ K with the spins confined to the $c$-plane on average. There is a certain degree of random disorder in their orientation in the $c$-plane (canted antiferromagnet). Attempts are underway [3] to determine the $T = 0$ groundstate by the equation-of-motion technique for a two-dimensional (2D) model. The random disorder has also strong influence on the shapes of the magnetic Bragg peaks and of the 2D Bragg rods. First investigations [4] on the line shapes of these 2D-rods in a sample of Rb$_2$Mn$_{0.75}$Cr$_{0.25}$Cl$_4$ has revealed a pronounced minimum of the temperature dependent inverse correlation length well below the transition temperature $T_a$. However, the corresponding energy width and the behaviour at very low temperatures has not been investigated previously.

2. Experimental and results

The experiment has been carried out on the 4F1 triple axis spectrometer viewing the cold source of the Orphée reactor at the C.E.N. Saclay, France. An incident neutron energy of $E_i = 14.4$ meV and a $60^\circ 30^\circ 30^\circ 20^\circ$ beam collimation has been used through our experiment. The brick-shaped sample ($15 \times 8 \times 6$ mm$^3$) was aligned with its [001] and [110] reciprocal lattice vectors to the scattering plane. For temperature setting between $T = 4.2$ K and $T = 70$ K a standard LHe-bath cryostat has been used. The temperature dependent intensities of the (1/2 1/2 0) afm Bragg peak and of the afm rod at $Q = (1/2 1/2 0.28$ have been investigated by energy- and (110)- and (001)-$Q$-scans. Further, no ferromagnetic scattering at the (1 1 0) rod positions was observed. Contours of the spectrometers resolution function have been measured carefully in all directions at the (1 1 0) and (0 0 6) nuclear Bragg peaks. These results have been used to calibrate the resolution calculation in the region of interest in $(Q, \omega)$-space.

Different types of scattering in Rb$_2$Mn$_{0.70}$Cr$_{0.30}$Cl$_4$ can be seen simultaneously: nuclear scattering occurs at Bragg peaks such as (110), (002), (112) etc. Further, there is a coexistence of two-dimensional (2D) and three-dimensional (3D) scattering at low temperatures. When lowering the sample temperature towards $T_0$, there is an increasing precursory scattering on afm intensity rods at $Q = (2n + 1)/2$, $(2m + 1)/2$, $\xi$; $n, m$ integer, $\xi$ real. Below $T_0$ afm Bragg peaks arise at rod-positions with $\xi$ integer from the 3D afm ordering. Accordingly the 2D rod intensity is decreasing, but not vanishing with decreasing temperature. The experimental data exhibit a linear temperature dependence of the integrated intensity of the afm Bragg scattering at $Q = (1/2 1/2 0$ (cf. Fig. 1), corresponding to a critical exponent $\beta = 1/2$. This may be considered to be unusual when compared to ordinary 2D antiferromagnets with $\beta = 1/8$ [5]. Regarding the peak width (FWHM), a considerable broadening in [0 0 1] -direction, i.e. in direction of the 2D-rods, has been observed (Fig. 1), indicating a finite out-of-plane correlation length.

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The integrated rod intensity is maximum at $T_0$, smoothly decreasing linearly with changing temperatures. At $T=4.2$ K still 20% of the maximum intensity value at $T_0$ is being observed, due to the coexistence of 2D and 3D afm order below $T_0$. A pronounced temperature dependence of the 2D rod widths has been found (Fig. 2). The 2D rod line shape is considerably broader than the instrumental resolution width, when scanning either the energy transfer of the quasi-elastic scattering or the momentum transfer $Q$ in (110) direction perpendicular to the rod extension. There is a maximum energy width at $T_0$ saturating to a value of $\Delta\nu_{\text{FWHM}} = 0.25$ THz. At temperatures below $T \approx 12$ K narrowing peak shapes have been found and at $T = 4.2$ K the peak width reaches the value of the instrument resolution without an evidence for a temperature hysteresis. The $Q$ (110) rod width behaves similar: again, there is a plateau region between $T_0$ and 11 K, well above the $Q$-resolution value (cf. Fig. 2). Below $T = 11$ K, however, there is an additional broadening of the $Q$ (110) width down to a temperature of $T = 8$ K and then a subsequent narrowing again. Further, a hysteresis effect has been found, showing the broadening effect more pronounced upon heating. The combination of these three characteristics are typical of spin glass behaviour with freezing effects and, thus, we may interpret our results as follows:

a) the 2D magnetic order is of final life time and, hence, of finite in-plane correlation;

b) below $T = 11$ K, the onset of freezing behaviour reduces the thermally activated motion of the spins, pinning them to their instantaneous orientation with an accordingly reduced correlation length;

c) at very low temperatures, the spins have got sufficient time to reassemble themselves in their individual potential minima. This time dependence is responsible for the temperature hysteresis of the correlation length and $Q$ (110) width.

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