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IDENTIFICATION OF FLUCTUATING SUSCEPTIBILITY COMPONENTS IN 
Rb$_2$CrCl$_4$: A QUASI-2-DIMENSIONAL EASY PLANE FERROMAGNET

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Abstract. – Inelastic neutron scattering with polarisation analysis has been used to investigate the critical long wavelength spin fluctuations in Rb$_2$CrCl$_4$. As well as spin wave and central peak components expected from fluctuations within the easy plane, weak central peak components are observed which may result from the diffusion of spin vortices.

The ionic ferromagnet Rb$_2$CrCl$_4$ adopts a slightly distorted K$_2$NiF$_4$ type structure in which the Cr$^{2+}$ ions (S = 2) lie in square planar arrays in well separated layers [1]. Since the Cr$^{2+}$ sublattice is pseudo tetragonal it is convenient to refer to cell constants in the I4/mmm space group, in which case a$_0$ = 5.086 Å and c$_0$ = 15.715 Å at 4.5 K. The exchange coupling is predominantly near neighbour Heisenberg (J/k_B ≈ 7.6 K), with relatively much weaker interplaner coupling J' (J'/J ≈ 10$^{-4}$) [2]. When a small canting is neglected, the spin wave behaviour is well described by an approximate Hamiltonian with dominant planar anisotropy (~ 1 K) which confines the spins to the (001) plane, and weaker four fold anisotropy (~ 0.1 K) which defines (110) as the easy axes of magnetisation. The magnetic dipolar interaction is generally negligible at a microscopic level.

Rb$_2$CrCl$_4$ orders three dimensionally at T_C ≈ 52.2 K as a result of the weak interplaner coupling. 2d, possibly XY-like, behaviour sets in above T_C + 3 K, whence the critical neutron scattering takes the form of rods of intensity parallel to [001] in reciprocal space [3]. The dynamic critical scattering in zero external field at (q, q, 2.9) was previously studied using unpolarised neutrons [4]. The data as q → 0 and T → T_C were successfully analysed in terms of sharp (resolution limited) spin waves and a broad Lorentzian central peak, although the polarisation of the fluctuations giving rise to these components could not be identified. The q = 0 spin wave mode was found to be a soft mode, renormalising rapidly and anomalously within ~ 1 K of T_C.

In the present experiment a crystal of ~ 0.27 cm$^3$ was mounted in a helium filled aluminium can attached to the cold finger of an Oxford Instruments vertical field cryomagnet, with temperature measured by a Pt resistance thermometer. The IN12 triple axis spectrometer on the cold neutron guide, ILL, Grenoble, was used in the "W" configuration with neutron wavelength $\lambda$ = 5.02 Å, and nominal collimation angles 60°.30' .30'.60°. The monochromator was pyrolitic graphite (002 plane) and a cooled Be filter minimised $\lambda$/2 contamination. Between this and the sample, a polarising mirror defined a vertical neutron spin polarisation. Polarisation analysis [5] was achieved by the combination of a polarisation sensitive Heusler alloy (111) analyser crystal plane, and a "flipper" solenoid mounted between this and the sample. Spin flip (SF) and non spin flip (NSF) cross sections were measured with constant incident energy and scattering vector Q, counting for typically 21 minutes per point. The resolution function was carefully determined, and the flipping ratio (~ 40) was regularly checked. In the scattering geometry used [110] was vertical and [110] and [001] lay in the scattering plane. These axes are labelled y, x, and z respectively, for consistency with Mertens et al. [6], who have predicted the dynamic critical behaviour of 2d XY magnetic systems.

Most scans were taken near to (0, 0, 2.9) in a vertical field of 2 KG, a value strong enough to prevent neutron depolarisation by the sample, but not so strong as to suppress the critical scattering. In this configuration the field $H$, the neutron polarisation $P$ and the magnetisation $M$ were all vertical, and perpendicular to $Q$. Therefore easy plane magnetic fluctuations longitudinal ($S^{yy}$, NSF) and transverse ($S^{zz}$, SF) to the spin direction were measured, along with background and incoherent scattering from various sources [5]. The measured intensity was analysed using the Harwell routine FITSQW. This convolves the calculated dynamic structure factor $S(Q, \omega)$ with the experimentally determined instrumental resolution function and enables parameters in the former to be fitted to the data. In all fits proper account was taken of the background and incoherent scattering. Typical results are shown in figure 1. The NSF (longitudinal) cross section fitted well to a central peak of Lorentzian form. The peak height was observed to reach a maximum at ~ T_C in zero field, and at progressively higher temperatures the larger the applied field. The energy width was approximately constant above T_C. The longitudinal polarisation of this central peak suggests that its origin is diffusive [7]. The SF (transverse) cross section was successfully analysed in terms of spin wave excitations which were sharp at all temperatures (50-60 K).
clear evidence of a Lorentzian or Gaussian central peak of approximate FWHM 0.024 THz (0.1 meV), and of intensity too great to arise from any source other than out-of-plane fluctuations.

In summary, the principal sources of dynamic critical scattering in Rb$_2$CrCl$_4$ are (i) sharp in-plane spin waves, as previously observed [4]; (ii) a strong in-plane longitudinal central peak, presumably due to spin diffusion; (iii) a weak central peak corresponding to in-plane transverse fluctuations; and (iv) a weak central peak corresponding to out-of-plane transverse fluctuations. All these features were observed in the temperature range where 2d XY-like behaviour is expected, and where according to Kosterlitz-Thouless theory [8, 9], vortices are the relevant spin configurations. Vortex diffusion has been predicted to give rise to both in- and out-of-plane transverse central peaks, with squared Lorentzian and Gaussian peak shapes respectively [6]. Although our data are insufficient to determine peak shapes unambiguously, central peaks of both polarisations are observed. The in-plane central peak was measured in a magnetic field, making direct comparison with the theory difficult. However, the out-of-plane transverse central peak width, calculated using expression (4) of reference [7], the parameters appropriate to Rb$_2$CrCl$_4$, and taking $b = 1.5$ [9], is found to be 0.028 THz (FWHM). This compares favourably with the observed width of $\sim 0.024$ THz. We may conclude that there is promising evidence of vortex diffusion in Rb$_2$CrCl$_4$.

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