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MAGNETISM OF A TWO-DIMENSIONAL XY SYSTEM: NEUTRON STUDIES OF CoCl$_2$ INTERCALATED GRAPHITE

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Abstract. - We present the results of magnetic neutron diffraction studies on a single crystal of a first stage CoCl$_2$ intercalated graphite by elastic and inelastic techniques versus temperature. We deduce the magnetic couplings and the presence of defects such as vortices, related to the 2D XY nature of the compound.

Introduction

The physics of 2D systems is still a challenge searching a good experimental magnetic system and CoCl$_2$ intercalated in graphite is a good candidate to test the Kosterlitz-Thouless (KT) ideas on 2D XY models [1]. Different works were performed on this system by many techniques (susceptibility [2, 3], specific heat [4, 5] elastic [6, 7] and inelastic [8] neutron scattering). But all of these on polycrystals and the results were not conclusive on the KT nature of the transition observed at around 8 K. Though high stages [9] are certainly better candidates for 2D systems, the lack of single crystals of these high stage compounds drives us to study first the first-stage compound. In this compound, the interlayers magnetic interaction should be strong enough to obtain a real magnetic order and hence to observe the magnons and to determine the magnetic coupling constants.

We have studied by neutron diffraction at the Institut Laue-Langevin on the two-axis cold source diffractometer D16 a single crystal of first stage CoCl$_2$ intercalated in graphite. The inelastic study was performed on the same sample on the thermal neutrons three-axis spectrometer IN8.

We have first checked the crystalline structure by measuring the (00$l$), (10$l$)$_{Co}$, (10$l$)$_{C}$ nuclear Bragg ridges at 20 K (above $T_N$) and confirmed the structure obtained previously by X-rays [10]. We have also measured the in-plane (200 Å) and out-of-plane (200 Å) correlation lengths and the stacking of CoCl$_2$ planes (ABC type with many stacking faults). The mosaicity of the c-axis is around 5° (half with at half maximum) and the in-plane mosaicity is less than 1° for the graphite and 2° for the CoCl$_2$. It should also be noticed that the respective orientation of CoCl$_2$ versus graphite are 0° or 30° in the same crystal, inducing a twining of the sample.

The magnetic structure

Figure 1 presents the (00$l$) and (10$l$)$_{Co}$ magnetic diffraction patterns at 4 K integrated over h, k directions. The (00$l$) diffusion pattern indicates a clear c-axis antiferromagnetism where the spins lay in the plane which is in agreement with previous studies on polycrystals [6, 7]. From a comparison between nuclear and magnetic intensities, we deduce that only 9 % of the spins are in-plane ordered ferromagnetically at low temperature (1.6 K) though it is saturated. The correlation length along c-axis is constant (100 Å) and no critical broadening was observed. This result differs from that of the second stage compound where the c-axis magnetic coupling is obviously smaller [7]. The
(10\overline{1})_{Co} diffraction pattern is flat which is characteristic of two dimensional order and indicates a complete absence of magnetic correlations between the layers. Only the resulting magnetization of each plane (9\% of the ferromagnet) is correlated antiferromagnetically and gives the observed 00l magnetic line.

In order to understand the nature of the defects which are responsible of this value of 9\%, we have undertaken numerical simulations of a single finite plane of a triangular cobalt lattice by a classical Monte Carlo method. The originality of the calculation was to introduce a demagnetization field due to the finite size effects in the sample (200 Å, dipolar effects). The importance of this effect was first pointed out by Flandrois and by Rancourt [11, 12]. Figure 2 presents one of our calculated configurations of the low temperature state exhibiting a clear vortex and a corresponding magnetization of 10\%. Since the vortex is the natural defect of the 2D XY system (it replaces the domains), the presence of vortices is not a surprise but it is not related directly to the KT model in which the number of vortices varies specifically with the temperature [1].

![Fig. 2. – A configuration of the low temperature state of the simulation.](image)

The inelastic study

The magnetic dispersion relation measures directly the magnetic couplings which are the fundamental parameters of all the calculations. Such a determination has been already performed in pristine CoCl$_2$ [13], and in a polycrystalline of second stage intercalated graphite [8], but the present study in a single crystal is the only way to determine the possible in-plane anisotropy. Figure 3 presents the dispersion relation in \{(100)\} and \{(110)\} directions. No in-plane anisotropy has been observed and the resolution of the spectrometer was not sufficient for measuring the interlayers interaction given by the gap in the zone center. A simple model with only the first neighbors in-plane coupling $J$ [14], gives the dispersion relation drawn in figure 3 with $J_1 = 15$ K. This value is similar to that of the second stage compound [8], but different from that of pristine CoCl$_2$ [13]: 28.5 K. This difference can be interpreted by the charge transfer, and the surrounding of the ion by changing the Cl-Cl distance in the plane and along the c-axis. The $T_N$ values are also different (6.5 K here, 24.9 K in pristine compound). The $\pi JS^2$ is also too high: 11.5 K [1]. Since the presence of an interlayer coupling decreases this value, it cannot interpret this discrepancy but the numerical simulation gives $T_c=2JS^2$ (7 K) indicating that dipolar effect decreases the energy of the vortex formation.

![Fig. 3. – The magnetic dispersion relation. The full lines are the model described in text.](image)

We have also studied the variation of the magnon dispersion relation with the temperature and observed a renormalization effect near the boundaries of the Brillouin zone. The existence of magnons at 40 K (5 $T_N$) is surprising but they need only strong ferromagnetic fluctuations. This observation is hence the proof of the existence of such fluctuations already observed by specific heat [5] and is characteristic of two dimensional systems.

[9] The stage is the number of graphite planes which separate two nearest intercalated layers.