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Neutron-Scattering Experiments on Well-Staged Graphite - $FeCl_3$

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Introduction

This paper reports results of a neutron diffraction study on both the magnetic and the lattice dynamic structure of graphite-$FeCl_3$. The $FeCl_3$ system was selected for this study because of the interesting magnetic properties previously reported for this system.[1,2,3] Of particular interest is the low temperature magnetic phase transition, which has been shown to be stage dependent.[2,3] For example, in the magnetic state, the spin alignment[1] has been reported to be in-plane with an assumption that within a single layer the spins were ferromagnetically aligned, but between planes the alignment could be either ferro (stage 2) or antiferromagnetic (stage 1). In addition, Mössbauer experiments [2,3] suggested that the stage 1 and 2 compounds showed very similar magnetic behavior[3], while the stage 4 and 6 compounds failed to show a low temperature in-plane spin alignment.

Special emphasis was given to the study of the low temperature magnetic state of the Fe^{3+} spins and to the longitudinal phonon modes for k_z axis acoustic branches. All experiments reported here were carried out on a stage 2 sample of graphite-$FeCl_3$. The sample was prepared by a two-zone vapor growth technique[4] and was characterized by (00ℓ) x-ray and neutron diffraction patterns to be > 95% stage 2, with only minor inclusions of other stages.

Magnetic State

The results of the (00ℓ) neutron scans for temperatures T > 80 mK showed no evidence for ferromagnetic spin alignment (either inter-planar or intra-planar) in a stage 2 compound. Static in-plane magnetic fields of up to ~6 Tesla were applied and fields of this magnitude failed to achieve ferromagnetic in-plane alignment by means of a spin flop transition. In this in-plane high field experiment at T ~0.1 K we were able to produce an in-plane magnetization which was only about 10% of the expected ferromagnetic saturation value. Some evidence of inter-planar magnetic correlations was however found. A weak (hk0) peak indicating a doubling of the in-plane unit cell was observed. However, from our analysis of the data we conclude that neither a ferro nor a simple 2 sublattice antiferromagnetic inter-planar alignment was compatible with our observations. The magnetic state inferred from the Mössbauer experiments[3] cannot definitively establish long-range order. A Mössbauer experiment is sensitive to the local spin environment and often is unable to distinguish a long-range ordering from a slowing down of a spin relaxation process.

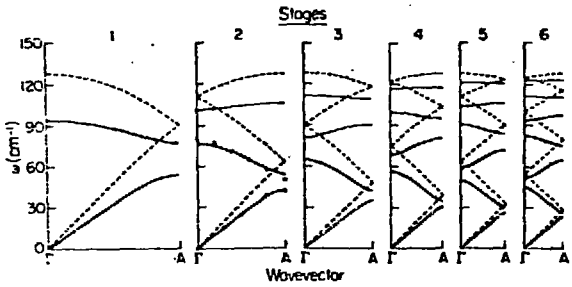

Fig. 1. Longitudinal phonon modes propagating in the z direction in graphite-$FeCl_3$ intercalation compounds. The dashed curve represents the zone folding of the pristine graphite branch. The points for stage 2 represent our inelastic neutron scattering measurements. The solid curves are calculated from the one-dimensional model discussed in the text.

These neutron diffraction results for our stage 2 sample are thus inconsistent with any of the previously proposed magnetic structures.[1,3] Though the results do not preclude a low temperature magnetic phase transition, neither do they confirm a conventional transition to a magnetically ordered state.

Lattice Modes

On the other hand, inelastic neutron diffraction peaks were observed in room temperature (00ℓ) k-space scans. These diffraction peaks were identified with the longitudinal acoustic branches. Only 3 branches were observed and these are shown in the stage 2 panel of Fig. 1 as the open circles. The high frequency optical branch (which occurs at ~868 cm^{-1} in pristine graphite) was not observed, nor were any of the $FeCl_3$ type intercalate vibrations observed. These intercalate $FeCl_3$ modes are expected to be IR-active and should occur in the $200 < \omega < 500$ cm^{-1} range.[5]

The calculated results for the low frequency longitudinal phonon branches for stage 1 to 6 graphite-$FeCl_3$ are shown in Fig. 1, in which the dashed curve is the folded graphitic longitudinal mode and the solid curves are the result of a one-dimensional model calculation with nearest neighbor elastic interactions. The calculation of these branches as a function of stage is achieved by introducing additional graphite layers into the c-axis unit cell and using the same basic set of force constants (see Table 1). Because of the assumption that the interactions between graphite interior layers are identical

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