MAGNETIC FIELD DEPENDENCE OF THE NÉEL TEMPERATURE OF THE ONE-DIMENSIONAL ANTIFERROMAGNET K₂FeF₅

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The dependence of the Néel temperature ($T_N$) of single crystal samples of K₂FeF₅ on the magnitude and direction of an applied magnetic field has been measured using Mössbauer spectroscopy. The measurements show that with fields applied perpendicular to the easy axis $T_N$ increases with increasing applied field, while with fields applied parallel to the easy axis $T_N$ decreases with increasing applied field until the spin-flop transition is reached, above which $T_N$ increases again. These data are consistent with theoretical results for pseudo one-dimensional Heisenberg systems.

INTRODUCTION

During the last decade low-dimensional magnetic systems have received considerable attention from both theoretical and experimental physicists. One-dimensional magnetic systems are of particular interest as they represent the simplest examples which show the collective behaviour characteristic of infinite systems. They also exhibit certain features which are only found in, or are particularly pronounced in, one-dimensional systems. In addition there have recently been a number of physical realizations of pseudo one-dimensional antiferromagnets.

Pseudo one-dimensional magnetic systems are those in which the intrachain exchange interaction $J$ is much greater than the interchain exchange interaction $J'$. In such systems the ratio $J'/J$ gives a measure of the degree of one-dimensionality. While a purely one-dimensional system cannot sustain long range three-dimensional magnetic ordering at any finite temperature (although at temperatures of the order of $JS(S+1)/k_B$ and below there will be a correlation of the spin directions for considerable lengths along the individual chains) the small interchain interactions lead to a magnetic ordering transition at a temperature (the Néel temperature, $T_N$) which is << $JS(S+1)/k_B$. Many of the properties of these systems are determined by the behaviour of the individual chains and this is particularly true in the intermediate temperature region between $T_N$ and the true paramagnetic state at high temperatures (>> $JS(S+1)/k_B$).

In addition to the effects of dimensionality, anisotropy plays an important rôle in determining the behaviour of these systems. The theoretical calculations usually consider the Ising, XY or Heisenberg case, although actual materials only approximate to these. In general with greater anisotropy the correlation length for spins within each chain will be larger and thus a transition to an ordered state will be more likely. Therefore all else being equal the transition temperature will be higher in a system with high anisotropy (e.g. Ising) than in a system with zero anisotropy (i.e. Heisenberg). The effect of an applied magnetic field is to induce an additional anisotropy term, which, depending on the direction of the applied field, may increase or decrease the total anisotropy and hence increase or decrease the transition temperature. In the case of an anti-
ferromagnetic system the spins will in general prefer to align perpendicular to any applied magnetic field. Thus if a field is applied parallel to the easy axis of magnetization the total anisotropy is decreased and therefore the Néel temperature is decreased. This decrease will continue with increasing applied magnetic field until at a certain magnitude of the applied field the spins reorientate (flop) to become perpendicular to the applied field and to their original direction and lie (in the case of orthorhombic anisotropy) along the intermediate axis, with the remaining orthogonal axis being known as the hard axis. For increasing applied field in the spin-flopped phase the total anisotropy increases and so does $T_N$. When the field is applied along the intermediate or hard axes the effect is to increase the total anisotropy and hence also $T_N$, the effect being more pronounced when the field is applied along the intermediate axis.

Various theoretical treatments of the behaviour which is discussed qualitatively above may be found elsewhere (de Jonge et al. [1, 2], Hjømnsen et al. [3], Villain and Loveluck [4]). This behaviour has now been observed in a number of systems using various techniques to observe the magnetic transition (e.g. Dupas and Renard [5], Boucher et al. [6], de Jonge et al. [7]).

K$_2$FeF$_5$, the subject of the present investigation, has a crystal structure which suggests that the magnetic behaviour should be one-dimensional in character (Vlasse et al. [7]). The lattice is orthorhombic with $a = 2.039\text{nm}$, $b = 1.284\text{nm}$ and $c = 0.740\text{nm}$. The structure is made up of infinite zig-zag chains of (FeF$_6$)$_3$-octahedra with common cis corners running along the $a$ axis. The intrachain Fe-Fe distance is $0.38\text{nm}$ while the interchain Fe-Fe distance is $0.57\text{nm}$ and thus the strongest exchange interaction ($J$) is along the $a$ axis. (N.B. The labelling of the axes given above is from Dance et al. [8]; in the previous publications of Vlasse et al. [7] and Gupta et al. [9] the axes were labelled differently.)

Previous measurements on K$_2$FeF$_5$ using magnetic susceptibility and neutron diffraction techniques (Dance et al. [8]) as well as Mössbauer spectroscopy (Gupta et al. [9], [10], [11] and [12].) have confirmed its one-dimensional antiferromagnetic character. In particular the Mössbauer measurements have shown that in the magnetically ordered state at low temperatures there is an anomalously low value of the saturation hyperfine field as well as a dependence of the hyperfine field on the applied field, both of which are due to spin reduction effects, which result from spin wave theory and are particularly pronounced in one-dimensional systems. Comparison of the experimental and theoretical data leads to an estimate of around $10^{-3}$ for $J'/J$ in K$_2$FeF$_5$. From the single crystal Mössbauer and neutron diffraction data the magnetic easy axis was determined to be the $b$ axis. An applied magnetic field of $3.7\text{T}$ at $4.2\text{K}$ produces spin reorientation to the intermediate axis which lies in the $a c$ plane between the $a$ and $c$ axes.

In the present investigation Mössbauer spectroscopy is used to monitor the transition to the magnetically ordered state in K$_2$FeF$_5$, using single crystal samples, as a function of the magnitude and direction of an applied magnetic field. Although other techniques, such as heat capacity measurements, may be more convenient for just measuring the value of $T_N$, Mössbauer spectroscopy has the advantage that it also provides information concerning the magnitude and direction of the magnetization both above and below the transition temperature.

**EXPERIMENTAL**

Single crystals of K$_2$FeF$_5$, prepared by flux growth (Vlasse et al. [7]) and with axes characterized by Laue back-reflection, were set in epoxy resin and thinned down to give samples suitable for Mössbauer spectroscopy. $^{57}$Fe Mössbauer spectra were recorded with a conventional constant acceleration spectrometer. The absorber was placed in a variable temperature insert which in conjunction with a SrTiO$_3$ capacitor thermometer and an electronic controller enabled temperatures between 5 and $20\text{K}$ to be measured and controlled to within $\pm 0.1\text{K}$ over periods of