^{125}Te MÖSSBAUER EFFECT STUDY OF MAGNETIC HYPERFINE STRUCTURE IN THE FERROMAGNETIC SPINEL $CuCr_2Te_4$ *

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The Mössbauer hyperfine spectrum of ^{125}Te has been measured in the ferromagnetic spinel $CuCr_2Te_4$. The hyperfine field was found to be 148 ± 5 kOe. The nuclear magnetic moment of the 35.6 keV excited state of ^{125}Te was determined to be + 0.74 ± 0.07 nm.

In this letter, we report on the measurement of a transferred hyperfine field at the tellurium anion site in the ferromagnetic spinel $CuCr_2Te_4$. This compound represents a unique opportunity for studying transferred hyperfine fields using Mössbauer techniques since Te is the only chalcogenide with a known Mössbauer nuclide and $CuCr_2Te_4$ is the only ferromagnetic spinel which has been successfully prepared using tellurium as an anion [1,2].

$CuCr_2Te_4$ is one of the interesting groups of chromium spinels which have shown anomalous ferromagnetic behaviour [2-4]. $CuCr_2Te_4$ has a Curie temperature T_C = 365°K [1]. The spin configuration of this particular spinel was recently investigated by neutron diffraction techniques [5]. From the results, it was concluded that $CuCr_2Te_4$ was a normal spinel with the magnetic Cr^{3+} ions occupying solely the B sites and diamagnetic Cu^+ occupying the A sites. A model proposed to describe the magnetic behaviour in the ferromagnetic spinels [2] involves a dominant exchange interaction of the 90° Cr-X-Cr superexchanges type, where X is the anion. For the $3d^3$ cations, this exchange interaction is ferromagnetic.

In this study, the magnetic hyperfine interaction at the Te anion site was investigated using the 35.6 Mössbauer gamma ray resonance in ^{125}Te. The magnetic hyperfine interaction causes the 35.6 keV $\frac{3}{2}$+ excited state to split into four levels and causes the $\frac{1}{2}$+ ground state to split into two levels. Since the gamma ray transition is

predominantly a magnetic dipole (M1) transition, the Mössbauer pattern will consist of six lines. The relative location of the six lines in the magnetic hyperfine pattern is completely determined by $R = \mu_1/\mu_0$, the ratio of the excited state nuclear magnetic moment to the ground state nuclear magnetic moment. The ground state moment is known from nuclear magnetic resonance measurements (μ_0 = - 0.8872 nm) [6]. The excited state moment has not been determined although previous Mössbauer measurements [7,8] have definitely established the moment as positive. From theoretical considerations, the excited state moment has been estimated to be $\mu_1 \approx$ + 0.7 nm [8].

The measurements were made using a source of ^{125}Te^m (58d). The ^{125}Te^m was obtained by neutron irradiation of Te enriched to 94% in ^{124}Te. The Te was in the form of cubic PbTe which provided a single resonance line source. The $CuCr_2Te_4$ powder was embedded in acrylic plastic for use as an absorber. The powder absorber thickness was 53.5 mg/cm^2.

The 35.6 keV gamma rays were detected using a xenon-nitrogen filled proportional counter. Selection of the escape peak provided separation of the gamma ray from the intense X-ray (27.4 keV and 31.2 keV) background. The resonant absorption spectrum was obtained using a time mode Mössbauer spectrometer. Both the source and absorber were cooled to liquid nitrogen temperature for the measurements.

A typical resonant absorption spectrum is shown in fig. 1. In order to determine whether this spectrum was a partially resolved six-line spectrum due to magnetic hyperfine splitting or whether it was a two-line spectrum due to quadrupole splitting, the data were analyzed assuming the follow-
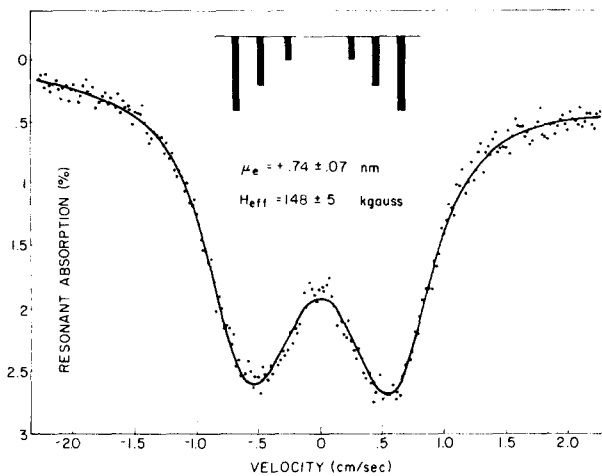
Fig. 1. Resonant absorption spectrum from a PbTe source and $CuCr_2Te_4$ obsorber (53.5 mg/cm^2) at liquid nitrogen temperature. The solid line is the "best fit" six-line spectrum. The bars denote the location and relative intensity of the six lines.

ing spectral shapes. First, it was assumed that the spectrum consisted of two Lorentzian-shaped lines with equal line widths, but different line intensities. The line locations, line intensities, and line widths were used as variables in the fitting. Second, it was assumed that the spectrum consisted of six Lorentzian-shaped lines with equal line widths. The line intensities were constrained to the ideal intensity ratio for a powder absorber (3:2:1:1:2:3 for the locations determined by $\mu_1 \approx + 0.7$ nm). The variables used in determining the line locations were R and H_{eff}, the effective hyperfine field. Other variables were the line width and a line intensity parameter. The result of the least squares computer fit with the six-line spectrum is shown as the solid line in fig. 1. The bars denote the location and relative intensity of the six lines.

On the basis of the fittings with the assumed spectra, it was concluded that the splitting was due to a magnetic hyperfine interaction. The reasons were as follows: 1) the χ^2 value for the "best fit" six-line spectrum was 25 percent smaller than the value for the two-line spectrum; 2) the line width for the two-line spectrum was 36% broader than the expected line width corrected for finite absorber thickness, whereas the line width for the six-line spectrum was in excellent agreement; and 3) the magnitude of the electric field gradient needed to produce the

observed splitting ($eq = 4.3 \times 10^{16}$esu) is 10 percent greater than that due to a single 5p electron [9]. Any reasonable bonding model for Te in $CuCr_2Te_4$ would yield less than the equivalent of one unbalanced 5p electron.

In the final six-line analysis, a small quadrupole interaction was taken into account since the symmetry of the tellurium site is not cubic. The average quadrupole coupling constant obtained from the computer fit to several measurements is $\frac{1}{2}e^2qQ/ = - 0.08 \pm 0.08$ mm/sec.
The average value of $R = \mu_1/\mu_0$ obtained from the data is $R = - 0.84 \pm 0.09$. Using the previously determined value of the ground state nuclear moment, we find the nuclear moment of the 35.6 keV excited state to be $\mu_1 = + 0.74 \pm 0.07$ nm, in good agreement with the estimated value. The average value of the effective hyperfine field is $H_{eff} = 148 \pm 5$kOe.

The nonmagnetic Te^{2-} ion would not normally be expected to have a hyperfine field at its nucleus. However, in a ferromagnetic compound such as $CuCr_2Te_4$, the Te 5s electrons can be spin polarized through interaction with the magnetic cations, leading to a hyperfine field at the Te nucleus through the Fermi contact interaction. The following two mechanisms are the most probable source of the polarization. First, covalent mixing of the Te 5s orbitals with the Cr 3d orbitals, and second, exchange polarization of the Te 5s electrons by the Cr 3d electrons. A 5s electron polarization of a few percent would provide a field of the right magnitude [10].

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