MÖSSBAUER STUDY OF THE PLANAR ANTFERROMAGNET FeTa$_2$O$_6$

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(Received 1 November 1994; accepted in revised form 5 January 1995 by C. E. T. Gonçalves da Silva)

We report on Mössbauer spectroscopy study of magnetic ordering in FeTa$_2$O$_6$. At temperatures above 10 K, the compound is paramagnetic; the spectrum obtained at 10 K shows a very asymmetric doublet, suggesting the onset of the magnetic ordering; between 9.75 K and 4.2 K the spectra consist of magnetic components. The temperature dependence of the sublattice magnetization, as determined from Mössbauer measurements, qualitatively resembles that calculated for the two-dimensional Ising model.

Keywords: A. magnetically ordered materials, B. chemical synthesis, D. phase transitions, E. nuclear resonances.

1. Introduction

In FeTa$_2$O$_6$ the sublattice formed by the iron atoms has the same symmetry as the Ni sublattice in the body-centered tetragonal K$_2$NiF$_4$ compound, the well-known two-dimensional Heisenberg antiferromagnet. This observation has motivated investigations to search for the existence of low-dimensional magnetic behavior in the title compound. Magnetic susceptibility measurements have suggested an antiferromagnetic ordering with the Néel temperature at about 15 K [1,2]. Heat capacity measurement [2] has shown a clear transition at about 8 K, suggesting short-range correlations at about 15 K, and a long-range ordering at about 8 K. Similar results have been obtained for the isomorphous compounds CoTa$_2$O$_6$ and NiTa$_2$O$_6$ [3]. Thus, the magnetic behavior of the FeTa$_2$O$_6$ family is analogous to that of the K$_2$NiF$_4$ one [4,5]. In both families the susceptibility has a broad maximum and falls off slowly in the paramagnetic region, a characteristic associated with planar magnets [6].

Mössbauer spectroscopy (MS) has been used to study the magnetic behavior of Rb$_2$FeF$_4$ [4], K$_2$FeF$_4$ [5] and FeTa$_2$O$_6$ [1,2,7]. For the K$_2$FeF$_4$ compound, the MS results have shown a gradual phase transition from the antiferromagnetic to the paramagnetic regime, with the temperature range extending from about 64 K to 70 K. In this range the spectrum consists of two fractions, belonging to antiferromagnetically ordered and paramagnetic K$_2$FeF$_4$, respectively. For Rb$_2$FeF$_4$ a similar result was observed, with the Néel temperature ranging from 55 K to 60 K. For the FeTa$_2$O$_6$ compound, as far as we know, only MS measurements at RT and at 4.2 K have been published [1,2,7]. In this communication we report preliminary results on the MS measurements of this compound at temperatures between RT and 4.2 K.
2. Experimental

Appropriate amounts of powdered Fe, Fe₂O₃, and Ta₂O₅ were mixed, ground, pelleted, encapsulated under vacuum (\(p \approx 10^{-3}\) Torr) and heated at 1320 K for 48 h, with an intermediate regrinding after 24 h. Subsequently the sample was cooled at 320 K/h and powdered to 320 mesh. X-ray powder diffraction pattern was obtained in a Siemens diffractometer with monochromated CuKα radiation (\(\lambda = 0.1542\) nm). Measurements were performed in the range of \(10 \leq T \leq 100\) with a scan rate of 2° /min. MS measurements were carried out at temperatures between 300 K and 4.2 K, using a constant acceleration electromechanical drive system with a multichannel analyzer for collecting and storing the data. \(^{57}\)Co in rhodium was used as at room temperature as a source. The temperature stability was better than 0.1 K.

3. Results and Discussion

As discussed elsewhere [7], the X-ray pattern and lattice parameters of our sample are quite similar to those published by other authors [1,2]. The hyperfine parameters obtained from the RT and from the 4.2 K MS measurements are also almost the same as those previously reported [1,2].

Typical spectra are shown in Fig. 1. The hyperfine parameters obtained by a least-squares procedure are shown in Table I. All the spectra obtained at temperatures \(T > 10\) K were fitted to a quadrupolar doublet. At 10 K the spectrum shows a very asymmetric doublet that can be fitted to a hyperfine Hamiltonian \(H_{HF} = H_M + H_Q\), where \(H_M\) is a very low effective magnetic field interaction and \(H_Q\) is the electric quadrupolar interaction, as given by [2]. The spectra obtained between 9.85 K and 4.2 K show relaxation broadening characteristic of magnetic phase transition [4,5]. These spectra were fitted to a field distribution with the same hyperfine Hamiltonian as before, and linewidth \(\Gamma\) fixed at 0.27 mm/s. This \(\Gamma\) value, obtained by fitting the RT spectrum, is indicative of the good quality of the sample; as the iron foil calibration resulted in \(\Gamma = 0.26\) mm/s, the early result suggests that there is no significant impurity effect on the Mössbauer spectra of our sample. The average values of the hyperfine fields are plotted in Fig. 2.

These results suggest that between RT and 11 K there is no indication of long-range order. Based on the fitting of the Mössbauer spectrum obtained at 10 K, this temperature was designed as the onset of long-range order. It is interesting to note that 10 K is about 0.67\(T_{\chi_{\text{max}}}\), where \(T_{\chi_{\text{max}}}\) is the temperature of the susceptibility maximum [1,2]. Such a behavior, also observed in Rb₂FeF₄ [4], is compatible with heat capacity measurements on FeTa₂O₆, as reported by Eichler et al [2]. Their data exhibit a relatively sharp peak at 8.5 K and have motivated them to assign a critical temperature of 8.5 K for the 3-d magnetic transition in FeTa₂O₆ and to attribute the susceptibility maximum at 15 K to short-range correlations.
TABLE I - Hyperfine parameters of FeTa₂O₆ measured at different temperatures. H is the average effective magnetic field at the iron sites; ΔH is the difference between the maximum and the minimum values of H used in the field distribution; ΔE_Q is the quadrupolar splitting; δ_α is the isomer shift relative to α-Fe; Γ is the linewidth at half-height; η is the asymmetry parameter of the EFG tensor.

<table>
<thead>
<tr>
<th>T (K)</th>
<th>4.2</th>
<th>8</th>
<th>9.75</th>
<th>10</th>
<th>15</th>
<th>RT</th>
</tr>
</thead>
<tbody>
<tr>
<td>H (kG)</td>
<td>254</td>
<td>251</td>
<td>207</td>
<td>5</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>ΔH (kG)</td>
<td>4</td>
<td>8</td>
<td>17</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>ΔE_Q (mm/s)</td>
<td>3.15</td>
<td>3.16</td>
<td>3.16</td>
<td>3.11</td>
<td>3.17</td>
<td>3.06</td>
</tr>
<tr>
<td>δ_α (mm/s)</td>
<td>1.23</td>
<td>1.26</td>
<td>1.28</td>
<td>1.25</td>
<td>1.26</td>
<td>1.12</td>
</tr>
<tr>
<td>Γ (mm/s)</td>
<td>0.27</td>
<td>0.27</td>
<td>0.27</td>
<td>0.27</td>
<td>0.27</td>
<td>0.27</td>
</tr>
<tr>
<td>η</td>
<td>0.33</td>
<td>0.32</td>
<td>0.31</td>
<td>0.33</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

In this sense it is interesting to discuss, in more detail, the spectra we have obtained below 10 K, which are qualitatively compatible with a slowing down of the electron-spin relaxation process in the critical region [8]. In this picture the Mössbauer hyperfine splitting is assumed to be proportional to the sublattice magnetization, and other relaxation mechanisms responsible for the line broadening are simulated by the field distribution used in the fitting algorithm. The increasing contribution from the slowing down mechanism as the temperature is decreased, is suggested by the decreasing of ΔH, as shown in Table I, but even at the lower temperature there is a small relaxation effect. The spectrum taken at 4.2 K was fitted to a field distribution with an average hyperfine field of 254 kG and a difference between the maximum and the minimum values of 4 kG; the obtained chi-square was 0.84.

We have tried to fit this spectrum with different strategies, having the chi-square as a criterion for the fitting goodness. For instance, it can be fitted to a single field with the linewidth as a free parameter; the obtained results were: H=254 kG, Γ=0.30 mm/s. Significantly, the obtained chi-square was 0.84. With Γ fixed at 0.27 mm/s, the chi-square changes to 1.04 in the single-field scheme. Thus, the fitting program systematically converges to a spectrum with broadened lines, or with a small-range field distribution. These results suggest a small relaxation effect, since the linewidth obtained at high temperatures ruled out impurities effects. In summary, fitting this spectrum to a field distribution, we are suggesting a long-range order with minor contribution from some kind of spin relaxation mechanism at 4.2 K.

As can be seen in Fig. 2, the sublattice magnetization vs T curve qualitatively resembles that one obtained for the two-dimensional Ising model, with a remarkable sharp transition at T_N=10 K. In this sense our results are compatible with the fitting of the susceptibility data performed by Eicher et al [2]. Their best result was obtained with a 2-d Ising model.

4. Conclusions

Although we do not have a more conclusive model of the magnetic behavior of the FeTa₂O₆ compound, as measured by MS, it is possible to establish, at least, a qualitative picture. From RT to about 11 K, the compound is clearly in the paramagnetic regime. Below 11 K the onset of long-range order is manifested by an asymmetry of the doublet, compatible with the emerging of a small hyperfine magnetic field. The local magnetization at the iron site increases suddenly up to about 8 K after
which it remains almost constant up to 4.2 K. A plot of the hyperfine magnetic field as a function of the temperature, shows a curve qualitatively quite similar to the one obtained for the two-dimensional Ising model. Further studies are in progress to obtain the model-related critical temperature $T_N$, as well as the critical exponent $\beta$.

A more striking result from our experiment is the absence of superposition of paramagnetic and magnetically ordered components, differently from what was observed in $\text{Rb}_2\text{FeF}_4$ [4] and in $\text{K}_2\text{FeF}_4$ [5].

Acknowledgment - We wish to thank Dr. L. Amaral and Dr. P.M. Mors for valuable comments on this manuscript. This work was supported in part by the Brazilian agencies CNPq and FINEP.

References