EVIDENCE OF MAGNETIC ORDERING IN TELLURIUM SUBSTITUTED $FeSb_2$

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We report on a ^{57}Fe Mössbauer study of tellurium substituted $FeSb_2$, $FeSb_{2-x}Te_x$ (x = 0.2, 0.4, 0.6), at temperatures between 4.2 K and 300 K. For all three alloys, the Mössbauer spectra at 4.2 K are characteristic of a magnetically ordered state. The hyperfine field at Fe site increases with increasing tellurium concentration. The magnetic character may be attributed to the existence of a very narrow band gap leading to fairly strong Coulomb and exchange interactions between holes in the valence band and electrons in the conduction band.

1. Introduction

$FeSb_2$ crystallizes in the orthorhombic C 18 type marcasite structure and is a non-magnetic semiconductor with a very small band gap ($E_g \approx 0.03$ eV) [1,2,3]. Its high susceptibility at 300 K (attaining a value of 5.8×10^{-4} emu/mole) may further be enhanced by the addition of selected impurities, leading to a magnetic ground state [4]. The $FeSb_{2-x}Te_x$ compounds with x = 0.2, 0.4 and 0.6 were found to exhibit metallic and temperature independent paramagnetic character [5]. ^{57}Fe Mössbauer experiments at 300 K for the same series revealed a continuous and smooth variation of the electric quadrupole splitting with Te concentration, suggesting that the marcasite-arsenopyrite-marcasite transitions are smooth and continuous [6]. We report here primarily on Mössbauer experiments carried out at 4.2 K $\leq T \leq$ 300 K for the compounds with x = 0.2, 0.4 and 0.6. A preliminary analysis of the data reveals unambiguously that these compounds order magnetically below 70 K.

2. Experimental Details

These ternary compounds prepared by a solid state reaction method were characterized by X-ray diffraction [5,7]. The Mössbauer spectra were recorded with a $^{57}Co(Rh)$ source at room temperature and the absorber was kept inside a liquid helium cryostat. The absorber temperature could be varied by applying suitable current to a heater. The temperature could be measured and controlled within $\pm$ 1 K. The spectra were fitted with superpositions of Lorentzian lines. In the presence of magnetic hyperfine interactions, the line positions and intensities were calculated by diagonalization of the full hyperfine Hamiltonian.

3. Results and Discussion

Fig. 1 shows the spectra measured at 4.2 K and 300 K for x = 0.2, 0.4 and 0.6. The spectra for $FeSb_2$ and $FeTe_2$ at 4.2 K have also been included in the figure to bring out clearly the differences between the end member compounds and the mixed compounds. The

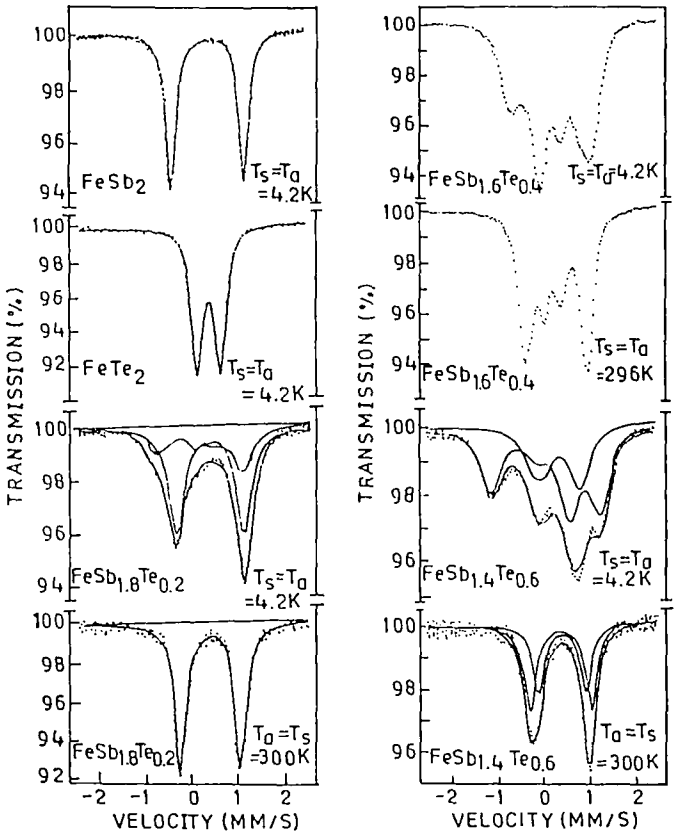

Figure 1: Mössbauer spectra of tellurium substituted $FeSb_2$. T_a and T_s refer to the absorber and source temperature, respectively.

Mössbauer spectra of $FeSb_2$ and $FeTe_2$ could be fitted with a quadrupole doublet, which shows that they are non-magnetic at 4.2 K. As the tellurium concentration in the mixed system increases, a magnetic hyperfine field at Fe site develops and reaches a maximum at about 6 Tesla for $x = 0.6$. The spectrum for $x = 0.2$ was fitted assuming two iron sites, of which one exhibits a magnetic splitting while the other is a quadrupole doublet. For the specimen with $x = 0.6$, the spectrum was fitted with two distinctly different, non-zero hyperfine fields (≈ 6 Tesla and 2 Tesla). For comparison, the room temperature spectra also shown in Fig. 1 are typical for a non-magnetic state.

Fig. 2 shows the temperature dependence of the Mössbauer spectra for $x = 0.4$ and 0.6. The fitting of these spectra using the full Hamiltonian is in progress. Nevertheless, the spectra clearly show that the magnetic hyperfine interaction sets in as the temperature is lowered. The transition temperature for $x = 0.4$ and 0.6 is about 70 ± 5 K and 65 ± 5 K.

The magnetic behaviour may be attributed to the existence of a very narrow band

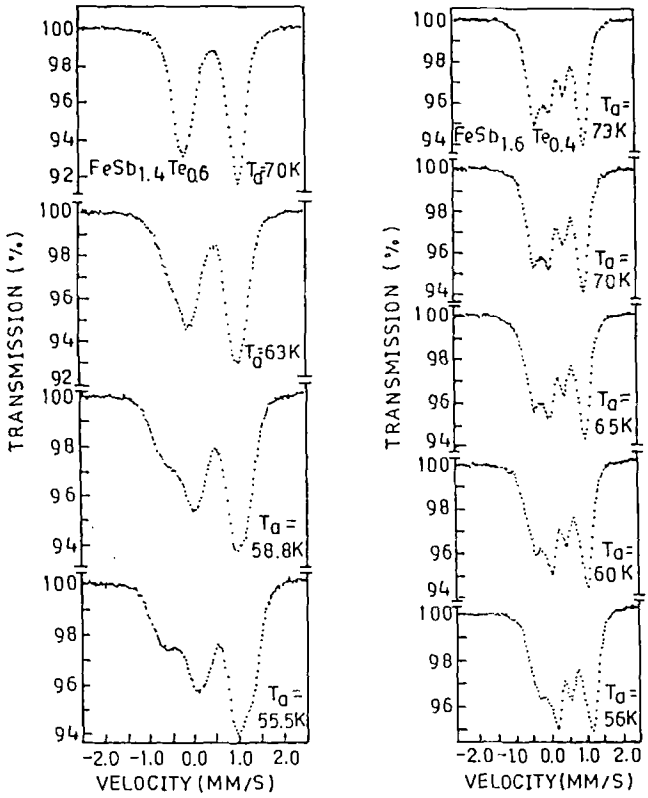

Figure 2: Temperature dependence of the Mössbauer spectra of tellurium substituted $FeSb_2$. T_a refers to the absorber temperature. All spectra were recorded with the $^{57}Co(Rh)$ source at 300 K.

gap in $FeSb_2$ which is further reduced by substitution of antimony by tellurium. Already in $FeSb_2$, both the valence and conduction band states are derived from d-like states which have fairly strong Coulomb and exchange interactions between holes in the valence band and electrons in the conduction band [4,8]. Moreover as the electrons are thermally excited from valence to conduction band when the temperature is raised, not only intra-band interactions will contribute but also inter-band Hund type coupling between electrons becomes important and gives rise to a stable magnetic ground state. In order to gain a better understanding of the magnetic character of the mixed compounds, systematic studies of the band gap, of the magnetic susceptibility and also neutron diffraction experiments are being planned. A detailed analysis of the Mössbauer spectra for the whole series is in progress and will be reported separately elsewhere [9].

4. Conclusion

The Mössbauer spectra of the compounds with $x = 0.2$, 0.4 and 0.6 of the series $FeSb_{2-x}Te_x$ reveal unusual magnetic behaviour below 70 K.

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