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MAGNETIC SUSCEPTIBILITY OF $\text{Cd}_{1-x}\text{Fe}_x\text{Te}_{1-y}\text{Se}_y$

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Magnetic susceptibility of $\text{Cd}_{1-x}\text{Fe}_x\text{Te}_{1-y}\text{Se}_y$ was found to consist of Van Vleck term (characteristic for the Fe^{2+} centers in tetrahedral crystal symmetry) and temperature-dependent contribution. The occurrence of the latter indicates an existence of additional magnetic moments. We ascribe their origin to the modification of the Fe^{2+} energy level spectrum induced by mixed Te-Se anion environment.

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Magnetic properties of diluted magnetic semiconductors (DMS) depend primarily on the type of the substitutional magnetic ion, and far less on the semiconductor host. Hence the behavior of different II-VI systems containing Fe^{2+} ions shows common characteristics.

The Fe^{2+} free ion ground state for the d^6 electronic configuration is 5D . When the $\text{Fe}^{2+}(3d^6)$ impurity is substitutionally incorporated in the cubic crystal, the 5D state is split under the influence of the tetrahedral crystal field and spin-orbit interaction in such a way that the resultant ground level is a magnetically inactive singlet Γ_1 . Thus Fe^{2+} ions reveal only a field-induced magnetic moment. Consequently at very low temperatures, where the thermal energy is much smaller than the distance between the ground and excited states, temperature-independent Van Vleck paramagnetism should occur. A corresponding saturation of the magnetic susceptibility of Fe^{2+} in several II-VI compound crystals have been actually observed [1].

In this paper we present an attempt to change the typical magnetic properties of the Fe^{2+} ion by using the ternary mixed-anion alloy $\text{CdTe}_{1-y}\text{Se}_y$ as the host material. The $\text{Cd}_{1-x}\text{Fe}_x\text{Te}_{1-y}\text{Se}_y$ crystals used in this study were grown by the Bridgman method. The crystal structure of the material established using powder and single crystal X-ray diffraction, was found to be zinc blende in the entire concentration range under investigation. The homogeneity and composition

of the samples were measured by an electron microprobe and X-ray fluorescence analysis. We studied the samples with selenium mole fraction y up to 0.13 and iron concentration x not exceeding 0.01.

The a.c. magnetic susceptibility measurements were performed using a standard mutual inductance method. An excitation field of amplitude 40 Oe and frequency 75 Hz was employed. Representative susceptibility data taken in the temperature range from 1.6 K to 4.2 K are shown in Fig. 1. The ternary compounds

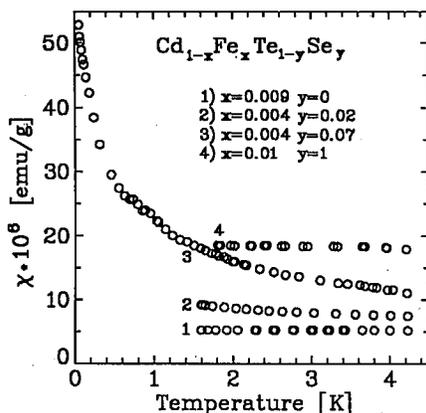


Fig. 1. Low-field magnetic susceptibility of the $\text{Cd}_{1-x}\text{Fe}_x\text{Te}_{1-y}\text{Se}_y$ crystals. The diamagnetic susceptibilities of the host crystals have been subtracted from the experimental data.

$\text{Cd}_{1-x}\text{Fe}_x\text{Te}$ and $\text{Cd}_{1-x}\text{Fe}_x\text{Se}$ reveal a temperature-independent paramagnetism. In the case of quaternary $\text{Cd}_{1-x}\text{Fe}_x\text{Te}_{1-y}\text{Se}_y$ alloy with small Se concentrations (for instance $y = 0.02$) the magnetic susceptibility resembles that observed in the "parent" compound. For higher Se concentrations the susceptibility increases as the temperature is decreased, and its temperature dependence becomes stronger with increasing Se concentration.

Such a behavior could be induced by a modification of the Fe^{2+} energy level spectrum in two alternative ways: (i) either the magnetically active state becomes the ground state, or (ii) the energy distance between the ground and the first excited state is significantly reduced. In order to resolve the above question, we have extended our magnetic susceptibility studies of a $\text{Cd}_{0.996}\text{Fe}_{0.004}\text{Te}_{0.93}\text{Se}_{0.07}$ sample to the millikelvin temperature range. A permanent increase of the susceptibility was observed down to 30 mK. As one can see in Fig. 1, the increase of the susceptibility is clearly enhanced at extremely low temperatures. The above result suggests that the first of the two possibilities — the change of the energy level sequence — occurs in the investigated quaternary crystals.

This can be understood as follows. Let us consider an isolated Fe^{2+} center in $\text{Cd}_{1-x}\text{Fe}_x\text{Te}_{1-y}\text{Se}_y$ in a ligand field of nearest anions. In this case a given Fe^{2+} ion can be surrounded by 0, 1, 2, 3 or 4 Se nearest-neighbors, the remaining ions in its tetrahedral surrounding being Te. Assuming a random distribution of

Fe and Se ions throughout the lattice, we can evaluate the probabilities of finding the Fe^{2+} ion in one of the above environments. In the crystal with low Se and very low Fe^{2+} concentrations (which is the case in point) only two configurations of nearest-neighbors are probable: the Fe^{2+} iron is (i) either surrounded by a Te quartet, or (ii) by one Se and three Te ions. In the former case the symmetry of the Fe^{2+} environment is obviously the same as that of Fe^{2+} in CdTe. In that case the energy level scheme of the Fe^{2+} ion should remain undisturbed, to a good approximation. In the case of a mixed anion environment the presence of a Se ion replacing one of the Te ions induces a strong distortion (contraction) in the bond direction (111), due to the 13% difference between Se and Te covalent radii.

The energy level spectrum of the Fe^{2+} ion was calculated as a function of the distortion parameter ν' , by using a model similar to that used by Slack et al. [2]. The results of our calculations for the lowest-lying energy levels is presented in Fig. 2. Note that for ν' lower than -175 cm^{-1} , the Γ_3 doublet becomes the ground state of the Fe^{2+} ion.

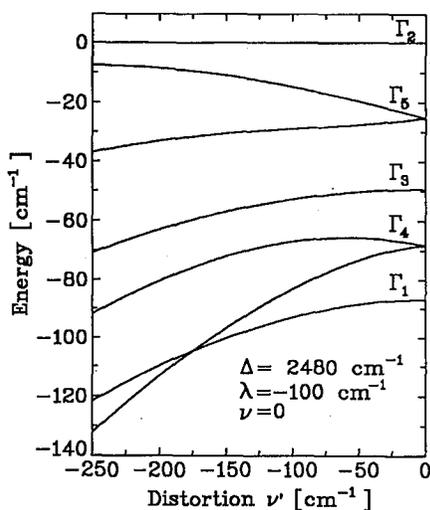


Fig. 2. The low-lying energy levels of the $Fe^{2+}(3d^6)$ ion in a tetrahedral environment distorted in the bond direction (111) as a function of the distortion parameter ν' .

We conclude that in $Cd_{1-x}Fe_xTe_{1-y}Se_y$ the magnetic susceptibility consists of two main components: a temperature-independent Van Vleck susceptibility of the Fe^{2+} centers in the environment of four-like anions (Te), and a second temperature-dependent term arising from magnetic moments of Fe^{2+} ions possessing a magnetically active ground state. We ascribe the existence of this latter to the mixed Te-Se anion environment.

Acknowledgments

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