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# The complex structure of $\text{Fe}^{2+} (3d^6)$ magnetism in $\text{HgSe}_{1-y}\text{S}_y$ mixed crystals<sup>†</sup>

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## Abstract

High magnetic field magnetization measurements of  $\text{Hg}_{1-x}\text{Fe}_x\text{Se}_{1-y}\text{S}_y$  ( $x = 0.002$ ,  $y = 0.17$ ;  $y = 0.28$ ) at temperatures ranging from 1.6 to 119 K and in magnetic fields up to 20 T are reported. The magnetization of the  $\text{Fe}^{2+} (3d^6)$  ion in the investigated mixed anion crystals is shown to be different from the Van Vleck behaviour. A theoretical description of the experimental data is based on an assumption that the electronic structure (and thus magnetic properties) of the  $\text{Fe}^{2+} (3d^6)$  ion in a mixed anion crystal depends on its local environment. In this model the total magnetization is calculated as a statistically weighted sum of magnetizations found for specific local configurations. The model provides a good quantitative description of the experimental data.

The electronic structure of the  $\text{Fe}^{2+} (3d^6)$  ion diluted in  $A_{II}B_{VI}$  compounds has been described in the literature within a framework of the crystal field model [1]. The calculated magnetization is in quantitative agreement with experimental data for compounds crystallizing in the zinc-blende as well as in the wurzite structure [2]. In the wurzite structure case the crystal field potential contains a trigonal distortion term which reflects  $C_{3v}$  symmetry of the nearest neighbourhood of a  $\text{Fe}^{2+} (3d^6)$  ion [3]. Usually in  $A_{II}B_{VI}$  semiconductors doped with iron the ground state of the  $\text{Fe}^{2+} (3d^6)$  ion ( $s = 2$ ,  $l = 2$ ) is a magnetically inactive singlet. Thus the  $\text{Fe}^{2+} (3d^6)$  ion exhibits only field induced magnetic moment, known as Van Vleck paramagnetism. The main features of such

a paramagnet are: temperature independent susceptibility (and magnetization) at low temperatures and a very slow saturation of magnetization in high magnetic fields.

It turns out that magnetic properties of *mixed anion* crystals  $\text{Cd}_{1-x}\text{Fe}_x\text{Te}_{1-y}\text{Se}_y$  are substantially different [4, 5]. In that case the susceptibility does not exhibit Van Vleck behaviour, which led the authors to the conclusion that the local environment of the  $\text{Fe}^{2+} (3d^6)$  ion may essentially influence its energy spectrum. This assumption allowed to describe the magnetization of  $\text{Fe}^{2+} (3d^6)$  in  $\text{CdTe}_{1-y}\text{Se}_y$  for small  $y$  [5] by taking into account only contributions from  $\text{Fe}^{2+} (3d^6)$  in 4 Te tetrahedral environment and 3Te1Se trigonally distorted tetrahedral environment. The remaining possible configurations were neglected.

In the present paper we analyse the magnetization of a similar random alloy  $\text{Hg}_{1-x}\text{Fe}_x\text{Se}_{1-y}\text{S}_y$ , but in a higher composition range. Thus we have to take into account the electronic structure of the  $\text{Fe}^{2+} (3d^6)$  ion for all possible types of its nearest neighbourhoods.

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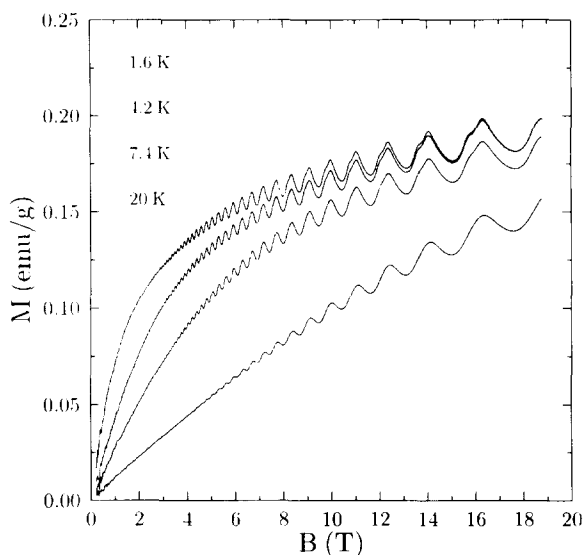


Fig. 1. The magnetization of  $\text{Hg}_{0.998}\text{Fe}_{0.002}\text{Se}_{0.83}\text{S}_{0.17}$  at a few temperatures.

The crystals were grown by the modified Bridgman method. The composition of samples used in the experiment were obtained by the energy dispersive X-ray fluorescence analysis ( $\text{Hg}_{1-x}\text{Fe}_x\text{Se}_{1-y}\text{S}_y$ ;  $x = 0.002 \pm 0.0005$ ,  $y = 0.17 \pm 0.01$ ;  $x = 0.002 \pm 0.0005$ ,  $y = 0.28 \pm 0.01$ ). The magnetization measurements were carried out at temperatures from 1.6 to 119 K in the magnetic field up to 20 T ( $B \parallel [110]$ ). A high precision capacitive torsional magnetometer has been used to collect the magnetization data continuously during the sweep of the Bitter magnet. The recorded traces of magnetization are presented in Figs. 1 and 2. The oscillatory behaviour superimposed on the magnetization of Fe ions in high magnetic fields is caused by the magnetization of free electrons. As can be noticed, the magnetization curves measured at 1.6 and 4.2 K do not coincide at low magnetic fields which implies that the  $\text{Fe}^{2+}(3d^6)$  ion does not exhibit the pure Van Vleck paramagnetism.

In order to describe the magnetization data we considered five possible configurations of a  $\text{Fe}^{2+}(3d^6)$  ion environment characterized by a different number of S and Se neighbours: 4Se, 3Se1S, 2Se2S, 1Se3S, 4S. The total magnetization was obtained as a sum of statistically weighted contributions of magnetizations calculated in each of these configurations. Calculations of the electronic structure of  $\text{Fe}^{2+}(3d^6)$  were performed in a way similar to that described in Ref. [3]. Our Hamiltonian always contained the tetrahedral crystal field potential, spin-orbit interaction and the Zeeman term. The presence of every sulphur atom in the neighbourhood of the

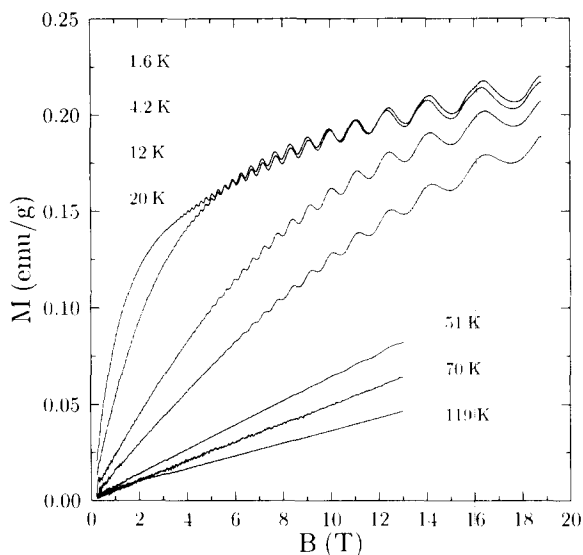


Fig. 2. The magnetization of  $\text{Hg}_{0.998}\text{Fe}_{0.002}\text{Se}_{0.72}\text{S}_{0.28}$  at a few temperatures.

$\text{Fe}^{2+}(3d^6)$  ion was modelled by an additional crystal field potential with the strength  $v'$  [6] (in Ref. [3] that parameter described the trigonal distortion due to the crystallographic structure). Depending on the number of S atoms in the nearest neighbourhood of the  $\text{Fe}^{2+}(3d^6)$  ion we deal with different types of symmetry. In particular we have  $C_{3v}$  symmetry for 1S or 3S atoms and  $C_{2v}$  for 2S atoms.

The eigenproblem for each configuration was solved numerically and the magnetization was calculated in the standard way. We used the following parameters: the crystal field splitting  $10Dq = -2430 \text{ cm}^{-1}$  [7], the spin-orbit interaction  $\lambda = -106 \text{ cm}^{-1}$  [8] and the best fitted  $v' = -180 \text{ cm}^{-1}$ .

As the probability of the particular neighbourhood at the given composition we used the binomial distribution. In Fig. 3 we showed, for two lowest temperatures, four main contributions to the magnetization originating from different  $\text{Fe}^{2+}(3d^6)$  ions' surroundings as well as the total magnetization compared to the experimental curves. The case of 4S atoms in an iron ion surrounding is not shown, because at higher magnetic field  $B = 20 \text{ T}$  its absolute contribution to the total magnetization is as small as  $0.003\mu_B$  for  $y = 0.17$  and  $0.02\mu_B$  for  $y = 0.28$ . Such theoretical description provides a good agreement with experimental data.

In conclusion, it is shown that the influence of sulphur atoms in the nearest neighbourhood of the Fe ion on its electronic structure can be described by one phenomenological parameter.

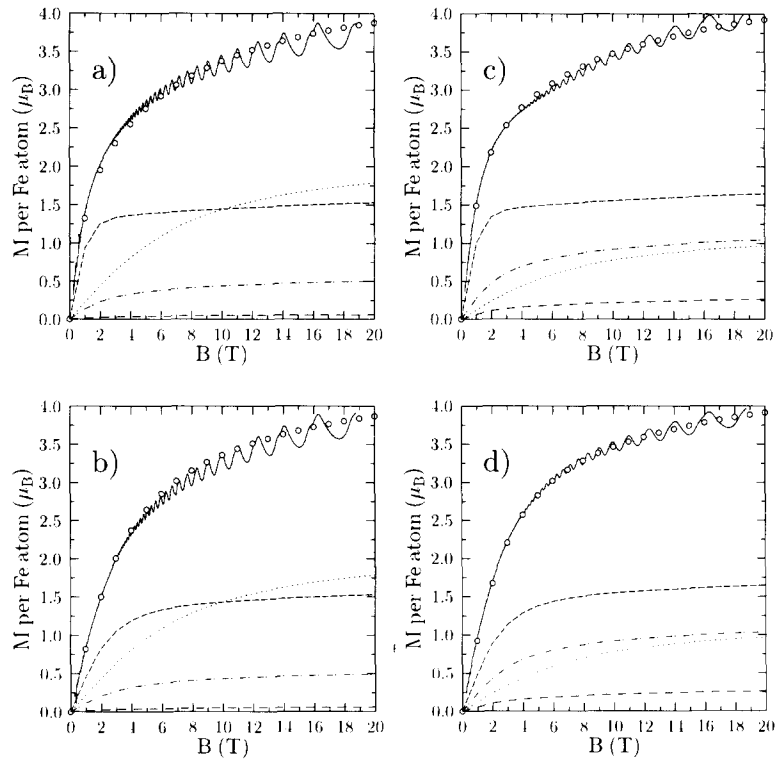


Fig. 3. The measured magnetization of  $\text{Hg}_{1-x}\text{Fe}_x\text{Se}_{1-y}\text{S}_y$  (solid line) compared to the results of calculations (open circles). The calculated magnetization was obtained as a sum of the following contributions:  $4\text{Se}(\text{T}_d)$  – dotted line,  $3\text{Se}1\text{S}(\text{C}_{3v})$  – dashed line,  $2\text{Se}2\text{S}(\text{C}_{2v})$  – dash-dotted line,  $1\text{Se}3\text{S}(\text{C}_{3v})$  – long dashed line; (a)  $x = 0.002$ ,  $y = 0.17$ ,  $T = 1.6$  K; (b)  $x = 0.002$ ,  $y = 0.17$ ,  $T = 4.2$  K; (c)  $x = 0.002$ ,  $y = 0.28$ ,  $T = 1.6$  K; (d)  $x = 0.002$ ,  $y = 0.28$ ,  $T = 4.2$  K.

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