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MAGNETIC SUSCEPTIBILITY STUDIES OF Fe-DOPED II–VI COMPOUNDS

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Low-field magnetic susceptibility χ of Fe-doped II–VI compounds has been studied for wide iron concentration range ($3\times10^{18}\leq N_{Fe}\leq10^{21}$ cm^{-3}). For the low iron content the Van Vleck-type paramagnetism is observed at low temperatures in all investigated materials (except for $Hg_{1-x}Fe_xSe$), which entirely confirms the theoretical prediction. For the higher Fe contents the experimental data still remain unexplained.

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The problem of the $Fe^{2+}(3d^6)$ ion in a tetrahedral crystal-field configuration has been treated in the literature [1]. The ground state of Fe^{2+} ion is a magnetically inactive singlet, which leads to a Van Vleck-type paramagnetism. A corresponding saturation of the magnetic susceptibility at low temperature was previously observed in several II–VI compound crystals, doped with small amounts of iron ($N_{Fe}\leq7\times10^{18}$ cm^{-3}) [2]. However, in samples containing higher iron mole fraction ($x\geq0.02$), the full saturation was never reached, even at very low temperatures [3, 4]. An observed increase of the magnetic susceptibility while lowering temperature has been attributed either to a presence of foreign paramagnetic impurities (such as Mn^{2+} ions) [4], or to a strong mixing between the conduction band states and $Fe^{2+}(3d^6)$ states in the case of $Hg_{1-x}Fe_xSe$ [3].

In order to shed some light on the above problem, we performed a detailed study of the low-field magnetic susceptibility for three types of II–VI compounds, characterized by a different energetic location of the $Fe^{2+}(3d^6)$ level: (i) Fe^{2+} level degenerate with the conduction band (HgSe:Fe), (ii) located in the energy gap (CdSe:Fe, CdTe:Fe), (iii) degenerate with the valence band (HgTe:Fe). We studied the samples with the Fe content changing in the range $0.0002\leq x\leq0.06$.

A standard mutual inductance method has been used. We applied the alternating magnetic field of amplitude 40 Oe and frequency 75 Hz. The absolute value

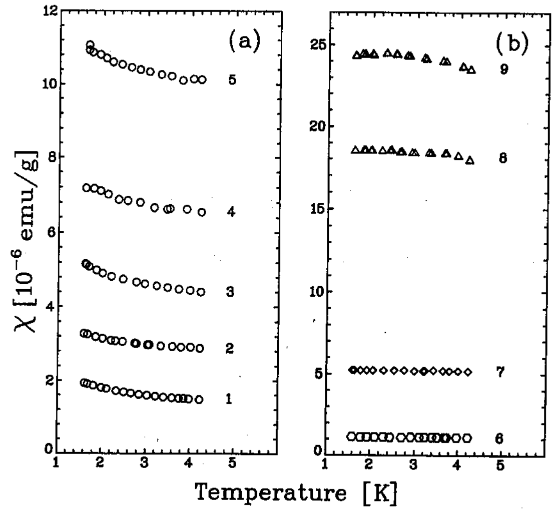

Fig. 1. The magnetic susceptibility (per gram) in the low temperature region for low iron concentration ($x \leq 0.02$):
(a) in $Hg_{1-x}Fe_xSe$: numbers label data obtained for samples with different iron mole fraction: 1) x = 0.0002, 2) x = 0.0003, 3) x = 0.003, 4) x = 0.006, 5) x = 0.01;
(b) in other investigated II–VI compounds: 6) $Hg_{0.99}Fe_{0.01}Te$, 7) $Cd_{0.99}Fe_{0.01}Te$, 8) $Cd_{0.99}Fe_{0.01}Se$, 9) $Cd_{0.98}Fe_{0.02}Se$.

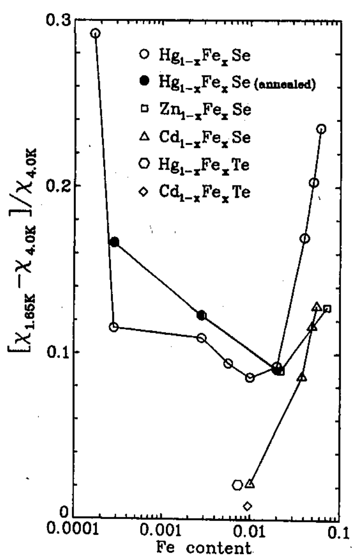

Fig. 2. The relative change of the magnetic susceptibility between 1.65 K and 4.0 K; the lines are guides for eye only; data for $Zn_{1-x}Fe_xSe$ are taken from [5].

of the magnetic susceptibility was determined with experimental accuracy better than 3 %.

As expected, at high temperatures all the samples reveal a typical Curie–Weiss behaviour, which was already discussed elsewhere (see for example [2, 3, 5]), thus we limit our presentation to the low-temperature results. The magnetic susceptibility as a function of temperature in the range from 1.6 K to 4.2 K is shown in Fig. 1(a),(b) for different compounds with low Fe concentration ($x \leq 0.02$). One can see that all the investigated crystals (except for $Hg_{1-x}Fe_xSe$) exhibit Van Vleck paramagnetism in the temperature range of interest. Hence we can rule out the presence of appreciable amounts of any paramagnetic impurities, which should give rise to Curie-like behaviour of the susceptibility even at very low temperature. The fact that the Fe^{2+} level is degenerate with the valence band, as occurs in $Hg_{1-x}Fe_xTe$, does not influence the magnetic properties of the crystal. This provides an argument against a mixing between the valence band and $3d$ states. As we mentioned above, the situation is quite different in the case of $Hg_{1-x}Fe_xSe$: we observed a permanent increase of the susceptibility with lowering temperature, even for extremely low concentrations of Fe ions. We assign this effect to the presence of Fe^{3+} ions in this compound [6].

For $x > 0.02$ the susceptibility behaviour is not understood: all the investigated crystals exhibit strong temperature dependence of χ. Moreover, the increase of the susceptibility in low temperature range becomes enhanced with the iron concentration.

In Fig. 2 we gathered together the results obtained for all investigated II–VI materials. The relative increase of χ between 1.65 K and 4.0 K is specified as a function of Fe content. It seems that the susceptibility behaviour depends on the kind of a host material, the system $Hg_{1-x}Fe_xSe$ being especially interesting. In this case three regions of the Fe concentration are distinguished:

(i) the very low iron concentrations ($x \leq 0.003$), for which nearly all of iron ions present in the crystal are in Fe^{3+} state. This results in a strong increase of the susceptibility with lowering temperature. The observed change of χ is smaller than expected, because the number of Fe^{3+} ions is reduced due to the presence of other (native) donors in our crystals [6]. Some of our samples were annealed in saturated Se vapour in order to diminish the concentration of native donors, and then the susceptibility was measured again. This procedure results in enhanced χ increase, as it was expected.

(ii) In the Fe concentration range $0.003 < x < 0.02$ the number of Fe^{2+} ions becomes considerably higher than the number of Fe^{3+} ions. Therefore, the Van Vleck term (due to Fe^{2+}) should predominate over Curie-like Fe^{3+} contribution to the susceptibility and the relative χ change with temperature should diminish, as it was observed.

(iii) For the Fe molar fraction $x > 0.02$ the fraction of Fe^{3+} ions becomes insignificant. The behaviour of χ is then similar to that observed for other materials. As mentioned, the temperature dependence of χ becomes stronger with the increasing iron concentration. It should be also mentioned that in samples with the still higher iron concentration the susceptibility curves show a broad maximum, related to a spin-glass formation [3].

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