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## MAGNETIZATION AND SUSCEPTIBILITY OF $\text{Cd}_{1-x}\text{Co}_x\text{Se}^*$

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The magnetization and magnetic susceptibility of Bridgman-grown  $\text{Cd}_{1-x}\text{Co}_x\text{Se}$  with values of  $x$  up to 0.04 have been measured over a temperature range from 2 to 390 K and in magnetic fields up to 23 T. A pair-exchange value  $J/k_B$  of about  $-3$  K was obtained from the magnetic field dependence of the magnetization. This exchange is probably due to the next-nearest-neighbor interaction. The high-temperature susceptibility data indicated a presence of the Curie-Weiss type paramagnetism and the temperature independent Van Vleck type paramagnetism.

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### 1. Introduction

The system  $\text{Cd}_{1-x}\text{Co}_x\text{Se}$  is a diluted magnetic semiconductor (DMS) with some significant differences from its more thoroughly studied counterpart  $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$ . The magnetic ion  $g$ -factor in  $\text{Cd}_{1-x}\text{Co}_x\text{Se}$  is anisotropic, with values at 78 K  $g_{\parallel} = 2.29$  and  $g_{\perp} = 2.3$  [1]. In  $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$   $g = 2$  and is isotropic. The spin of the Co-ion in DMS equals 1.5, while the spin of the Mn-ion equals 2.5. Lewicki et al. investigated the magnetic susceptibility of  $\text{Cd}_{1-x}\text{Co}_x\text{Se}$  and found the exchange interaction parameter  $J/k_B$  of  $-37$  K, about five times larger than in  $\text{Cd}_{1-x}\text{Mn}_x\text{Se}$  [2]. Shapira et al. measured the high-field magnetization in  $\text{Zn}_{1-x}\text{Co}_x\text{Se}$  and  $\text{Zn}_{1-x}\text{Co}_x\text{S}$  and from the steps observed in magnetization they determined an exchange parameter of an order of  $-3$  K [3]. They attributed this value to the next-nearest-neighbor interaction.

In the present paper we report some results of high-field low-temperature magnetization and low-field high-temperature susceptibility measurements in  $\text{Cd}_{1-x}\text{Co}_x\text{Se}$ .

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## 2. Experiment

The crystals of  $\text{Cd}_{1-x}\text{Co}_x\text{Se}$  were grown by the Bridgman technique and the Co concentrations were determined by electron microprobe analysis. The estimated uncertainties in the  $x$ -value, including variation throughout the sample, were about 20 %.

The susceptibility measurements were carried out on a SQUID magnetometer, over a temperature range from 2 to 390 K, and the maximum field was 0.05 T (for details see Ref. [4]). The high-field studies up to 23 T at 4.2 K were carried out on hybrid magnets in the High-Field Laboratory for Superconducting Materials of Tohoku University. The experimental details were described previously [5].

## 3. Results and discussion

Magnetization as a function of magnetic field is shown in Fig. 1. The experimental data were fitted to the expression

$$M = M_S + M_P + \chi_0 H, \quad (1)$$

where  $H$  is the magnetic field,  $M_S$  and  $M_P$  are contributions from magnetic singles and pairs, respectively, and  $\chi_0$  is the susceptibility of the host lattice. In CdSe we determined  $\chi_0 = -4 \times 10^{-7}$  emu/g. The complete expressions for  $M_S$  and  $M_P$  are given in Ref. [5]. The fitting parameters were  $\bar{x}_1$  in  $M_S$ ,  $\bar{x}_2$  and  $J_P$  in  $M_P$ , where  $\bar{x}_1$  and  $\bar{x}_2$  are effective numbers of magnetic ion singles and pairs, respectively, and  $J_P$  is the pair exchange parameter value. The results of fits are shown in Fig. 1 as solid lines.

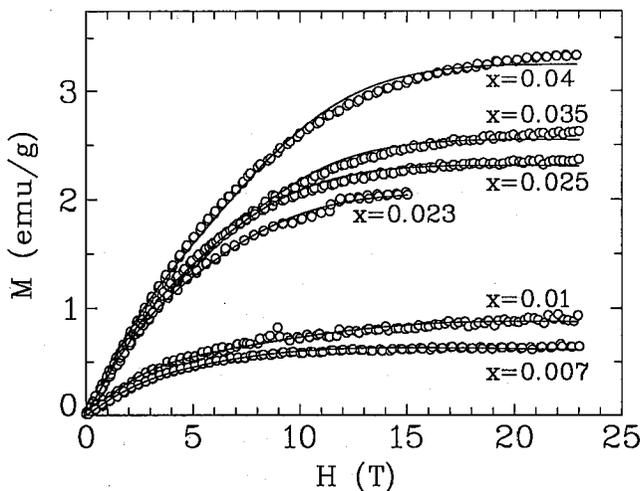


Fig. 1. Magnetization of  $\text{Cd}_{1-x}\text{Co}_x\text{Se}$  at 4.2 K. Solid lines are fits to the Brillouin function and pair-exchange interaction.

From the fits we obtained the average value of  $J_P/k_B = -2.6 \pm 0.5$  K. This is more than an order of magnitude less than the exchange parameter value reported

by Lewicki et al. [2]. However, the first step in magnetization for an exchange value of 40 K would appear at field of more than 60 T. The magnetization below that field would be dominated mostly by the next-nearest-neighbor interaction. Therefore, we believe that our value of  $J_P$  is the exchange parameter value of the next-nearest-neighbor interaction. Shapira et al. also obtained a low value of the exchange parameter in Co-doped zinc chalcogenides and interpreted it as the next-nearest-neighbor interaction [3].

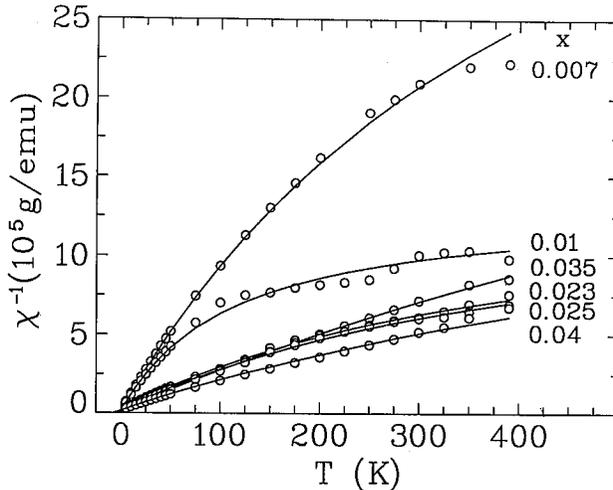


Fig. 2. Inverse susceptibility for  $Cd_{1-x}Co_xSe$ . Solid lines are fits to the Curie-Weiss law and a temperature-independent Van Vleck type paramagnetism.

Inverse susceptibility as a function of temperature is shown in Fig. 2. The data are different from those commonly observed in DMS. The apparent saturation of  $\chi^{-1}$  at high temperatures, particularly for small  $x$ -values, indicates a presence of a temperature-independent paramagnetism. Lewicki et al. pointed out the existence in  $Cd_{1-x}Co_xSe$  of a temperature-independent paramagnetic Van Vleck contribution, depending on the spin-orbit splitting [2]. Following this concept we fitted the susceptibility data to the expression

$$\chi = \frac{P_1}{T + \theta} + \chi_0 - 2G, \quad (2)$$

where  $T$  is the absolute temperature,  $P_1$  is the Curie constant,  $\theta$  is the Curie temperature, and  $2G$  is the Van Vleck type temperature-independent term depending on the magnetic ion  $g$ -factor and the spin-orbit coupling  $\lambda$ . The fitting parameters were  $P_1$ ,  $\theta$ , and  $G$ , and the results of fits carried out from 5 to 390 K are shown in Fig. 2 as solid lines.

Unfortunately, the parameters  $P_1$  and  $\theta$  depended strongly on the temperature range over which the relation (2) was applied and we could obtain almost any value of  $J/k_B$  from 4 to 100 K, depending upon the fit range. The fits carried out over the whole temperature range gave reasonable (though not quite reliable)

values of the effective numbers of magnetic ions and the average value of  $J/k_B$  obtained from those fits was about  $-6.5 \pm 1.5$  K, in between the high value reported by Lewicki et al. and the low value obtained from high-field magnetization data. The temperature-independent paramagnetic contribution was about an order of magnitude larger than that estimated by Lewicki et al., and corresponded to a value of  $\lambda$  an order of magnitude smaller than the free ion value of spin-orbit interaction.

We see that the high-temperature susceptibility data are difficult to interpret. It was not possible to get from them unambiguous results on the nearest-neighbor exchange. From the low-temperature high-field magnetization data we obtained a low value of the exchange parameter, possibly due to the next-nearest neighbor interaction.

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