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Anisotropic magnetic susceptibility of hexagonal Co-based diluted magnetic semiconductors

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We present magnetic-susceptibility data for two new cobalt-based diluted magnetic semiconductor systems with the wurtzite lattice structure: $Cd_{1-x}Co_xSe$ and $Cd_{1-x}Co_xS$. At low temperatures, the magnetic susceptibility of both systems is anisotropic. We analyze the low-temperature data within the framework of the crystal-field model, including pair interaction. The analysis yields the value of the zero-field splitting D/k_B of the Co^{2+}-ion orbital ground state as 1.0 ± 0.1 K for the selenides, and as 0.85 ± 0.1 K for the sulfides. From quantitative analysis of the high-temperature data, we obtain the value of the nearest-neighbor Co^{2+}-Co^{2+} exchange integral J/k_B for $Cd_{1-x}Co_xSe$ to be -37 ± 5 K.

I. INTRODUCTION

The most extensively studied diluted magnetic semiconductors (DMS's)—also referred to as semimagnetic semiconductors—are alloys based on II-VI semiconductor compounds, in which a fraction x of the cations (Zn^{2+}, Cd^{2+}, or Hg^{2+}) has been randomly replaced by Mn^{2+} or Fe^{2+} ions. The results of experimental and theoretical investigations on these materials have been compiled in many reviews, for example Refs. 1–3. Recently, DMS alloys containing Co^{2+} ions have also been successfully synthesized, and results concerning electronic Raman scattering in $Cd_{1-x}Co_xSe$ (Ref. 4), inelastic neutron scattering,[5] and magnetic susceptibility of the cubic $Zn_{1-x}Co_xSe$ and $Zn_{1-x}Co_xS$ (Ref. 6) have already been published.

In this paper we report a study of the magnetic properties of two new Co-based alloys, $Cd_{1-x}Co_xSe$ and $Cd_{1-x}Co_xS$, both of which have a hexagonal (wurtzite) lattice structure. The hexagonal structure possesses a polar direction (c axis), corresponding to the trigonal symmetry axis. In the wurtzite lattice (as in the zinc blende structure), each cation is surrounded by four anions situated on the corners of a tetrahedron. Wurtzite materials, however, usually crystallize with small c-axis distortions, i.e., the c/a ratio is not exactly equal to the value $\sqrt{8/3}$ predicted by ideal hcp arrangement. This small lattice distortion leads to a significant change of the magnetic properties, manifesting itself as an anisotropy of the magnetic susceptibility at low temperatures. This is the most distinctive feature differentiating wurtzite DMS's containing Co^{2+} ions from their zinc-blende counterparts, e.g., $Zn_{1-z}Co_xS$ or $Zn_{1-x}Co_xSe$ (Ref. 6). We will discuss this point more fully later in the paper.

This paper is organized as follows. In Sec. II we give a brief outline of two theoretical models describing the magnetic susceptibility of Co^{2+} ions in a wurtzite lattice. We also present a calculation of pair contribution to the total susceptibility. In Sec. III we present the low-temperature, single-crystal experimental data for the two systems under investigation, along with the best fits of theoretical curves for the two principal orientations of the magnetic field, i.e., $\mathbf{H}\parallel\hat{c}$ and $\mathbf{H}\perp\hat{c}$. In Sec. IV we analyze the high-temperature regime of the magnetic susceptibility, which provides a determination of the exchange integral J between nearest Co^{2+} neighbors.

II. THEORETICAL MODEL

It is well known that the crystal field plays an important role in determining the magnetic properties of transition metal ions. In the case of Co-based DMS materials with wurtzite structure, the crystal field potential consists of two terms. The first—and dominant—is the tetrahedral-symmetry T_d term. The second, and much smaller term, results from the lattice distortion mentioned earlier, and from electrostatic interactions with the more distant ions. This latter term has trigonal symmetry. The details of the solution of the crystal field Hamiltonian which accounts for these effects have been presented by several authors,[7–10] the most exhaustive of these being Ref. 10.

The energy-level diagram for a Co^{2+} ion occupying a lattice site in a wurtzite crystal is shown in Fig. 1. Under the action of the tetrahedral crystal field the free-ion ^4F ground state splits into a $^4A_2$ ($^4\Gamma_2$) orbital singlet and two orbital triplets, $^4T_2$ ($^4\Gamma_5$) and $^4T_1$ ($^4\Gamma_4$). The $^4T_2$ excited state is separated from the $^4A_2$ ground state by the amount $\Delta\simeq3300$ cm^{-1} (Ref. 7). The value of Δ is much larger than the thermal energy at room temperature, and thus the occupations of the $^4T_2$ and the $^4T_1$ states are much smaller than that of the ground state. In the presence of the trigonal term and of spin-orbit interaction, the energy levels undergo further splitting. The fourfold degenerate ($S=\frac{3}{2}$) $^4A_2$ ground state divides into two Kramers doublets. At zero magnetic field the doublet

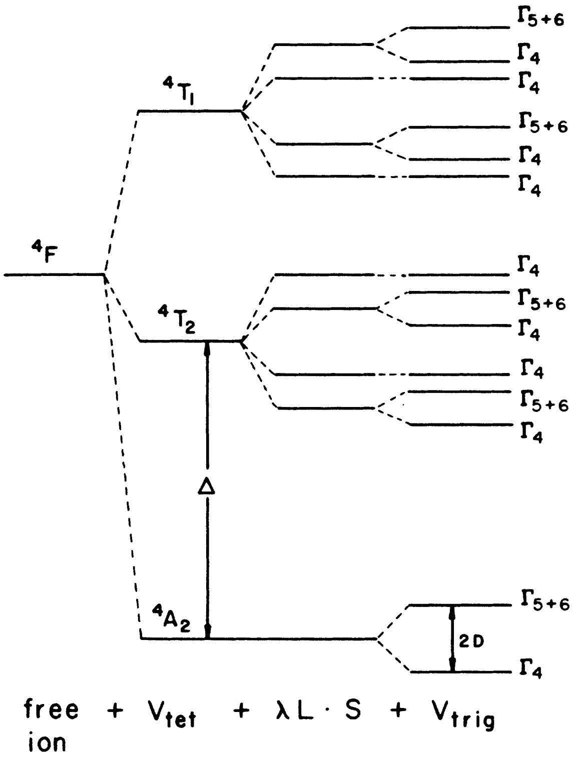

FIG. 1. Schematic diagram of the energy levels of Co^{2+} ion in a tetrahedreal (V_{tet}) and trigonal (V_{trig}) crystal field (after Ref. 10). For the sake of clarity, distances between the levels are not in scale.

states are separated by a value $2D$, as shown in Fig. 1.

These levels can be described by the following Hamiltonian[10] (neglecting a constant term which does not contribute to the magnetic susceptibility):

$$H_S=D(S_z^2-\tfrac{5}{4})+g_\perp\mu_B S_x H_x+g_\parallel\mu_B S_z H_z+G_\perp H_x^2+G_\parallel H_z^2 \;, \tag{1}$$

where

$$D=\frac{\lambda}{2}(g_\parallel-g_\perp),\quad G_\perp=\frac{\mu_B^2}{\lambda}(\tfrac{1}{2}g_\perp-1),\quad G_\parallel=\frac{\mu_B^2}{\lambda}(\tfrac{1}{2}g_\parallel-1)\;, \tag{2}$$

μ_B is the Bohr magneton, λ is the spin-orbit coupling constant, S_x, S_z, H_x, and H_z are the components of spin and of the magnetic field, and $g_\perp$ and $g_\parallel$ are g factors for $\mathbf{H}\perp\hat{\mathbf{c}}$ and $\mathbf{H}\parallel\hat{\mathbf{c}}$, respectively. The two quadratic terms come from the admixture of $^4T_2$ to the $^4A_2$ states. A straightforward calculation gives the energy eigenvalues for $\mathbf{H}\parallel\hat{\mathbf{c}}$:

$$E_{1,2}=-D\pm\tfrac{1}{2}g_\parallel\mu_B H+G_\parallel H^2 \;, \tag{3}$$

$$E_{3,4}=D\pm\tfrac{3}{2}g_\parallel\mu_B H+G_\parallel H^2 \;; \tag{4}$$

and for $\mathbf{H}\perp\hat{\mathbf{c}}$:

$$E_{1,2}=\tfrac{1}{2}g_\perp\mu_B H\pm(g_\perp^2\mu_B^2H^2-g_\perp\mu_B HD+D^2)^{1/2}+G_\perp H^2 \;, \tag{5}$$

$$E_{3,4}=-\tfrac{1}{2}g_\perp\mu_B H\pm(g_\perp^2\mu_B^2H^2+g_\perp\mu_B HD+D^2)^{1/2}+G_\perp H^2 \;. \tag{6}$$

The magnetic susceptibility can be readily calculated by the standard Van Vleck formula,[11] which yields for $\mathbf{H}\parallel\hat{\mathbf{c}}$

$$\chi_{S\parallel}=\frac{g_\parallel^2\mu_B^2}{4k_BT}\,\frac{9+\exp(2D/k_BT)}{1+\exp(2D/k_BT)}-2G_\parallel \;, \tag{7}$$

and for $\mathbf{H}\perp\hat{\mathbf{c}}$

$$\chi_{S\perp}=\frac{g_\perp^2\mu_B^2}{k_BT}\,\frac{1}{1+\exp(-2D/k_BT)}+\frac{3g_\perp^2\mu_B^2}{4D}\tanh(D/k_BT)-2G_\perp \;. \tag{8}$$

In the high-temperature limit, the above formulas reduce to a Curie law for $S=\frac{3}{2}$ minus a temperature-independent term $2G$, as follows:

$$\chi_{S\parallel}=\frac{5g_\parallel^2\mu_B^2}{4k_BT}-2G_\parallel=\frac{g_\parallel^2\mu_B^2S(S+1)}{3k_BT}-2G_\parallel \;, \tag{9}$$

$$\chi_{S\perp}=\frac{5g_\perp^2\mu_B^2}{4k_BT}-2G_\perp=\frac{g_\perp^2\mu_B^2S(S+1)}{3k_BT}-2G_\perp \;. \tag{10}$$

Assuming the free Co^{2+} ion value of the spin-orbit coupling constant[7] ($\lambda=-178\ \text{cm}^{-1}$), and taking g-factor values from electron-spin-resonance (ESR) data[12] ($g_\parallel=2.295$ and $g_\perp=2.303$), we can estimate the temperature-independent term of the magnetic susceptibility. At room temperature this term contributes about 4% of the total susceptibility. It is therefore significant only for $T\geq300$ K, and may be neglected at very low temperatures. This conclusion can be understood in terms of crystal field theory. Large splitting, i.e., large Δ, leads to a weak mixing of the higher orbital states with the ground state, and yields small values of $G_\parallel$ and $G_\perp$.

Many authors (e.g., Refs. 13 and 14) use an empirical spin Hamiltonian in their analysis of magnetic susceptibility data. This Hamiltonian, in the form

$$H=D[S_z^2-\tfrac{1}{3}S(S+1)]+g_\perp\mu_B S_x H_x+g_\parallel\mu_B S_z H_z \;, \tag{11}$$

is equivalent to Eq. (1) except for the two quadratic terms, $G_\perp H_x^2$ and $G_\parallel H_z^2$. Note that in Eq. (11) the mixing of states has been neglected. From the estimates presented above, it is clear that the empirical spin Hamiltonian applies well at low temperatures.

Up to this point we have discussed only the properties of an *isolated* Co^{2+} ion, i.e., an ion without exchange interactions with other magnetic ions. This approximation is correct only at very low concentrations of Co^{2+} ions. Within the nearest-neighbor cluster model,[15] one can easily calculate the probabilities that a magnetic atom is isolated, in a pair, or in a three-atom cluster. In the concentration range that is of particular interest here, ($x=0.002$–0.010) open and closed three-atom cluster probabilities[15] are small enough (<0.01) to be neglected, so that the dominant contributions to χ arise from isolated Co^{2+} ions and from pairs.

The magnetic susceptibility of isolated ions can be calculated using formulas (7) and (8). For pairs we have adopted the method proposed by Owen.[16] The pair Hamiltonian can be expressed as

$$H_P = H_{S1} + H_{S2} - 2J\mathbf{S}_1 \cdot \mathbf{S}_2 \ , \tag{12}$$

where H_{S1} and H_{S2} are single ion Hamiltonians [Eq. (1)], J is the exchange integral, and $\mathbf{S}_1$ and $\mathbf{S}_2$ are spin operators. If $|J|$ is much larger than the zero-field splitting $2D$ of the ground orbital state, then the pair is most conveniently described in terms of the total spin $\mathbf{S}' = \mathbf{S}_1 + \mathbf{S}_2$, where S' can take values $S' = S_1 + S_2, S_1 + S_2 - 1, \ldots, 0$. For $Cd_{1-x}Co_xSe$, $J/k_B = -37 \pm 5$ K (see Sec. IV) and $D/k_B = 1.0 \pm 0.1$ K (Sec. III), so that this condition is fulfilled. Equation (12) can then be explicitly written as[16]

$$\begin{aligned} H_P = {} & W_{S'} + g_\perp \mu_B H_x S'_x + g_\parallel \mu_B H_z S'_z \\ & + \beta_{S'} D[S'^2_z - \tfrac{1}{3}S'(S'+1)] + 2G_\perp H_x^2 + 2G_\parallel H_z^2 \ , \end{aligned} \tag{13}$$

where

$$\begin{aligned} W_{S'} &= -J[S'(S'+1) - 2S(S+1)] \ , \\ \beta_{S'} &= \frac{3S'(S'+1) - 4S(S+1) - 3}{(2S'-1)(2S'+3)} \ , \end{aligned} \tag{14}$$

and

$$S' = 3,2,1,0; \quad S = \tfrac{3}{2} \ . \tag{15}$$

Eigenvalues of the pair Hamiltonian have been found analytically. Applying the Van Vleck formula (cf. Ref. 11), one can now calculate the magnetic susceptibility χ_P of Co^{2+} ion pairs. We will not present this complicated equation in explicit form. The molar magnetic susceptibility is given by

$$\chi = x N_A (P_S \chi_S + \tfrac{1}{2} P_P \chi_P) \ , \tag{16}$$

where x is the concentration of Co^{2+} ions, N_A is Avogadro's number, and $P_S = (1-x)^{12}$ and $P_P = 12 \times (1-x)^{18}$ are respective probabilities for single ions and for ion pairs.

We should comment on two important assumptions made above. First, we completely neglected exchange interaction with the more distant neighbors. Unfortunately, exact values (or even estimates) of J_2, J_3, etc., have not been determined, and are not yet available. Also, we have disregarded triple ion and large clusters. While—as indicated earlier—this is justified in the case of samples used in the present study, we cannot apply the above formulas to DMS's with high concentrations of Co^{2+} ions.

III. EXPERIMENTAL RESULTS AT LOW TEMPERATURES AND DISCUSSION

Single-crystal samples of $Cd_{1-x}Co_xS$ and $Cd_{1-x}Co_xSe$ were prepared by the vertical Bridgman method. The x-ray powder diffraction analysis confirmed the wurtzite structure for all samples studied. The samples were oriented using standard Laue diffraction. The specimens were then attached to the magnetometer sample holder, and the sample orientation was again checked. This procedure provided crystal orientation to within ±3°.

The static magnetic susceptibility measurements were carried out as a function of temperature using a vibrating-sample magnetometer, EG&G model 155. The system was calibrated against a standard Ni sample having a known value of the magnetic moment. The sample was placed in a helium cryostat, and the sample temperature was stabilized between 4.2 and 300 K using a cryogenic temperature controller, Lake Shore DRC-91C. The diamagnetic contributions[17] for CdSe ($\chi_d = -0.334.10^{-6}$ cm^3/g) and CdS ($\chi_d = -0.37.10^{-6}$ cm^3/g) were subtracted from the susceptibility data. The experimental error depended on the size of the detected signal and varied from $\Delta\chi/\chi \cong 0.03$ to $\Delta\chi/\chi \cong 0.05$. For samples with concentrations of Co^{2+} ions $x < 0.01$ we could not make measurements above 40 K, because the signal-to-noise ratio decreases rapidly and becomes less than 20 for $T > 30$ K.

The molar magnetic susceptibility obtained for $Cd_{1-x}Co_xSe$, $x = 0.0065$, is shown in Fig. 2 (in cgs units). The continuous lines represent the best fit of the theoretical formulas [Eqs. (7), (8), and (13)–(16)] for both orientations of the magnetic field $\mathbf{H}$. It can be seen from Sec. II that these expressions contain four parameters: the concentration of Co^{2+} ions x, the zero field splitting D, the exchange integral J, and the g factors $g_\parallel$ or $g_\perp$. It was not necessary to extract all of these parameters from our data, because the exchange integral could be obtained with better accuracy from the high-temperature susceptibility (Sec. IV), and the most precise values of the g factors could be obtained from the ESR results available in the literature. We therefore fixed these parameters at $J/k_B = -37$ K (Sec. IV), $g_\parallel = 2.295$, and $g_\perp = 2.303$ (Ref. 12). The remaining parameters affecting the magnetic susceptibility at low temperatures are the concentration x of the Co^{2+} ions, and the difference between $\chi_\perp$ and $\chi_\parallel$, which is sensitive to the zero-field splitting D. These two quantities were used as the fitting parameters in the present analysis.

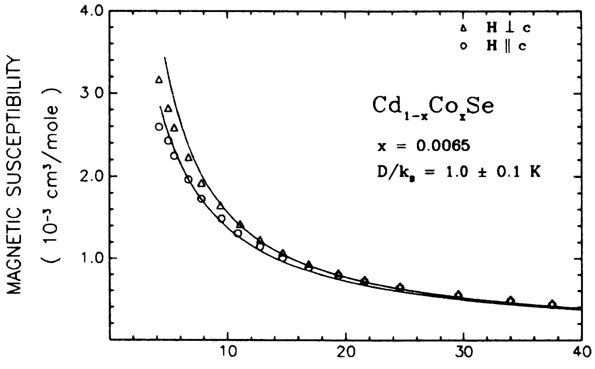

FIG. 2. Molar magnetic susceptibility of $Cd_{1-x}Co_xSe$, $x = 0.0065$, as a function of temperature for two orientations of the magnetic field, $\mathbf{H} \parallel \hat{\mathbf{c}}$ and $\mathbf{H} \perp \hat{\mathbf{c}}$.

With this procedure, our data for $Cd_{1-x}Co_xSe$, $x=0.0065$, yielded $D/k_B=1.0\pm0.1$ K. This value is quite close to the result $D/k_B=1.1$ K obtained from ESR measurements (cf. Ref. 12). Using the expression

$$\lambda=\frac{2D}{g_{\parallel}-g_{\perp}}\ , \tag{17}$$

one can now estimate the spin-orbit coupling constant λ. The result, $\lambda=-174\ \mathrm{cm}^{-1}$, is very close to the free-ion value of $\lambda=-178\ \mathrm{cm}^{-1}$ (cf. Ref. 7).

The data for $Cd_{1-x}Co_xS$, $x=0.002$, shown in Fig. 3, are analyzed in the same way as above. We set $J/k_B=-37$ K, $g_{\parallel}=2.269$, and $g_{\perp}=2.286$ (Ref. 18). One comment should be made about the exchange integral J. At present, we do not know the value of J for $Cd_{1-x}Co_xS$, because samples with large concentrations of Co^{2+} ions, where J could be reliably determined from the high-temperature susceptibility, are not yet available (see the discussion at the beginning of this section). In the case of $Cd_{1-x}Mn_xSe$ and $Cd_{1-x}Mn_xS$, the exchange integrals are almost equal to each other: $J/k_B=-10.6$ K (Ref. 19) and $J/k_B=-10.5$ K (Ref. 20), respectively. With this in mind, we decided to use the same value of J for $Cd_{1-x}Co_xS$ as for $Cd_{1-x}Co_xSe$, at least as a close estimate. One should also note that only the pair contribution χ_P [Eq. (16)], which is less than 10% of the total susceptibility, depends on J, so that a small error in the value of J is not expected to affect the value of D deduced from the data. With these assumptions, the best fit of the theoretical curves yielded $D/k_B=0.85\pm0.1$ K for $Cd_{1-x}Co_xS$, which is close to the value $D/k_B=0.95$ K extracted from ESR measurements.[18]

In Fig. 4 we present the angular dependence of the magnetic susceptibility of $Cd_{1-x}Co_xS$, $x=0.002$, at a fixed temperature $T=4.2$ K. As mentioned earlier (Sec. II), the admixture of the trigonal potential to the tetrahedral crystal field results in an axial symmetry of the physical properties of the crystals. It is well known that in this case (and with $\chi_{\perp}>\chi_{\parallel}$) the anisotropy of χ obeys the following angular dependence:

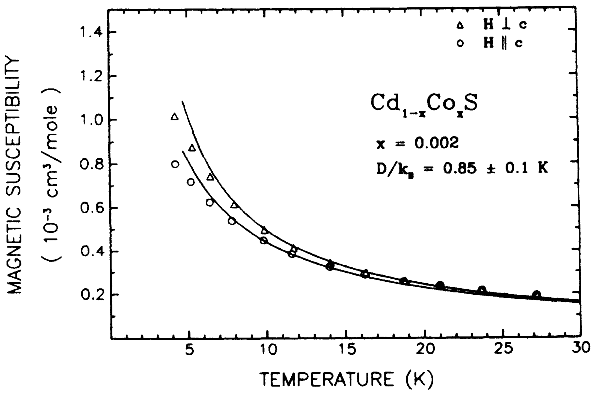

FIG. 3. Molar magnetic susceptibility of $Cd_{1-x}Co_xS$, $x=0.002$, for magnetic field parallel or perpendicular to the c axis.

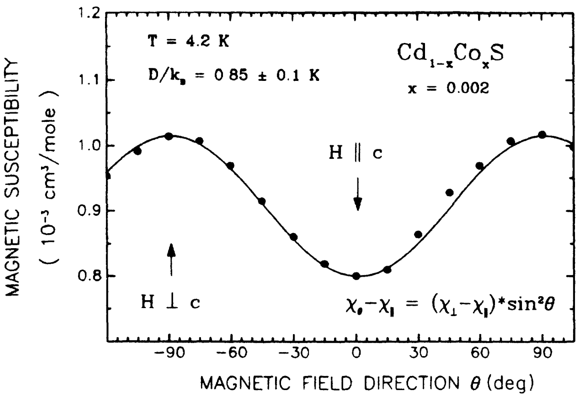

FIG. 4. Angular dependence of the magnetic susceptibility of $Cd_{1-x}Co_xS$, $x=0.002$, at a fixed temperature $T=4.2$ K.

$$\chi_\theta-\chi_\parallel=(\chi_\perp-\chi_\parallel)\sin^2\theta\ , \tag{18}$$

where θ is the angle between the direction of the magnetic field $\mathbf{H}$ and the c axis. One sees that the data points fit the theoretical curve quite well. Similar agreement was observed for all other samples studied.

IV. HIGH-TEMPERATURE MAGNETIC SUSCEPTIBILITY OF $Cd_{1-x}Co_xSe$

In this section we present the results for $Cd_{1-x}Co_xSe$ with concentrations of Co^{2+} ions $x=0.062$ and $x=0.085$. In order to determine the exchange integral J we performed measurements in the temperature range up to 300 K.

The model presented in Sec. II neglects contributions to the magnetic susceptibility resulting from all clusters larger than nearest-neighbor (NN) pairs. That model is therefore not adequate for samples with $x>0.01$. At high temperatures an alternative approach to χ has been used. For $k_BT>J$, the exchange interactions can be readily incorporated into the susceptibility by using a high-temperature expansion for a randomly dilute Heisenberg antiferromagnet[19] or the mean-field approximation. For this case, the magnetic susceptibility is expressed in the form of the Curie-Weiss law, plus a small temperature-independent term χ_∞ (Ref. 21),

$$\chi(T)=\frac{C(x)}{T-\Theta(x)}+\chi_\infty\ , \tag{19}$$

with the Curie constant per mole given by

$$C(x)=xN_A(g\mu_B)^2S(S+1)/(3k_B)\ , \tag{20}$$

and the Curie-Weiss temperature

$$\Theta(x)=2xS(S+1)zJ/(3k_B)\ , \tag{21}$$

where z is the number of cations in the first coordination sphere (12 for the wurtzite structure), and all other symbols are as previously defined.

In the case of $Cd_{1-x}Co_xSe$, the difference between $g_\perp$ and $g_\parallel$ (cf. Ref. 12) is very small $(g_\perp-g_\parallel)/g_\perp\approx3.5\times10^{-3}$. Thus in the high-temperature regime

$\chi_\perp \approx \chi_\parallel$ (within experimental accuracy), and we no longer need specify the orientation of the magnetic field.

As we mentioned earlier (Sec. II), the temperature-independent term χ_∞ results from the admixture of higher orbital states to the ground state. Although the formulas derived in Sec. II are not quantitatively applicable in the range $x > 0.01$, they can still be used for estimating χ_∞:

$$\chi_\infty \approx -2G = xN_A \frac{\mu_B^2}{2D}(g_\parallel - g_\perp)(g-2)\,, \qquad (22)$$

where $g = (g_\perp + g_\parallel)/2$. This value of χ_∞, and the diamagnetic contributions for Cd^{2+} and Se^{2-} ions,[17] were subtracted from the measured susceptibility values, which were then analyzed in terms of the Curie-Weiss formulation.

The inverse molar susceptibility of $Cd_{1-x}Co_xSe$ (*after* subtraction of the aforementioned constant terms) is shown in Fig. 5. The straight lines represent the least-squares fit of the Curie-Weiss law to the experimental data in the high-temperature regime. Using Eq. (21), we then determined the exchange integral J. For $x = 0.062$ we have obtained $\Theta(x) = -68 \pm 10$ K and $J/k_B = -37 \pm 5$ K; and for $x = 0.085$, $\Theta(x) = -96 \pm 10$ K and $J/k_B = -38 \pm 5$ K. One should note that the value of J is much larger than that for $Cd_{1-x}Mn_xSe$ ($J/k_B = -10.6$ K, cf. Ref. 19). Likewise, the values of J for Co-based DMS's with zinc blende structure were also found to be much larger than those of their Mn-based counterparts (cf. Ref. 6).

It should be noted, finally, that the value of J calculated from the Curie-Weiss law is not entirely a result of nearest-neighbor exchange, but also contains contributions from interactions between second-, third-, and higher-order neighbors. These contributions are usually small, but not negligible,[22] and may amount to perhaps 10% of the total value of J.

Unfortunately we were unable to determine the value of J for $Cd_{1-x}Co_xS$, because the value of x in the samples available to us was too small to carry out reliable χ measurements above $T \approx 40$ K, where the Curie-Weiss procedure could be reliably applied.

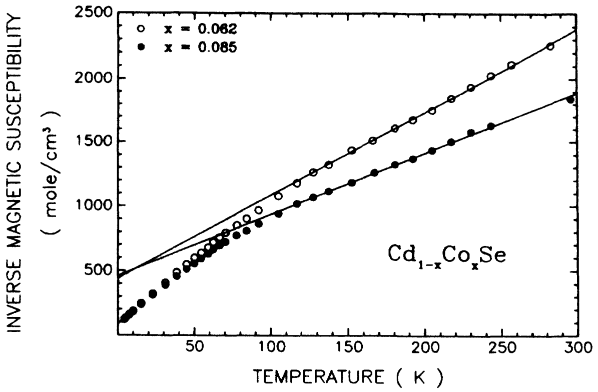

FIG. 5. Inverse molar susceptibility of $Cd_{1-x}Co_xSe$ as a function of temperature for $x = 0.062$ and $x = 0.085$.

V. CONCLUSION

In conclusion, we have observed the anisotropy of the magnetic susceptibility of $Cd_{1-x}Co_xSe$ and $Cd_{1-x}Co_xS$ at low temperatures. A simple theoretical model, including single ion and pair contributions to χ, has been successfully used for the interpretation of the experimental data and for the calculation of the zero-field splitting D of the orbital ground state. We have also pointed out that the differences between the results of the point charge crystal field model and the empirical spin Hamiltonian are insignificant at low temperatures. The exchange integral J between Co^{2+} ions has been determined from the fit of the Curie-Weiss law to the high-temperature data. Its value is approximately three times larger than that for corresponding Mn-based DMS's. The mechanism responsible for such a high value of J in Co-based DMS alloys has not yet been explained.

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