PHYSICAL REVIEW B VOLUME 44, NUMBER 12 15 SEPTEMBER 1991-II

# Spin freezing in cobalt-based diluted magnetic semiconductors

P. M. Shand, A. Lewicki,* I. Miotkowski,† and B. C. Crooker
*Department of Physics, Purdue University, West Lafayette, Indiana 47907*

J. K. Furdyna
*Department of Physics, University of Notre Dame, Notre Dame, Indiana 46556*

(Received 6 December 1990; revised manuscript received 8 April 1991)

We have measured the magnetic susceptibility of the cobalt-based diluted magnetic semiconductors $\mathrm{Zn}_{1-x}\mathrm{Co}_x\mathrm{S}$, $\mathrm{Cd}_{1-x}\mathrm{Co}_x\mathrm{S}$, and $\mathrm{Cd}_{1-x}\mathrm{Co}_x\mathrm{Se}$ in the temperature range 33 mK to 4.2 K, and for concentrations $0.03 \leq x \leq 0.103$. All samples exhibit a low-temperature transition from paramagnetism, which is ascribed to a spin-freezing process. The spin-freezing temperatures are well described by a power-law function of the concentration. This implies that the spatial dependence of the exchange interaction is of the form $J(R) \sim (R/R_1)^{-n}$, for a nearest-neighbor distance $R_1$. The value $n = 6.3 \pm 1.2$ is obtained, which is close to the result $n = 6.8$ obtained in a previous analysis of manganese-based materials. We compare our results to the manganese-based data and discuss the properties of the long-range interaction which leads to spin freezing in both cases.

## I. INTRODUCTION

Over the past decade, much research has been conducted in the field of diluted magnetic semiconductors (DMS's). DMS's are semiconducting alloys in which a part of the cation sublattice of a "parent" semiconductor has been replaced by randomly substituted magnetic ions, which are usually transition-metal ions that have partially filled *d* orbitals. The manganese-based alloys of the form $A^{\mathrm{II}}_{1-x}\mathrm{Mn}_x C^{\mathrm{VI}}$, with $A^{\mathrm{II}} \equiv \mathrm{Cd}$, Zn, and Hg and $C^{\mathrm{VI}} \equiv \mathrm{S}$, Se, and Te, have been the most thoroughly investigated materials thus far and several good reviews can be found in the literature (see Refs. 1–3, for instance).

These materials are interesting, in part, due to their magnetic properties. Low-field ac magnetic-susceptibility measurements performed on Mn-based DMS's have established the existence of two magnetic phases: a high-temperature paramagnetic phase and a low-temperature phase resembling a spin glass. The latter phase is identified by a cusp or kink in the ac susceptibility. Other measurements, including heat capacity,[4] remanent magnetization,[5] field cooled, and zero-field cooled susceptibilities,[6] also yield results similar to those seen in spin glasses.

The exact nature of the transition in these materials remains, however, a topic of debate. Based on neutron-scattering data,[7] Furdyna and Samarth[8] have proposed a qualitative cluster-locking model in order to explain the low-temperature behavior of the magnetic susceptibility of DMS's. The neutron-scattering experiments show the existence of short-range type-III antiferromagnetic order in Mn-based DMS's. The correlation length $\xi$ of these ordered regions increases as the temperature is decreased, and saturates in the vicinity of the spin-freezing transition. Furdyna and Samarth interpret these ordered regions as clusters that grow with decreasing temperature, eventually becoming large enough to overlap, giving rise to the cusp in the susceptibility and the slow dynamics associated with a frozen state. Cluster-blocking models similar to the above usually display activated dynamics.[9–11] However, a number of recent frequency-dependent susceptibility measurements have found power-law scaling in the relaxation time, which is typically associated with a canonical spin-glass transition.[12–14]

It was originally thought that nearest-neighbor (NN) antiferromagnetic exchange interactions[4,15] produced the spin-freezing transition.[16] However, the discovery that a spin-freezing transition exists for concentrations $x$ below the NN percolation threshold for these lattices[17,18] has turned attention to mechanisms by which long-range exchange interactions among widely separated spins at low concentrations could lead to a spin-frozen state. The exact nature of these long-range interactions remains an open question.

Twardowski *et al.*[19] have made a single log-log plot of spin-freezing temperature $T_f$ versus $x$ for several families of Mn-based wide-band-gap II-VI DMS materials. A striking universality emerged, with virtually all the data points falling on a single straight line. This suggests that the *same* long-range exchange interaction is responsible for the spin-freezing transition in all the Mn systems that were investigated. However, the key point here is that the data constitute a universal function of $x$. This means that the spin-freezing transition is occurring at the same value of $R_m/R_1$, where $R_m$ is the average distance between magnetic ions and $R_1$ is the NN distance in each family, and that it does not scale with the strength of the NN exchange.

A class of DMS materials has recently been prepared, with cobalt being the magnetic substitute. The isolated (i.e., noninteracting) $\mathrm{Co}^{2+}$ ion, under the influence of a tetrahedral crystal field (as exists in a II-VI environment), has a $^4A_2$ orbital singlet ground state, with the first excited state being a $^4T_2$ orbital triplet.[20] Though the $^4T_2$

state is further split by the spin-orbit interaction, the energy separation between the first excited state and the ground state far exceeds room-temperature thermal energy. Therefore, the behavior of the susceptibility should be determined mainly by ground-state properties, and apart from differences in $g$ factor ($g \approx 2.3$ for $Co^{2+}$) and spin ($S=\frac{3}{2}$ for $Co^{2+}$) should be similar to that of Mn ($g \approx 2$ and $S=\frac{5}{2}$).

Comparisons between the cobalt- and manganese-based DMS's provide us with a unique opportunity to gain insight into the long-range interactions leading to spin freezing in these materials. Since the host lattices are identical, and differences can be attributed to the different magnetic ions. Cobalt is a particularly attractive substitutional ion because of its relatively simple ground state. In addition, recent magnetic-susceptibility and neutron-scattering measurements have established that the nearest-neighbor exchange constants for the Co-based alloys studied so far are three to four times larger than those of the corresponding Mn-based materials.[21,22] In this paper, we present the results of low-temperature (33 mK $\leq T \leq 4.2$ K) ac susceptibility measurements on the Co-based materials $Zn_{1-x}Co_xS$, $Cd_{1-x}Co_xS$, and $Cd_{1-x}Co_xSe$. Concentrations of $Co^{2+}$ ions in the samples studied were in the range $0.03 \leq x \leq 0.103$. Preliminary results on the $Zn_{1-x}Co_xS$ system have been presented previously.[23]

## II. EXPERIMENTAL DETAILS

Two methods were used in the preparation of our samples. The $Zn_{1-x}Co_xS$ samples were prepared using the chemical vapor transport technique, and the $Cd_{1-x}Co_xS$ and $Cd_{1-x}Co_xSe$ samples were fabricated by means of the vertical Bridgman method.

Electron microprobe analysis was used to determine the concentration of $Co^{2+}$ in our samples. Further analysis using atomic absorption (AA) spectroscopy was performed on eight of our samples, and two of our samples were studied using energy-dispersive analysis of x rays (EDAX). The AA method provides a measure of the total concentration in the sample, whereas the other two methods measure the concentration within approximately 1 $\mu$m of the surface. In general, we found that the concentration over the analyzed surface of each sample varied by 10% or less, but a few samples had variations in concentration of up to 15%, indicating substantial inhomogeneity. We were concerned that the sample inhomogeneity might be much larger, since the microprobe analyzes only the surface. However, when both microprobe and AA analyses were done on the same sample, the average microprobe value agreed with the AA value to within 10%. This suggests that the microprobe provides a reasonable measure of the inhomogeneity. The reported concentrations are averages of the values determined with the various methods.

Magnetic-susceptibility measurements were performed with the aid of a standard mutual inductance bridge operating at 26 Hz. A superconducting quantum interference device was used as a null detector. In order to exclude external fields, a superconducting lead shield was used to surround the entire coil assembly. In addition, another superconducting shield and a Mumetal shield (Mumetal is a standard high-permeability material used to screen out magnetic fields) were placed around the evacuated chamber in which the coil assembly was housed. This enabled us to use very small measuring fields, the oscillating field at the sample having an amplitude of approximately 0.002 Oe. The error $\Delta\chi/\chi$ in our susceptibility measurements depended on the sample mass, concentration, DMS family, and the error in our determination of the background susceptibility. The mass of a typical sample was approximately 100 mg. At 4.2 K, where the signal was smallest, the experimental error had a maximum value $\Delta\chi/\chi \approx 0.07$. At lower temperatures the signal increased rapidly, especially for low $x$. At the transition, $\Delta\chi/\chi$ was always $<0.01$.

Our samples were cooled by thermal contact with a cold finger attached to the mixing chamber of a dilution refrigerator operating in the temperature range 33 mK$\leq T \leq 4.2$ K. Using GE 7031 varnish, the sample was glued to one end of a sheet of fine copper wires. The other end of the wire foil was in good thermal contact with the mixing chamber. Use of the wire foil minimized eddy current effects.

Temperature was measured with a Speer 100 carbon resistor. This resistor was calibrated against an Oxford Instruments thermometer in the previously mentioned temperature range. The quoted accuracy of the Oxford thermometer was ±5%. We estimate the maximum error in our temperature to be ±10%. In order to measure the susceptibility at a particular temperature, the temperature was held constant until the susceptibility signal was stable for at least 15 min. Especially at the lowest temperatures, this waiting time was necessary to ensure that the sample was in thermal equilibrium with the refrigerator.

## III. RESULTS

Shown in Fig. 1 are susceptibility versus temperature curves for several samples. We interpret the rounded cusp in the $\chi$ versus $T$ plot as a spin-freezing transition. While a sharper cusp might be expected at the spin-freezing transition, we note that sample inhomogeneity can lead to rounded curves such as we find. In fact, curves similar to those presented here are common in previous measurements of Mn-based materials, especially at low values of $x$.[17,18,24,25] In order to determine the spin-freezing temperature $T_f$ in a consistent manner, straight lines were drawn through the data points on either side of the transition and the point of intersection of these lines was taken to be the transition temperature. The values of $T_f$ for all of our samples are compiled in Table I. We will interpret our data in terms of a spin-freezing transition based on a comparison with the work on Mn-based DMS's. We emphasize here that further experiments (e.g., zero-field cooled and field cooled susceptibility and frequency-dependent susceptibility) are necessary in order to characterize the nature of the transition.

In Fig. 2 we present a log-log plot of $T_f$ versus $x$ for all our samples. The uncertainty in $T_f$ is ±10% as men-

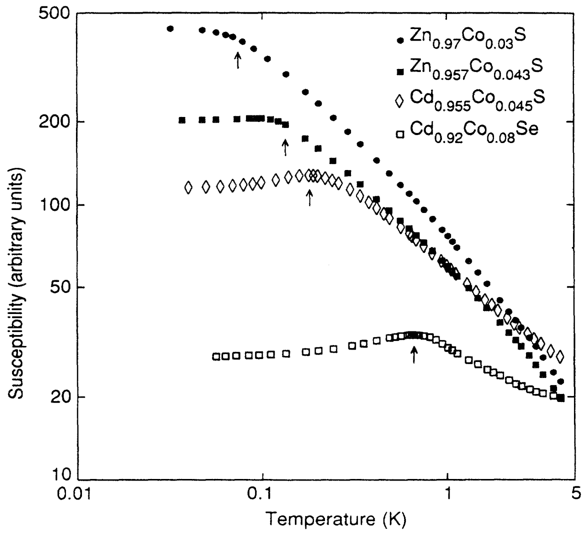

FIG. 1. Susceptibility vs temperature plots for several DMS materials for different values of $x$.

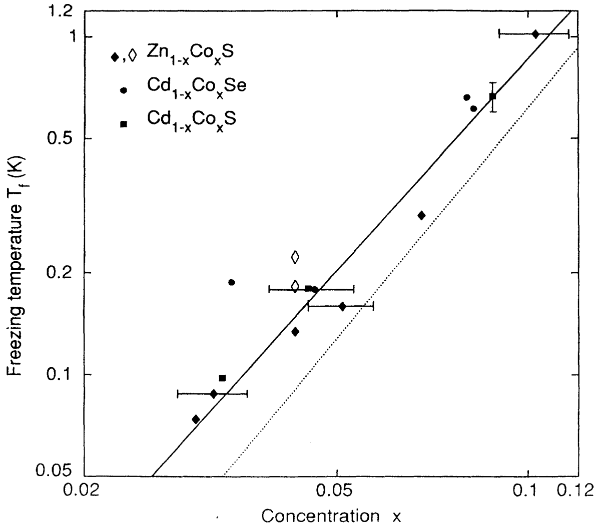

FIG. 2. Plot of spin-freezing temperature $T_f$ vs concentration $x$ for all samples. The solid line is the best fit to the data. The dashed line represents the fit to the Mn-based data (from Twardowski *et al.* in Ref. 19). The concentration error bars shown represent uncertainties in $x$ in the range $10\% \leq \Delta x / x \leq 15\%$. The relative error in every other $x$ value is less than 10%. The temperature error bar shown corresponds to $\Delta T/T = 10\%$, which is the uncertainty in all our temperature measurements, as mentioned in the text. The two open diamonds represent measurements done on the same sample at 1 kHz (upper point) and 26 Hz (lower point). All other points were taken at 26 Hz.

tioned in the preceding section. The uncertainty in $x$ is also $\pm 10\%$ or less, unless otherwise indicated by error bars. The straight line shown in this figure is a good fit to a large majority of the data points. The single point that is not well fit corresponds to a $Cd_{1-x}Co_xSe$ sample that showed anomalous behavior in optical measurements of the susceptibility anisotropy as well.[26] We have also included in Fig. 2 measurements performed at two different frequencies (26 Hz and 1 kHz) on a single sample. These points are represented by the open diamonds. We will comment on the significance of these measurements in Sec. IV.

The solid line shown in Fig. 2 is a linear least-squares fit to a log-log plot of the data. A slope of $2.1 \pm 0.4$ and a $y$ intercept of 4.64 were obtained.[27] Also shown as a dotted line in the figure are the results for the Mn-based DMS's presented by Twardowski *et al.*[19] We will compare the two sets of data in the next section.

TABLE I. Freezing temperatures $T_f$ and concentrations $x$ for the materials investigated in this paper.

| Material | $x$ | $T_f$ (K) |
|---|---|---|
| $Cd_{1-x}Co_xSe$ | 0.034 | 0.187 |
| | 0.046 | 0.178 |
| | 0.080 | 0.658 |
| | 0.082 | 0.609 |
| $Cd_{1-x}Co_xS$ | 0.033 | 0.098 |
| | 0.045 | 0.180 |
| | 0.088 | 0.667 |
| $Zn_{1-x}Co_xS$ | 0.030 | 0.074 |
| | 0.032 | 0.088 |
| | 0.043 | 0.134 |
| | 0.051 | 0.159 |
| | 0.068 | 0.296 |
| | 0.103 | 1.021 |

## IV. DISCUSSION

The spin-freezing transition temperatures for all of the Mn-based DMS's and for the Co-based DMS's studied here are well described by the relation

$$T_f = \kappa x^{n/3} , \tag{1}$$

with $n = 6.3 \pm 1.2$ for the cobalt data, $\kappa$ being a positive constant. It has been argued previously for Mn-based DMS's (Refs. 19 and 28) and metallic spin glasses[9,29] that the relationship between $T_f$ and $x$ is a direct consequence of the spatial dependence of the exchange interaction between magnetic ions. If we assume a collective phase transition to a spin glass,[28] then for spin freezing to take place, the thermal energy of neighboring moments $S_i$ and $S_j$ separated by $R$ should be of the order of the exchange energy $|2J(R)\mathbf{S}_i \cdot \mathbf{S}_j|$. At the freezing temperature, we can express this condition as

$$J(R_m)S^2=\alpha k_B T_f \ , \tag{2}$$

where $R_m$ denotes the mean distance between magnetic ions, $S$ is the spin quantum number of the magnetic ion, and $\alpha$ is a constant that depends on the mechanism of the spin freezing. The volume per magnetic ion is given by

$$\tfrac{4}{3}\pi\left[\frac{R_m}{2}\right]^3=\frac{a^3}{4x} \ , \tag{3}$$

since the volume per ion in a fcc (or hcp) unit cell of length $a$ is $\frac{1}{4}a^3$. From (1), (2), and (3) one readily obtains

$$J(R)=J_*\left[\frac{R}{R_1}\right]^{-n} \ , \tag{4}$$

where $J_*$ is a constant giving the strength of the interaction. We note that it is also possible to obtain Eq. (4) if one assumes a percolating cluster model as was shown by Smith for metallic spin glasses.[9] We expect that the cluster-locking model of Furdyna and Samarth would also lead to Eq. (4), since it is very similar to the percolation problem.

Both the data presented here and previous data on the Mn-based DMS's are indicative of a spin-spin interaction of the form shown in Eq. (4). The exponent of 6.3 obtained in this work is close to the value of 6.8 obtained in Mn-based materials[19] and also close to the value of 6.0 obtained from heat-capacity measurements on a $Cd_{1-x}Co_xS$ sample.[30] As pointed out in Ref. 31, direct measurements of the second- and third-nearest-neighbor exchange constants in high-field magnetization experiments on $Cd_{1-x}Mn_xTe$ also exhibit a $(R/R_1)^{-6.8}$ dependence.[32] We point out that the value of the exponent $n$ obtained in the heat-capacity and the high-field magnetization experiments does not depend on the existence of a spin-freezing transition. The fact that the value of $n$ obtained from the spin-freezing transiton agrees with these other determinations supports our use of Eq. (4).

These results suggest that the same long-range interaction leads to spin freezing below the NN percolation threshold in both the Mn-based and Co-based systems. The fact that the exponents are similar in these two systems is not surprising. For the low-concentration samples studied, the magnetic ions are far apart, and we expect that the value of the exponent will depend primarily on the intervening host lattice, which is identical. We expect that the different magnetic ions would only affect the overall strength of the interaction (i.e., $J_*$), not its variation with separation.

In fact, there is evidence in our data for such an effect on the strength of the interaction in going from the Co-based to the Mn-based materials. As shown in Fig. 2, $T_f$ at fixed $x$ for the Co-based alloys is 1.5–2 times greater than the corresponding value for the Mn-containing materials. In fact, this difference is probably somewhat larger, since the Mn data were typically taken at higher frequencies, which for spin glasses is known to shift $T_f$ to higher values. As can be seen in Fig. 2, the measurement done at 1 kHz lies above the measurement done at 26 Hz on the same sample. 1 kHz is well within the range of frequencies used in the Mn data.[19]

This general increase in $T_f$ going from $Mn^{2+}$ to $Co^{2+}$ is most likely due to the large increase (3–4 times) in the value of $J_1$ (and hence presumably $J_*$) for Co-based alloys compared to their Mn counterparts.[21,22] This can be seen if one examines Eqs. (2) and (4). If $J_*$ is proportional to $J_1$, then the strength of the exchange interaction increases by a factor of approximately 3.5 in going from Mn-based systems to Co-based systems. On the other hand, $S^2$ decreases by a factor of $\frac{25}{9}$. The net result is that the exchange energy of the Co-based systems is roughly 1.3 times greater than that of their Mn counterparts and hence the spin-freezing temperatures should be systematically higher, as we have found.

It is generally believed that indirect exchange between magnetic ions through the intervening lattice leads to the spin-freezing transition. Larson *et al.*[16,33] have performed extensive theoretical calculations on exchange interactions in DMS materials, which establish superexchange as the dominant interaction up to third- or fourth- (fcc) nearest-neighbor distances. Recent experimental results also support the superexchange theory.[34,35] The theory predicts that the $d$-$d$ exchange should have a functional form $f(R/a)$ for the dependence on distance, which is consistent with our results. However, Larson *et al.* obtained

$$f(r)\sim\lambda e^{-\beta r^2} \ , \tag{5}$$

where $r\equiv R/a$, and $\lambda$ and $\beta$ are constants. The functional form given by Eq. (5) does not describe our data well. There is a large deviation at lower concentrations and our best fit gives $\beta=0.6$ compared to $\beta=4.89$ obtained by Larson *et al.* for $Cd_{1-x}Mn_xTe$.

The lack of a good fit to the exponential is perhaps not surprising in view of the fact that the concentrations studied in this work have average distances between magnetic ions that range from fifth to tenth nearest neighbors. It is expected that a crossover will occur from superexchange at higher concentrations to Bloembergen-Rowland exchange[36] at lower concentrations.[33] The sum of two exponentials with different decay constants would provide a better fit to our data.

Recent theoretical work by Yu and Lee[37] calculated the total exchange due to both mechanisms in $Cd_{1-x}Mn_xTe$. They find that the indirect exchange is highly anisotropic. Their calculated values along the most favorable direction fall off roughly as $(R/R_1)^{-7}$ from $R/R_1=\sqrt{2}$ out to at least $R/R_1=\sqrt{8}$, which covers the range of our experimental data.[38] The similarity between their calculated dependence on $R$ and that observed experimentally is encouraging, although it is not clear how the anisotropy they observed would appear in the experimental results.

A key discrepancy between theory and experiment remains concerning the strength of the long-range interaction. It is expected theoretically[33] that this strength should scale with the value of $J_1$. Experimentally, data on all of the wide-band-gap II-VI Mn-based materials indicate that this strength is a constant in spite of large differences in NN exchange constants $(-6.6K \le J_1/k_B \le -13\ \mathrm{K})$.[2] We also see no evidence in our data on the Co-based materials for a dependence on

$J_1$. The data in Fig. 2 show that none of the alloys measured have consistently higher or lower freezing temperatures than those given by the solid line, although the variation in NN exchange constants is smaller [$J_1/k_B = -47.5$ K for $Zn_{1-x}Co_xS$ (Refs. 21 and 22) and $-37.5$ K for $Cd_{1-x}Co_xSe$ (Ref. 39)] and hence it would be harder to detect such trends.

It is possible that lattice distortions introduced when the magnetic ions are nearest neighbors account for much of the variation in $J_1$ among the Mn-based alloys, and also among the Co-based alloys.[34] This would make $J_1$ a poor predictor of the strength of further-neighbor exchange. In this regard, measurements of $J_2$ and $J_3$ for other alloys might provide more insight. As discussed above, however, our data do suggest that the change in $J_1$ when cobalt is substituted for manganese does affect the strength of the long-range interactions. This suggests that nearest-neighbor lattice distortion does not cause the factor-of-3 difference in $J_1$ between the Co-based materials and the Mn-based materials.

Finally, an interesting feature of the cobalt systems is that a distortion along the $c$ axis in the wurtzite (hcp) lattice structure splits the $^4A_2$ ground state of the $Co^{2+}$ ion into two Kramers doublets separated by an energy $2D$ in zero magnetic field.[20] The lower doublet has $S_z = \pm\frac{1}{2}$ and the upper one has $S_z = \pm\frac{3}{2}$. For $Cd_{1-x}Co_xS$, $2D/k_B = 1.7$ K and for $Cd_{1-x}Co_xSe$, $2D/k_B = 2.0$ K.[39] The splitting in the Mn-based wurtzite materials is much smaller and zinc-blende (fcc) materials (e.g., $Zn_{1-x}Co_xS$) undergo no such splitting. Despite the difference in energy level structure caused by the splitting, there is no apparent difference in the variation of $T_f$ with respect to $x$ in our data (see Fig. 2). We do not at present understand this behavior, and further experiments are planned in order to shed some light on this problem.

## V. CONCLUSIONS

Low-temperature ac magnetic-susceptibility measurements have been performed on the $Zn_{1-x}Co_xS$, $Cd_{1-x}Co_xS$, and $Cd_{1-x}Co_xSe$ DMS systems. All samples showed spin-freezing transitions for $Co^{2+}$ concentrations well below the percolation threshold. A log-log plot of $T_f$ versus $x$ revealed that the data from all three families could be described well by a single straight line. The empirical power-law exponent $n$ that characterizes the spatial dependence of the exchange interaction $J(R)$ was found to have a value of $6.3\pm1.2$, which is close to the value $n = 6.8$ found for the equivalent II-VI Mn-based systems. Therefore, the long-range interactions that lead to the spin-frozen state in both $Mn^{2+}$ and $Co^{2+}$ DMS systems are not significantly different in their spatial properties. This in turn suggests that the fundamental electronic processes that give rise to these interactions are very similar. Further, the explicit dependence of the exchange interaction on $R/R_1$ suggests that the exchange process involves the underlying lattice and is therefore reminiscent of superexchange.

It was also found that the magnitude of $T_f$ at fixed $x$ was 1.5–2 times greater in the Co-based materials than in the Mn-based materials. The large increase (factors of 3–4) in $J_1$ that occurs when the $Mn^{2+}$ ion is replaced by the $Co^{2+}$ ion may be partially responsible for this elevation in $T_f$. The data also gave no indication that at low temperatures ($T \ll 2D/k_B$), the spins in the wurtzite materials were in an effective $S = \frac{1}{2}$ state, in contrast to zinc-blende spins, which are always in the $S = \frac{3}{2}$ state. This result is not understood at present.

## ACKNOWLEDGMENTS

This work was supported by the National Science Foundation (NSF) under Materials Research Group Grant No. DMR-89-13706 and NSF Grant No. DMR-86-03889.

---

[*] Permanent address: Department of Solid State Physics, Akademia Gorniczo-Hutnicza (AGH), PL-30-059 Krakow, Poland.

[†] Permanent address: Institute of Physics, Polish Academy of Sciences, PL-02-668 Warszawa, Poland.

[1] N. B. Brandt and V. V. Moshchalkov, Adv. Phys. **33**, 193 (1984).

[2] J. K. Furdyna, J. Appl. Phys. **64**, R29 (1988).

[3] H. Keesom, in *Semiconductors and Semimetals*, edited by J. K. Furdyna and J. Kossut (Academic, Boston, 1988), Vol. 25. This volume contains reviews of several different subfields of the subject of DMS's. For a discussion of magnetic properties, see S. Oseroff and P. H. Keesom, p. 73.

[4] R. R. Galazka, S. Nagata, and P. H. Keesom, Phys. Rev. B **22**, 3344 (1980).

[5] S. P. McAlister, J. K. Furdyna, and W. Giriat, Phys. Rev. B **29**, 1310 (1984).

[6] S. B. Oseroff, Phys. Rev. B **25**, 6584 (1982).

[7] T. M. Giebultowicz and T. M. Holden, in *Semiconductors and Semimetals* (Ref. 3), p. 125.

[8] J. K. Furdyna and N. Samarth, J. Appl. Phys. **61**, 3526 (1987).

[9] D. A. Smith, J. Phys. F **4**, L266 (1974).

[10] S. Shtrikman and E. P. Wohlfarth, Phys. Lett. **85A**, 467 (1981).

[11] S. Geshwind, A. T. Ogielski, G. Devlin, J. Hegarty, and P. Bridenbaugh, J. Appl. Phys. **63**, 3291 (1988).

[12] A. Mauger, J. Ferré, M. Ayadi, and P. Nordblad, Phys. Rev. B **37**, 9022 (1988).

[13] Y. Zhou, C. Rigaux, A. Mycielski, M. Menant, and N. Bontemps, Phys. Rev. B **40**, 8111 (1989).

[14] S. Geshwind, David A. Huse, and G. E. Devlin, Phys. Rev. B **41**, 4854 (1990).

[15] S. Nagata, R. R. Galazka, D. P. Mullin, H. Akbarzadeh, G. D. Khattak, J. K. Furdyna, and P. H. Keesom, Phys. Rev. B **22**, 3331 (1980).

[16] This antiferromagnetic exchange interaction has been attributed to superexchange. See B. E. Larson, K. C. Hass, H. Ehrenreich, and A. E. Carlsson, Solid State Commun. **56**,

347 (1985).

[17]M. A. Novak, S. Oseroff, and O. G. Symko, Physica **107B**, 313 (1981).

[18]M. A. Novak, O. G. Symko, D. J. Zheng, and S. Oseroff, Physica **126B**, 469 (1984).

[19]A. Twardowski, H. J. M. Swagten, W. J. M. de Jonge, and M. Demianiuk, Phys. Rev. B **36**, 7013 (1987).

[20]M. Villeret, S. Rodriguez, and E. Kartheuser, Physica **162B**, 89 (1990).

[21]A. Lewicki, A. I. Schindler, J. K. Furdyna, and W. Giriat, Phys. Rev. B **40**, 2379 (1989).

[22]T. M. Giebultowicz, P. Klosowski, J. J. Rhyne, T. J. Udovic, J. K. Furdyna, and W. Giriat, Phys. Rev. B **41**, 504 (1990).

[23]P. M. Shand, A. Lewicki, B. C. Crooker, W. Giriat, and J. K. Furdyna, J. Appl. Phys. **67**, 5246 (1990).

[24]A. Twardowski, C. J. M. Denissen, W. J. M. de Jonge, A. T. A. M. de Waele, M. Demianiuk, and R. Triboulet, Solid State Commun. **59**, 199 (1986).

[25]H. J. M. Swagten, A. Twardowski, W. J. M. de Jonge, M. Demianiuk, and J. K. Furdyna, Solid State Commun. **66**, 791 (1988).

[26]D. U. Bartholomew, E-K. Suh, A. K. Ramdas, S. Rodriguez, U. Debska, and J. K. Furdyna, Phys. Rev. B **39**, 5865 (1989).

[27]It was stated in Ref. 23 that a log-log plot of $T_f$ versus $x$ for the $Zn_{1-x}Co_xS$ samples could not be fit by a single straight line. A power law in $x$ plus a dipole term was used to fit the data. Based on the results of more extensive analyses of the concentration of cobalt ions in our samples, we now believe that a single power-law fit (as in this paper) provides a reasonably good description of data, inasmuch as the uncertainty in the concentration is fairly large.

[28]M. Escorne, A. Mauger, R. Triboulet, and J. L. Tholence, Physica **107B**, 309 (1981).

[29]A. Mookerjee and D. Chowdhury, J. Phys. F **13**, 431 (1983).

[30]A. Lewicki, A. I. Schindler, I Miotkowski, B. C. Crooker, and J. K. Furdyna, Phys. Rev. B **43**, 5713 (1991).

[31]B. E. Larson, K. C. Hass, and R. L. Aggarwal, Phys. Rev. B **33**, 1789 (1986).

[32]W. J. M. de Jonge, A. Twardowski, and C. J. M. Denissen, in *Diluted Magnetic (Semimagnetic) Semiconductors,* edited by R. L. Aggarwal, J. F. Furdyna, and S. Von Molnar, MRS Symposia Proceedings No. 89 (Materials Research Society, Pittsburgh, 1987), p. 153.

[33]B. E. Larson, K. C. Hass, H. Ehrenreich, and A. E. Carlsson, Phys. Rev. B **37**, 4137 (1988).

[34]J. Spalek, A. Lewicki, Z. Tarnawski, J. K. Furdyna, R. R. Galazka, and Z. Obuszko, Phys. Rev. B **33**, 3407 (1986).

[35]A. Lewicki, J. Spalek, J. K. Furdyna, and R. R. Galazka, Phys. Rev. B **37**, 1860 (1988).

[36]N. Bloembergen and T. J. Rowland, Phys. Rev. **97**, 1697 (1955).

[37]Shinn-Sheng Yu and Ven-Chung Lee, Phys. Rev. B **40**, 10 621 (1989).

[38]Except for the samples with the smallest two $x$ values ($x=0.003, 0.032$), $R/R_1=\sqrt{8}$ is greater than $R_m/R_1$ for all our samples. The distance $R=\sqrt{8}R_1$ is therefore large enough to adequately describe the behavior of most of our samples.

[39]A. Lewicki, A. I. Schindler, I. Miotkowski, and J. K. Furdyna, Phys. Rev. B **41**, 4653 (1990).