

Percolation effects and magnetic properties of the randomly diluted fcc system $\text{Co}_p\text{Mg}_{1-p}\text{O}$

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(Received 1 December 1986)

Temperature-dependent, low-field (50 Oe) dc magnetic susceptibility (χ) studies are reported for the randomly diluted fcc system $\text{Co}_p\text{Mg}_{1-p}\text{O}$. The studies, using powder samples, covered the temperature range 1.6–300 K and the composition range $0.10 \leq p < 1$. Variations of the Néel temperature T_N , the Curie-Weiss temperature Θ , the Curie constant C , the low-temperature magnetization M , and the lattice constant a_0 with p are reported. The observed percolation threshold $p_c = 0.13$ agrees well with the predicted value of $p_c = 0.136$ for fcc system with nearest-neighbor and next-nearest-neighbor interactions included. The variation of $\Theta(p)$ and $C_M(p)$ with p is found to be linear whereas that of $t = T_N(p)/T_N(1)$ is more complex, showing a crossover near $p = 0.45$, where d^2t/dp^2 changes sign. Calculations are needed to explain this observed t versus p variation. For $p_c < p < 0.45$, χ below T_N depends upon how the sample is cooled through T_N , and the hysteresis and time dependence (relaxation) of χ are observed at 4.2 K if the sample is cycled through a higher field (6 kOe). These observations are not fully understood. An estimate of the exchange constants for the $\text{Co}_p\text{Mg}_{1-p}\text{O}$ system is given.

I. INTRODUCTION

In recent years, there has been considerable interest in randomly diluted magnetic systems since these systems simulate percolation phenomenon and complex magnetic ordering such as spin-glass states are sometimes observed near the percolation threshold p_c .^{1–3} In a recent paper,¹ de Jongh has reviewed the experimental magnetic studies in the diluted systems. So far, experiments have been reported in quadratic layer systems,⁴ cubic systems,⁴ tetragonal systems [such as MnF_2 : Zn (Ref. 5) and FeF_2 : Zn (Ref. 6)], and the fcc ferromagnetic system EuS :Sr.² However, there is a lack of theoretical and experimental studies in fcc antiferromagnetic systems. Since the percolation phenomenon depends strongly on the crystal symmetry and magnetic properties depends on the sign of the exchange interactions, it is of interest to study fcc antiferromagnetic systems such as $\text{Co}_p\text{Mg}_{1-p}\text{O}$. It is known that (i) CoO is a type-II antiferromagnet with $T_N \cong 289$ K,⁷ and (ii) earlier studies of Arkhipov,⁸ Dyrek,⁹ Bielanski *et al.*,¹⁰ and Cimino *et al.*¹¹ show that it is possible to prepare solid solutions of CoO in MgO over the entire composition range, i.e., $\text{Co}_p\text{Mg}_{1-p}\text{O}$ retains the fcc structure for $0 < p < 1$. However, these earlier magnetic studies were all limited to temperatures above 77 K and in some cases only a few compositions were investigated.^{8–11} Consequently, the interesting results on the variation of the Néel temperature T_N with p on approach to p_c , the nature of magnetic states near p_c , and the magnitude of p_c itself were not available from the earlier studies.^{8–11} In this paper we report the results of our detailed study using temperature-dependent dc magnetic-susceptibility measurements on the $\text{Co}_p\text{Mg}_{1-p}\text{O}$ system. About 20 samples were investigated in the composition range $0.1 < p < 1$ and in the temperature range 1.6–300 K. From these studies, variations of several quantities, viz., T_N , $t = T_N(p)/T_N(1)$, dt/dp , Θ (Curie-Weiss tempera-

ture), and C (Curie constant) with p are determined and $p_c \cong 0.13$ is estimated. This estimate of p_c agrees with the theoretical $p_c = 0.136$ for a fcc lattice with nearest-neighbor (NN) and next-nearest-neighbor (NNN) interactions included.¹² History-dependent magnetic behavior is observed for compositions near p_c . Although a preliminary report of some of these results was made in a recent communication,¹³ details of these results, their analysis, and their interpretation is given in this paper.

II. PERTINENT EXPERIMENTAL DETAILS

There are three important experimental aspects to this work: (i) preparation of $\text{Co}_p\text{Mg}_{1-p}\text{O}$ samples, (ii) characterization of the samples, and (iii) temperature-dependent magnetic-susceptibility studies. Each of these aspects is now considered below.

All our samples are powder specimens which were prepared starting from appropriate amounts of $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (obtained from Alfa Products) as follows.^{9–11} The mixture of nitrates is warmed in a beaker in air to form a clear solution. The solution is then slowly heated in air to yield dry flakes. The flakes are removed, powdered, and then sintered at 400°C in air for about 2 h in an alumina-silica boat to drive away NO_2 , resulting in $\text{CoO}:\text{MgO}$ multiphase powder. A part of this powder ($\cong 150$ mg) is pressed into a cylindrical pellet with a brass die (a stainless-steel die was not used to avoid magnetic contamination). Both the pellet and the remaining powder were then sintered at 1400°C for 8 h in air, followed by rapid quenching in liquid nitrogen. To avoid contamination, a new alumina-silica boat was used for each composition. The powder is used for characterization by x-ray-diffraction and atomic-absorption studies and the pellet is used in the magnetic studies. For two compositions, viz., $p = 0.13$ and 0.17, attempts to prepare samples with the above procedure did

not yield proper compositions. These samples were therefore prepared by starting from single-phase $\text{Co}_p\text{Mg}_{1-p}\text{O}$ powders with $p=0.05$ and 0.25 prepared as above and sintering the appropriate mixture at 1400°C , followed by quenching as described above.

To characterize the samples, an initial test used was magnetic measurements at room temperature for any hint of magnetic contamination. In almost all cases, this was not a problem. Next, using a standard Debye-Scherrer camera, x-ray patterns were obtained for each sample after 4-h $\text{Co } K\alpha$ exposure. Only samples which showed clean fcc lines were further studied. The line positions were measured and, using standard procedures, the lattice parameter was calculated for different Bragg angles α . Then, extrapolating the straight line to $\sin^2\alpha=1$, the experimental value of the lattice constant a_0 was determined for each composition.

The next step was to determine the composition parameter p by measuring the atomic absorption of Co^{2+} and Mg^{2+} . For all samples, the evaluated p values so determined were within 3% of the nominal p values. The graph of experimental a_0 (in \AA) against experimental p is shown in Fig. 1, where the straight line is a least-squares fit (correlation 99.92%) with the equation

$$a_0(p) = 4.2139 + 0.0476p \quad (1)$$

It is noted that our values of a_0 are in good agreement with a few compositions studied by Dyrek⁹ and Bielanski *et al.*¹⁰

The x-ray and atomic-absorption measurements do not necessarily ensure thermodynamic random dilution of the samples. Under experimental results we argue that the observed Curie-Weiss temperatures Θ attest to the random site dilution of our samples. This is an important consideration for percolation studies since any chemical clustering would raise the measured T_N and, hence, the measured p_c .

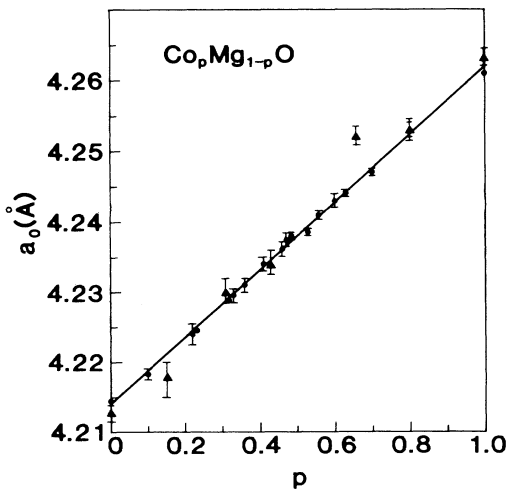


FIG. 1. Variation of the lattice constant a_0 with p in $\text{Co}_p\text{Mg}_{1-p}\text{O}$ system. Solid triangles are the data from Ref. 9 and the solid circles represent this work. The straight line is a least-squares fit [Eq. (1)] to our data.

The dc magnetic-susceptibility (χ) measurements were carried out with a Faraday balance, some details of which have been reported earlier.¹⁴ Two notable changes from the earlier procedures¹⁴ are (i) a sample holder was not needed since the pellets could be suspended in the balance directly, avoiding background corrections, and (ii) a calibrated digital temperature indicator (Air Products model APD-E) was used instead of a potentiometer. Most temperature-dependent measurements were carried out at 50 Oe, the lowest stable field available from the Varian 9-in. electromagnet. Measurements were made either in the zero-field-cooled (ZFC) or field-cooled (FC) mode. In the ZFC (FC) mode, the samples were cooled to the lowest temperature, 4.2 or 1.6 K, in zero-field (50 Oe) followed by $\chi = M/H$ measurements at 50 Oe with increasing temperatures.

III. MOLECULAR-FIELD THEORY

In order to allow a suitable discussion of the experimental results presented in Sec. IV, we briefly review the major results of the molecular-field theory (MFT) for type-II antiferromagnets¹⁵ and for diluted antiferromagnets.¹⁶ Assuming J_1 and J_2 as the NN and NNN exchange interactions with coordination numbers $Z_1 (=12)$ and $Z_2 (=6)$, respectively, for the fcc lattice, MFT yields the following results with the usual notation:

$$3k_B T_N = J_2 Z_2 S(S+1), \quad (2)$$

$$3k_B \Theta = (J_1 Z_1 + J_2 Z_2) S(S+1), \quad (3)$$

and

$$\chi(T_N) = Ng^2 \mu_B^2 / (J_1 Z_1 + 2J_2 Z_2). \quad (4)$$

For the diluted Heisenberg Hamiltonian, Spalek *et al.*¹⁶ have recently shown that in the high-temperature limit ($T > T_N$) the leading term for χ yields the Curie-Weiss law

$$\chi(p) = \frac{C(p)}{T + \Theta(p)}, \quad (5)$$

where $C(p)$ is the Curie constant and $\Theta(p)$ is the Curie-Weiss temperature. Here we wish to note that one usually measures χ_g , the susceptibility per gram, and the variations of $C_g(p)$ and the molar $C_M(p)$ with p are different. It can be shown that

$$C_M(p) = C_M(1)p, \quad (6)$$

$$C_g(p) = C_M(p)/W, \quad (7)$$

and

$$\Theta(p) = \Theta(1)p, \quad (8)$$

where W is the molecular weight of $\text{Co}_p\text{Mg}_{1-p}\text{O}$. Thus, although $C_M(p)$ and $\Theta(p)$ vary linearly with p , $C_g(p)$ varies nonlinearly with p because W depends on p . Also, $\chi(p)$ does not vary linearly with p , but by defining $\chi_M^* = \chi_M(p)/p$ and plotting $1/\chi_M^*$ against T , we can determine $C_M(1)$ and $\Theta(p)$. The importance of $\Theta(p)$ in a randomly diluted system is that since it measures the sum of all magnetic interactions [Eq. (3)] it should vary linearly

with p if there is no chemical clustering (assuming no significant change in lattice constant). Thus the experimental variation of Θ with p should provide a check on the quality of random dilution of the samples. If Θ is too large, it would indicate chemical clustering. Since the maximum change in the lattice constant between $p=0$ and 1 in $\text{Co}_p\text{Mg}_{1-p}\text{O}$ is about 1% (Fig. 1), changes in J_1 and J_2 with dilution should be quite negligible.

IV. EXPERIMENTAL RESULTS AND DISCUSSION

A. Temperature dependence of magnetic susceptibility

The temperature dependence of the magnetic susceptibility for several representative compositions is shown in Fig. 2, where we have plotted $\chi_g^* = \chi/p$, and $\chi = M/H$ is the measured gram susceptibility at 50 Oe. For some compositions, viz., $p < 0.45$ we have shown the data for both the FC and ZFC modes. It is evident that the temperature corresponding to the peak in χ_g^* decreases with decreasing p . The peak in χ or χ_g^* is usually associated with T_N . However, a more accurate measure of T_N is the maximum in $d(\chi_g^*T)/dT$ since this quantity is proportional to the specific heat near T_N .¹⁷ Usually, $d(\chi_g^*T)/dT$ peaks at a temperature few percent lower than the peak position in χ_g^* and, in this work, T_N is determined from $d(\chi_g^*T)/dT$. Another observation from Fig. 2 is that, for $p < 0.45$, χ for the FC mode is higher than that for the ZFC mode below T_N , although in both cases the position of T_N is evident and nearly coincident. For $p > 0.45$, a slight increase in χ_g^* at lower temperature is observed.

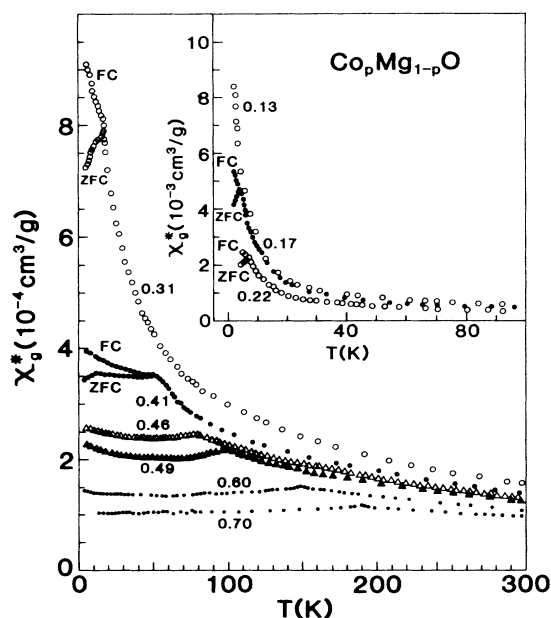


FIG. 2. Temperature dependence of $\chi_g^* = \chi/p$ for several samples of $\text{Co}_p\text{Mg}_{1-p}\text{O}$ with p values indicated. χ is the measured susceptibility per gram at 50 Oe.

B. Percolation effects

To check the validity of the CW (Curie-Weiss) law in $\text{Co}_p\text{Mg}_{1-p}\text{O}$ and to determine $\Theta(p)$, $C_g(p)$, and $C_M(p)$, we have plotted $1/\chi_M^*$ against T in Fig. 3 for several compositions. For $p=0.8, 0.9$, and 1, the data are not plotted since for these compositions T_N is close to the room temperature (the highest temperature of our measurements) and CW law is valid only well above T_N . From the linear fits, the values of $\Theta(p)$ and $C_M(1)$ and hence $C_M(p) = pC_M(1)$ can be determined. The variations of $C_M(p)$ and $\Theta(p)$ so evaluated are shown in Figs. 4 and Fig. 5, respectively. Least-squares fits to the data yield the following variations:

$$C_M(p) = 3.38p \quad (9)$$

and

$$\Theta(p) = 379p - 2 \quad (10)$$

In the above analysis we have included estimated diamagnetic and Van Vleck contributions to χ_g even though their contributions are quite small (e.g., for CoO, the combined estimated contribution is few percent of χ_g at most temperatures¹⁸). From Eqs. (9) and (10), for pure CoO with $p=1$, one obtains $C_M(1) = 3.38$ and $\Theta(1) = 377$, and for MgO with $p=0$, $C_M(0) = 0$ and $\Theta(0) = -2$ are obtained. The magnitude of $\Theta(0) = -2$ should really be considered to be zero because of the uncertainties, whereas $C_M(0) = 0$, as expected. The magnitude of $C_M(1) = 3.38$ corresponds to magnetic moment $\mu = 5.22\mu_B$, a value in agreement with an earlier estimate by Singer.¹⁹ The magnitudes of estimated $\Theta(1)$ for CoO determined by Dyrek,⁹ Singer,¹⁹ and Arkhipov⁸ from high-temperature χ are, respectively, 405, 330, and 280 K. Our extrapolated $\Theta(1)$ nearly equals the average of the values given in Refs. 9 and 19 (Fig. 5). Also, as seen in Fig. 5, the magnitudes of $\Theta(p)$ determined by Dyrek⁹ for some other compositions agree with our values. Since $\Theta(p)$ varies linearly with p over an extended range, our extrapolation procedure to determine $\Theta(1)$ is likely to be quite accurate. Also, as

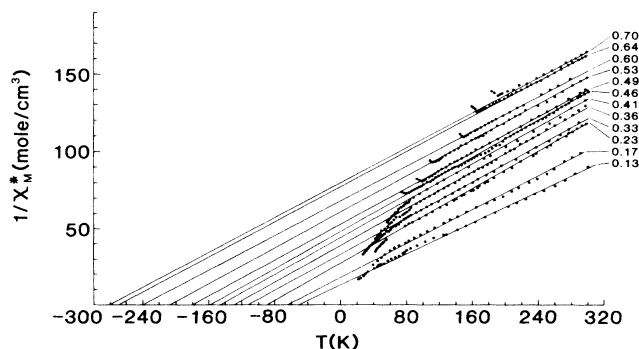


FIG. 3. Plots of $1/\chi_M^*$ against temperature for several samples of $\text{Co}_p\text{Mg}_{1-p}\text{O}$ with p values indicated, where $\chi_M^* = \chi_M/p$ and χ_M is the molar susceptibility. The straight lines are least-square fits to the data for the high-temperature region.

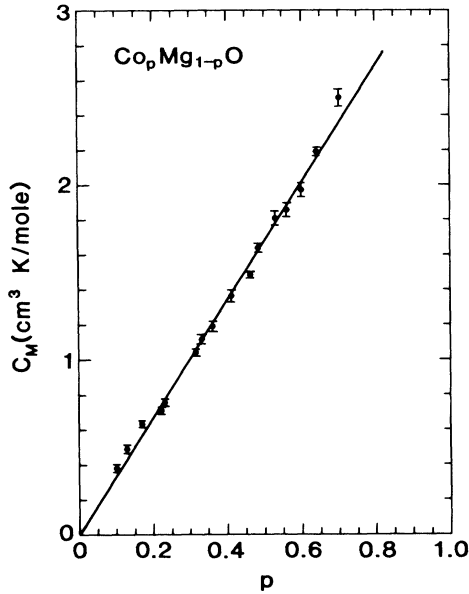


FIG. 4. Variation of the molar Curie constant $C_M(p) = C_M(1)p$ plotted against p , where $C_M(1)$ is determined from slopes of Fig. 3. The solid line is a least-squares fit, Eq. (9).

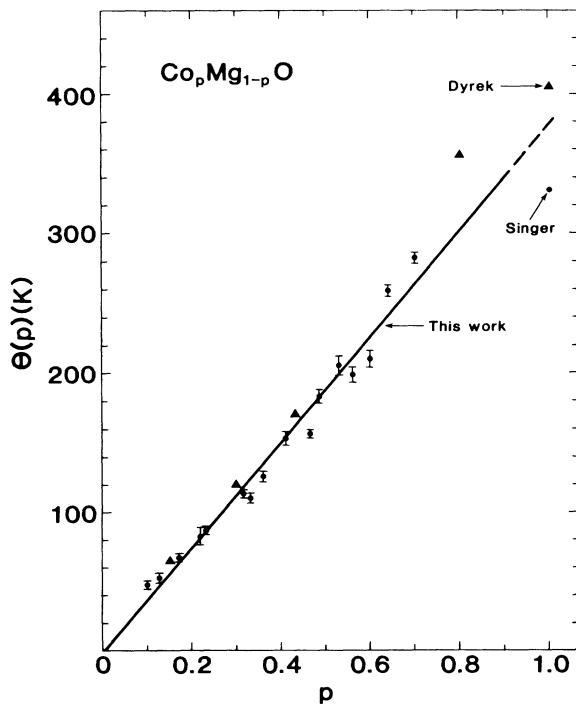


FIG. 5. Variation of the Curie-Weiss temperature $\Theta(p)$ plotted against p for $\text{Co}_p\text{Mg}_{1-p}\text{O}$, where solid triangles are from Ref. 9. The solid line is a least-squares fit to our data, Eq. (10).

noted earlier, the linearity of $\Theta(p)$ with p is an important indication of the quality of the random dilution of our samples.

From the plot of $1/\chi_g^*$ against temperature, C_g can be determined for each composition. The variation of C_g with p is shown in Fig. 6, where the dotted curve is Eq. (7) with $C_M(1)=3.38$. As noted earlier, the variation of C_g with p is nonlinear. To highlight this nonlinearity in a diluted system is the primary objective of showing Fig. 6.

The variation of the reduced Néel temperature $t = T_N(p)/T_N(1)$ against p and that of computed dt/dp against p is shown in Fig. 7. Here we have also plotted the data points of Dyrek⁹ and Arkhipov⁸ for $p > 0.5$. These values are consistent with our measurements. The lowest p value for which T_N was observed in our work is $p=0.17$ for which $T_N=3.5$ K (Fig. 2). For $p=0.13$, no peak in χ was observed down to 1.6 K, the lowest temperature of our experiments obtained by pumping on liquid helium. Similarly, for $p=0.10$ no T_N could be detected down to 1.6 K. Although one cannot rule out a $T_N < 1.6$ K in these samples ($p=0.13$ and 0.10), it is our conjecture, using the extrapolation in Fig. 7, that these materials do not order at a nonzero T_N . By extrapolation in Fig. 7 we deduce $p_c=0.13\pm 0.01$.

A second estimate of the experimental p_c in $\text{Co}_p\text{Mg}_{1-p}\text{O}$ is obtained in Fig. 8, where we have plotted the magnetization M measured at the lowest temperature (1.6 or 4.2 K) of our experiments for the ZFC mode against p . M at 1.6 K peaks at $p=0.13$, and $p=0.14$ for 4.2 K. These peak positions determine p_c and the reasoning is as follows.²⁰ For $p < p_c$, the sample consists of isolated Co^{2+} clusters and spins which may be super-

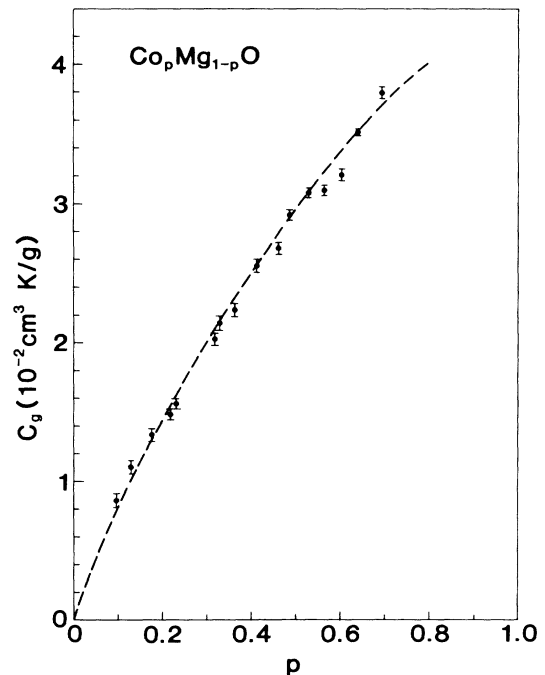


FIG. 6. Variation of the gram Curie constant C_g plotted against p . The dotted curve is Eq. (7).

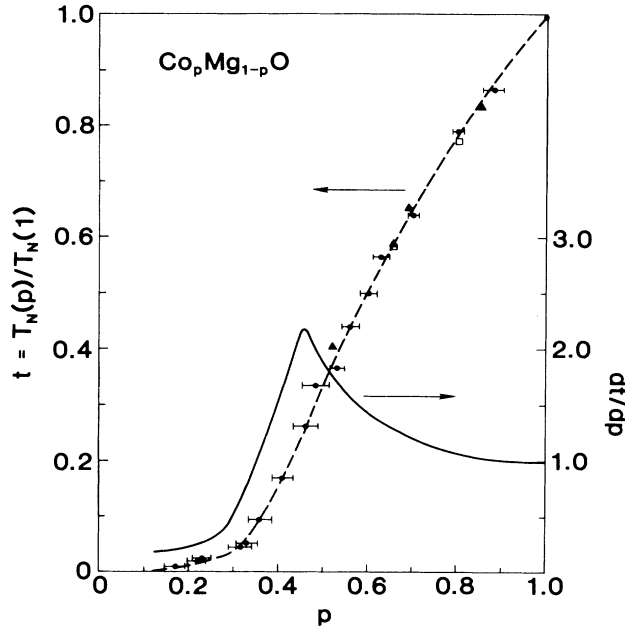


FIG. 7. Variation of the reduced Néel temperature t and computed dt/dp plotted against p for $\text{Co}_p\text{Mg}_{1-p}\text{O}$. The solid triangles are the data from Ref. 8, the open squares from Ref. 9, and solid circles from this work. The dotted line is drawn for visual aid. $t \rightarrow 0$ for $p = p_c = 0.13$.

paramagnetic or at least not compensated for antiferromagnetically. As p increases ($p < p_c$), the number and size of these clusters increases, thereby resulting in increasing M with increasing p . For $p > p_c$, the infinite antiferromagnetic cluster sets in, and with increasing p , the number of spins in the infinite cluster increases, whereas the number and size of isolated clusters decreases. Since the infinite cluster is compensated for antiferromagnetically, M decreases with increasing p for $p > p_c$. Also, the compensation of the spins in the infinite cluster is complete only at 0 K. Therefore the peaking of M at $p = p_c$ should be most pronounced at the lowest temperature. This is evident in Fig. 8. Thus the results in Figs. 7 and 8 fairly unambiguously show that p_c is close to 0.13 in $\text{Co}_p\text{Mg}_{1-p}\text{O}$.

The above experimental value of $p_c = 0.13$ is in excellent agreement with the theoretical magnitude of $p_c = 0.136$ for a fcc lattice with both NN and NNN interactions included.¹² This is quite consistent since it is known that for CoO, only J_1 and J_2 are significant,⁷ and it is not necessary to include NNNN for which the predicted $p_c = 0.061$.¹²

C. Crossover effect

We next examine the variation of computed dt/dp with p (Fig. 7). Near $p = 1$, $dt/dp = 1$, as expected for three-

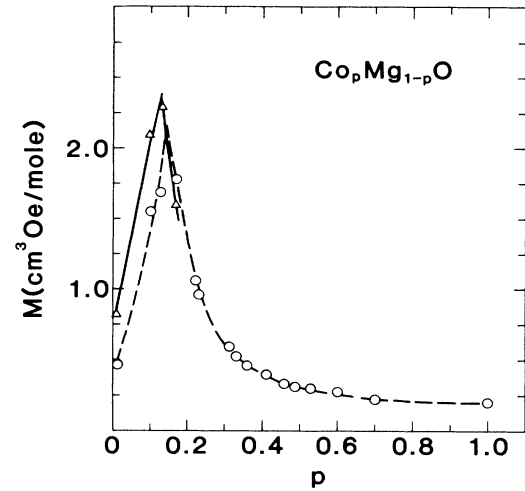


FIG. 8. Variation of the low-temperature magnetization M plotted against p at 1.6 K (open triangles) and 4.2 K (open circles). Lines are drawn through the points for visual aid.

dimensional (3D) Ising systems.¹² In contrast, for $\text{Mn}_p\text{Mg}_{1-p}\text{S}$, another fcc antiferromagnetic system, the observed $dt/dp \approx 1.4$ near $p = 1$,²⁰ close to the predicted value of $\frac{4}{3}$ for a Heisenberg system.¹² Our observations are quite reasonable since it is known that CoO is almost Ising-like near T_N .⁷ The increase in dt/dp with further dilution (decreasing p) towards the Heisenberg value may be due to a decrease in the crystal-field anisotropy. For $\text{Mn}_p\text{Zn}_{1-p}\text{F}_2$, McGurn and Tahir-Kheli²¹ have argued that the anisotropy energy decreases linearly with p . Although the Co systems are more complicated because of the contribution from the spin-orbit interaction, somewhat similar phenomenon may be operative in $\text{Co}_p\text{Mg}_{1-p}\text{O}$. The crossover in d^2t/dp^2 near $p \approx 0.45$, where d^2t/dp^2 changes sign, is perhaps one of the most interesting results of our work. It may be related to the lowering of the dimensionality of the infinite cluster since near p_c the cluster is essentially one dimensional.^{1,22-24} Kumar *et al.*²³ and Stinchcombe²⁴ have calculated the variation of t versus p for simple-cubic Heisenberg and Ising systems and McGurn and Tahir-Kheli²¹ have calculated T_N vs p for $\text{Mn}_p\text{Zn}_{1-p}\text{F}_2$ system. However, no similar calculation exists for the fcc antiferromagnetic system with which we can compare our results of Fig. 6. Such a calculation would therefore be most welcome since detailed experimental results are now available.

D. Field- and time-dependent effects

The observation of Fig. 2 shows that, below T_N , χ for the FC mode is higher than that for the ZFC mode, and that is observed only for compositions with $p < 0.45$. For compositions in the range $0.45 < p < 0.60$, there is an increase in χ with decreasing T at lower temperatures. This latter effect is probably due to isolated spins which should

give a Curie-law- $(1/T)$ type contribution to χ . The fact that for $p > 0.70$ this contribution is negligible suggests that the number of isolated spins is comparatively negligible for these compositions.

In order to understand the field-dependent behavior below T_N for $p < 0.45$, some additional experiments were carried out on one representative composition, viz., $p = 0.23$ with $T_N = 8.9$ K. The sample was cooled to 4.2 K in the ZFC mode and then the magnetization was measured first with increasing field (up to 6 kOe) and then with decreasing fields (6 kOe to 100 Oe). In the decreasing field, M is higher (Fig. 9). After reaching 100 Oe, the time dependence of M was measured and the results are shown in Fig. 10. It is seen that M decreases with time, as if it is trying to approach the initial magnitude measured with increasing field (this is indicated by an arrow in Fig. 10). For the same sample cooled in the FC mode in a separate experiment, hysteresis is again observed (Fig. 9). Also, a somewhat similar time dependence as in Fig. 10 is observed after cycling the field. On the other hand, no time dependence is observed at 4.2 K either in the ZFC or FC mode. The time dependence is observed only after the sample is cooled below T_N (to 4.2 K), the field is increased to a higher value (6 kOe), and then lowered to a lower value (100 Oe). This suggests that the magnetic states, obtained by cooling through T_N in zero field or in a small field (50 Oe), although different, are stable. However, the magnetization process produces an unstable state which relaxes towards a state we started from. A plot of $\ln M$ vs t yields a time constant of about 167 min.

At present, it is difficult to resolve the true nature of the magnetic state for $p < 0.45$ from the observations reported here alone. It is possible that for $p_c < p < 0.45$ the

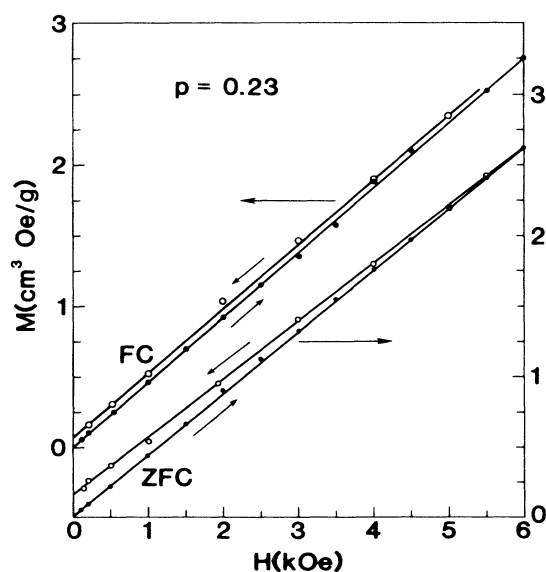


FIG. 9. Field dependence of the magnetization M plotted against magnetic field for the FC and the ZFC modes at 4.2 K in a sample with $p = 0.23$. Ordinate scales have been shifted for the two cases for clarity. The arrows point to the curves obtained for increasing and decreasing fields.

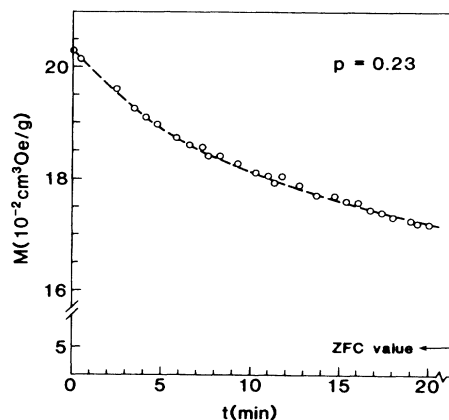


FIG. 10. Time dependence of the magnetization M at 4.2 K and in 100 Oe after field cycling as discussed in the text. The initial value of M at 100 Oe after cooling in the ZFC mode is indicated.

infinite cluster contains frustrated or weak bonds leading to field-dependent susceptibility, as Singh *et al.*²⁵ have argued. For $p < 0.45$, the rapid increase of χ_M^* with decreasing temperatures in Fig. 3 and the crossover in dt/dp in Fig. 7 may be related to the same phenomenon. Of course, for $p < p_c$ the observations have to be dominated by finite clusters. ac-susceptibility measurements at different frequencies are likely to be very useful in deciding whether the magnetic state for $p_c < p < 0.45$ is dominated by frustrated bonds or by superparamagnetic clusters of different size.²⁶

E. Exchange constants

We have seen that $\Theta(p)$ and $C_M(p)$ scale linearly with p in $\text{Co}_p\text{Mg}_{1-p}\text{O}$ and that the changes in lattice constants with dilution are negligible. These observations suggest that J_1 and J_2 are not affected significantly by dilution. For pure CoO, it is estimated that $J_1 = 5.5$ K and $J_2 = 27.4$ K (from spin-wave dispersion curves).⁷ Since reasonably accurate values of Θ and μ are now available from this work, one can attempt to use Eqs. (2)–(5) to evaluate J_1 and J_2 , if we can estimate g and S . Now, $\mu = g\mu_B\sqrt{S(S+1)} = 5.2\mu_B$. It is customary to use the S value from Hund's rule neglecting spin-orbit coupling and then lump the spin-orbit contribution in g . This procedure gives $S = 1.5$ and $g = 2.70$ for $\mu = 5.2\mu_B$. On the other hand, expressions (2)–(5) are based on a Heisenberg Hamiltonian assuming $g = 2$. Therefore we shall assume $g = 2$ and $S = 2.16$ to correspond to $\mu = 5.2\mu_B$. This procedure, with $\Theta = 377$ K and $T_N = 289$ K, yields $J_2 = 21.2$ K and $J_1 = 3.2$ K from Eqs. (2) and (3). Using Eq. (4) with $\chi(T_N) = 4.72 \times 10^{-3}/\text{mol}$ (Ref. 18) yields $J_1 + J_2 = 26.5$ K, a value that is consistent with the above estimates. These estimates of J_1 and J_2 are not too different from those determined from spin-wave dispersion curves. It is noted that if we use $g = 2.7$ and $S = \frac{3}{2}$, then $J_2 = 38.5$ K and $J_1 = 5.8$ K are obtained. The primary difficulty in determining J_1 and J_2 in CoO, besides the

well-known limitations of MFT, is how to include the spin-orbit contribution to the magnetic moment in a simple manner. The Green's-function analysis of Lines and Jones,¹⁵ so successful for MnO, therefore cannot be used for CoO since it is based on the Heisenberg Hamiltonian.

V. CONCLUDING REMARKS

In this work very accurate data of T_N versus p for the $\text{Co}_p\text{Mg}_{1-p}\text{O}$ system has been presented for the whole composition range for the first time. Although the observed percolation threshold, $p_c=0.13$, agrees extremely well with the predicted value, no calculation yet exists with which the observed variation of T_N versus p could be compared. We hope that these results would encourage such a calculation, for there are many interesting features (notably, a crossover near $p=0.45$) of this curve that are not seen in simple-cubic and bcc Heisenberg and Ising systems. The role played by isolated spin clusters needs

further experimental studies, particularly using ac-susceptibility measurements. Although single crystals of $\text{Co}_p\text{Mg}_{1-p}\text{O}$ are somewhat difficult to prepare because of their high melting points, this work shows that much can be learned from studies with powder specimens in this very interesting system.

ACKNOWLEDGMENTS

The authors are grateful to Professor A. S. Pavlovic for assistance with x-ray-diffraction studies and to Ms. Kenna Johnson of West Virginia Geological Survey for assistance with atomic-absorption work. The technical assistance of J. Dean, C. Sicina, J. Johnson, D. Feathers, C. Weber, and Karen Adams is also acknowledged. The authors benefited greatly from discussions with Professor M. F. Ferer and Professor J. M. Honig. This work was supported in part by the National Science Foundation under Grant No. DMR-81-06620.

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