Nearest-neighbor Mn antiferromagnetic exchange in $Ga_{1-x}Mn_xN$

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We analyze the magnetic behavior of well-characterized, precipitate-free $Ga_{1-x}Mn_xN$ thin films containing Mn at higher levels than previously attained; up to $x=0.36$. This level is above the percolation threshold x_c for nearest-neighbor cations, such that exchange between nearest neighbors will dominate the magnetic response. The susceptibility decreases as the Mn content increases up to and beyond x_c, as an increasing fraction of the Mn experiences antiferromagnetic exchange. The dominance of antiferromagnetic behavior at higher Mn concentrations and the total lack of evidence for ferromagnetic ordering even above x_c demonstrates that the nature of the exchange between Mn^{2+} ions in GaN is antiferromagnetic.

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I. INTRODUCTION

The magnetic behavior of the dilute magnetic semiconductor (DMS) $Ga_{1-x}Mn_xN$ has come under close scrutiny in recent years due to the important potential utility of a magnetic wide band-gap semiconductor in incorporating spin-based processing into electronic devices.[1–3] Despite this attention, experimental investigations of the magnetic behavior have as yet failed to find a single accepted magnetic character in $Ga_{1-x}Mn_xN$, variously reporting paramagnetism,[4,5] ferromagnetism,[6–8] superparamagnetism,[9] or antiferromagnetism[10–12] in thin films or crystals prepared with Mn concentrations with $0.0001<x<0.075$. In the films exhibiting ferromagnetic behavior, Curie temperatures range from ~10 K (Refs. 13 and 14) to over 900 K.[15] There is no fully agreed explanation for the wide variety of observed magnetic behaviors in the literature, which has led to uncertainty in the mechanism of magnetic interaction between the Mn ions in GaN.

A central question that still has at best an ambiguous answer is the exchange strength and sign as a function of the Mn-Mn separation. The response to this question is the key to determining which of the proposed exchange mechanisms are active in $Ga_{1-x}Mn_xN$: indirect Ruderman-Kittel-Kasuya-Yoshida (RKKY) via carriers, the formation of bound magnetic polarons, superexchange, or double exchange via the N ions have all been suggested as suitable models.[16] One obvious route to an answer is via studies spanning the dilute to the concentrated limit, working across the range where there is a rapid increase in probability of finding another Mn ion among the 12 neighboring cations around any given Mn. Recent theoretical work has suggested that $Ga_{1-x}Mn_xN$ requires doping levels of greater than $x=0.20$ to attain room-temperature ferromagnetic behavior,[17] corresponding to the percolation limit assuming exchange between nearest neighbors only. However, crystalline samples containing concentrations as high as this have been plagued by phase separation problems[18–21] or Mn clustering issues.[22,23] Among the detected phases are Mn_3GaN,[19,24,25] $Mn_{4-x}Ga_xN$,[26] and $Mn_{1-x}Ga_x$,[27,28] which include antiferromagnetic, ferromagnetic, ferrimagnetic, and spin-glass magnetic behaviors, further complicating the debate about the intrinsic properties of the single-phase material.

In order to address exactly the nearest-neighbor exchange interaction we have investigated the magnetic behavior of phase-pure $Ga_{1-x}Mn_xN$ films with substitutional Mn at much higher concentrations than have been achieved previously. Our magnetic measurements were performed on polycrystalline films prepared by ion-assisted deposition,[29,30] showing a Mn bonding configuration that rigorously substitutes for Ga. Thus the Mn ions are bonded to four N ions in a local tetrahedral coordination, with a second ionic shell of 12 cations; the Mn ions are then found in an environment that follows that of Ga ions in GaN. The material is single phase in the sense that it is an alloy in which the Mn appears randomly substituted for Ga in the GaN structure. As the Mn concentration is increased up to and beyond the percolation threshold, we observe a decrease in the magnetization, a result beyond that of GaN doped with Mn to lower concentrations. It will be demonstrated that the exchange interaction, dominated at these high Mn concentrations by the nearest-neighbor interaction, is clearly antiferromagnetic.

To understand the inferences we draw from our results it is useful to contrast, under ferromagnetic and antiferromagnetic nearest-neighbor exchange, the magnetic behavior that can be expected as the Mn concentration approaches and surpasses the percolation threshold. The number of Mn ions in the nearest cation shell to any specific cation (Mn or Ga) is 12, and above the percolation threshold x_c there is the formation of a macroscopic sample-spanning exchange-coupled cluster of nearest-cation neighbor coupled Mn ions. For concentrations below this threshold the typical number of nearest-neighbor connected Mn ions rises with increasing Mn concentration, ultimately diverging at the threshold. It immediately follows that above x_c the sample will show a macroscopic moment in the ground state if the nearest-neighbor exchange is ferromagnetic. It is notable that this argument is also exactly as has been described for Mn-doped GaAs; that system also shows percolation behavior,[31,32] though the long-range nature of the ferromagnetic RKKY interaction pushes the threshold to much lower Mn concen-

TABLE I. Elemental compositions of investigated $Ga_{1-x}Mn_xN$ films, reported in atomic percent. The results are determined from a combination of Rutherford backscattering spectroscopy and nuclear reaction analysis measurements. Secondary-ion mass spectroscopy shows that the low, unintentional O doping in the films is localized in the top few nanometers.

Sample	at. % Mn	at. % Ga	at. % N	at. % O
$x=0.086$	4.3	39.2	54.2	2.4
$x=0.18$	9	34.5	55.5	1
$x=0.36$	18	26	52	4

tration. Below x_c one might expect to see superparamagnetic behavior from ferromagnetically coupled clusters.

We now consider the magnetic order to be expected under antiferromagnetic exchange conditions with the majority of the moments within a nearest-neighbor exchange-coupled cluster aligned in opposition to their neighbors. Even an antiferromagnetically coupled cluster can be expected to have incomplete spin balance and a small net moment, but nonetheless the antiferromagnetic exchange between coupled Mn reduces the potential moment available for alignment in an applied field, and the susceptibility falls as the concentration approaches x_c. Such a reduced magnetic response near x_c is in direct contrast to the behavior under ferromagnetic nearest-neighbor exchange described in the previous paragraph. We will see below that this reduced response is exactly the behavior observed in our $Ga_{1-x}Mn_xN$ films, providing clear evidence for antiferromagnetic nearest-neighbor exchange.

II. EXPERIMENTAL DETAILS

The $Ga_{1-x}Mn_xN$ films investigated here were grown to between 80 and 220 nm thicknesses on Si, SiO_2, and thin Mylar film substrates using ion-assisted deposition.[29] Both the composition and structure of these films have been carefully examined.[30] The compositional details were determined by a combination of Rutherford backscattering spectroscopy, nuclear reaction analysis, and secondary-ion mass spectroscopy, with the results summarized in Table I. The surface of the films is partially oxidized, resulting in the few percent O content that is detected in some samples. The incorporated Mn content is as high as $x=0.36$ in largely O-free GaN.

The possibility of doping such very high levels of Mn into GaN without altering the overall structure is supported by the calculations of Chan *et al.*[33] who suggest that up to 65% of the Ga may be replaced by Mn. The high Mn concentration is, however, gained at a price; these films are all nanocrystalline with typically 3 nm diameter crystallites. Their x-ray diffraction patterns show the nanocrystals to be random stacked,[34] a mixture of the stacking sequences expected for fcc and hcp lattices. Such a structure does not alter the ionic coordination out to the second shell (the first cation shell about a central cation). Furthermore, the site percolation threshold, which we exploit in our interpretation below, is $x_c=0.2$ in both of the close-packed structures.[35]

The Mn ion distribution is of special importance for this study. In previous work we reported that the Mn ions show an extended x-ray absorption fine structure pattern that establishes they have a coordination that is identical to that of the Ga in these and in Mn-free GaN.[30] They have four N nearest neighbors and 12 cation second-nearest neighbors. We see no evidence at all for secondary phases such as rocksalt-structure MnN, which is an antiferromagnetic metal with $T_N=400$ °C.[36] These conclusions are further supported by optical and dc conductivity measurements showing a gapped spectrum with the Mn ions progressively increasing the density of localized states in the gap;[37] again there is no evidence of metallic MnN. The conductivity evolves from variable-range hopping between states localized in the gap at low Mn concentrations toward weak localization at the highest Mn content, where the localized states begin to overlap. X-ray absorption spectroscopy clearly identifies the Mn as being in the 2+ oxidation state.

The magnetic properties of the films were measured using a Quantum Design magnetic property measurement System (MPMS) superconducting quantum interference device (SQUID) magnetometer in fields from 0–60 kOe and at temperatures from 2–300 K. Temperature-dependent measurements were made with both zero-field-cooled (ZFC) and FC procedures. Prior to the measurements, the films were cooled to 2–5 K in zero applied field. The ZFC measurements were then made while heating the films in the presence of a fixed magnetic field. Subsequently, FC measurements were made by cooling the films from 300 K in the presence of the same field as for the ZFC measurements. All measurements were performed with the magnetic field applied parallel to the surface of the films. The $Ga_{1-x}Mn_xN$ films investigated in this study were the samples deposited on diamagnetic Mylar films, allowing for a much larger volume of sample to be inserted into the typical SQUID measurement holder than possible with deposition onto rigid substrates. Except where specifically noted, the substrate magnetic susceptibility is negligible in comparison with the measured susceptibility of the $Ga_{1-x}Mn_xN$ films.

III. RESULTS AND DISCUSSION

The ZFC temperature dependence of the magnetic susceptibility χ is shown in Fig. 1 for a $Ga_{1-x}Mn_xN$ film with $x=0.086$. The convex shape of the temperature dependence and the lack of saturation, even at the lowest temperature of 2 K, resemble a paramagnetic response, with no clear phase transition into an ordered magnetic phase. Furthermore, there is no irreversibility between the ZFC and FC curves (not shown). The low-temperature field dependence of magnetization in Fig. 2 similarly shows none of the signs of ferromagnetism, such as saturation or a remanent magnetization, and instead resembles more closely the Brillouin function behavior of paramagnets.

However, the values of χ shown in Fig. 1 decrease as the measuring field increases. This behavior is observed at all temperatures and thus it cannot simply be attributed to the onset of saturation of the Brillouin function in large fields. In fact, the decrease in χ can be explained by the presence of a

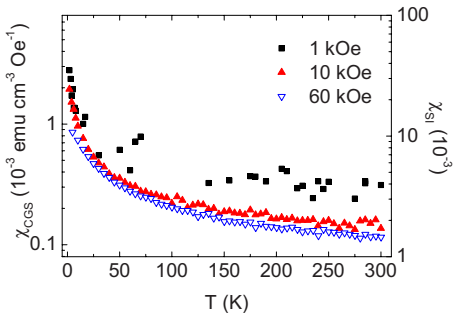

FIG. 1. (Color online) Magnetic susceptibility $\chi=M/H$ versus temperature for a $x=0.086$ thin film measured in various applied fields. Note the decrease in χ as the field increases, which we attribute to the presence of a near temperature-independent contribution to the magnetization resulting from nearest-neighbor antiferromagnetic correlations among some of the Mn ions.

small, positive, roughly temperature-independent contribution to the magnetization which persists up to the highest measurement temperatures. Indeed, the value of the magnetic moment at room temperature in an applied field of 10 kOe is approximately three times that which would be expected from a purely paramagnetic film containing Mn^{2+} at $x=0.086$, indicating that a fraction of the Mn ions experience strong magnetic correlations that persist to high temperature. Thus we are led to a model of the system in which some of the Mn ions experience strong magnetic exchange, which we argue below to be antiferromagnetic, and the remainder of the ions experience much weaker exchange interactions.

The field-dependent magnetization of both $x=0.086$ and $x=0.18$ $Ga_{1-x}Mn_xN$ samples in Fig. 2 are well fit with the function

$$M = \alpha N g \mu_B J B_J(H, T_{\text{eff}}), \tag{1}$$

where α is the effective paramagnetic fraction of the N Mn ions in the film, μ_B is the Bohr magneton, J is the total

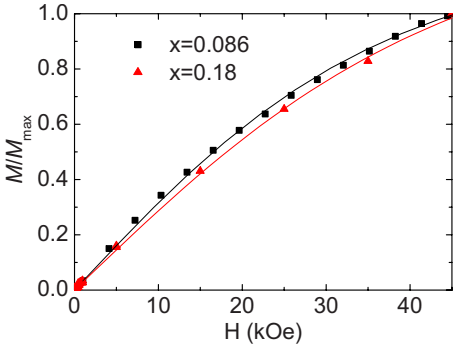

FIG. 2. (Color online) Magnetization versus field for the $x=0.086$ and $x=0.18$ $Ga_{1-x}Mn_xN$ films measured at 5 K, normalized to the values at $H_{max}=45$ kOe. The points are the measured data, the lines are fits to these data using the modified Brillouin function from Eqs. (1) and (2) with $T_0=+1.4\pm0.2$ K($x=0.086$) and $T_0=+2.7\pm0.2$ K($x=0.18$). The small but positive values for T_0 indicate weak antiferromagnetic correlations among the largely isolated Mn ions in these samples.

angular momentum quantum number of the magnetic particles, and $B_J(H, T_{\text{eff}})$ is the Brillouin function

$$B_J(H, T_{\text{eff}}) = \frac{2J+1}{2J}\coth\left[\left(\frac{2J+1}{2J}\right)y\right] - \frac{1}{2J}\coth\left[\left(\frac{1}{2J}\right)y\right] \tag{2}$$

with

$$y = \frac{Jg\mu_B H}{k_B T_{\text{eff}}}, \quad T_{\text{eff}} = T + T_0,$$

dependent on the applied field H and measurement temperature T. The value of J was set to 5/2 consistent with previous x-ray absorption measurements on these films.[37] Note that Eq. (2) contains the full form of the semiclassical expression for the field- and temperature-dependent magnetization of an assembly of magnetic moments, whereas the more commonly used Curie or Curie-Weiss approximation is applicable only where $g\mu_B H \ll k_B T$. The use here of the full form allows fits to be made to the data even at the highest measurement fields. At this point we have not included any additional term to represent the roughly temperature-independent contribution described above, as the magnitude of this term is small and it becomes significant only at higher temperatures.

In the argument of the Brillouin function, we have introduced an effective measurement temperature $T_{\text{eff}}=T+T_0$, as used by Zajac *et al.*[10] and others.[38] Positive values of T_0 imply antiferromagnetic interactions, which cause the Mn^{2+} spins to align a little less effectively with an applied field than would be the case for a system of totally noninteracting paramagnetic moments. For the $x=0.086$ sample the best fit T_0 value is only $+1.4\pm0.2$ K, with the small value here implying rather weak interactions among the relatively widely separated Mn ions. The value of a few kelvin for T_0 is in good agreement with that found for dilute paramagnetic $Ga_{1-x}Mn_xN$ crystals fitted with the Curie-Weiss law.[10]

The $x=0.18$ sample displays qualitatively similar behavior in the field and temperature dependence of the magnetization, and is also well fit with Eqs. (1) and (2). The higher fitted value of $T_0=+2.7\pm0.2$ K reflects an increased effect of the antiferromagnetic interactions at higher Mn concentration. Notably, although the film contains over twice as many Mn ions as the $x=0.086$ sample, the magnitude of the magnetization is actually smaller, and the magnetization shows less tendency to saturate at high fields, as shown in Fig. 2. This decrease in overall magnetization is another clear signal that the underlying magnetic exchange interaction between closely spaced Mn ions is antiferromagnetic in these films.

To further understand the origin of the different contributions to the magnetization, we note that the average exchange interaction experienced by each Mn ion depends on its local environment. A Mn concentration of $x=0.086$ is below the percolation limit for the fcc and hcp lattices ($x_c=0.20$),[35] but in a random distribution over cation sites, some Mn ions will have one or more Mn occupying nearest-neighbor Ga sites (Mn_{Ga}). It is important to note that such a random distribution does not involve a segregation of Mn cations, either through a spinodal decomposition mechanism

TABLE II. Parameters for fitting the temperature-dependent magnetization measurements of $Ga_{1-x}Mn_xN$ using the model of Eq. (3).

Mn (at. %)	H (kOe)	α	T_0 (K)	M_0 (emu cm^{-3})
4.3(x=0.086)	1	0.36	1.9 ± 0.4	0.29 ± 0.02
4.3(x=0.086)	10	0.48	5.0 ± 0.1	1.08 ± 0.02
4.3(x=0.086)	60	0.57	12.4 ± 0.4	3.94 ± 0.07
9(x=0.18)	2	0.16	8.1 ± 0.2	0.24
9(x=0.18)	5	0.21	11.7 ± 0.3	0.07 ± 0.01
18(x=0.36)	10	0.002	24.8 ± 0.7	−0.10

or with the formation of a crystallographically distinct phase. Considering the Mn ions to be randomly distributed on hcp lattice sites, the equations of Behringer[39] are used to calculate the proportions of Mn ions with zero, one and two nearest neighbors. For x=0.086, these proportions are 0.34, 0.205, and 0.135, respectively. Katayama-Yoshida *et al.*[40] have calculated the intrinsic exchange couplings in $Ga_{1-x}Mn_xN$ to be only very short range, in sharp contrast to the longer-range interactions in other Mn-doped III-V semiconductors, where ferromagnetism is well established.[41] Thus, we expect a contribution to the magnetization from the more isolated Mn ions which shows only weak correlations, and a contribution exhibiting much stronger correlations from the ions which lie in close proximity to one or more other Mn.

To compare the behavior of films with Mn content below, close to, and above the percolation threshold we extend the model of Eqs. (1) and (2) to the temperature-dependent data, including also a temperature-independent offset magnetization to model the contribution of the more strongly correlated Mn ions which will present a much weaker temperature dependence of magnetization than the isolated Mn. We thus model the temperature dependence of magnetization as

$$M = \alpha N g \mu_B J B_J(H, T_{\text{eff}}) + M_0. \quad (3)$$

The model of Eq. (3) results in better fits to the measured data than other model forms previously applied to DMS,[42,43] such as Curie-Weiss or simultaneous Curie-Weiss plus Curie terms. The parameters T_0, α, and M_0 from fitting to Eq. (3) are displayed in Table II.

As the concentration of Mn increases, more of the Mn ions experience strong exchange interactions with other nearest neighbor Mn_{Ga}, and thus fewer of the ions are free to respond to an applied field, which accounts for the decrease in the paramagneticlike fraction α. Furthermore, the effects of the antiferromagnetic exchange become more prevalent even among the less strongly coupled Mn ions, leading to an increase in the values of T_0 as Mn content increases.

At the same time the values of M_0, representing the strongly coupled Mn fraction, actually decrease as Mn content increases, representing a significant stiffening of the strongly correlated moments. We imagine this contribution to the magnetization to originate from the canting of moments in antiferromagnetically aligned Mn_{Ga} clusters.[44] In fact, the most highly doped x=0.36 film is far above the percolation limit for the fcc or hcp crystal structures, so an unbroken exchange-coupled cluster of the strongly coupled Mn_{Ga} ions extends throughout the film. This will substantially inhibit the ability of the Mn moments to respond to an applied field and the net negative value of M_0 for the x=0.36 film shows that the contribution from these strongly coupled Mn ions is reduced enough to be dominated by the weak diamagnetism of the substrate. In Fig. 3 we show the values of χ for the $Ga_{1-x}Mn_xN$ films at all three Mn concentration values. The temperature-independent component M_0, as per Eq. (3), has been removed so as to better show the change in the temperature-dependent magnetization with increasing Mn concentration. The values of χ decrease as the Mn content increases, clearly showing the increase in the amount of the paramagneticlike fraction of the Mn that experiences antiferromagnetic exchange. The decrease in χ is especially dramatic above the percolation threshold x_c=0.20.

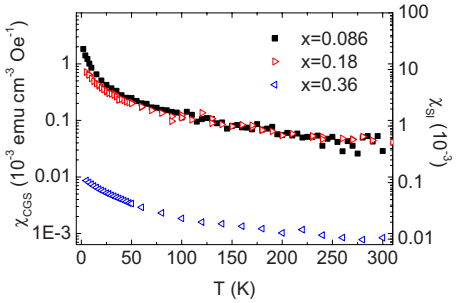

FIG. 3. (Color online) χ vs temperature for $Ga_{1-x}Mn_xN$ films with different values of x.

The above results demonstrate that at low Mn concentrations the magnetization closely resembles a paramagnet with only a small contribution from more strongly correlated Mn moments. Only as the Mn content increases close to and beyond the percolation threshold does the antiferromagnetic exchange affect a more significant fraction of the Mn with a consequent weakening of the temperature and field dependence of the magnetization.

Our data clearly signal that the exchange interaction in these $Ga_{1-x}Mn_xN$ films is antiferromagnetic and very short ranged, showing appreciable strength only when the Mn concentration is high enough that a significant fraction of Mn ions have one or more Mn_{Ga} among their nearest neighbors. This result is in contrast to many predictions in the literature that find ferromagnetic coupling between the Mn ions.[40,45,46] However, studies predicting ferromagnetism usually assume Mn^{3+} ions, in contrast to the Mn^{2+} ionic state more commonly found via experimental investigation[10,37,47] in noncodoped material. The assumed charge state of the Mn ions has a strong influence on the predicted exchange, and indeed calculations find the antiferromagnetic state to be more stable when the Mn ions are in the 2+ state,[48] and when the Mn-N and Mn-Mn bond lengths are allowed to relax to equilibrium.[49,50] Calculations for Mn^{2+} ions in fully relaxed configurations often find good agreement with experimental determinations of the positions of the magnetically active Mn d levels within the GaN band gap.[51] It is also interesting to note that a recent calculation finds that Mn ions within

groups of two or more couple antiferromagnetically for all but the highest Mn oxidation states.[52]

Our analysis supports the results of Ref. 14 that show that the most structurally perfect, well-characterized films of $Ga_{1-x}Mn_xN$ result in ferromagnetism at low temperatures ($\sim$10 K) only. In order to produce such films, great care was needed to ensure epitaxial growth conditions and that unintentional defect states such as N vacancy donors were reduced. Only with such stringent conditions on the growth process and the use of several complementary techniques for characterizing the structural ordering of the Mn is a phase-pure state of the material ensured and the Mn incorporated as Mn^{3+} ions, rather than the more commonly observed Mn^{2+} state. As mentioned earlier, the films of the present study contain concentrations of Mn^{2+} above the condition for percolation in the fcc or hcp structure. Ferromagnetic exchange between the Mn^{2+} ions would necessarily result in a net ferromagnetic character in the above-percolation films, however we find only the signatures of antiferromagnetic coupling. The evidence of this study and Ref. 14 calls into question the supposedly intrinsic origin of the high-temperature magnetic properties of $Ga_{1-x}Mn_xN$ found in numerous other studies.

IV. CONCLUSIONS

In summary, we have reported magnetic measurements of single-phase, well-characterized $Ga_{1-x}Mn_xN$ films at up to $x=0.36$. Even above the percolation threshold, there is no sign of a long-range ferromagnetically ordered phase down to 2 K and up to 60 kOe applied fields. Instead, the magnetization of the films is due to a continuum of coexisting magnetic contributions owing to the nature of a random distribution of Mn ions on cation sites. We have fitted the magnetization successfully with the two most dominant contributions: the fraction of isolated Mn^{2+} spins in a paramagneticlike state with weak antiferromagnetic exchange and Mn^{2+} that is more strongly antiferromagnetically correlated with more closely spaced neighbors. This characteristic shows that the exchange between nearest-neighbor Mn^{2+} located on cation sites in GaN is antiferromagnetic.

In the context of a recent study on Mn-doped GaN carefully prepared to be free of precipitates, our results support the notion that it is likely that the $Ga_{1-x}Mn_xN$ ferromagnetism reported elsewhere is generally due to extrinsic effects such as Mn clustering or unintentional defects. It appears that intrinsic ferromagnetism may be best achieved in $Ga_{1-x}Mn_xN$ through careful codoping with elements designed to compensate the unintentional *n*-type carriers and drive the phase-pure material *p* type. The codoping not only generates holes as possible mediators of exchange between Mn ions but also results in the presence of a larger concentration of Mn^{3+}. Indeed, a number of experiments have shown that an increased hole concentration in $Ga_{1-x}Mn_xN$ from codoping with *p*-type elements such as Mg improves the ferromagnetic properties.[28,53–55] The difficulty in achieving an efficient *p*-type doping of GaN ensures that this method of encouraging ferromagnetism in $Ga_{1-x}Mn_xN$ remains a significant challenge.

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