Scaling behavior of the magnetization of insulating Si:P

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We report measurements of the magnetization of insulating phosphorus-doped silicon for fields up to 50 kG and temperatures down to 1.25 K, for dopant concentrations $N=6.7\times10^{17}$ to 2.8×10^{18} cm^{-3}. The data are found to be in quantitative agreement with a generalization, to finite fields, of a scaling calculation of the magnetic properties of the insulating phase, and support a model of hierarchically coupled spin pairs.

Experimental studies have shown[1–4] that the magnetic behavior of lightly doped semiconductors (a random amorphous antiferromagnet) is rather different than that of spin glasses, including ones with predominantly antiferromagnetic interactions.[5] Despite the possibility of frustration, the large quantum fluctuations in this spin-$\frac{1}{2}$ system appear to drive the system to a ground state consisting of localized singlets, much like the quasi-one-dimensional organic salts[6] which are their one-dimensional analogs. The magnetic susceptibility varies as $T^{-\alpha}$ at low temperature with α almost independent of temperature but varying weakly with concentration of donors in the experimental temperature[3] range 30 mK–4 K. Such behavior has been modeled in terms of coupled spin clusters[2–3] as well as a hierarchy of coupled spin pairs.[3,7,8] A renormalization-group scheme has substantiated the basic philosophy of the latter scheme. It should be noted, however, that renormalization effects are non-negligible.[9] Further, though the calculations have been carried out only for the spin-$\frac{1}{2}$ (donor) case, similar behavior (magnetic susceptibility $\sim T^{-\alpha}$) has also been observed[10] for silicon doped with boron, an acceptor, whose ground state is described by an angular momentum $j=\frac{3}{2}$.

Though various calculations have been compared with each other,[7,11] their fits to individual experimental curves have involved adjustable parameters. It is clearly desirable to have an experimental check of the theoretical picture which is free of this limitation. In the present study we use measurements of the nonlinear magnetization in insulating Si:P to achieve this end. We have studied a range of concentrations $N=6.7\times10^{17}$ to 2.8×10^{18} cm^{-3} (as determined by room-temperature resistivity measurements[12]—on this scale, the metal-insulator transition density $N_c=3.5\times10^{18}$ cm^{-3}). Our measurements cover a number of temperatures in the range 1.2 K–10 K, and fields up to 50 kG.

Within the picture of hierarchically coupled spin pairs with renormalized interactions,[9] the distribution of (renormalized) exchanges[13] can be obtained directly from the temperature dependence of the susceptibility. The model can then be generalized to finite fields and, with *no further adjustable parameters,* predicts the magnetization at all fields and temperatures. Our main result is that the theoretical model fits our experimental data well at all the concentrations, temperatures, and fields studied. According to recent NMR relaxation-time measurements,[14] the magnetic behavior on either side of the metal-insulator transition appears to be qualitatively similar. Thus an understanding of the insulating behavior on a quantitative level is rather important in seeing any crossover effects as the metallic phase is approached.

Four Si:P samples, three (A–C) from Mulab and one (D) from Virginia Semiconductors (all cut from single crystals grown by the Czochralski technique) were used in the present study. Their room temperature resistivities, measured using the van der Pauw method,[15] and the corresponding phosphorus concentrations deduced from the Thurber curve,[12] are listed in Table I.

Magnetic measurements were made in a Faraday balance on samples weighing between 200 and 500 mg. The donor magnetization $M(H,T)$ was determined to a precision of 2×10^{-7} emu G/g between 1.25 K and room temperature, by subtracting the measured magnetization for a zone-floated high-resistivity n-type silicon sample. Magnetization curves were obtained for fields up to 50 kG at the lower temperatures, and the susceptibility was de-

TABLE I. Room-temperature resistivity, donor concentration (obtained using Ref. 12) and the parameter α deduced from fits to susceptibility and magnetization data (see text), for samples A to D.

Sample	Room Temperature ρ (Ω cm)	N (cm^{-3})	α
A	0.0285	6.7×10^{17}	0.78 ± 0.02
B	0.0205	1.30×10^{18}	0.64 ± 0.03
C	0.0145	2.4×10^{18}	0.64 ± 0.02
D	0.0134	2.8×10^{18}	0.64 ± 0.02

duced from the initial linear part, to a precision of 2×10^{-10} emu/g.

Figure 1 shows the susceptibility per donor as a function of temperature for samples A and C between 1.25 and 10 K on a double-logarithmic plot. In agreement with earlier experimental studies[3,4] to much lower temperatures, the susceptibility at low temperatures obeys quite well the relation $\chi \propto T^{-\alpha}$, with the values of α listed in Table I. For comparison, the Curie law for noninteracting moments is also shown in Fig. 1.

Assuming that the low-temperature behavior of the system in the insulating phase can be described in terms of spin pairs coupled hierarchically according to a *renormalized* distribution of antiferromagnetic Heisenberg exchange, $P(J)$, the susceptibility is given by

$$\chi(T)=\frac{4C}{T}\int_0^{J_c}\frac{P(J)dJ}{3+e^{J/k_BT}}\ , \tag{1}$$

where C is the Curie constant $C=Ng^2\mu_B^2S(S+1)/3k_B$ with $S=\frac{1}{2}$ (the Curie susceptibility $\chi_C(T)=C/T$), and J_c is the upper cutoff of the exchange distribution, which for our system is ~ 1 Ry ~ 500 K. One recognizes immediately from Eq. (1) that a power-law form $\chi(T)\sim T^{-\alpha}$ for $k_BT \ll J_c$ follows from a power-law distribution of the renormalized exchange, i.e., $P(J)\sim J^{-\alpha}$. Consequently, within our experimental accuracy, we are able to parametrize our low-temperature susceptibility data in terms of a single parameter α (<1). The same would follow from earlier experiments,[3,4] as well as the numerical results,[9] to the extent that the small curvature on a $\log\chi$ versus $\log T$ plot can be neglected.

Within the same model of coupled spin pairs with an exchange distribution $P(J)$, the magnetization can be expressed as

$$M(H,T)=\frac{4Ck_B}{g\mu_B}\int_0^{J_c}\frac{P(J)dJ\,\sinh(g\mu_BH/k_BT)}{1+e^{J/k_BT}+2\cosh(g\mu_BH/k_BT)} \tag{2}$$

and for $P(J)\sim J^{-\alpha}$, we may write for $g\mu_BH$, $k_BT \ll J_c$, $J \ll J_c$,

$$M(H,T)=(k_BT/g\mu_B)\chi(T)f_\alpha(g\mu_BH/k_BT)\ , \tag{3}$$

where $\chi(T)$ is the measured susceptibility, and

$$f_\alpha(y)=\left[\int_0^\infty dx\frac{x^{-\alpha}\sinh y}{1+e^x+2\cosh y}\right]\Bigg/\left[\int_0^\infty\frac{dx\,x^{-\alpha}}{3+e^x}\right]. \tag{4}$$

Thus, for a given α, a plot of $M(H,T)/T\chi(T)$ versus H/T should yield a universal curve for all H and T well below the cutoff J_c. Figure 2 shows such a plot for each of the four samples studied. As can be seen, all the data for each sample at low temperatures ($T<4.2$ K) lie on a

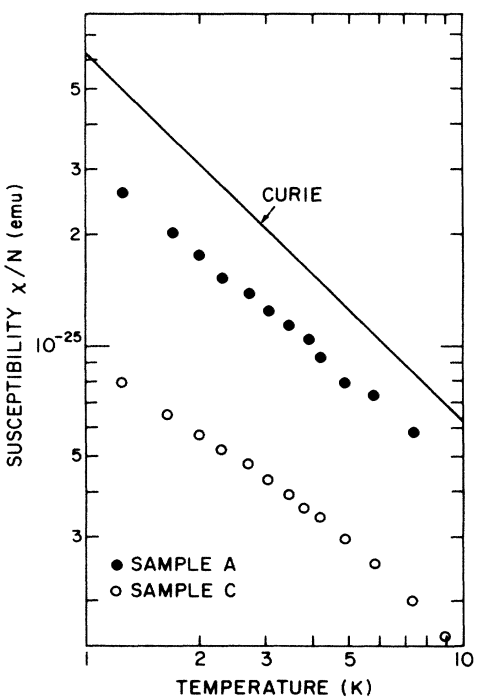

FIG. 1. Double-logarithmic plot of magnetic susceptibility versus temperature for samples A ($N=6.7\times10^{17}$ cm^{-3}) and C ($N=2.4\times10^{18}$ cm^{-3}).

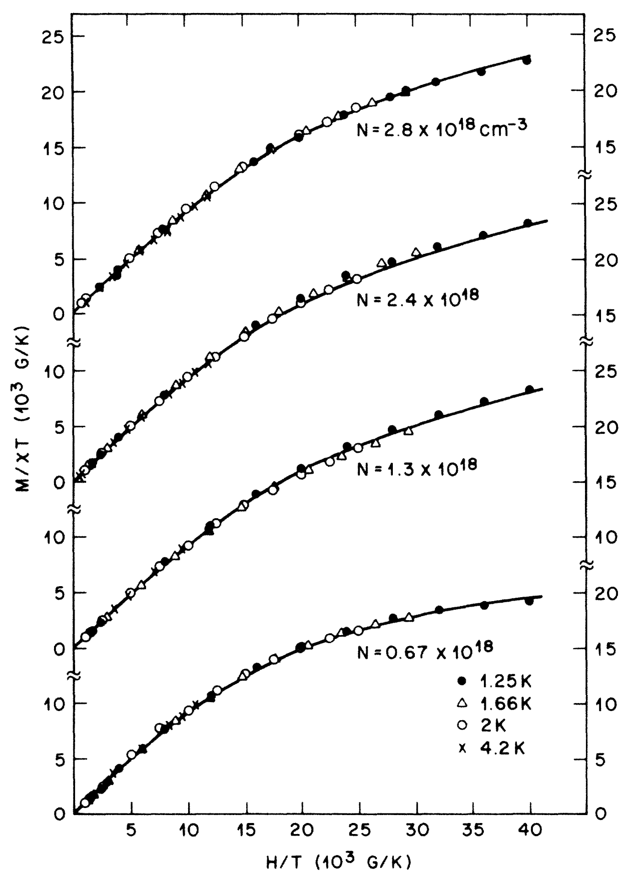

FIG. 2. $M(H,T)/T\chi(T)$ versus H/T for each of the four samples ($N=6.7\times10^{17}$ cm^{-3} to 2.8×10^{18} cm^{-3}) for different temperatures and magnetic fields up to 50 kG. Symbols correspond to the data at the temperatures indicated; solid curves are the theoretical expression [Eq. (4)] based on the picture of hierarchically coupled pairs of spins with a renormalized distribution of exchange.

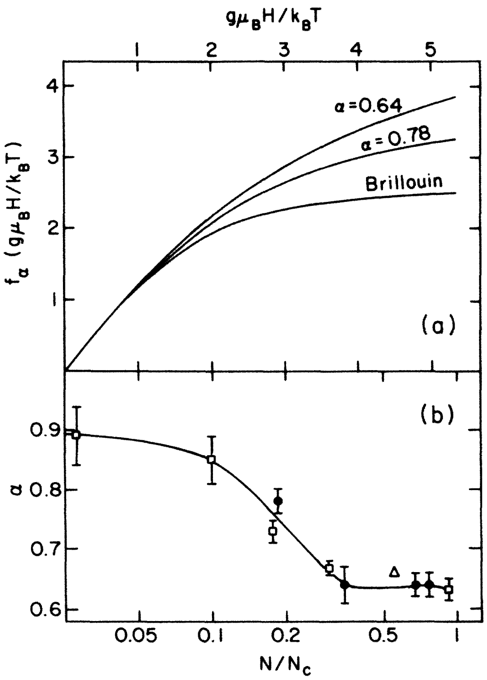

FIG. 3. (a) Plot of $f_\alpha(g\mu_B H/k_B T)$ versus $(g\mu_B H/k_B T)$ according to Eq. (4) for values of $\alpha=0.78$ and 0.64 (corresponding to samples A and C), as well as the free-spin Brillouin function. (b) Values of α deduced from the current experimental data (points), as well as susceptibility measurements to millikelvin temperatures (Ref. 3, squares; Ref. 4, triangles), as a function of phosphorus concentration.

single curve which deviates significantly from the low-field linear behavior. Further, the solid lines drawn in Fig. 2 are curves *calculated* numerically from Eq. (4), for the values of α listed in Table I, which are consistent with the susceptibility data. On the basis of the excellent agreement between the theoretical curves and experimental data, we conclude that the scenario of hierarchically coupled pairs of spins with renormalized interactions, which emerges from the scaling studies of Bhatt and Lee,[9] provides a *quantitative* description of the magnetic properties of the insulating phase.[16]

To contrast the system with free spins, we have plotted in Fig. 3(a) the function $f_\alpha(g\mu_B H/k_B T)$ for the α values for samples A and C and the Brillouin function, which is the corresponding quantity for free spins. As can be seen, the experimental curve does not saturate, and can be clearly differentiated from the free-spin case. The lower part of the figure, Fig. 3(b), plots the values of α as a function of phosphorus concentration from Table I along with values deduced from earlier experiments[3,4] down to millikelvin temperatures. The results are consistent with each other, and approach a concentration-independent value $\alpha \approx 0.65$ at the higher concentrations. We speculate, based in part on the work of Bhatt and Lee,[9] that this is a characteristic signature of the susceptibility in the insulating phase at sufficiently low temperatures where $\chi(T) \ll \chi_C(T)$, the Curie value.

We would like to mention in passing that a scaling similar to Eq. (3) is observed when we analyze our earlier data[10] for the acceptor system Si:B, except the function $f_\alpha(H/T)$ is expected to be different because the acceptor ground state has an angular momentum $j=\frac{3}{2}$. This universality of the scaling form seems to imply a universal nature of the magnetic structure of the insulating phase, though measurements to lower temperatures are necessary to decide that issue. Earlier susceptibility measurements[3,4] of the donor system down to millikelvin temperatures have pointed to a $T=0$ critical point. Our measurements, though admittedly restricted to higher temperature, show that the nonlinear magnetization obeys a scaling form given by Eq. (3), in which the scaling variable is (H/T), consistent with $T_c=0$.

Our measurements show that even for concentrations as high as 2.8×10^{18} cm^{-3} (about 20% below the metal-insulator transition density), the temperature and field dependence of the magnetization at temperatures $T<4$ K is quantitatively fit by the localized-electron-spin Hamiltonian. This is in contrast to a recent suggestion by Jerome *et al.*[17] that for $N \gtrsim 1\times10^{18}$ cm^{-3} and $T\sim1$ K in Si:P, the electrons are delocalized. We too have seen departures from insulating behavior to diamagnetic[18] metallic behavior, but at higher temperatures and concentrations. An analysis of the crossover to metallic behavior and comparison with the work of Ref. 17 is underway and will be reported elsewhere. Clearly, a quantitative characterization of the insulating behavior is a prerequisite for any systematic analysis of the behavior as the transition is approached. The present measurements of the magnetization for a range of temperature and fields have helped accomplish that goal. They constitute the first experimental check, free of parameters, that scaling studies[9] provide a quantitatively accurate description of the magnetic behavior of the insulating phase.

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