Susceptibility of Si:P across the metal-insulator transition. II. Evidence for local moments in the metallic phase

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For donor concentrations on both sides of the metal-insulator transition, the paramagnetic component of the susceptibility of Si:P at low temperature is described by a power law $\sim T^{-\alpha}$, modified by a term which represents thermal activation to higher-energy states which are magnetically inert. For insulating samples, the power law is associated with the random Heisenberg antiferromagnet. The smooth continuation of the same behavior onto the metallic side of the transition suggests that local moments exist in metallic Si:P. We further suggest that this may imply that the exchange interaction between the magnetic moments does not undergo any abrupt or major change in character as the transition is crossed, in spite of the appearance of delocalized electrons.

INTRODUCTION

The zero-temperature conductivity of doped semiconductors such as Si:P has been shown to change very sharply, albeit not discontinuously, as the metal-insulator transition is crossed by varying the dopant concentration[1] or by applying uniaxial stress.[2] In contrast, the magnetic response of these materials appears to change smoothly over the same range of concentration and, to date, no evidence of a transition has been detected in measurements of the magnetic susceptibility. An anomalous and unexplained temperature dependence has been known[3-9] for some time to exist for just-metallic samples down to 1 K. Recent ESR measurements of Paalanen *et al.*[10] and of Ikehata and Kobayashi,[11] although their results differ substantially in detail, nevertheless establish beyond doubt that the susceptibility of just-metallic Si:P continues to rise sharply even as the temperature is reduced to 10 or 20 mK.

One explanation of this behavior, recently invoked by Gan and Lee[12] to account for the anomalous NMR relaxation observed by Paalanen *et al.*,[13] is that local magnetic moments are present in just-metallic doped semiconductors due perhaps to singly occupied electron states below the Fermi energy,[14] or due to clusters[12] with an odd number of electrons. Alternatively, recent work of Castellani, Lee, and co-workers[15] based on work by Finkelstein[16] suggests that the strong temperature dependence of the susceptibility of samples with concentrations just above the transition arises from a slowing down of spin diffusion due to electron-electron interactions in the presence of strong disorder.

In this article we use the analysis of high-temperature susceptibility data discussed in the preceding companion paper to extract and examine in detail the paramagnetic component at low temperature or, in other words, the term which one normally associates with interacting moments in the insulating material. We show that the data are qualitatively similar on both sides of the transition, and fit the same expression over our available range of temperature. We present plausibility arguments which can account for the behavior in the metallic phase in terms of singly occupied states below the Fermi level, or clusters containing an odd number of electrons.

EXPERIMENTAL RESULTS AND DISCUSSION

As described in more detail in the preceding article, the susceptibility was measured between 1.25 and 300 K for nine samples of Si:P with donor concentrations spanning the metal-insulator transition which occurs at 3.5×10^{18} cm^{-3}. In units of 10^{18} cm^{-3}, the concentrations[17] are 0.67, 1.26, 2.42, 2.76, 3.4, 3.61, 3.83, 4.27, and 4.6. The susceptibility of the host silicon crystal was determined from measurements on a zone-floated sample and was subtracted to yield the susceptibility associated with the phosphorus impurities. The donor susceptibility per phosphorus atom is plotted in Fig. 1 as a function of the logarithm of the temperature.

Based on the discussion and analysis of the preceding paper, the dotted curve shown in Fig. 1 represents the baseline relative to which one would extract the low-temperature contribution which is normally associated with local moments in insulating material, and relative to which one should examine the anomalous temperature dependence in just-metallic samples. One should note that of the six curves plotted for concentrations ranging from 30% below to 30% above the critical concentration, the two steeper curves are for insulating material and the remaining four are on the metallic side of the transition.

The susceptibility relative to the dotted baseline of Fig. 1 is plotted as a function of temperature on a double logarithmic plot in Fig. 2. The most striking feature of these data is the smooth and continuous progression across the transition. These curves show no evidence at all that a transition has taken place, and are qualitatively similar in every way.

Measurements to very low temperatures by Andres *et al.*,[18] and subsequently confirmed by others,[9,19] have established that the susceptibility of insulating Si:P obeys

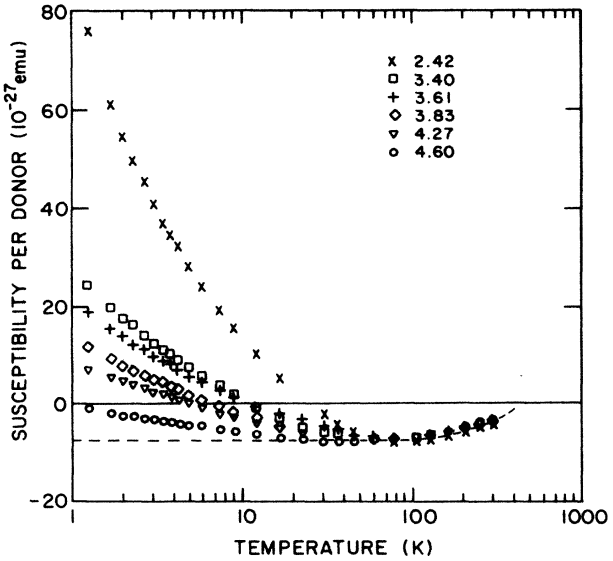

FIG. 1. Susceptibility per donor as a function of temperature for samples with concentrations indicated in units of 10^{18} cm^{-3}. The dotted curve is the baseline relative to which the susceptibility at low temperatures is examined.

the approximate relation $\chi \sim T^{-\alpha}$. This is attributed to the behavior of the random Heisenberg antiferromagnet (RHA), which is an ensemble of localized spins distributed randomly in space, that interact antiferromagnetically with each other via short-range direct Heisenberg exchange. For hydrogenic wave functions (which are appropriate envelope wave functions for donors in silicon), the exchange energy is given by[20]

$$J(r)=J_0(r/a_B)^{5/2}\exp(-2r/a_B)\ , \tag{1}$$

where r is the distance between the localized spins, and a_B is the Bohr radius. The exponential dependence of the exchange on the separation between the interacting spins, coupled with the random spatial distribution of those spins, gives rise to an extremely broad distribution of exchange energies. It is this broad distribution of exchange energies of the RHA which distinguishes it from ordered quantum antiferromagnets, and gives rise to its unique low-temperature properties. In considering the magnetic response of the RHA, the distribution of exchange can be taken into account by calculating the magnetic response of pairs of spins, coupled hierarchically in order of decreasing exchange energy. This has been done by considering the distribution of spin pairs as a function of increasing separation (and therefore decreasing exchange), $P(r)$,[21–23] or directly as a function of decreasing exchange, $P(J)$.[24] The latter method, as implemented with renormalized interactions in the scaling calculation of Bhatt and Lee,[24] has proven to be particularly successful. It shows, in accordance with experimental findings, that the low-temperature susceptibility of Si:P in the insulating phase is well described by the relation

$$\chi(T)=nA(n)T^{-\alpha}\ , \tag{2}$$

where $0<\alpha<1$, and both A and α decrease with increasing concentration.

The data of Fig. 2 clearly follow $AT^{-\alpha}$ behavior in the restricted temperature range up to 4 K, irrespective of whether the samples are insulating or metallic. One does expect this for the insulators (the three top curves), and one also expects departures from this simple behavior at higher temperatures due to thermal activation and the consequent disappearance of some of the localized moments. This would involve activation of electrons to nonmagnetic states, such as extended states at energies above the mobility edge or, more likely, to doubly occupied states at the Fermi level.[14] A simple model which incorporates the $AT^{-\alpha}$ behavior of the random Heisenberg antiferromagnet and a reduction in the number of electron moments by thermal depopulation characterized by an activation energy Δ is given by

$$\begin{aligned}\chi(T)&=nAT^{-\alpha}[1-\exp(-\Delta/k_BT)]\\&=nAT^{-\alpha}[1-f(T)]\ .\end{aligned} \tag{3}$$

A fit to this expression yields the parameters A, α, and Δ listed for the insulating samples in the first three rows of Table I. The quality of the fit is exhibited in Fig. 3(a),

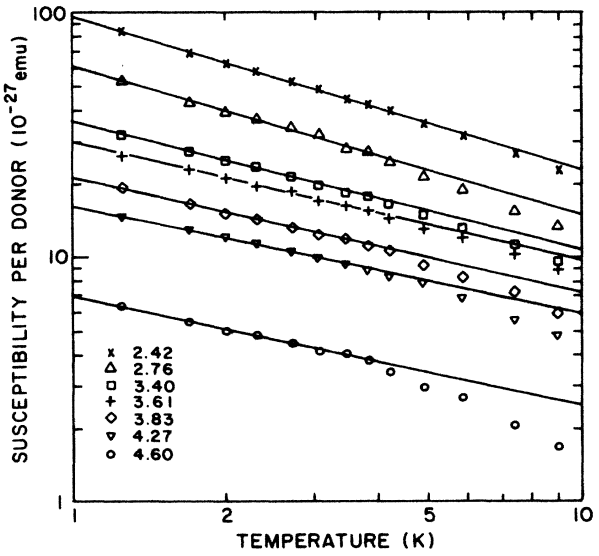

FIG. 2. Double-logarithmic plot of the susceptibility per donor versus temperature. For each sample, the straight line is a least-squares fit of Eq. (2) to the data below 4 K.

TABLE I. Values of the parameters A, α, and Δ as deduced from fits to Eqs. (2) and (3).

Sample	Concentration (10^{18} cm^{-3})	A (10^{-27} emu)	α (± 0.02)	Δ (K)
D	2.42	96.2	0.62	26
E	2.76	60.5	0.60	13
F	3.40	36.1	0.53	18
G	3.61	29.9	0.49	17
H	3.83	21.3	0.48	16
I	4.27	16.3	0.45	17
J	4.60	6.9	0.44	13

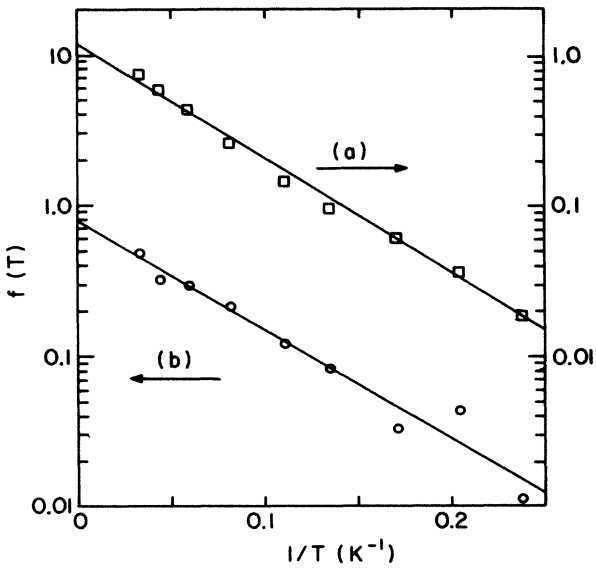

FIG. 3. Plot of $\log_{10} f(T)$ vs $1/T$ for: (a) an insulating sample ($n = 3.40 \times 10^{18}$ cm^{-3}); (b) a just-metallic sample ($n = 3.61 \times 10^{18}$ cm^{-3}). The parameter Δ of Eq. (3) is determined from the slope of the least-squares best-fit line.

where the function $f(T)$ of Eq. (3) is plotted on a logarithmic scale as a function of the inverse temperature for the sample containing 3.4×10^{18} cm^{-3}.

It is striking that the behavior of the just-metallic samples is in every way similar to that of the insulating material. Figure 2 shows that a $T^{-\alpha}$ term apparently accounts correctly for the behavior below 4 K, and the susceptibility appears to deviate at higher temperature in much the same way. This similarity prompted us to attempt to fit the model of Eq. (3) to the metallic material, and the resulting parameters are listed in rows 4–7 of Table I. Figure 3(b) shows $f(T)$ as a function of T for a typical just-metallic sample, and shows that a good fit does result from this procedure. It is interesting to note that the values for the activation energy Δ are quite reasonable at ~ 16 K, or ~ 1 meV, and show no abrupt change at the transition.

We have thus demonstrated the apparent validity of Eq. (3) for both insulating and just-metallic Si:P. The meaning of this model is clear for concentrations below the transition. Thus, the $T^{-\alpha}$ term originates with the random antiferromagnet which has been clearly established to be relevant for the insulators by experiments over an extended range of temperature[18,19] and measurements of the nonlinear magnetization.[9] The remaining temperature dependence is due to the depletion of electronic moments by activation to nonmagnetic states. On the other hand, it is not clear why the same expression applies so well for the metallic side of the transition.

A possible explanation for the behavior of the susceptibility of the just-metallic samples at low temperature, as has been advanced to account for NMR data,[12] is that the temperature dependence is caused by the persistence of local moments into the metallic phase. If this is the case, then Δ is a measure of the energy separation between the local moment states and the Fermi level. The question still remains, however, as to why the susceptibility continues to vary as $T^{-\alpha}$ for $n > n_c$. A necessary and central ingredient in the model which explains the $AT^{-\alpha}$ behavior for insulators is the presence of an extremely broad distribution of exchange energies. We suggest that if sites exist in the metallic phase which can support a net local moment (either singly occupied sites, or clusters with an odd number of electrons), then it is plausible that interactions between the moments exist which are sufficiently short range in nature to give rise to a magnetic response at low temperatures that is similar to that of the random Heisenberg antiferromagnet. The exchange interaction between the local moments, in general, will not be given by the simple expression of Eq. (1). Even if the direct exchange between the localized states varies as $\exp(-2r/a)$, where a is the localization length, effects of indirect exchange via delocalized electrons may result in a very complicated net exchange interaction. Nevertheless, it is possible that the interaction between local moments will still be a strong function of the distance between them.[12] In addition, the spatial distribution of the local moment sites is expected to be statistically random, at least for the longer length scales. Thus, even in the metallic phase, it is possible to retain the broad distribution of exchange energies that is key to the magnetic behavior of the random Heisenberg antiferromagnet. This distribution of exchange would result, just as in the insulating phase, in a susceptibility that varies as $T^{-\alpha}$.

Inasmuch as the details of the behavior of the RHA, such as the value of α, may be expected to depend on the detailed form of the exchange interaction, the apparently monotonic progression of the $AT^{-\alpha}$ form across n_c could possibly arise from a finite-temperature effect, namely, the effect of delocalized electrons which exist on both sides of n_c at $T \geq 1$ K.[25] The presence of these electrons could ensure that the form of the net exchange interaction between local moments varies smoothly across n_c, giving rise to a smooth variation in the $AT^{-\alpha}$ behavior. If this is the case, then one would expect that measurements at sufficiently low temperatures ($T \ll 1$ K) would show an abrupt change in the susceptibility near n_c. However, we find that if we use an appropriate baseline, as discussed in detail in the preceding companion paper, to analyze recent ESR measurements for $n > n_c$,[10] and earlier static measurements for $n < n_c$,[18] that the $AT^{-\alpha}$ behavior is continuous across n_c for temperatures down to 30 mK. In this sense, it would seem that the exchange interaction between the local moments, if they do exist in metallic samples, is little affected by indirect exchange via delocalized electrons, for concentrations near n_c.

SUMMARY AND CONCLUSIONS

Application of the analysis which is presented and discussed in detail in the preceding paper has allowed a separation of the low-temperature paramagnetic contribution which is normally associated with interacting local moments on the insulating side of the transition, and whose persistence onto the metallic side is not under-

stood. A quantitative fit is obtained to a power law $\sim T^{-\alpha}$, modified by a term $[1-\exp(-\Delta/kT)]$ which represents the disappearance of local moments through activation to nonmagnetic states. This behavior is well understood for insulating material, where short-range exchange among randomly placed moments results in the wide distribution of coupling energies which leads to the $\sim T^{-\alpha}$ behavior of the random Heisenberg antiferromagnet. The smooth continuation of the same behavior across the transition is surprising, and may imply that local moments exist in metallic Si:P. Furthermore, by introducing an agent which allows for indirect exchange, one might expect that the appearance of charge carriers would alter the character and range of the exchange interaction between the local moments. We argue instead that the observed $\sim T^{-\alpha}$ form of the susceptibility of metallic Si:P implies that the nature of the exchange interaction varies smoothly across the transition and that the distribution of exchange energies remains broad.

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