ESR Studies of Compensated Si:P,B near the Metal-Insulator Transition

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We have studied the ESR properties of three compensated n-type Si:P,B samples near the metal-insulator transition covering the low-temperature regime from 30 mK to 10 K. We find that both the susceptibility and the ESR linewidth increase dramatically as the temperature is lowered, and in the metallic Si:P,B samples the susceptibility increase is more than in similar uncompensated Si:P samples. We compare results for the insulating phase with numerical calculations, then discuss the metallic region in light of various theoretical models for the low-temperature thermodynamic behavior.

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In a simple picture of the metal-insulator (MI) transition the low-temperature excitations in the insulating phase involve localized electrons, while in the metallic phase they involve itinerant electrons. Within this picture one expects that the magnetic properties of these two phases are drastically different—the Fermi-liquid-like metallic phase is expected to have a small, nearly temperature-independent susceptibility, while the insulating phase should have weakly interacting local magnetic moments with a large Curie-type susceptibility. However, numerous ESR [1–4], NMR [5–8], and thermodynamic measurements [9–15] have shown that this simple picture is inadequate, at least in the extensively studied case of phosphorus-doped silicon (Si:P). The magnetic properties of Si:P are found to be surprisingly similar in the metallic and insulating phases near the MI transition and show characteristics of local magnetic moments. This observation suggests that local magnetic moments persist into the disordered metallic phase and dominate the low-temperature thermodynamic properties [1,9,12–15] as well as the spin dynamics [7,8]. Milovanovic, Sachdev, and Bhatt [16] have numerically solved a disordered Hubbard model with randomly placed centers within a self-consistent-field approximation and found local moment instabilities at a few percent of the sites in the metallic phase, in rough agreement with the experimental result in Si:P. A Fermi-liquid theory incorporating the local moments has been formulated by Sachdev [17]. Bhatt and Fisher [18] have very recently given arguments that these local moments persist down to the lowest temperatures, notwithstanding the Kondo effect or the local-moment–local-moment interaction. However, these theoretical approaches do not address all aspects of the effects of local moments in the disordered metal, e.g., how they affect the critical conductivity behavior at the MI transition.

Attempts to clarify the above issues by comparing compensated and uncompensated semiconductors near the MI transition have raised more questions. Recent NMR measurements in boron-compensated Si:P [19] are in qualitative agreement with similar earlier measurements in uncompensated Si:P [7] and are interpreted in terms of interactions of ^{29}Si nuclei with local moments. However, the critical conductivities of Si:P [20] and Si:P,B [21,22] behave quite differently and one finds a steeper conductivity onset for the uncompensated case. Fitting the conductivity near n_c by the critical form $\sigma \propto (n-n_c)^{\mu}$, the critical exponent is $\mu = 0.5$ and 0.9 for Si:P and Si:P,B, respectively.

In this Letter we report low-temperature ESR measurements for three Si:P,B samples in the vicinity of the MI transition and compare both their spin susceptibility and ESR linewidth with previously determined values for uncompensated samples [4]. The three samples had electron concentrations of $(2.6, 4.9, \text{and } 8.0)\times 10^{18}$ cm^{-3} and compensation values, defined as the ratio of acceptor to donor doping densities, of 0.6, 0.5, and 0.5, respectively. The electron density n was determined from the room-temperature Hall coefficient and the compensation from the value of n and the donor doping density N_D measured by neutron activation. This procedure is described in detail elsewhere [21,22]. The samples were in the form of a stack of ten thin slabs, $12\times 9\times 0.4$ mm each, cut thinly to minimize eddy-current losses. The doping profile of each slab was measured at room temperature with a resistivity technique and found to span $\pm 7\%$. This is somewhat more than the $\pm 2\%$ density variation of the uncompensated Si:P samples used in the earlier control experiment [4]. However, the density variation is not expected to significantly affect the susceptibility, which is found to vary gradually as n goes through n_c. The ESR techniques in this experiment and in the earlier Si:P study [4] were identical. Again the ^{29}Si NMR signal was used for calibrating the ESR spectrometer in absolute susceptibility units. We estimate an absolute accuracy of $\pm 20\%$ for our susceptibility values.

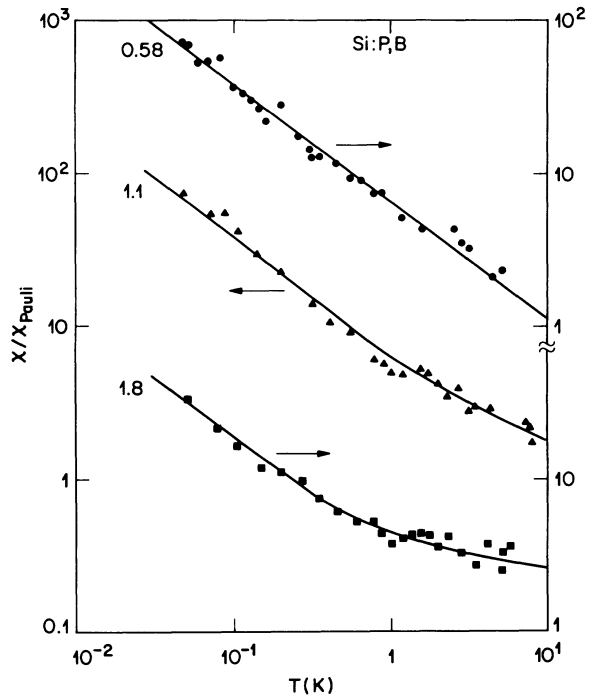

FIG. 1. Temperature dependence of normalized susceptibility χ/χ_{Pauli} of three Si:P,B samples with different normalized electron densities, $n/n_c = 0.58$, 1.1, and 1.8. Solid lines through data are a guide to the eye.

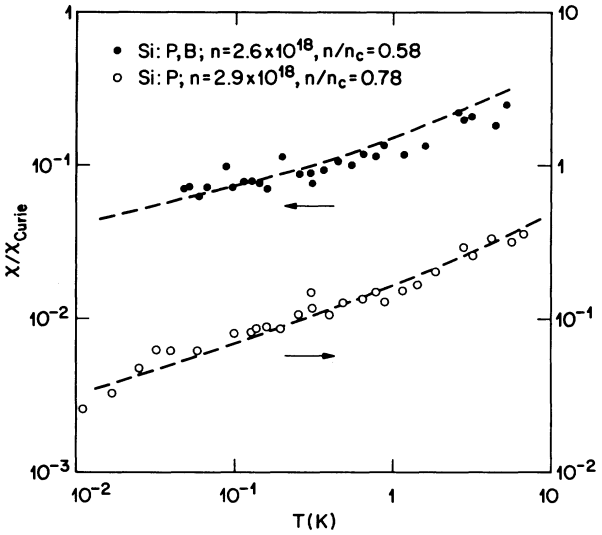

FIG. 2. Comparison of measured normalized susceptibilities χ/χ_{Curie} (circles) of insulating Si:P and Si:P,B samples with theoretical calculation (dashed lines) described in the text.

In Fig. 1 we show the enhancement of the susceptiblity χ (relative to $\chi_{Pauli} = 3n\mu_B^2/2k_BT_F$) as a function of temperature for all three compensated samples. These data are qualitatively similar to the uncompensated Si:P data [4], i.e., the susceptibility increases towards lower temperatures approximately as a power law $\chi \propto T^{-\alpha}$. As shown in Fig. 2, this temperature dependence is observed over our entire temperature range for insulating samples. In this figure we have compared the normalized susceptibilities $\chi/\chi_{Curie} \propto T^{1-\alpha}$ ($\chi_{Curie} = n\mu_B^2/3k_BT$) of compensated and uncompensated Si:P and find, using least-squares fits, that the exponent $\alpha = 0.75 \pm 0.05$ for Si:P,B is somewhat larger than the value of 0.62 ± 0.03 for Si:P. The dashed lines in Fig. 2 represent a quantitative theoretical calculation of the susceptibility using no adjustable parameters as explained below.

The susceptibility of uncompensated Si:P for $n < n_c$ was explained by Bhatt and Lee (BL) using a quantum spin-$\frac{1}{2}$ random Heisenberg antiferromagnetic Hamiltonian [23]. We have performed a similar computer calculation of the susceptibility of a model appropriate for a *compensated* doped semiconductor deep in the insulating phase. The model consists of distributing donor and acceptor sites at random in a 3D continuum. The negatively charged acceptors provide a fixed random Coulomb potential while the electrons are allowed to occupy the donor sites with the lowest-self-consistent energies, neglecting quantum-mechanical (hopping, exchange) terms. The ground state is tested for stability against one- and two-electron hops. The susceptibility is then calculated in the manner of BL using the antiferromagnetic spin-$\frac{1}{2}$ Heisenberg exchange Hamiltonian:

$$H = \sum_{i,j} J(r_{ij}) \mathbf{S}_i \cdot \mathbf{S}_j , \qquad (1)$$

where the sum over i and j includes the electron occupied donor sites. For the exchange constant we use the asymptotic hydrogenic result [24] $J(r) = J_0(r/a)^{5/2}\exp(-2r/a)$, where $a = 16$ Å ($n_c^{1/3}a = 0.25$ for Si:P) and $J_0 = 140$ K. The high-temperature curvature of the theoretical lines in Fig. 2 is due to the asymptotic formula chosen for $J(r)$. This formula underestimates J at small r and these are the values relevant at high temperatures. A theoretical estimate of the exponent α, obtained from the low-temperature behavior of the dashed lines in Fig. 2, is found to be slightly larger in the compensated case. This is due to the rearrangement of the electron occupied donor sites, which results for the compensated case in a distribution differing from the Poisson distribution at short distances. In summary, the theory with no adjustable parameters is in remarkable agreement with the experimental results for the insulating phase.

The difference between Si:P and Si:P,B is more dramatic on the metallic side of the MI transition—the susceptibility enhancement is unexpectedly large in Si:P,B at the lowest temperatures even for the very metallic sample $n/n_c = 1.8$. As shown in Fig. 3, comparing Si:P and Si:P,B samples with similar values of $n/n_c \approx 1.1$, the compensated system shows a factor of 3 to 5 larger local moment fraction than the uncompensated one for $T < 0.1$ K. This is in contrast to the theoretical results of Milovanovic, Sachdev, and Bhatt [16] who find for the disordered Hubbard model that the fraction of local moments

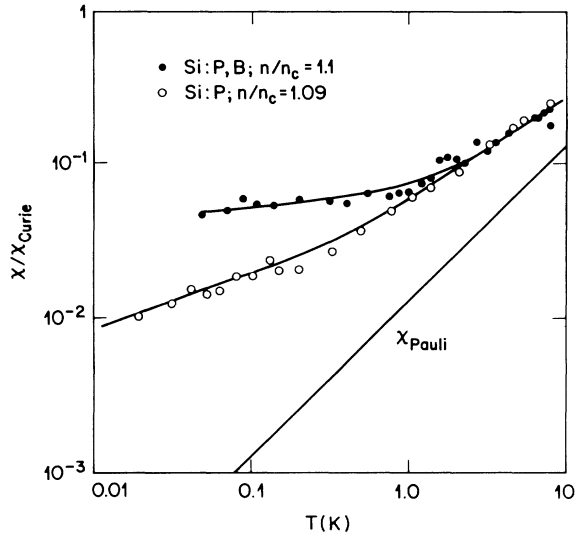

FIG. 3. Comparison of normalized susceptibilities χ/χ_{Curie} of uncompensated and compensated Si:P in the metallic regime.

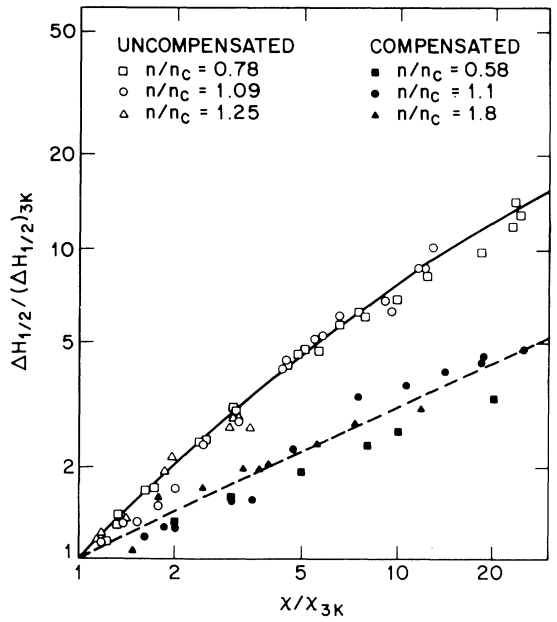

FIG. 4. Log-log plot of ESR linewidth vs susceptibility for uncompensated and compensated Si:P. Both quantities have been normalized by their values at a temperature $T=3$ K.

decreases at carrier concentrations away from half filling of the band. One possible cause of this discrepancy is the presence of random fields in the experimental system arising from the negatively charged acceptors and positively charged donors. These are not included in the disordered Hubbard model calculation. Another possibility is the emergence of ferromagnetic exchange interactions due to the departure from half filling.

Assuming that the increased χ is due to an increase in the number of local moments rather than to ferromagnetic interactions and further that the strength of spin-flip scattering of itinerant electrons increases with the number of local moments we would expect the spin-flip rate τ_s^{-1} to be higher in compensated samples. This increased spin-flip rate could account for the critical-exponent difference between uncompensated Si:P and compensated Si:P,B, *provided* τ_s^{-1} is just below or equal to $k_B T/\hbar$ in Si:P within the T range from 3 to 100 mK. With this particular value of τ_s^{-1}, the higher spin-flip rate of Si:P,B (which should be checked by the magnetoresistance measurements) could put it (but not Si:P) into the spin-flip universality class [25,26]. However, within the same scaling approach for the spin-flip universality class, χ is not expected to diverge as T tends to zero, in contrast to the experimental evidence in Si:P,B. Thus, the one-component (i.e., the long-wavelength mode) scaling theory requires a coincidence of parameters to explain the critical-conductivity differences between Si:P and Si:P,B, and even then needs local moments to explain the susceptibility of Si:P,B.

The ESR line is broadened by the hyperfine interaction between the electrons and the P nuclei (hyperfine interaction with the nuclei of negatively charged B^- acceptors is very small due to the Coulomb repulsion) but motionally narrowed by the rapid electron spin motion. In the Fermi-liquid picture the spin motion is diffusive and can be tied to the Fermi-liquid properties of the sample [27]. In the localized moment picture the low-temperature linewidth is dominated by the fast spin-lattice relaxation of the local moments which then relax the itinerant electrons through the exchange processes [28]. The localized moments form clusters and spin motion via exchange processes determines their motional line narrowing [29]. In previous measurements of the uncompensated samples [4] the linewidth was observed to increase towards lower temperatures with the increase apparently proportional to the susceptibility increase. This simple proportionality is also predicted in the lowest-order disordered Fermi-liquid model [4,27] and it was argued that this observation supports the validity of the scaling theory description.

We find a broader ESR line in the compensated samples than in the uncompensated samples with similar values of n/n_c. The extra linewidth of Si:P,B samples might be expected, given the electron repulsion away from the B^- ions. This leads to an increase of both the electron wave function and the hyperfine interaction at P^+ sites. In Fig. 4 we have plotted the normalized linewidth as a function of normalized susceptibility for both the compensated and uncompensated Si:P. The normalization was done with the $T=3$ K values of the linewidth $(\Delta H_{1/2})_{3\,\text{K}}=0.9$, 1.1, and 1.6 G (0.45, 0.45, and 0.60 G) and the susceptibility $\chi_{3\,\text{K}}/\chi_{\text{Pauli}}=3.0$, 3.2, and 3.4 (4.5, 2.9, and 2.5) in the compensated (uncompensated) low-, medium-, and high-density samples, respectively. We find several interesting features in the data of Fig. 4. First, for the compensated samples the linewidth is *not*

proportional to the susceptibility. Second, the similar results for the insulating and metallic samples suggest that the same mechanism, presumably the local moment relaxation, is causing the line broadening in both phases.

A zeroth-order estimate of the linewidth within the localized spin model, assuming that the linewidth comes from the spin-lattice relaxation of the local moments, can be obtained as follows. The half-width $\Delta H_{1/2} \approx 1/T_1 \approx \frac{1}{2}\gamma_e H_{\rm hf}^2 \tau_c$, where the hyperfine field $H_{\rm hf}=21$ G [30] and τ_c is the correlation time for the hyperfine interaction when a spin excitation diffuses within a given cluster. The correlation time τ_c is expected to scale inversely with the characteristic exchange interaction J, which has a wide distribution in our disordered system. If we make the drastic assumption that the distribution of J values (including further neighbor interactions) can be described by a single parameter α, we can obtain τ_c by averaging J^{-1} over the distribution $P(J)=J^{-\alpha}$ from the hyperfine energy $\hbar\gamma_e H_{\rm hf}$ to the thermal energy $k_B T$. This will give a linewidth of

$$\Delta H_{1/2} \propto H_{\rm hf}(\hbar\gamma_e H_{\rm hf}/k_B T)^{1-\alpha}. \quad (2)$$

In qualitative agreement with Fig. 4, Eq. (2) predicts a linewidth which for $\alpha > 0.5$ has a weaker temperature dependence than the temperature dependence of the susceptibility, $\chi \propto T^{-\alpha}$. An exact proportionality between the linewidth and susceptibility is accidentally achieved when $\alpha = \frac{1}{2}$. Computer calculations such as those done for the uncompensated system [29] are necessary to check the merits of this rather simplified argument.

In conclusion, our ESR studies of boron-compensated Si:P near the MI transition support the two-fluid model of localized spins and itinerant electrons in the metallic phase. The interaction between the two "fluids" leads to the spin-flip scattering of the itinerant electrons. The strength of this spin-flip scattering may affect the critical conductivity exponent. In the insulating phase the susceptibility is found to be in quantitative agreement with the computer simulations of a generalization of the Poisson distributed random antiferromagnet to take into account the effect of compensation.

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