

## Spin Dynamics of Nearly Localized Electrons

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(Received 16 June 1986)

The electron-spin-resonance signal of metallic Si:P near the metal-insulator transition has been measured down to a temperature of 30 mK. The paramagnetic spin susceptibility and the resonance linewidth are found to increase sharply with decreasing temperature. We argue that these effects are due to the enhancement of spin fluctuations, and the accompanying slowing down of spin diffusion near the metal-insulator transition. The results are compared with the predictions of recent theories for the susceptibility and linewidth of disordered interacting-electron systems.

PACS numbers: 71.30.+h, 72.15.Cz, 76.30.Pk

Studies of uncompensated phosphorus-doped silicon (Si:P) have yielded a great deal of information about the properties of electrons in disordered systems.<sup>1</sup> Transport measurements have demonstrated that the disorder-enhanced interactions are important in determining the magnetic field and temperature dependence of the conductivity in the metallic phase.<sup>2</sup> The behavior close to the metal-insulator transition at density  $n_c$  is not very well understood. It has been suggested<sup>3</sup> that near  $n_c$ , uncompensated Si:P could be viewed as a strongly correlated liquid with disorder, in which the main effect of electron correlations is to strongly enhance the local spin fluctuations. However, a quantitative generalization of the Brinkman-Rice<sup>4</sup> calculation to include disorder or finite temperatures has not yet been accomplished. Recent progress in theories<sup>5,6</sup> which take disorder into account to lowest order (while treating interactions to all orders) also suggests the importance of spin fluctuations near  $n_c$  and the scaling behavior is found to be strongly dependent upon external perturbations that couple to the electron spin (e.g., magnetic fields, spin-flip and spin-orbit scattering). Unfortunately since the scaling equations flow to strong coupling, the connection with experiment near  $n_c$  is not clear.

Information about spin fluctuations can be obtained from nuclear magnetic resonance (NMR) and electron-spin resonance (ESR). Recent NMR relaxation results were argued to signal the existence of slow spin fluctuations near the metal-insulator transition at millikelvin temperatures.<sup>7</sup> While previous ESR measurements<sup>8,9</sup> have indicated that the spin susceptibility in the metallic phase is somewhat temperature dependent well below the degeneracy temperature  $T_F \approx 100$  K, the ESR linewidth was found to level off at a constant value below 10 K. In this Letter, we report an ESR study of Si:P just on the metallic side of the metal-

insulator transition down to 30 mK. We find a strong, temperature-dependent susceptibility enhancement and ESR line broadening at the lowest temperatures, qualitatively as found on the insulating side.<sup>10</sup> Similar behavior has recently been reported by Ikehata and Kobayashi<sup>11</sup>; however, our susceptibility data differ quantitatively from their published results, and no quantitative results for the linewidth are given in Ref. 11. We determine the electron-spin diffusivity and spin-flip scattering rate from the linewidth, and compare the results with predictions of recent theories.<sup>5,6,12</sup>

Two samples of Si:P with P concentrations  $n/n_c = 1.09$  (Sample A) and 1.25 (Sample B) were studied. The samples were in the form of a stack of five thin slabs,  $10 \times 15 \times 0.8$  mm<sup>3</sup> each, to minimize eddy-current losses. The doping density  $n$ , determined from the conductivity of several adjacent test samples, was found to be macroscopically homogeneous within  $\pm 2\%$ . The ESR and the <sup>29</sup>Si NMR signals were observed with a conventional  $Q$ -meter continuous-wave spectrometer operating at 18.4 MHz. The latter allowed an absolute calibration of the spectrometer from the known Curie susceptibility of <sup>29</sup>Si nuclei.<sup>13</sup> The weak resonance signals were recovered by signal averaging of about 200 static magnetic field sweeps over the absorption line. From the Kramers-Kronig relation, the static susceptibility is an integral of the dynamic susceptibility as a function of frequency. For Lorentzian lines (as well as for narrow resonance lines of other shapes) this can be shown to be equivalent to an integral of the ESR signal as a function of the static magnetic field.<sup>13</sup> This was applicable to the data above 30 mK reported here. It is important to note that we measure only the paramagnetic spin susceptibility in contrast to static techniques which measure a sum of both the spin and the diamagnetic contributions.

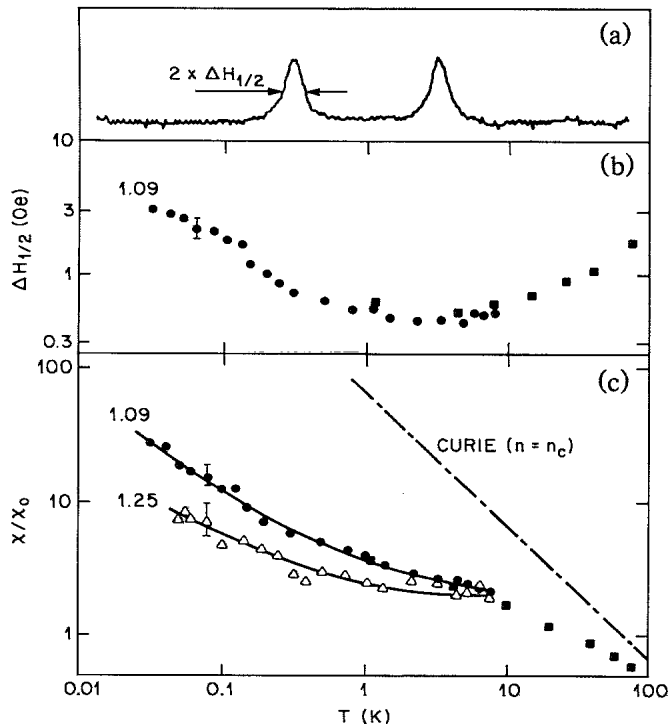


FIG. 1. (a) A typical ESR spectrum—sample A, 200 mK; (b) temperature dependence of the linewidth of sample A (circles,  $n/n_c = 1.09$ ) at 18.4 MHz, and of an  $n \approx n_c$  sample of Ref. 8 (squares) at 9.1 GHz; (c) spin susceptibility  $\chi$ , normalized to the Pauli value for samples A (circles) and B (triangles,  $n/n_c = 1.25$ ) compared with the sample from Ref. 8 (squares).

A typical ESR spectrum is shown for Sample A in Fig. 1(a). The linewidth (half width at half maximum) determined from such spectra as a function of temperature for Sample A is shown in Fig. 1(b). Qualitatively similar results are found in Sample B. Our linewidths agree quite well with previous measurements at high temperature.<sup>8,9,14</sup> Figure 1(c) shows the enhancement of the spin susceptibility  $\chi$  (relative to the Pauli value,  $\chi_0 = 3n\mu_B^2/2k_B T_F$ ) for both samples as a function of  $T$ . While these measurements are in qualitative agreement with other data,<sup>8,9,11</sup> the absolute values differ considerably, presumably because of calibration difficulties in previous experiments due to low signal levels. Agreement (within  $\approx 10\%$ ) is found only with the high-temperature results of Quirt and Marko.<sup>8</sup>

We find several interesting qualitative features in the susceptibility data. At temperatures above 30 K the susceptibility approaches the Curie law. Well below the Fermi temperature in a noninteracting metal we would expect a temperature-independent susceptibility. Instead we find no evidence of susceptibility saturation down to  $T/T_F \approx 3 \times 10^{-4}$ , and the susceptibility of the sample closer to the metal-insulator transi-

tion increases to a value more than an order of magnitude greater than the Pauli spin susceptibility  $\chi_0$ . This is qualitatively as expected from the theories of Finkelstein<sup>5</sup> and Castellani *et al.*<sup>6</sup> as well as from the Brinkman-Rice<sup>4</sup> picture. Further, the susceptibility and linewidth appear to cross over smoothly to the behavior found on the insulating side of the transition (where the system has been modeled as an amorphous antiferromagnet<sup>15</sup>), and so an explanation along these lines is not ruled out. Unfortunately none of these theories, as they stand, provides a *quantitative* comparison with experiment.

The linewidth measures the longitudinal spin fluctuations at low frequencies and can be expressed formally in terms of Kubo formulas.<sup>16</sup> There are two sources of spin dephasing leading to the linewidth: The first, which dominates at high temperatures, arises from the spin-orbit mixing in the conduction band of Si. The strong dependence of the linewidth on donor type<sup>14</sup> leads us to conclude that the main sources of spin-orbit scattering are the impurity core potentials.<sup>17</sup> The second mechanism, which was shown to dominate<sup>18</sup> on the insulating side of the transition ( $n \approx n_c/2$ ), is the hyperfine interaction between the electron and nuclear (P) spins. For both the spin-orbit and hyperfine processes, the linewidth can be understood as arising from two contributions: (i) that due to short-wavelength spin fluctuations, in which case the relaxation occurs during the ballistic motion of an electron past an impurity; and (ii) the long-wavelength diffusive motion of the electron before the return to a given site. In clean metals at high temperature (i) is the main dephasing mechanism, whereas in dirty metals ( $k_F \approx 1$ ) the diffusive contribution dominates. In conventional spin-orbit scattering<sup>19</sup> in metals, the orbital angular momentum is carried by the ballistic electrons; consequently its diffusive contribution vanishes. In the case of Si:P, however, spin-orbit scattering is a natural consequence of impurity scattering between the spin-orbit-mixed states of the Si conduction-band minima,<sup>17,20</sup> and the orbital angular momentum is due to the  $p$ -wave character of the electron wave function. In such a situation both the ballistic and diffusive contributions are present.<sup>17</sup> In the limit where the diffusive contribution dominates, and not too close to the metal-insulator transition, the Kubo formula yields the estimate

$$\Delta H_{1/2}(T) \approx [D_0/D_s(T)](\Delta H_{1/2})_0, \quad (1)$$

implying an enhancement of the linewidth if there is a suppression of the (temperature-dependent) spin-diffusion coefficient  $D_s(T)$  from the bare Ioffe-Regel spin-diffusion coefficient  $D_0 \approx v_F l/3$ . The quantity  $(\Delta H_{1/2})_0$  is of the same order as the short-wavelength ballistic contribution. Estimating it as a sum of hyper-

fine and spin-orbit terms, we have

$$(\Delta H_{1/2})_0 = \frac{3\pi\gamma A^2\hbar}{2E_F} + \frac{1}{\gamma\tau_{iv}}(\Delta g)^2\alpha, \quad (2)$$

where  $\gamma$  is the gyromagnetic ratio of the electron;  $A = 21$  G, the hyperfine interaction in the atomic limit;  $\tau_{iv}$ , the intervalley scattering time;  $\Delta g = 0.003$ , the known shift in the  $g$  value of the electron in the conduction band of silicon; and  $\alpha$ , a numerical constant which depends on details of the silicon band structure and the impurity core potential. A simple theoretical estimate<sup>21</sup> of  $\tau_{iv}$  gives  $(\Delta H_{1/2})_0 \approx 0.03$  G. However, analysis of Pifer's ESR linewidth data<sup>14</sup> well above  $n_c$ , where the linewidth is  $T$  independent and should be well described by Eq. (2), suggests a value of  $1/\tau_{iv}$  that is larger than the theoretical estimate by a factor of  $6 \pm 2$ . This would imply  $(\Delta H_{1/2})_0 \approx 0.2$  G near  $n_c$ .

The analysis can be pushed further, in analogy with the Einstein relation for the conductivity, by introduction of a spin-transport coefficient  $\lambda_s$ , defined by  $D_s = \lambda_s/\chi$ . Away from  $n_c$  on the metallic side, one expects, both on physical grounds and from the scaling analysis of Refs. 5 and 6, that the spin-transport coefficient ( $\lambda_s$ ) is equal to the conductivity in appropriate units. Since the latter is only weakly temperature dependent (less than 10% for our samples<sup>2</sup>), this argument suggests that the temperature dependence of the linewidth is determined by the magnetic susceptibility. This result has been obtained explicitly<sup>12</sup> in the weak disorder limit within the framework of the Finkelstein

approach to disordered interacting electrons.

We plot in Fig. 2 the ratio of the linewidth to its value at a reference temperature  $T_M$  (where the linewidth is a minimum) versus the corresponding ratio for the susceptibility. The data are only shown for  $T < T_M$  beyond which phonon-assisted processes dominate the linewidth. The straight line through the data points in Fig. 2 has a slope of 0.9, in good agreement with the expected slope of unity. The absolute values of  $\chi$  and  $\Delta H_{1/2}$  at  $T_M$  are also consistent with theory, both being larger by a factor of 3 than  $\chi_0$  and  $(\Delta H_{1/2})_0$ , respectively [if we use the estimate of  $(\Delta H_{1/2})_0$  based on Ref. 14].

Our results also provide useful information regarding various cutoffs in scaling theories of interactions and localization. The important cutoffs are  $\tau_s^{-1}$ , the electron spin-flip time;  $\tau_{ee}^{-1}$ , the inelastic electron scattering time; and the temperature  $k_B T/\hbar$ . If  $\tau_s^{-1} > \tau_{ee}^{-1}$ , the single-particle localization effects at lowest order in  $1/k_F l$  are suppressed. The interaction effects are normally cut off by  $\max(\tau_{ee}^{-1}, k_B T/\hbar)$ . However, if  $\tau_s^{-1} > \max(\tau_{ee}^{-1}, k_B T/\hbar)$  the contributions from the triplet particle-hole channel are suppressed. In Fig. 3 we have plotted  $\tau_s^{-1}$ ,  $\tau_{ee}^{-1}$ , and

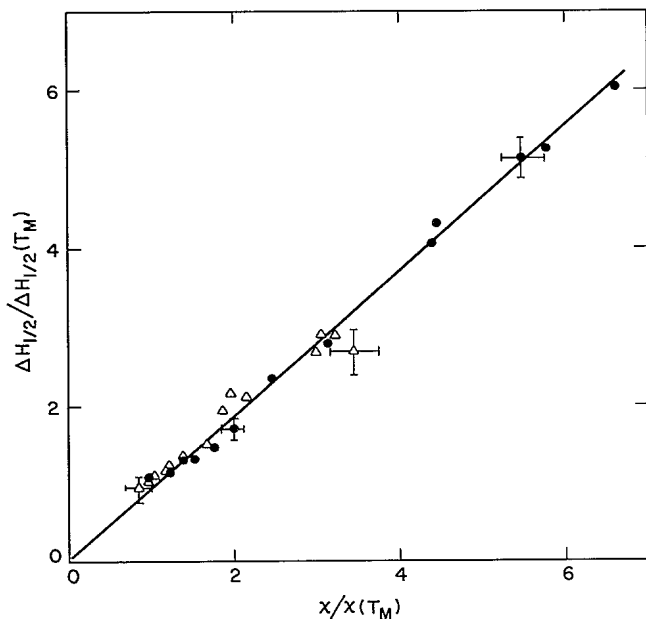


FIG. 2. Temperature dependence of the linewidth vs that of the susceptibility, normalized to their values at  $T_M$ , the temperature at which the linewidth is a minimum [Fig. 1(b)].

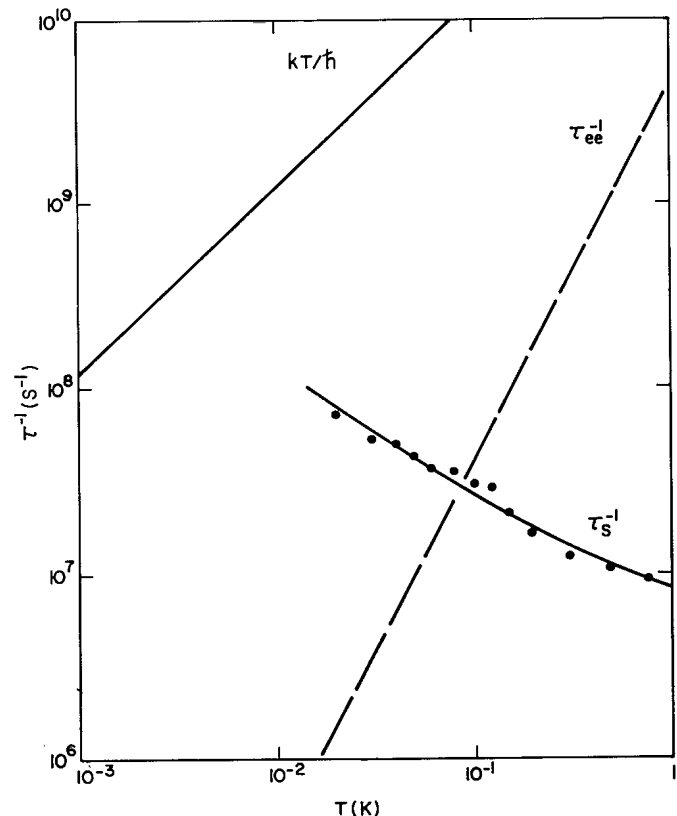


FIG. 3. Spin-flip scattering rate  $\tau_s^{-1} = \gamma \Delta H_{1/2}$  compared to a theoretical estimate of the inelastic rate  $\tau_{ee}^{-1}$  and the thermal rate  $k_B T/\hbar$ , as a function of temperature.

$k_B T/\hbar$  for sample 1 as a function of temperature. Because of the small single-electron localization effects in Si:P near the metal-insulator transition,<sup>1,2</sup> there are no measurements of  $\tau_{ee}^{-1}$  available and we use theoretical estimates.<sup>22</sup> From Fig. 3 we are able to make the following estimates: (a) The lowest-order single-particle localization effects are suppressed around 100 mK when  $\tau_s^{-1}$  becomes larger than  $\tau_{ee}^{-1}$ . (b) By extrapolating the data for  $\tau_s^{-1}$  we find that the triplet particle-hole channel fluctuations are not eliminated until around 3 mK. The susceptibility is expected to saturate at this temperature.

In conclusion, we have studied the ESR line in metallic Si:P and found that the paramagnetic spin susceptibility increases towards the metal-insulator transition and with falling temperatures. No saturation of the susceptibility was detected down to  $T/T_F \approx 3 \times 10^{-4}$ . The linewidth measurements are consistent with recent theories<sup>5,6,12</sup> and help us deduce a spin diffusivity which is over an order of magnitude slower than the bare diffusion at the lowest temperatures. The fluctuations in the triplet particle-hole channel which contribute to the enhancement of the susceptibility should also yield an *increase* in the conductivity with falling temperatures. A conductivity increase around 10% has indeed been observed in similar samples below 1 K.<sup>1,2</sup> We have also estimated that the contributions from the triplet channel will ultimately be cut off by nuclear spin-flip scattering in Si:P below 3 mK. These measurements have been possible because even close to  $n_c$  the ESR linewidth of Si:P is narrower than in most clean metals. Consequently, we expect that in typical disordered metal-semiconductor alloy systems, the spin scattering will be considerably higher than in Si:P. If so, the triplet channel would be suppressed at a higher temperature, and the temperature dependence of the susceptibility would be correspondingly weaker. We speculate that this is a major factor in the rather different conductivity onset behavior seen in doped semiconductors compared to metal-semiconductor alloy systems.

We thank S. Geschwind, P. A. Lee, and Y. Yafet for useful discussions. One of us (A.E.R.) was partially supported by U.S. Office of Naval Research Young Investigator Award.

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