Spins in Si:P Close to the Metal-Insulator Transition

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The spin-lattice relaxtion time T_1 of ^{29}Si nuclei is measured near the metal-insulator transition as a function of temperature and magnetic field. The observed relaxation rate in the metal is up to 10^3 times larger than that for nuclei interacting with free, degenerate electrons. The enhanced relaxation rate and its magnetic field dependence suggest the existence of intrinsic, quasistatic spins with an ω^{-1} spectrum. These spins may cause strong spin-flip scattering and may therefore be responsible for the anomalous critical exponent of the electrical conductivity.

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The electrical conductivity of many disordered systems, including amorphous metal alloys[1] and compensated semiconductors,[2] increases smoothly and is proportional to $n/n_c - 1$ above the metal-insulator (MI) transition. This behavior, which is predicted by extrapolations of both the localization theory for noninteracting electrons[3] and the theory of disordered interacting electrons without localization,[4,5] does not seem to apply to an important class of doped semiconductors. For example, the critical conductivity exponent in uncompensated Si:P and Ge:Sb has been measured[2,6,7] to be $\simeq 0.5$. We expect that Coulomb interactions play a larger role in these uncompensated semiconductors which are characterized by one scattering center per electron than in compensated systems with more scatterers. The full problem including both Anderson localization and the Coulomb interactions has not been solved, but it has been speculated that the Coulomb interactions could give rise to quasistatic spins which would act as strong scattering centers for individual electrons.[8] In the absence of Coulomb interactions and in the weak localization limit the problem of spin-flip scattering has been solved.[9] In this perturbation theory, the spin-flip scattering destroys the first-order term and restores the second-order term which gives the exponent 0.5 if it can be extrapolated into the critical region. Motivated by these ideas we have studied the electron spins in Si:P by measuring the spin-lattice relaxation time of ^{29}Si.

In Si:P the scalar hyperfine interaction is the dominant coupling mechanism between electrons and nuclei,[10] and at high donor concentrations, i.e., $n \gg n_c$, the Korringa law[11]

$$T_{1K}^{-1} = \frac{4\pi k_B T}{\hbar}\left(\frac{\gamma_n}{\gamma_e}\right) K^2 \propto \frac{k_B T}{E_F}\chi^m \qquad (1)$$

is valid.[12,13] In Eq. (1), $K = \Delta B/B$ is the Knight shift of the resonance frequency caused by the hyperfine field, γ_n and γ_e are the nuclear and electronic gyromagnetic ratios, E_F is the Fermi energy, and χ^m is the electronic susceptibility of the clean metal. Using the concentration dependence of Eq. (1) and the high-density results of Sundfors and Holcomb,[12] we extrapolate the Korringa prediction to $n = n_c = 3.75 \times 10^{18}/\text{cm}^3$ and get $T_{1K}T = 140$ min K. Although the Korringa prediction works well for clean metals, as we will see below, it underestimates the rate by three orders of magnitude close to the MI transition at low temperatures.

This rate enhancement can be parametrized in terms of the spin-correlation time τ (in a simple picture, the time one electron spin spends at a given donor site):

$$T_1^{-1} \propto k_B T \chi \tau/[1 + (\omega\tau)^2], \qquad (2)$$

where $\omega = \gamma_n B_0$ is the nuclear Larmor frequency and χ is the effective, transverse susceptibility for the dirty metal under the experimental conditions of finite magnetic field. Qualitatively, T_{1K}^{-1} is small due to the short correlation time $\sim\hbar/E_F$ of the free (ballistic) electrons and the small χ^m. The enhancement of T_1^{-1} is approximately $T_{1K}/T_1 \approx (\tau E_F/\hbar)(\chi/\chi^m)$. In a dirty metal with $k_F l \sim 1$ the main relaxation mechanism would be due to spin diffusion, where $\tau \sim D_s^{-1}(\omega) \times k_F^{-2}$ is the diffusion time of the electron spins at ω.

Two Si:P samples were studied by the standard cw NMR technique. One slightly insulating sample was installed inside a uniaxial stress device[6] and driven by the stress through the MI transition. In this way a density range from $n/n_c \simeq 0.9$ to 1.03 was covered continuously. A larger and slightly metallic sample ($n/n_c \simeq 1.035$) was prepared with an rf coil designed to extend our measurements to lower NMR frequencies where the effects are larger. The static magnetic field B_0 was created by a superconducting solenoid and the homogeneity of B_0 was estimated to be better than 10^{-5} over our small samples. In agreement with previous high-temperature studies[12,13] the low field ($B_0 < 0.5$ T) NMR line was found to have homogeneous dipolar broadening of about 20 μT. At a 2-T field and 30 mK temperature the linewidth was 85 μT in the insulator ($n/n_c \simeq 0.9$) and 70 μT in the metal ($n/n_c \simeq 1.03$) and in both cases decreased slightly at higher temperatures. The asymmetric high-field line shape is attributed to the distribution of Knight shifts due to the variation of electronic wave function around the P donors.[12,13] The temperature depen-

dence can be attributed to the transverse χ, and the slight narrowing of the line in the metallic phase to motional narrowing by delocalized electrons. T_1 was measured by following the relaxation of the NMR line after spin inversion by adiabatic fast passage. The main NMR line was found to relax with a single time constant T_1 within our resolution. Our signal-to-noise ratio was not high enough to study the high-frequency tail of the NMR line, which has been demonstrated[10] to relax faster than the main line, in agreement with the dependence of T_1^{-1} on K in Eq. (1).

In Fig. 1 we show T_{1K}/T_1 of sample 1 as a function of uniaxial stress, S. The MI transition, indicated by the arrow in the figure, was located by a separate conductivity measurement equivalent to that reported previously.[6] The upper scale of the figure shows approximate density values, and is nonlinear over the wide stress region shown. We notice two interesting features in the data: T_1^{-1} is strongly enhanced over T_{1K}^{-1}, and it increases smoothly (and only slightly) through the MI transition. (In comparison, the dc electrical conductivity σ at $T=0$ K and $H=0$ T decreases by over 4 orders of magnitude in the same stress region.) In terms of the simple model of Eq. (2), χ and τ are both enhanced but the data do not prove that they behave critically at the transition. In other words the metallic (itinerant) electrons and localized electrons have similar spin dynamics at finite, low temperatures ($T/T_F \simeq 10^{-4}$), while σ shows distinct differences.

In Fig. 2 we have plotted T_{1K}/T_1 as a function of temperature at a magnetic field of 0.844 T (note that $T_{1K}^{-1} \propto T$). Both in the metal and in the insulator we find a steep increase of the scaled rate at the lowest temperatures. Over the measured temperature region the increase is fitted by a power-law temperature dependence; in the insulator we find $T_{1K}/T_1 \propto T^{-0.7}$ and in the metal $T_{1K}/T_1 \propto T^{-0.5}$. The power law in the insulating phase is similar to that of the static susceptibility, $\chi \propto T^{-0.67}$, measured previously.[14] However, the diverging temperature dependence on the metallic side is puzzling, because it suggests that either τ or χ is diverging. As noted above, we would expect some

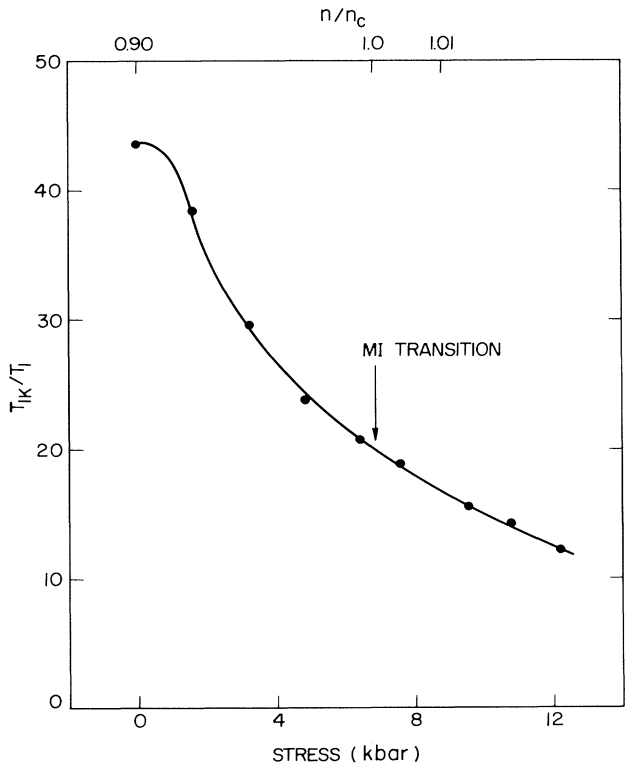

FIG. 1. The normalized spin-lattice relaxation rate T_{1K}/T_1 as a function of the applied uniaxial stress S at 30 mK temperature and 0.844 T magnetic field. The density scale n/n_c at the top of the figure is only approximate and nonlinear. However, the MI transition is accurately known from a separate electric conductivity measurement.

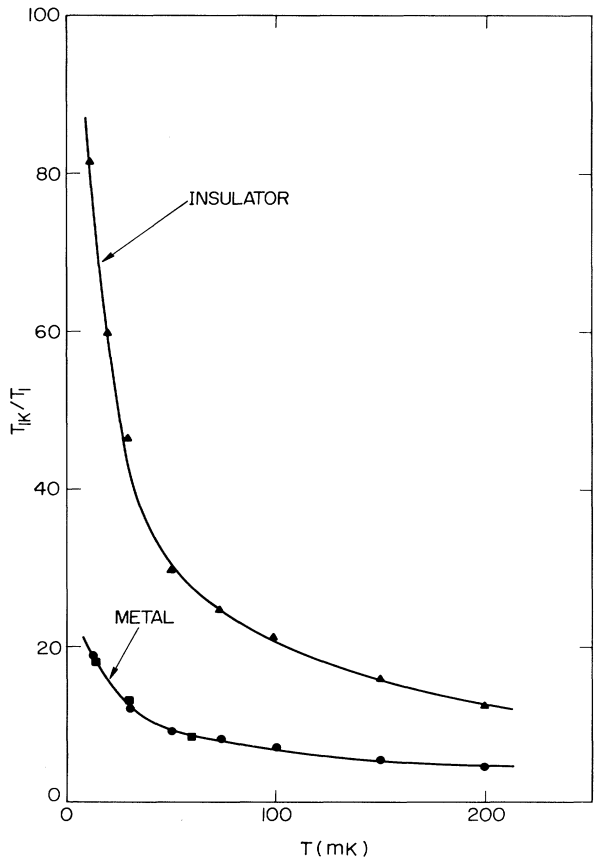

FIG. 2. The temperature dependence of the normalized relaxation rate T_{1K}/T_1 at 0.844 T field: Triangles, sample 1, $n/n_c \simeq 0.90$ (zero stress); circles, sample 1, $n/n_c = 1.03$ (maximum stress); squares, sample 2, $n/n_c \simeq 1.035$ (no stress).

temperature-dependent enhancement of χ but also a Pauli-type saturation at the lowest temperatures. When interpreted in terms of a T dependence of χ, our measurements suggest a surprisingly low saturation temperature, $T_{sat}/T_F \lesssim 10^{-4}$.

Although T_{1K} [Eq. (1)] is independent of the applied magnetic field B_0 we find a field dependence in T_1^{-1} of Si:P on both sides of the transition. In Fig. 3 we present T_{1K}/T_1 as a function of B_0 at $T = 13.5$ mK. For the metallic samples T_1^{-1} is approximately proportional to B_0^{-1} over two decades in field. The maximum measured enhancement over T_{1K}^{-1} is 380 at the lowest field $B_0 \simeq 0.03$ T. In the upper part of the figure we also show $T_{1\rho}^{-1}$, the relaxation rate at extremely low effective magnetic field $B_{eff} = (B_1^2 + B_D^2)^{1/2} \simeq 10$ μT, where $B_1 \lesssim 10$ μT is the applied rf field and $B_D \sim 10$ μT is the dipolar field.[15] This $T_{1\rho}^{-1}$ is over 3 orders of magnitude larger than T_{1K}^{-1} and nearly independent of B_0. Since, for our low rf fields, $B_1 \lesssim B_D$, we also found that $T_{1\rho}$ is independent of B_1. $T_{1K}/T_{1\rho}$ was found to decrease towards higher temperatures approximately as T_{1K}/T_1.

We do not have a quantitative explanation for our results. However, there appear to be two classes of models both of which require strong electron-spin effects. The first of these is based on an extension of calculations developed for localized electron spins and the second on a substantial slowing down of metallic spin diffusion. The point of view implied by Eqs. (1) and (2) is that for a metal, but the problem might be theoretically tractable with either approach. The first class would be an extension of ideas developed to describe the insulating state,[14] such as the analysis of Blumberg[10,16] for dilute paramagnetic centers. In this case we would include a significant contribution from localized states below the Fermi level.[17] This model cannot be applied directly near the transition because the P donors do not form a dilute system—the interactions are strong enough to produce the metal-insulator transition and the interelectron spacing (~ 60 Å) is only a few times the Bohr radius (20 Å) of the hydrogenlike donor wave function.

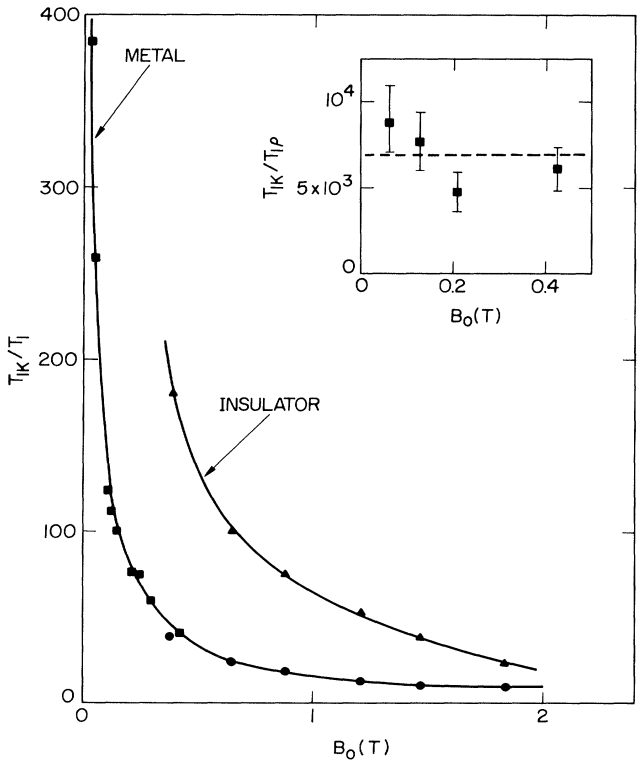

FIG. 3. The magnetic field dependence of T_{1K}/T_1 at 13.5 mK temperature. The data symbols are the same as in Fig. 2. The inset shows $T_{1\rho}$, the zero-field relaxation time, as a function of static field B_0. $T_{1\rho}$ was measured only for sample 2.

Another important factor in chosing an appropriate approximation is that the magnetization of a typical donor might be expected to saturate under our experimental conditions where $\mu_B B_0/k_B T > 1$. This simple criterion appears to be inadequate because a linear field dependence similar to ours has been seen[10] in Si:P at higher T where $\mu_B B_0/k_B T < 1$. In this context, we would reiterate that $T_{1\rho}$ does not have a strong field dependence as might be expected from dilute paramagnetic centers.

The metallic approach to the problem involves a drastic slowing of the spin diffusion, D_s, compared to the charge diffusion, D_c. An example of substantial slowing of the spin fluctuations has recently been observed near a magnetic percolation threshold with use of neutron scattering.[18] Within this picture we suggest that the field dependence of T_1 arises from the distribution of correlation times for the spin fluctuations. By assuming a τ distribution $P(\tau)d\tau \propto d\tau/\tau$ (corresponding to a spin-fluctuation spectrum $\propto \omega^{-1}$), we get from Eq. (2) that $T_1^{-1} \propto \chi/\omega$ for $\omega > \tau_{max}^{-1}$ and $T_1^{-1} \propto \chi$ for $\omega < \tau_{max}^{-1}$. This gives the desired ω (or B_0) dependence of T_1 and also gives a field-independent $T_{1\rho}$ with a similar T dependence to that of T_1. From $T_{1\rho}$ we also estimate $\tau_{max} \simeq 10^{-5}$ sec, which is considerably longer than the transport scattering time $\simeq 10^{-13}$ sec. This indicates that, in this strongly correlated electron system, the effective spin diffusion, $D_s \propto \tau_{max}^{-1}$, is considerably slower than the charge diffusion, $D_c \propto \sigma$.

We have speculated that the enhanced magnetic susceptibility and the slowing down of spin diffusion relative to charge diffusion on the metallic side are due to strong Coulomb correlations in the disordered electron liquid. Both these effects can be found in a simple model of the Mott transition discussed by Gutzwiller and by Brinkman and Rice[19] for an order system. For example, if one includes only the geometric effects of impurities but ignores localization altogether, D_s/D_c

$\rightarrow 0$ near the MI transition, independent of the impurity scattering time while χ diverges. The Brinkman-Rice paramagnetic liquid also has a T-dependent enhancement of the specific heat qualitatively similar to that observed[20] in Si:P. A quantitative discussion of these ideas would, however, require a finite-temperature generalization of the Gutzwiller approach to a disordered metal including localization effects.

In conclusion, our principal results are the observation of anomalously fast spin-lattice relaxation in a disordered metal near the metal-insulator transition, and unexpected temperature and magnetic field dependences of this relaxation. These results indicate quasistatic spin states that are intrinsic to a correlated, disordered metal. These quasistatic spins change the symmetry of the scattering of the charge degrees of freedom and thus may be responsible for the conductivity exponent of $\frac{1}{2}$ near the metal-insulator transitions in Si:P.

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[15]$T_{1\rho}$, the rotating-frame relaxation time, was measured by stopping the adiabatic frequency sweep at $\omega_0=\gamma_n B_0$ for a time Δt and then reversing the sweep to the starting point ω; $\omega_0-\omega >> \gamma B_1$. During the time Δt, the nuclear magnetization $\mathbf{M}$ rotates in the plane perpendicular to $\mathbf{B}_0$ in an effective perpendicular field $B_{\rm eff}=(B_1^2+B_D^2)^{1/2}$ and relaxes with time constant $T_{1\rho}$. $T_{1\rho}$ was determined by comparing M before and after the adiabatic sweeps.

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