PHYSICAL REVIEW B VOLUME 46, NUMBER 16 15 OCTOBER 1992-II

# Knight shifts in Si:P and Si:(P,B) in the vicinity of the metal-insulator transition

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(Received 25 November 1991; revised manuscript received 17 April 1992)

Recent experimental and theoretical work has suggested that a two-fluid model can be used to describe the magnetic properties of the just-metallic phase of metal-insulator systems such as Si:P. Alternative descriptions based on a Fermi liquid approach have also been put forward to explain the magnetic and other behaviors of these systems. In the present work $^{29}$Si Knight-shift measurements have been made on a number of samples of Si:P and Si:(P,B) in the just-metallic region as a function of temperature and electron concentration. The magnetization of the $^{29}$Si spin system follows Curie law behavior down to 50 mK. The mean Knight shift is found to be temperature independent in the range 50 mK–4.2 K for both compensated and uncompensated systems. This result is consistent with a proposal that the $^{29}$Si spins that are observed in the Knight-shift measurements interact only with the itinerant electron fluid, even though the susceptibility is controlled by the other fluid of localized moments.

## INTRODUCTION

Much experimental and theoretical effort has been devoted to the study of metal-insulator (MI) systems, in particular Si:P, on the just-metallic side of the transition. While it appears that the insulating state is fairly well understood, the situation in the metallic state close to the transition is less clear.

Experiments on the $^{31}$P Knight shift in Si:P, carried out by Alloul and Dellouve,[1] have been interpreted[1,2] as favoring a phenomenological two-fluid model in the MI transition region. The two "fluids" are regarded as being made up of localized and itinerant electrons, respectively. Further support for this model has come from $^{29}$Si spin-lattice relaxation-time measurements[3] on Si:P and Si:(P,B) and from specific-heat measurements[4] as a function of applied magnetic field on Si:P.

Theoretical developments in the field of disordered electronic systems appear to be concentrated in two main directions. In the disordered Fermi-liquid approach[5] electron-electron interactions are properly taken into account and the disorder effects are treated as a perturbation using renormalization procedures. In a recent calculation Belitz and Kirkpatrick[6] have extended these ideas in an effort to understand the behavior of the magnetic susceptibility in the vicinity of the transition. They suggest that the MI transition is preceded on the metallic side by another transition which involves a change from diffusive to subdiffusive spin transport. In this approach, long-lived correlations in spin orientations can give rise to susceptibility enhancements compared to the Pauli susceptibility. An alternative approach has recently been suggested by Milovanović, Sachdev, and Bhatt.[7] They treat the noninteracting Anderson-Hubbard Hamiltonian exactly and account for electron-electron interactions in a Hartree-Fock effective-field approximation. The results suggest that the disordered electron system displays an instability towards the formation of local moments on a length scale much shorter than the coherence length of the electrons' Hartree-Fock wave functions. In a slightly earlier paper Sachdev[8] has examined a phenomenological Hamiltonian for the two-fluid model which includes interactions between the fluids. He finds that the itinerant electron spin susceptibility is enhanced by the presence of the local moments. The enhancement is temperature independent and arises from a decrease in a Fermi-liquid parameter.

In recent experimental work magnetic susceptibility measurements have been made on both Si:P (Ref. 9) and Si:(P,B) (Ref. 10) using the Hebel-Slichter technique, which gives the total spin susceptibility $\chi_s$. A marked temperature dependence is observed in contrast to the normal Pauli temperature-independent behavior for metals. The measured values of $\chi_s$ as a function of $n/n_c$ exhibit enhanced values compared to the Pauli value. The compensated samples exhibit a more marked temperature dependence than the uncompensated samples.

More recently, Bhatt and Fisher[11] have argued in a general way that the low-temperature thermodynamics of metal-insulator systems is dominated by spins on isolated sites in the metallic phase. This leads to a divergent susceptibility as $T \rightarrow 0$. They suggest that coupling of the rare sites to the surrounding Fermi fluid is too weak to quench the local moments even at very low temperatures.

Compensation increases the disorder of the system and it is therefore of interest to make measurements on both Si:P and Si:(P,B). Electrical conductivity measurements at low temperatures[12] lead to apparently different values for the critical exponent $\mu$ for compensated and uncompensated samples. Thomas[13] has reviewed the literature on critical exponents and finds that values of $\mu$ tend to cluster around either 0.5 or 1. This suggests that there are two universality classes. Thomas also observes that $\mu=0.5$ in systems where there is one electron per scattering site and $\mu=1$ in more disordered materials in which there are several scattering sites per electron. A number of theoretical approaches,[14–16] based on Fermi-liquid ideas, take into account electron-electron interactions and suggest that the scattering mechanism can affect $\mu$. Strong scattering leads to $\mu=1$, while weak scattering gives lower values. There is, however, no generally accepted theory which predicts $\mu=0.5$ in Si:P. Recent experimental work by Thomanschefsky[17] provides evidence that for lightly compensated samples of Si:(P,B) the critical exponent is between 0.5 and 1. It must be added that it is difficult, in many experiments, to be certain that measurements are being made in the critical region sufficiently close to $n_c$. Nevertheless, the available evidence points to the conclusion that compensated and uncompensated systems belong to different universality classes. The present work is concerned with Knight-shift measurements on both kinds of system. The results are considered in relation to available magnetic susceptibility data.

## KNIGHT SHIFTS IN DISORDERED SYSTEMS

Knight-shift measurements on MI systems may be regarded as complementary to magnetic susceptibility measurements. While local moments may play a role in determining the total spin susceptibility, it is the itinerant electrons which govern the Knight shifts. For a disordered metallic system the Knight shift for a nucleus at site $i$ may be expressed as

$$K_i=\frac{8\pi}{3}\langle |u(r_i)|^2\rangle_{E_F}\chi_i \ , \tag{1}$$

where $\chi_i$ is the local susceptibility at the $i$th lattice site and $\langle |u(r_i)|^2\rangle_{E_F}$ is the electron probability density at the nucleus averaged over the Fermi surface. Because of the disordered nature of MI systems, a distribution of Knight shifts is obtained, giving rise to an asymmetric line shape. It is important to note that $\chi_i$ in Eq. (1) is not to be identified with the total spin susceptibility $\chi_s$. Kaveh and Liebert[18] have obtained an expression for the Knight shift using the tight-binding approximation. For $^{29}$Si in Si:P the expression for the mean Knight shift may be written in the form

$$\langle K\rangle=A\chi_p e^{-R_0/a_B} \ , \tag{2}$$

where $R_0$ is a measure of the average distance between a silicon atom and its nearest-neighbor phosphorus atoms. We take $R_0=cn^{-1/3}$, with $c$ a constant of order unity. $\chi_p$ is the spin susceptibility per unit volume for the electrons in the impurity band and $a_B$ is the effective Bohr radius of a P impurity center. $A$ is a constant which can be calculated. $\chi_p$ depends on the density of states $N(E_F)$ at the Fermi level. Specific-heat data for Si:P, as well as numerical tight-binding calculations quoted by Kaveh and Liebert,[18] suggest that $N(E_F)$ varies approximately as $n^{1/3}$ for $n/n_c>1$. For compensated Si:(P,B) the density of states may be expected to be lower than for Si:P because of the broadening effect of disorder on the impurity band. [Nishio *et al.*[19] have inferred values of the specific-heat coefficient $\gamma$ from specific-heat measurements made on Si:P and fast neutron irradiated Si:P, which is a compensated material. For $n<n_c$, $\gamma$ for the compensated samples is greater than for the uncompensated samples. No specific-heat measurements appear to have been reported for Si:(P,B).] Using the tight-binding approach, the ratio of the density of states for the uncompensated and compensated materials may be written as

$$\frac{N_u(E_F)}{N_c(E_F)}=\frac{(B^2+V_c^2)^{1/2}}{(B^2+V_u^2)^{1/2}} \ , \tag{3}$$

where $B$ is the bandwidth without disorder and $V_u$ and $V_c$ are, respectively, the amplitudes of potential fluctuations in the uncompensated and compensated materials.

A modified version of the Kaveh-Liebert calculation is given below. In particular, the averaging over the phosphorus sites is carried out in a different way. A tight-binding wave function of the form

$$\psi(r)=(n\Omega)^{-1/2}\sum_j C_j\phi(r-R_j) \tag{4}$$

is chosen for the ground state of the electrons at low temperature. $R_j$ locates the phosphorus atoms, $\Omega$ is the volume of the material, and $n$ is the phosphorus atom density. $\phi(r)=(8\pi a_B^3)^{-1/2}\exp(-r/a_B)$ is a hydrogenlike wave function and the coefficients $C_j$ have a random phase and amplitude as a result of the disorder and associated scattering processes. To calculate $K$ for a nucleus at position $R$, Eq. (1) is used in the following form:

$$K=\frac{8\pi}{3}|\psi(R)|^2\Omega\chi_p \ . \tag{5}$$

$|\psi(R_{S_i})|^2$ is obtained by summing over all phosphorus atoms $j$ and is given by

$$|\psi(R_{S_i})|^2=\frac{1}{n\Omega}\sum_j |C_j|^2\frac{1}{8\pi a_B^3}\exp(-2|R_{S_i}-R_j|a_B^{-1}) \ . \tag{6}$$

Since the coefficients $C_j$ are of random phase and amplitude, the sum over cross terms $j\neq k$ is taken as zero. This is an approximation which implies that wave-function coherence is not maintained over distances larger than the phosphorus atom spacing. At low temperatures this may not be a very good approximation. For simplicity, we shall ignore any corrections introduced by the neglected terms. Converting the sum to an integral and assuming, as an approximation, a uniform distribution of phosphorus atoms gives for the mean value

$$\langle |\psi(R_{S_i})|^2\rangle=\frac{1}{\Omega}\frac{1}{8\pi a_B^3}4\pi\int_{R_0}^{\infty}r^2\exp(-2ra_B^{-1})dr \ , \tag{7}$$

where we put $R_{S_i}=0$.

Performing the integral gives

$$\langle |\psi(R_{S_i})|^2\rangle = \frac{1}{8\Omega}(2c^2a_B^{-2}n^{-2/3}+ca_B^{-1}n^{-1/3}+1)$$
$$\times\exp(-2ca_B^{-1}n^{-1/3})\ , \qquad (8)$$

where the lower limit $R_0$ in the integral is taken as the average distance between a silicon atom and its nearest-neighbor phosphorus atom, as defined above. Substituting into Eq. (5) gives

$$\langle K\rangle = \frac{8\pi}{3}\chi_p(2c^2a_B^{-2}n^{-2/3}+ca_B^{-1}n^{-1/3}+1)$$
$$\times\exp(-2ca_B^{-1}n^{-1/3})\ . \qquad (9)$$

The susceptibility $\chi_p$ may be written as $\chi_p = 3\mu_B^2\gamma_S\gamma/\pi^2k_B^2$, where $\gamma_S$ is the Stoner enhancement factor. Equation (9) is similar to Eq. (2) but predicts that $\langle K\rangle$ falls off more rapidly with $n$ below $n_c$.

## KNIGHT-SHIFT MEASUREMENTS

The $^{29}$Si Knight shift has previously been measured[20,21] across the metal-insulator transition for uncompensated materials. The aim of the present work is to measure $K$, as a function of concentration and temperature, for selected Si:P and Si:(P,B) samples in the just-metallic region.

For the concentration-dependent measurements characterized wafers were crushed into a powder ( ~150 μm) and the samples mounted in small Teflon ampoules. These could be moved through a fixed nuclear magnetic resonance (NMR) coil immersed in liquid helium in a Janis Dewar. Movement of the train of two samples and a reference sample (insulating Si:As) permitted measurements to be made rapidly, minimizing possible effects of drift of the magnetic field of 1 T supplied by a regulated electromagnet. Pulsed Fourier-transform NMR methods were employed for data capture and analysis. A preliminary account of these measurements has been given previously.[22] The critical concentration values are based on the Thurber scale. For Si:P $n_c = 3.7\times10^{18}$ cm$^{-3}$, while for the Si:(P,B) samples, with a compensation ratio $N_B/N_p \simeq 0.5$, the value $n_c = 5.3\times10^{18}$ cm$^{-3}$ was used. Thomanschefsky[17] has given a detailed discussion of the methods involved in determining $n$ and $n_c$.

An Oxford Model 400 helium dilution refrigerator was used to obtain temperature-dependent measurements down to 50 mK at a field of 1 T provided by a high-homogeneity superconducting solenoid magnet. In this case the sample consisted of a stack of $0.5\times5\times10$ mm$^3$ slices cut from characterized wafers. The Si:(P,B) sample used ($n/n_c = 1.1$) was the same as that used by Hirsch *et al.*[10] Care was taken to achieve good thermal anchoring to the oxygen-free high-conductivity copper tail attached to the mixing chamber. Temperatures were measured using a calibrated germanium thermometer and the amplitude of the $^{29}$Si NMR signal was used as a check on the temperature. The reference Si:As sample was mounted near the measured sample. A control experiment was carried out with reference samples in both coils in order to calibrate the system and obtain absolute Knight-shift values. All quoted values are for the mean (first moment) Knight shift $\langle K\rangle$.

## RESULTS AND DISCUSSION

Figure 1 shows a plot of the mean $^{29}$Si Knight shift $\langle K\rangle$, normalized to the value at $n/n_c = 10$, versus $n/n_c$ for both Si:P and Si:(P,B) at 4.3 K based on values taken from Hirsch and Holcomb[21] (5.85 T) and Hoch, Thomanschefsky, and Holcomb[22] (1.0 T). The Kobayashi *et al.*[20] data (0.91 T), which are not plotted, are consistently higher on the metallic side than the values shown in Fig. 1 and appear to show a more marked decrease through the MI transition. The reason for the discrepancy must lie in some unknown systematic error. The data of Fig. 1 were obtained in separate experiments on samples derived from the same source and characterized in the same way. The consistency of the measurements gives some grounds for confidence in their reliability. The measurements on Si:(P,B) are taken from Hoch, Thomanschefsky, and Holcomb[22] and represent the only available data for compensated samples. The values were obtained using exactly the same equipment and procedures that were used for the uncompensated samples at 1 T. While there are only three data points, it appears that the Knight shift for the compensated samples decreases more rapidly than for the uncompensated samples as the transition is approached from the metallic side.

We suggest the following explanation for the data of Fig. 1. At high concentrations ($n \gg 10^{19}$ cm$^{-3}$) both compensated and uncompensated systems approximate dirty metals and the Knight shifts follow the Pauli susceptibility behavior ($\chi_p \propto n^{1/3}$). At lower concentrations on the metallic side of the transition the measured spin susceptibility $\chi_s$ is enhanced by local moments. The Knight shifts continue to decrease, in contrast to $\chi_s$, which suggests that the $\langle K\rangle$ values are sensitive to only the delocalized electron-nucleus interactions. For Si:(P,B) the decrease in $\langle K\rangle$ with decreasing $n$ is more rapid than for Si:P. If it is assumed that $\langle |u(0,i)|^2\rangle_{E_F}$ is roughly the same for both compensated ($c$) and uncom-

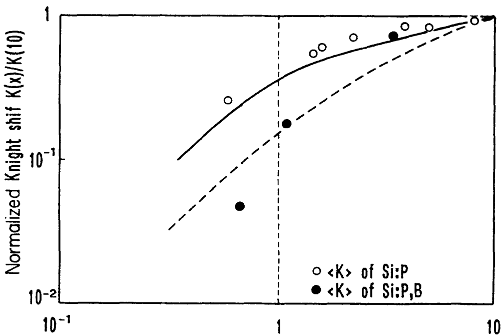

FIG. 1. The normalized mean Knight shift of $^{29}$Si in Si:P (open circles: Refs. 21 and 22) and Si:(P,B) (closed circles: Ref. 22) as a function of the relative electron concentration $n/n_c$. The solid curve through the Si:P points is based on the tight-binding approach of Kaveh and Liebert (Ref. 18). The dashed curve represents an attempt to account for the Knight shift in Si:(P,B). Details are given in the text.

pensated ($u$) samples, then this implies that, for the susceptibilities of the delocalized electrons, $\langle \chi_p^c \rangle < \langle \chi_p^u \rangle$. This, in turn, suggests a lower density of states for the delocalized electron fluid in the compensated system than in the uncompensated system. It is, however, not clear that $\langle |u(0,i)|^2 \rangle_{E_F}$ does not change somewhat with compensation, and the conclusion concerning the density of states given above must be regarded as tentative.

It is interesting to note that the results of Paalenen *et al.*[9] and Hirsch *et al.*[10] show that, for metallic samples with similar $n/n_c$ values, $\chi_s^c > \chi_s^u$. The difference is strongly temperature dependent. The decrease in the average delocalized electron fluid susceptibility with compensation, as inferred from the present Knight-shift results, is therefore to be contrasted with the corresponding increase in the total susceptibility, which, in the two-fluid model, is dominated by the localized fluid component.

Compensation increases the degree of disorder and strengthens the Anderson character of the transition. The present results suggest that this extra disorder is associated with a lower density of states at the Fermi level, which may be linked with the enhanced fluctuations in the lattice potential produced by compensation.

We have used the tight-binding Knight-shift expression to fit the normalized experimental data for Si:P. Use of the normalized values simplifies the expressions used as a number of factors cancel out. (Absolute calculations for the uncompensated material are in fair agreement with the measured $K_{\text{pk}}$ values.) For the normalized results the agreement between the fitted curve in Fig. 1 (full line) and the experimental points is quite acceptable for Si:P. For the uncompensated material the Anderson condition for localization produced by potential fluctuations was used to set $(V_u/B)_{n_c} = 1.6$. For the compensated material the ratio $V_c/B$ in Eq. (3) may be expected to be larger than $V_u/B$ because of the enhanced disorder produced by compensation. We have tried to allow for this by increasing the ratio $(V_c/B)_{n_c}$ from the theoretical value of 1.6 to 3.2. It is not clear that this procedure is valid but no simple alternative suggests itself. It can be seen from Fig. 1 (dashed line) that calculated values for $\langle K \rangle$ for Si:(P,B) are reduced below those for Si:P but agreement with the three experimental points is not very good. The experimental points fall off more rapidly than the theoretical curve as $n/n_c$ approaches 1 from the metallic side.

Measurements of the mean Knight shift $\langle K \rangle$ of the NMR line at different temperatures are shown in Fig. 2(b). The samples examined were Si:P ($n/n_c = 1.6$) and Si:(P,B) ($n/n_c = 1.1$). It appears that $\langle K \rangle$ is temperature independent for both types of samples within experimental uncertainty. The measurements at 1 K and below agree well with the previous 4.3-K measurements.

Figure 2(a) shows the measured $\chi$ values for Si:P (Ref. 9) and Si(P,B) (Ref. 10) as a function of temperature for comparison with the present Knight-shift results. A marked temperature dependence is found for $\chi_s$. In normal metals the Pauli susceptibility $\chi_P$ is temperature independent for $T \ll T_F$. In our samples $T_F \sim 100$ K. As mentioned above, it is believed, on the basis of $\chi_s$ measurements, that localized spins persist into the metallic phase. These local moments play a dominant role in determining $\chi_s$. Exchange or Ruderman-Kittel-Kasuya-Yosida (RKKY) -type[11] couplings between the localized moments may be expected to lead to departures from Curie law behavior, as observed.

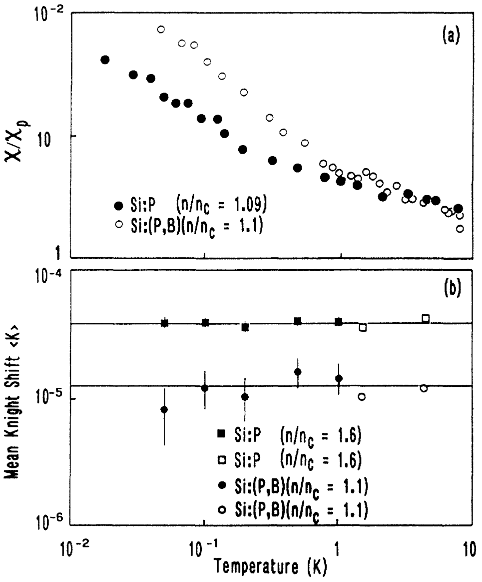

FIG. 2. (a) The relative spin susceptibility $\chi/\chi_P$ as a function of temperature for Si:P and Si:(P,B) based on the data of Refs. 9 and 10. (b) The mean Knight shift of $^{29}$Si in samples of Si:P and Si:(P,B) as a function of temperature. [For Si:(P,B) the sample is the same as that shown in (a)].

Previous work[20,23] has provided evidence that the $^{29}$Si NMR line shape has a slight temperature dependence. Corresponding to the change in line shape, it is suggested that there is a slight increase in the mean Knight shift $\langle K \rangle$ as the temperature is decreased. The peak Knight shift $K_{\text{pk}}$ was, however, not found to change with temperature. These early measurements used continuous-wave NMR methods. According to the Korringa relation,[24] $T_1K^2T = \text{const}$ and $T_1$ varies across the line at any given temperature. It is, therefore, possible that portions of the line with longer $T_1$'s were slightly saturated compared to the shorter $T_1$ portions. Such effects would produce a slight distortion of the line shape. The present results shown in Fig. 2(b) do not support any temperature dependence of $\langle K \rangle$.

In their $^{31}$P Knight-shift measurements Alloul and Dellouve[1] found that the fraction of $^{31}$P nuclei which could be observed in their experiments decreased markedly as $n$ approached $n_c$ from the metallic side. Furthermore, the fraction of observed $^{31}$P nuclei decreased as the temperature was lowered from 4.2 to 1.65 K. We have not observed any change in the fraction of $^{29}$Si nuclei observed in our experiments. Figure 3 shows plots of the nuclear temperature, obtained using the sample magnetization and Curie's law, versus the refrigerator mixing chamber temperature, measured using the germanium thermometer, for both Si:P and Si:(P,B). The linearity of the plots provides evidence that, within experimental error, Curie's

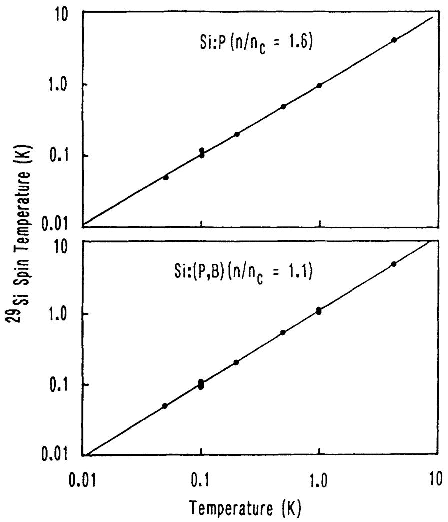

FIG. 3. (a) $^{29}$Si temperature for Si:P ($n/n_c = 1.6$), inferred from the magnetization, as a function of refrigerator mixing chamber temperature, measured using a germanium thermometer. (b) $^{29}$Si temperature as a function of mixing chamber temperature for Si:(P,B) ($n/n_c = 1.1$).

law is obeyed over the range 50 mK–4 K and that thermal contact of the sample with the mixing chamber is maintained.

The present results, taken together with the theoretical Knight-shift predictions based on the Kaveh and Liebert[18] approach, suggest that no significant enhancement of the Knight shift occurs through the interaction of delocalized spins with localized spins. If such effects were present, they might be expected to be most pronounced in the Si:(P,B) ($n/n_c = 1.1$) sample, which has the largest susceptibility enhancement. The absence of any detectable temperature dependence of $\langle K \rangle$ suggests that the two groups of spins interact either rather weakly or in such a way that no temperature variation of the local susceptibility of the itinerant electrons occurs. The latter possibility is in agreement with a recent theoretical prediction.[8] It is likely that the two sets of spins occupy spatially distinct regions in the sample and the two "fluids" should not be thought of as interpenetrating.

## CONCLUSION

The present concentration- and temperature-dependent Knight-shift measurements for Si:P and Si:(P,B) samples are consistent with the phenomenological two-fluid model for MI systems in the vicinity of the MI transition. They do not appear to support the disordered Fermi-liquid approach. The lack of any detectable temperature dependence of the Knight shift down to 50 mK suggests that interactions between localized and delocalized spins are weak. This result is consistent with a recent theoretical prediction[8] for the itinerant electron spin susceptibility.

## ACKNOWLEDGMENTS

We gratefully acknowledge the contributions made by Professor D. F. Holcomb at Cornell University, who supplied the samples used in the work and who also made valuable comments on the results. Helpful discussions with Dr. D. Joubert are also acknowledged. Financial support was provided by the Foundation for Research Development.

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