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1991 J. Phys.: Condens. Matter 3 8095

(http://iopscience.iop.org/0953-8984/3/41/008)

J. Phys.: Condens. Matter 3 (1991) 8095–8101. Printed in the UK

Uniaxial-stress effects on the magnetic properties of Si : P close to the metal–non-metal transition

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Received 17 May 1991

Abstract. A sample of Si : P with a doping density n such that $(n/n_c) = 0.85$ at zero stress, where n_c is the critical density for the metal–non-metal transition, has been studied at temperatures ranging from 15 mK up to 4.2 K using electron spin resonance techniques at very low magnetic fields. Some results have been obtained with uniaxial stress applied up to 0.2 GPa, implying a stress-tuning effect moving (n/n_c) close to 0.9. The results indicate that the ESR linewidth and the electron spin susceptibility are proportional to one another when the temperature is varied at zero stress, that the susceptibilty shows no sign of 'flattening off' at low temperature and that, within experimental error, there is no effect of stress on the magnetic susceptibility in this sample. The effect of stress on the linewidth is also anomalously small. The electrons appear to behave more like a metallic system than previous studies in Si : P in this density regime have demonstrated.

1. Introduction

There is a continuing problem in understanding the metal–non-metal transition in Si : P, connected with the anomalous scaling behaviour of the conductivity with doping density [1]. The anomaly may have magnetic origins [2], so a detailed understanding of the smooth passage in the magnetic behaviour of the electron spins, from Curie-like at $n \leqslant 10^{23}\,\mathrm{m^{-3}}$ to Pauli-like at $n > 2 \times 10^{25}\,\mathrm{m^{-3}}$, is imperative. Evidence has been presented [3] that the effect of uniaxial stress on Si : P in this region of doping density is to tune n_c, by effectively mixing into the ground-state donor wave-functions an admixture of higher-energy, more extended, functions. Such an increase in the size of the wave-function is of course connected, via the Mott criterion [4], to a reduction in n_c, the critical density of doping for metallic behaviour. The method gives access to high-resolution tuning of (n/n_c) in a way not possible by doping variation alone.

The electron spin resonance (ESR) method is accepted as the ideal way of accessing magnetic properties of electron spins, in that it gives discrimination against all magnetic properties other than the important one of the electron spin susceptibility. Unwanted impurity spins will, in general, produce spectra clearly distinguished from the electron spins of interest; one can therefore be sure that the technique is producing data that are a property of the relevant spins. There have been related ESR studies in a similar regime of phosphorus concentration by other groups; by Muruyama *et al* [5] at $n \simeq 2 \times 10^{24}\,\mathrm{m^{-3}}$, by Paalanen and co-workers [6, 7] in the just-metallic region, and by Ikehata *et al* [8]. There has also been a classic, SQUID-based, magnetic study of these materials in this density range at ultra-low temperature [9]. Our work connects with all these other projects. Several theoretical papers have been published pertaining to such measurements [10, 11].

The application of stress to modify ESR spectra of phosphorus donors in silicon has proved extremely useful in the low-donor-density limit [12, 13], where g-shifts, hyperfine splitting, and relaxation rates of the resonant donor electrons have been monitored. These isolated impurity studies have shown that the modest stresses applied in our experiment ($\approx$0.15 GPa) are sufficient to cause drastic changes in the various parameters measured. The Wilson and Feher experiment [12], for example, reveals that a stress of 0.1 GPa is sufficient to cause a diminution of the HFS of about 40%; they characterize this as indicating that the electron is then spending 60% of its time in the lower two of the six conduction valleys. Well below n_c then, the electron donor wave-functions are drastically altered by stress. Just above n_c, we also know that the electrical conductivity experiments [3] have demonstrated that stress causes large changes in the conductivity.

2. Experimental details

We first specify the density of our bar of Si : P. The sample was cut from a wafer supplied by Wacker, with a room temperature resistivity of 12.4 mΩ cm. We also measured the resistance ratio of the wafer ($\rho(4.2)/\rho(300)$) at several different positions across the wafer, obtaining numbers varying from 70 down to 12. At this density the ratio R is extremely sensitive to dopant concentration ($\Delta R/\Delta n = -2400$, i.e. a change of n from 3.16×10^{24} to 3.15×10^{24} m^{-3} produces a change of resistance ratio from 12 to 36). The room temperature resistivity value, corresponding to zero stress, leads, on the Mousty *et al* scale [14], for which $n_c = 3.75 \times 10^{24}$ m^{-3}, to $n = 3.2 \times 10^{24}$ m^{-3}. Other more recent scales [15, 16] produce different numbers for both n and n_c, but the ratio of $n/n_c = 0.85$ is insensitive to the choice of scale.

Our experiments have involved measurement of the ESR spectra at a sequence of temperatures from 15 mK up to 4.2 K. The experiment was carefully constructed in a bottom-loading, large-cooling-power dilution refrigerator, using a low-excitation Q-meter system for detection [17]. The solenoidal sample coil was thermally isolated from the sample. Great care was taken with the construction of the tail-piece of the refrigerator, with gold-plated, polished, surfaces and pressurized contacts used throughout. The sample itself, 15 mm × 1 mm × 1 mm, with the long axis parallel to ⟨100⟩, had polished ends, and sat in solder pods at each end on Be–Cu anvils. The geometry of the stress experiment was such that the stress was applied along the long axis, the coil monitored the ESR across the middle section of the sample (away from strain inhomogeneities) and the main magnetic field for the ESR experiment was applied in a horizontal direction, perpendicular to the long axis of the sample. The magnetic field generated by Helmholtz coils was swept in a sawtooth pattern, first positive then negative, for up to 16 h, typically averaging 300 sweeps to obtain one spectrum. A nuclear orientation thermometer was used to measure temperature below 50 mK, and a germanium resistance thermometer for higher temperatures.

3. Results and discussion

The measurements of the spin susceptibility are presented in figure 1, taken at 30.4 MHz, i.e. resonance is at about 0.1 mT. This is the integrated area of the ESR spectrum, which

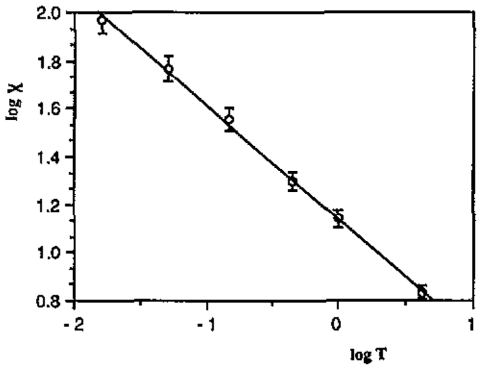

Figure 1. The temperature variation of the electron spin susceptibility (as measured by the ESR line area). No absolute calibration of the spin susceptibility has been undertaken, so the units of the susceptibility are arbitrary. From the fit, the value of α comes out to be 0.47 ± 0.02.

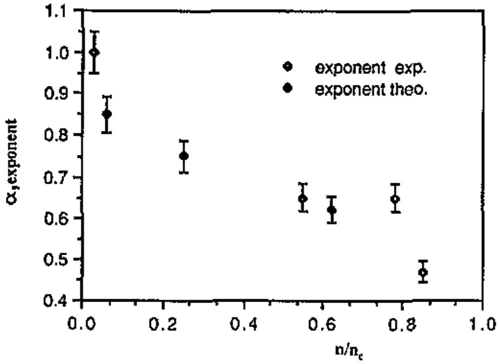

Figure 2. The exponent α, in $\chi \propto (T^{-\alpha})$, as fitted to the experimental and theoretical [18] curves. The low-density experimental point is from [9], and the next point is from [5].

consisted of a single line of roughly constant intensity that broadened dramatically as the temperature was lowered. These measurements were taken with a small residual $\langle 100 \rangle$ stress of 0.03 GPa applied to the sample to maintain the thermal link to the pressure cell. An important feature of this log–log plot is its lack of evidence for any saturation effect at the low-temperature end; our lowest temperature is well into the region where Andres *et al* [9] observed such an effect. As these authors discussed themselves [9], it may well be that some form of sample heating was going on in their experiment at the low end of their temperature scale. The behaviour of the susceptibility, figure 1, is fitted by a $\chi = (\beta/T^{\alpha})$ dependence with $\alpha = 0.47 \pm 0.02$. In a similar experiment, although at higher temperature, Muruyama *et al* [5] found $\alpha = 0.66$ for $n/n_c = 0.55$. Andres *et al* [9] show that the full Curie temperature dependence of $\alpha = 1$ is not obtained until $n/n_c = 0.027$. The just-metallic data of Paalanen *et al* [6] shows a shallow concave-up curvature on a similar $\log \chi : \log T$ plot, but a best fit straight-line to the 4.2 K to 30 mK data for their $n/n_c = 1.09$ sample gives $\alpha = 0.54$ and their $n/n_c = 1.25$ sample has a much smaller slope, $\simeq 0.33$, in the same temperature range. In the later work [7], at $n/n_c = 0.78$, they find $\alpha = 0.65$. The Bhatt and Lee numerical analysis of the susceptibility [18] for a random Heisenberg antiferromagnet is plotted in figure 2, together with the known experimental data discussed above. It is noticeable, in extracting values of α from their theoretical curves, that there is always a slight, convex-up, curvature to their curves of $\log \chi : \log T$. It is very slight, and we have taken straight-line fits to their lowest-temperature data. In figure 2, there does seem to be a significant discrepancy between the theory and experiment at values of n just below n_c. This is not unexpected, given the expectation of dielectric divergence and wave-function expansion as the metal–non-metal transition approaches.

A noticeable extra feature of our data is the almost rigid constancy of the ESR signal intensity as a function of temperature; we highlight this in figure 3, where the line area is plotted against linewidth, with temperature as the implicit parameter. (The data here were all taken at low uniaxial stress.) The slope of this line is 0.96 ± 0.10! The spin-flip scattering rate scales with the susceptibility over almost three orders of magnitude in the

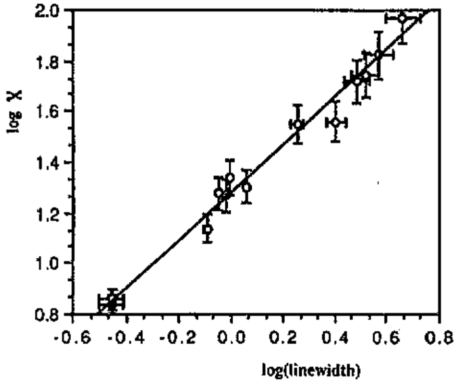

Figure 3. The correlation between ESR line area (again in arbitrary units) and linewidth, with temperature as the implicit variable. The linewidth has been defined as the value at half-maximum height. The gradient is 0.96 ± 0.10.

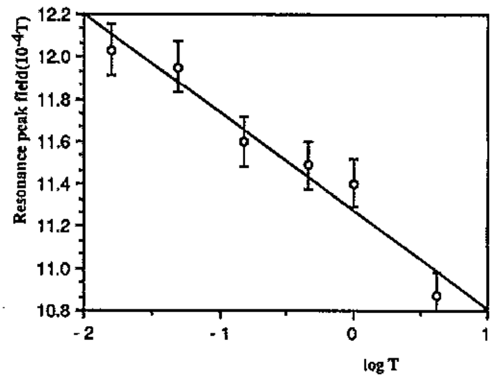

Figure 4. The field at which the resonance attains its maximum as a function of temperature. The full line is a guide to the eye.

temperature. In interpreting this data, it is difficult not to stray across the demarcation line of $n = n_c$, to make use of the theory [10] relevant to the metallic regime, even although our sample is just in the $n < n_c$ category. There a scaling of linewidth and susceptibility of just this type is predicted, with the broadening to low temperature diagnosed as a critical slowing down of the electron spin diffusion rate. A theory of interactions in disordered metals [19, 20] links the diffusivity of the electron spin and the electron spin susceptibility. We are to think, then, of the strong interactions as creating a clustering tendency (incipient localization) in the electron gas, at a stroke reducing electron spin diffusion and enhancing the electron spin susceptibility by giving it more local character. At the same time the reduced diffusion inhibits motional narrowing of the ESR linewidth and the line broadens. (The elementary possibility that we are looking at a dipolar coupled system, with the dipole moments having a temperature-dependent magnitude $\sim T^{-\alpha}$ has not escaped us; the magnitudes of the fields that arise from this mechanism are not big enough.) Is there any justification for using a theory for $n > n_c$ in considering an experiment where $n < n_c$? If there is, and we use it to explain linewidths for $n < n_c$, can the same theory also explain the susceptibility trends noted earlier? We have already noted some divergence between theory ($n < n_c$) and experiment ($n < n_c$) on the topic of electron spin susceptibility. We note also in the context of our initial questions that Alloul and Dellouve [21] have produced significant evidence that delocalized electrons, as shown by a metallic-like P^{31} NMR behaviour, exist at $n/n_c = 0.75$. They also demonstrate that large numbers of P^{31} nuclei, in samples with $n > n_c$, are seeing quasi-localized electron magnetic moments in their immediate environment. On the second question above, theory [19] ($n > n_c$) predicts a stronger temperature dependence $\chi \sim T^{-4/3}$ than we observe, although it is only fair to refer to the caveats inserted [19] about the dangers of extrapolating weak-coupling theory into regions of instability.

Sachdev and Bhatt [11] demonstrate that the Bhatt and Lee model [18] of interacting localized spins can explain ESR data in a system with $n < n_c$. It utilizes numerical simulation methods and gives a physical picture of the hopping of electron spins between sites being confined to smaller and smaller numbers of sites as the temperature is

Table 1.

Pressure (GPa)	Area (χ) (arbitrary units)	Peak intensity (mV)	Peak position (mT)	Linewidth (G)
0.03	73 ± 3	6.3 ± 0.3	1.05 ± 0.05	0.45 ± 0.03
0.10	75 ± 3	6.6 ± 0.3	1.04 ± 0.05	0.48 ± 0.03
0.15	69 ± 3	6.9 ± 0.3	1.00 ± 0.05	0.39 ± 0.03

lowered. More detail is needed to make a quantitative comparison between this theory and our experiment. The reduced number of hopping partners at $T \rightarrow 0$ K mirrors the reduced diffusivity of $n > n_c$ theories. There is some disagreement between our data on the scaling of the linewidth and the area of the ESR spectra and the data of Paalanen *et al* [7]. Their data was taken on a sample of $n/n_c = 0.78$. The comparison of the α-exponents is also rather different; in this work we derived a value of 0.47, whereas they quote a figure of 0.65. These differences may be correlated; our sample, being slightly more metallic, would be expected to have a smaller value of α, and if the temperature variation of the linewidth was much the same in the two samples, then their result, that the linewidth grows more slowly than α at the lowest temperatures, is derived from the faster variation of χ in their sample.

In figure 4, we show our measurements of the field at the resonance peak as a function of temperature. A possible explanation of this shift [11] as due to Breit–Rabi [22] coupling of the electron spin and the P^{31} nuclear spin has been advanced. The low-field ESR of the isolated P impurity resonates at $h\nu = \beta B$, i.e. it is shifted up-field, at constant frequency, from the normal resonance condition, $h\nu = 2\beta B$. At low temperature, Sachdev *et al* demonstrate that the reduction of range of electron tunnelling as T decreases is a possible cause of the shift up-field in resonance position. We have not attempted any quantitative evaluation of these measured shifts.

The stress measurements were taken at 15 mK, and consisted of positive field sweeps taken at three different stresses. A word first about our certainty that stress is being applied. The compressive stress was transmitted by the sample from the pressurizing diaphragm to a sensing diaphragm, linked to a capacitance plate. The measured stress was therefore stress transmitted through the sample. This stress-meter was calibrated with dead-weight measurements at 4.2 K before mounting it in the dilution refrigerator. The characteristics of the three spectra are assembled in table 1.

There has been [23] a stress–(n/n_c) calibration in Si : P for a very similar density from which we may deduce that n/n_c at 0.1 GPa in our sample is about 0.87 and at 0.15 GPa is 0.885. In an experiment [6] at n just greater than n_c a change of n/n_c from 1.25 to 1.09 can be estimated to produce a change of χ by a factor of 5 at 15 mK. This had therefore led us to expect a considerable change in χ under the modest stress of our experiment. Further, for Si : P, previous NMR and ESR studies have demonstrated that the change in the behaviour of the electron spin magnetization from pure Curie-type to pure Pauli-type occurs as the doping density is increased from about 10^{23} m^{-3} up to just over 10^{25} m^{-3}, i.e. by a range of two orders of magnitude in density. At zero stress and 10 mK, using the approximation that at any particular density the theoretical values are related by $\chi_{\mathrm{Pauli}} = \chi_{\mathrm{Curie}}(T/T_F)$, we can easily estimate that the susceptibility has to fall by more than three orders of magnitude over these two orders of density change. A simple linear modelling of this dependence on a log–log scale produces therefore a $\chi \sim n^{-3/2}$ behaviour in Si : P at this temperature. This is a lower limit, since our sample is in the middle of the density range considered, where changes will be at their most rapid. The effect of stress-tuning should also take into account the further feature that n is constant. In the analysis above χ *dropped* in spite of an *increase* in density. No such density increase occurs in the stress-tuning case; we may safely anticipate a $\chi \sim n_c^2$ dependence as a lower limit to the behaviour with respect to stress. A 4% decrease of n_c should produce at a minimum an 8% decrease of the susceptibility, and the change could be very much more (we estimate up to 20%). To summarize, then, we have experimental evidence that in the just-metallic density range the susceptibility is extremely sensitive to dopant concentration [6], whilst our own estimations of the potential effects of stress on χ in our sample imply a more modest, although still detectable, sensitivity.

The tabulated data in table 1 on the variation of the area of the ESR line (column 2) then represents a new result; χ is independent of stress, within experimental error, for stress just below n_c, i.e. at constant n, whereas χ is very sensitive to a minute change in n/n_c at constant n_c just above n_c. Packing the donors closer together does not produce the same effect as expanding the donor wavefunction, when using the magnetic susceptibility χ as the characteristic physical property. In this context we should remember that the electrical conductivity does appear to move with stress in much the same way as with density [3]. The electrical and magnetic properties of the interacting electron fluid do not move together in this system. The charge and spin degrees of freedom are uncoupled. Within the context of the Bhatt and Lee [18] model of the insulating side of the transition, not so many electron spin singlets are created by tuning from below with stress as are created by increasing the density of donors. However, it has to be admitted that the measurement of the ESR area in the stress experiment is not as yet totally convincing as a negative result. If we ignore the error bars on the measurement of area then the drop in the area at the highest stress is about equal to the minimum theoretical prediction. Clearly the experiment needs to be refined, most easily by repeating the measurement at a higher stress.

On the other hand, the fact that the line taken at the highest pressure has the smallest linewidth, is shifted to the lowest field and has the highest intensity is a convincing demonstration of the admixing of higher excited states, having reduced hyperfine interaction with the P nucleus, into the ground state by the stress. In an isolated impurity, however, a stress equal to our maximum stress, 0.15 GPa, would have reduced the hyperfine interaction by greater than 50%. Again the effect is less than expected.

4. Conclusion

In conclusion, we have explored to lower temperature than previous studies the electron spin susceptibility of a just-non-metallic Si : P sample. We produce evidence that down to 15 mK there is no saturation effect in the susceptibility, that it scales with temperature as $\chi \sim T^{-\alpha}$ with $\alpha \simeq 0.47$, and that the linewidth scales with temperature in exactly the same way. Comparison with theory is ambiguous; the best developed theory is for $n > n_c$ and it predicts the χ–linewidth correlation rather well, but fails to explain the temperature variation of χ. Finally the stress behaviour indicates a surprising independence of χ with respect to changes in n_c. Exchange and Coulomb interactions in this highly disordered system are not simply tuned by stress in the same way as they are tuned by density changes. A correct description of the just-localized electron gas may well have to be based on a non-percolating metallic-island model, rather than on the amorphous antiferromagnet model.

Acknowledgments

Throughout this work we have been generously helped by Dr D M Finlayson, both on technical experimental matters and through useful discussions. We also acknowledge gratefully the financial assistance of the SERC.

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