# Stress tuning of the magnetic properties of a near-metallic sample of silicon doped with phosphorus

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We report measurements down to 16 mK of the electron-spin resonance (ESR) of a sample of silicon doped with phosphorus, of doping density  $n/n_c$  (at ambient stress) of 0.85. Stress tuning to 0.28 GPa along the  $\langle 100 \rangle$  direction is used to "tune" the critical doping density  $n_c$  at the metal-nonmetal transition and thus vary  $n/n_c$  up to the value 0.91. The electron-spin susceptibility and ESR linewidth vary with temperature below 4.2 K in the same way, and the change in this variation with stress over most of the temperature range shows a strong magnetic-field and frequency dependence at the low fields of our experiments ( $\sim 10-20$  G). This implies that, as the metal-nonmetal transition approaches, the effect of magnetic field in polarizing the electron-spin pairs becomes rapidly weaker. However, at the lowest temperatures, with or without stress, a cusp develops in the susceptibility versus temperature behavior; we speculate that, so close to the metal-nonmetal transition, the exchange interactions are becoming sufficiently long range for spin-glass behavior to develop. Alternatively, the "rare-spins" scenario, coupled by Ruderman-Kittel-Kasuya-Yosida interactions, of Bhatt and Fisher may be driving the spin-glass behavior. At the low end of the temperature range only about 7% of the electron spins remain magnetically active. The rate of variation of the susceptibility with stress (which tunes  $n_c$ ) at the lowest temperatures is rather fast, with  $\chi$  varying as  $n_c^4$ .

#### **INTRODUCTION**

For many years the metal-nonmetal (MNM) transition in silicon doped with phosphorus has been the proving ground for competing theories of the mechanisms of the transition.<sup>1,2</sup> There is a recent review.<sup>3</sup>

The interpretation of the low-temperature electrical properties is controversial,<sup>4</sup> and the magnetic properties on the metallic side are anomalous, the susceptibility being not at all Pauli-like.<sup>5</sup> Similarly the magnetic and electric properties on the insulating side of the transition are unusual<sup>6,7</sup> and difficult to interpret. The possibility that the unusual electrical properties are driven by the magnetic interactions between the electron spins has long been questionable (see, for example, Finlayson<sup>2</sup>).

Our particular focus has been on understanding the buildup of magnetic interactions between the donor electrons as the donor density increases towards the metalnonmetal transition on the insulating side. In the lowdensity limit these magnetic properties should resemble a set of noninteracting, Curie, moments, with  $\chi$ , the electron-spin susceptibility, equal to C/T, where C is the Curie constant and T the temperature. At densities of P donors around  $10^{17}$  cm<sup>-3</sup> this behavior is observed.<sup>8</sup> As the density of donors increases, exchange interactions between donors build up, electrons can no longer be associated with individual sites, and for a fixed temperature the susceptibility dependence on n, the donor concentration, becomes weaker. Bhatt and Lee<sup>9</sup> have formulated a theory of the susceptibility in this concentration range leading up to the metal-nonmetal transition, based on renormalized interacting pairs. They<sup>9</sup> do not suggest any upper limit to the donor concentration for the applicability of their theory, except to indicate that it should not be relevant for  $n > n_c$ , where  $n_c$  is the critical density for the MNM transition in Si:P;  $n_c$  takes the value at ambient uniaxial stress of  $3.75 \times 10^{18}$  cm<sup>-3</sup>.

As the donor density increases into the metallic region, the susceptibility eventually becomes Pauli-like, although there is an extensive range of n before that where non-Fermi-liquid behavior is exhibited, with the susceptibility strongly temperature dependent while the system is metallic.<sup>5</sup>

A few years ago we published<sup>10</sup> a largely negative report on the effects of uniaxial stress on the magnetic properties of a sample of Si:P, doped to  $n = 3.2 \times 10^{18}$  cm<sup>-3</sup>, at mK temperatures. At stresses up to 0.15 GPa and temperatures down to 16 mK we were unable to confirm any significant effects of uniaxial stress on the susceptibility of the sample. Stress is known to "tune" the critical density  $n_c$  by expanding the extent of the donor wave function.<sup>11</sup>

A further feature of our previous paper<sup>10</sup> was that, at any stress, we could find no low-temperature evidence for deviation from a power-law dependence of  $\chi$  on temperature,  $\chi \sim T^{-0.47}$ , although other studies,<sup>8</sup> in similar concentration and temperature ranges, had measured such a low-temperature deviation.

In this paper, we readdress those former experiments with greater experimental accuracy and higher uniaxial stresses. We confirm many of the features of our previous paper, <sup>10</sup> but show that there is a significant dependence of the susceptibility on stress, and that there is a small cusplike feature in  $\chi$  at the lowest temperatures.

In addition we include some measurements of the frequency dependence of the data, and an absolute calibration of the magnitude of the susceptibility. The consequences of these measurements are explored in the context of available theories of the strongly interacting, near-metallic, state.

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Three papers provide the theoretical framework for the analysis of our experimental results.<sup>9,12,13</sup> Bhatt and Lee<sup>9</sup> provide a description of the highly disordered spin- $\frac{1}{2}$  antiferromagnet. The theory involves a hierarchical scheme of pairwise exchange interactions, with each pair having a ground-state singlet and an excited-state triplet, and successive pairs get "frozen-out" into their ground-state singlet as the temperature is lowered. Clusters of spins become effectively spin pairs via a renormalization technique.<sup>9</sup> The biggest exchange interactions get frozen-out at the highest temperatures, corresponding to the most closely coupled pairs. As the temperature lowers then increasingly more distant pairs get frozen-out. The numerical solutions<sup>9</sup> are presented as  $\log_{10}\chi:\log_{10}T$  curves for four different donor densities,  $n/n_c = 0.06$ , 0.25, 0.62, and 2.46. The  $\log_{10}\chi$ : $\log_{10}T$  curves are approximately linear; we have fitted the lowest temperature points9 to derive values of  $\alpha$  in  $\chi \sim T^{-\alpha}$  for each of the four densities (Table I).

We can be sure that as  $n/n_c \rightarrow 0$ ,  $\alpha \rightarrow 1$ , and that as  $n/n_c$  becomes large  $\alpha \rightarrow 0$ . (Clearly the value in Table I for  $n/n_c = 2.46$  is physically unreasonable; experimentally in this density range the variation of  $\chi$  with T is very slow, corresponding to very small  $\alpha$ .) For the three concentrations  $n/n_c = 0.06$ , 0.25, and 0.62,  $\alpha$  appears to vary approximately linearly with  $n/n_c$ , Fig. 1. Furthermore, the predicted values are corroborated by experiment.<sup>6,8</sup> The slope of Fig. 1 yields  $[d\alpha/d(n/n_c)] = -0.42$ , giving us a clear target for our stress experiment, if the linearity at Fig. 1 can be extended into the density range of our sample,  $n/n_c \sim 0.85$ .

The important predictions of Bhatt and Lee<sup>9</sup> are (1) the slopes of the  $\chi$ :*T* curves for several concentrations, (2) the distribution of *J* couplings for  $n/n_c = 0.22$ , showing that a significant number exceed 100 K, and (3) there should be no magnetic-ordering transition in this system, if the interactions are short range.

The other two theory papers<sup>12,13</sup> adapt the techniques of Ref. 9 to a more detailed understanding of the electron-spin-resonance spectra observed in insulating doped semiconductors. Since electron-spin resonance (ESR) is the experimental method used in our study for observation of the electron-spin susceptibility, <sup>14</sup> these papers are important and enable us to glean more information from the spectra than just the electron-spin susceptibility. Briefly, the authors<sup>12,13</sup> prove that the essential factor in deciding the linewidth varies as  $N^{-1/2}$ , where N is the number of P nuclei in the hopping cluster. The observe hopping spins are, of course, those not yet locked

TABLE I. The theoretical values of the exponent  $\alpha$  from Bhatt and Lee (Ref. 9). These are also plotted in Fig. 1.

$n/n_c$	α
0.06	-0.85
0.25	-0.75
0.62	-0.62
2.46	-0.515



FIG. 1.  $n/n_c$  plotted against the power-law exponent  $\alpha$ . The line is a linear fit to three points from Bhatt and Lee (Ref. 9) and the extrapolation takes the graph into the density region of our sample.

into their ground-state singlet by temperature. In this model, particular cluster sizes have particular dependences on ESR frequency, when that frequency v is of the order of the hyperfine coupling A between the phosphorus nucleus and the electron spin (A is 120 MHz in Si:P). Unfortunately numerical solutions are required; particular examples are shown in Refs. 12 and 13. Nonetheless certain semiquantitative predictions relevant to our measurements can be gleaned from these papers.

(i) When the Zeeman frequency of the ESR experiment is 60 MHz, the predicted behavior<sup>12,13</sup> of the linewidth is that it should vary as  $A/(2\sqrt{N})$ .

(ii) At the lower ESR frequency (30 MHz) of our measurements, the theory predicts substantially narrower linewidths than at 60 MHz, perhaps 2 G smaller at cluster size N = 4, about 1 G smaller at cluster size N = 20.

(iii) There is clearly a low-temperature cutoff beyond which the ESR linewidth can increase no further. This is associated with the minimum size of a cluster. Thus the linewidth can never exceed about 20 G.

#### EXPERIMENTAL DETAILS

We present ESR measurements at temperatures from 15 mK to 4.2 K in a sample of Si:P of doping density  $3.2 \times 10^{18}$  cm<sup>-3</sup>. This is the same sample as used in our earlier work.<sup>10</sup> All details are as before,<sup>10</sup> except that we access a different range of uniaxial stress. Here our range is from 0.06 to 0.28 GPa; this tunes  $n/n_c$  from 0.86 to 0.91,<sup>14</sup> a reduction of  $(n_c - n)/n_c$  of about 35%.

The ESR absorption, taken at two frequencies 30 and 60 MHz, is observed by sweeping the Zeeman magnetic field, typically from -50 to 0 to 50 G and down again. Signal averaging is needed. The integrated ESR response then gives directly the electron-spin susceptibility.<sup>15</sup>

An important new feature is the calibration of the absolute magnitude of the spin susceptibility at 4.2 K. A few milligrams of DPPH (supplied by Aldrich) powder was carefully weighed into a gelatin capsule and its magnetic properties measured on the superconducting quantum interference device (SQUID) at the Royal Institution. Dr. M. Dalton kindly ran these experiments for us. He used a temperature range of 1.5-20 K to calibrate C in  $\chi = C/T$ .

Using a larger amount of the same DPPH powder, set in a quartz tube of 1 mm diameter such that the powder occupied 7 mm of the length of the tube, an ESR sample was formed. With powder taken from the same wafer as our experimental Si:P sample, an identical tube was filled to the same dimensions. These sample sizes more than filled the ESR coil, which was 6 mm long, so that, during the ESR experiment, the question of how much of the sample is activated by the radio frequency is uncertain; however, because of the identical geometries of the samples with respect to the coil, we can be sure that the same fraction is activated in the Si:P as in the DPPH experiment. The final link in the chain of the calibration is closed by the relative weights of the DPPH and Si:P ESR powders, by the molecular weights of DPPH and Si:P, and by running ESR spectra at 4.2 K at 62 MHz in the spectrometer for both DPPH and Si:P.

A plausible objection to this procedure is that the powdering process introduces paramagnetic impurities into the Si:P. However, at 4.2 K, the ESR spectra from the powder are almost identical to those obtained from the single crystal, in shape and in width. A small difference in width of about 20% we ascribe to one of two reasons: (i) the two samples, powder and crystal, are cut from the same wafer, but in this concentration range the width is a very sensitive function of concentration, so that a very small change of concentration across the wafer could lead to such a change in width, and (ii) the single-crystal measurements are taken with at least a low stress applied, whereas the powder measurements are, of course, taken at zero stress, and this low stress produces its own narrowing effect. It seems implausible, therefore, that any paramagnetic centers introduced from the powdering process would have such identical ESR spectral characteristics.

If R is the integrated ESR signal at 4.2 K observed when DPPH is in the rf coil of the spectrometer, r milligrams of DPPH are used in this sample,  $R_{mol}$  is the molecular weight of DPPH,  $\chi^{R}_{mole}$  is the molar susceptibility of DPPH from the SQUID measurement, and f is the fraction of sample activated in the ESR experiment, then

$$R \propto (fr\chi_{\rm mole}^R)/R_{\rm mol}$$

Mutatis mutandis, we can write down for the silicon experiment

$$S \propto (f_S \chi^S_{\text{mole}}) / S_{\text{mole}}$$

The proportionality constant is determined by the gain of the electronic circuits, and will be the same in the two experiments. Thus

$$\frac{R}{S} = \frac{r}{s} \frac{S_{\text{mol}}}{R_{\text{mol}}} \frac{\chi_{\text{mole}}^{R}}{\chi_{\text{mole}}^{S}}$$

and a value of  $\chi^{S}_{mole}$  emerges.

The dilution refrigerator used in the ESR experiments was an Oxford Instruments 400 (bottom loading) instrument.



Field (gauss)

FIG. 2. The ESR spectra at four representative temperatures.

### THE EXPERIMENTS

Figure 2 shows some representative ESR absorption spectra, taken at low (30 MHz) frequency and low pressure. (Because of thermal contraction of the probe our low pressure is actually 0.06 GPa.) The broadening and shifting as the temperature cools are clearly visible. Another feature of Fig. 2 is the constancy of the intensity; this means that the width of the ESR is approximately proportional to the integrated area, if the line shape does not change (and it does not appear to do so).

Figure 3 shows the effect of stress; the ESR absorption at 18 mK is narrowed by the high stress, and again the data are marked by a constancy of the intensity.



FIG. 3. The pressure dependence of the ESR. The broader spectrum was taken at a uniaxial stress of 0.06 GPa. The narrower spectrum was taken at a uniaxial stress of 0.28 GPa. Both spectra were taken at 30 MHz and 18 mK.



FIG. 4. The ESR linewidth against temperature, on a log-log plot to base 10. Both sets of data were taken at 30 MHz, the filled symbols at 0.28 GPa, and the open symbols at 0.06 GPa.

Figure 4 shows, on a log-log plot, the variation of ESR absorption width with temperature at the lowest uniaxial stress, and at the maximum stress. Both low- and highstress data show power-law behavior down to 30 mK; the effect of stress is greatest ( $\sim 20\%$ ) at the low end of the temperature scale, although it is still significant ( $\sim 10\%$ ) even at 4.2 K, and both sets of data exhibit a cusplike feature at the very lowest temperatures, around 20 mK. This last feature is perhaps our most exciting result; realizing its importance we have spent some time verifying its validity. The effect is small, right at the extreme end of our temperature range; if the cusp indicates a new phase of the random  $S = \frac{1}{2}$  antiferromagnet then we only have access to a few mK of that phase. As far as can be experimentally determined, the temperature of the cusp is the same at the low as at the highest pressures.

Figure 5 is a log-log plot illustrating the width variation with temperature at low pressure but at two frequencies, i.e., the Zeeman frequency of the ESR spectrometer was set to 30 and 60 MHz. Here the indication is that there is no effect of frequency at the high end of the temperature range but that a frequency dependence opens up at the low end of the temperature scale. The cusp feature is approximately independent of frequency.

Finally we present the result of our calibration experiment; at ambient pressure and 4.2 K the susceptibility of our sample  $n = 3.2 \times 10^{18}$  cm<sup>-3</sup> is within experimental error the same as that of a noninteracting set of spins with  $\chi^{\text{Curie}} = C/4.2$ ,  $C = n\beta^2/k$ . More precisely  $\chi^{\text{sample}}/\chi^{\text{Curie}} = 1.04 \pm 0.05$  at ambient stress. The ESR measure-



FIG. 5. The ESR linewidth against temperature, on a log-log plot to base 10. Both sets of data were taken at 0.06-GPa stress. The closed symbols represent 30-MHz data, the open symbols 60-MHz data.

ments for the calibration were undertaken at a frequency of 62 MHz.

### DISCUSSION

In our density range  $n/n_c = 0.86 - 0.91$ , theory<sup>9</sup> predicts an approximate power-law behavior. If we leave out points below 30 mK, where the cusp is starting to form, then we do observe power-law behavior in the ESR width (Fig. 4) and in the electron-spin susceptibility (not shown because its behavior is so similar to that of the ESR width since the intensity remains roughly constant). If we do linear fits to our data points above 30 mK, the exponent parameters shown in Table II emerge. The error bars are generally bigger on the susceptibility exponents because integrating the area of the ESR line depends rather critically on clear base-line identification, and this is not always easy for broad ESR lines. We include in Table II the exponent  $\alpha$  values obtained from the linear extrapolation of the theoretical calculations, Fig. 1.

We have analyzed the ratio of the susceptibilities at low and high stress and plotted the logarithm of this ratio against the logarithm of the temperature, for T > 30 mK. If  $\chi \sim T^{-\alpha}$  and  $\chi' \sim T^{-\alpha'}$ , then  $\chi/\chi' \sim T^{\alpha'-\alpha}$ . The slope of the best-fit straight line gives  $\alpha' - \alpha = 0.010 \pm 0.007$ , and this corresponds to a slope  $d\alpha/d(n/n_c)$  $= -0.20\pm0.14$  for the 30-MHz data, while at 60 MHz this slope becomes  $-2.0\pm0.4$ . (The error bars in the analysis are based on the standard statistical formula for linear regression fits.)

TABLE II. The exponent  $\alpha$ , from the experimental graphs of log width and  $\ln \chi$  against  $\ln T$ , and from Bhatt and Lee (Ref. 9) via the extrapolation in Fig. 1.

Frequency (MHz)	Slope of ESR parameter	Lowest stress	Highest stress
30	Width	$0.50 {\pm} 0.01$	0.49±0.01
30	Area	$0.45 {\pm} 0.01$	$0.41 \pm 0.04$
60	Width	$0.55 {\pm} 0.05$	$0.52 {\pm} 0.03$
60	Area	$0.57 {\pm} 0.01$	0.47±0.04
Theory	Theory	0.52	0.50



FIG. 6. The magnetic-energy levels of an exchange-coupled spin pair as a function of magnetic field. The vertical arrows drawn at a particular field illustrate the developing difference between energies of intratriplet transitions as the field increases.

We compare these experimental values of the dependence of  $\alpha$  on stress with the extrapolated theoretical value predicted,  $d\alpha/d(n/n_c) = -0.42$  (Fig. 1). At the lower frequency the experimental value is significantly smaller than the extrapolated theoretical prediction, while at the higher frequency the opposite holds. The first point to make about this disparity is that some frequency dependence is expected in this parameter, which we now discuss.

For ease of interpretation, we show in Fig. 6 the typical field dependence of the singlet-triplet energy-level diagram of an electron-spin pair, interacting with a particular J coupling. Clearly a magnetic field B, such that  $\beta B \sim J$ , brings the lowest triplet level down towards the ground-state singlet; thus an already frozen-out pair could be unfrozen by application of a magnetic field. This is the origin of the larger susceptibilities at 60 MHz compared to 30 MHz (again intensities of the ESR lines are roughly constant, so that linewidth data, Fig. 5, provide a good representation of susceptibility data also). Similarly, for the unfrozen, still magnetically active, spin pairs, with J coupling  $\langle kT$  and the excited triplet occupied, the effect of the larger magnetic field, Fig. 6, is to produce a larger difference between the two splittings within the triplet; it is these two splittings that form the basis of the broadening of the ESR. In a random system there will be a juxtaposition of many such splittings, so that the effect of the splitting will appear as a broadening on a single, inhomogeneous, resonance line. The effect of frequency and magnetic field is thus similar for both the width and the susceptibility. It is also easy to see that at 4.2 K, if no spins are frozen-out, as our calibration indicates, then the effect of higher frequency on the unfreezing of the susceptibility cannot function.

Looking at Table II, it is clear that the root of the widely differing values of  $d\alpha/d(n/n_c)$  at the two frequencies appears to be the substantially greater effect of increased field and frequency at the lowest stress, i.e., the "unquenching" generated by the field is strongest here. It may be that the driving mechanism for the effect is

particularly sensitive to the proximity of the metalnonmetal transition, with the data taken at  $n = 0.91n_c$ being much less sensitive to magnetic field and frequency than the data taken at  $n = 0.86n_c$ .

However, if this slope  $d\alpha/d(n/n_c)$  can vary by an order of magnitude between 30 and 60 MHz the question arises as to what is the true experimental value to compare with theory in the density region of our sample. We need a value of the slope where the unquenching effects are negligible, which would indicate a need for measurements at even lower frequencies than 30 MHz. The value taken at 30 MHz is thus to be regarded as an upper limit to the experimental value, and the conclusion must be that the variation of  $\alpha$  with  $n/n_c$  is significantly slower in our sample than in samples at lower densities and significantly slower than extrapolated theoretical prediction.<sup>9</sup>

Another important feature of the data is the calibration of the absolute spin susceptibility. Apparently, at ambient stress, 0% of spin pairs are frozen-out<sup>9</sup> at our sample density at 4.2 K, in measurements taken at 62 MHz (no frequency dependence would be expected at this high temperature, and Fig. 5 provides experimental support that there is none). Bhatt and Lee<sup>9</sup> show a  $P(\ln J)$ against  $\ln J$  curve for a sample of density  $8 \times 10^{17}$  cm<sup>-3</sup>. about a quarter of the density of our sample, and on this curve 4.2 K is the temperature at which half of the spins would be frozen-out. Clearly at the substantially higher density of our sample a much larger fraction of spins should be frozen-out at this temperature. Given the rather accurate predictions of the theory<sup>9</sup> with respect to the temperature variation of the susceptibility, this quantitative failure is surprising. Apparently the very strong Jcouplings, which random siting of P impurities would predict, are absent. Anticlustering of P impurities would be one explanation. Alternatively theory breaks down for the close pairs of P impurities; it is still surprising, however, that such close pairs continue to contribute a Curie component to the susceptibility.

Before considering the sophisticated theories of the linewidth<sup>12,13</sup> we may attempt a semiquantitative check on the earlier qualitative understanding of the effect on the linewidth of increased magnetic field. The data of Fig. 5 at, say 30 mK, show a width at 60 MHz of 8.8 G and at 30 MHz of 7.2 G. Now 30 mK expressed as a frequency is equivalent to 600 MHz. At a Zeeman field of 11 G (30 MHz) the Breit-Rabi equations<sup>16</sup> predict a twoline spectrum split by 0.6 G for a pair with J = 600 MHz, while at 22 G (60 MHz) this splitting rises to 2.2 G. Of course, as J decreases, these figures will increase [e.g., at J = 300 MHz the equivalent splittings are 1 G (30 MHz) and 4.4 G (60 MHz)]. Since the sample should contain magnetically active pairs with J couplings ranging from 600 MHz downwards, it seems therefore that the fieldinduced broadening is rather small compared to that predicted by the simplest pair model.

Moving on to the Sachdev and Bhatt<sup>12,13</sup> theories of linewidth we can use the prediction that at 60 MHz the linewidth should be close to  $A/(2\sqrt{N})$ , to predict that at 30 mK the cluster size over which the hopping electron is moving has  $N \sim 6$ . At 100 mK,  $N \sim 25$ . The frequency

TABLE III. A comparison of linewidths (in G) between the data of Muruyama, Clark, and Sanny (Ref. 6) on a sample of donor doping intensity  $n=2.05\times10^{18}$  cm<sup>-3</sup> and this work,  $n=3.5\times10^{18}$  cm<sup>-3</sup>.

T (mK)	Muruyama, Clark, and Sanny (Ref. 6)	This work (low stress)	
30	9	8.8	
100	5.5	4	
1000	3	1.1	

dependence of the linewidth closely matches the analysis.<sup>12,13</sup>

Another comparison of linewidths can be made with the data of Muruyama, Clark, and Sanny<sup>6</sup> on a sample of  $n=2.05\times10^{18}$  cm<sup>-3</sup>. Table III shows the comparison. As a general rule, the higher doping density of our sample should have a much narrower line. One must also take into account that the stress applied, even though low, is sufficient to narrow the line. However, at low temperature the widths are much the same, perhaps indicating that, as the temperature falls, the residual exchange couplings of the still magnetically active pairs are similar no matter what the overall density of donors.

The temperature variation in the susceptibilities and widths implies that, if all spins are contributing at 4.2 K to the magnetic properties, then since  $\gamma$  (low stress)  $\sim T^{-0.45}$  only about 7% of the spins are contributing at 30 mK. [However, the statement at 4.2 K that all spins are contributing refers to ambient pressure. Since at 4.2 K the difference in susceptibilities at the two stresses (high and low) is about 10%, we may anticipate that perhaps only 97% of the spins are contributing to the magnetization at our lowest stress. This small correction at 4.2 K means little to the estimate for 30 mK.] Thus at 30 mK only the tail of the  $P(\ln J)$ : J curve is still magnetically active, corresponding to spins with weak couplings, and pairs of large separation. The sample at this temperature can be described as a dilute density of spin triplets bathed in a sea of spin singlets.

The cusp at and below 25 mK is present in both the integrated area and the linewidth (Fig. 4). Theoretical calculations<sup>9</sup> rule out any spin-glass behavior for shortrange interactions. This leaves the possibility that for  $n=3.2\times10^{18}$  cm<sup>-3</sup> the interactions become long range and spin-glass behavior emerges. Alternatively, Bhatt and Fisher<sup>17</sup> have developed a model where the competition between ferromagnetic and antiferromagnetic interactions drives the system towards a spin-glass phase. Unfortunately it is difficult to experimentally confirm spin-glass behavior when only 4 or 5 mK of the frozen phase is accessible (16 mK is our low-temperature limit).

We have discussed earlier<sup>10</sup> the expected variation of susceptibility with uniaxial stress close to the metalnonmetal transition. By the application of stress the sample value of  $n_c$  is varied at constant n. As a lower limit we<sup>10</sup> estimated that  $\chi$  might vary as  $n_c^2$  at 10 mK in this sample. Our measurement at 20 mK shows a 20% decrease of  $\chi$  for a 5% decrease of  $n_c$ ; thus  $\chi \sim n_c^4$  is the experimental prediction for the power-law dependence of the susceptibility on  $n_c$  close to the metal-nonmetal transition. Again, the proximity of the transition is evident, in that the observed sensitivity to stress is substantially greater than the calculated lower limit.

#### CONCLUSION

We have carefully measured, via the power-law dependence of the spin susceptibility on temperature, the uniaxial-stress dependence of the coefficient  $\alpha$  in a randomly doped semiconductor. The system is perhaps a realization of the random  $S = \frac{1}{2}$  antiferromagnet. Since the sample density *n* is  $0.85n_c$ , there is some doubt as to whether theory based on more dilute regimes will be applicable; we find that experiment so close to the metalnonmetal transition demonstrates a weaker dependence of  $\alpha$  on  $n/n_c$  than the extrapolated theory.

A calibration of the susceptibility in our sample indicates that the full Curie susceptibility in our sample is measured at 4.2 K and that it is only below this temperature that the susceptibility grows more slowly as T decreases. For the random antiferromagnet the couplings depress the susceptibility below the Curie value only below 4.2 K at this high density of donors. By 30 mK only about 7% of the spins remain magnetically active.

The frequency dependence of the ESR spectra appears to follow expectation for the random  $S = \frac{1}{2}$  antiferromagnet at least qualitatively, although the stress coefficient  $d\alpha/d(n/n_c)$  exhibits a very strong frequency dependence at the low fields and frequencies of these experiments. Close to the metal-nonmetal transition it appears that the effects of increased magnetic field and frequency are suppressed.

Finally, the appearance of a cusp in the susceptibility against temperature, but only at the lowest temperature, perhaps indicates that the range of the interactions between spins becomes long as n approaches  $n_c$ , and a spin-glass state<sup>9</sup> emerges. Another possibility is that the "rare-spin" ideas of Bhatt and Fisher<sup>17</sup> can be carried over from the just-metallic to the just-nonmetallic regime. There, the competition between ferromagnetic and antiferromagnetic interactions drives the system towards a spin-glass phase.

Overall, we can therefore highlight three particular features that indicate that the density of our sample makes its magnetic behavior qualitatively and quantitatively different from the strongly interacting, random,  $S = \frac{1}{2}$  antiferromagnet: (i) the gradient of  $\alpha$  with respect to  $n/n_c$  is weaker, (ii) the exchange interactions are weaker, as evidenced by the calibration experiment, and (iii) the appearance of the cusp indicates that the exchange has taken on a longer-range character.

Note added in proof. We have just completed a further check on the susceptibility of the powdered Si:P sample, measuring its susceptibility in a SQUID magnetometer between 150 and 170 K. This is a final verification of our calibration procedure. Unfortunately, the sample shows a paramagnetic susceptibility at high temperature, and its temperature dependence in the helium temperature range is much faster than our single-crystal measurements in the same temperature range. Accordingly we now believe that the powdering process during the calibration has introduced further paramagnetic defects. None of our single-crystal measurements are affected by this correction.

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