²⁹Si spin relaxation in Si:P and Si: (P,B) near the metal-insulator transition

M. J.R. Hoch* and D. F. Holcomb

Laboratory of Atomic and Solid State Physics and Materials Science Center, Cornell University, Ithaca, New York 14853

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The spin-lattice relaxation time T_1 of ²⁹Si nuclei in Si:P near the metal-insulator transition is known to have a marked magnetic field dependence. Various models have previously been proposed to account for this behavior. We present new measurements on a dilute sample with electron concentration well below the metallic range and on compensated material, Si:(P,B). We show that these data, as well as previous measurements, favor a local-moment model for the electron-spin relaxation agents rather than a Fermi-liquid model, even for samples just on the metallic side of the transition. The model, which includes effects of nuclear-spin diffusion, electron spin-spin interactions via the exchange coupling J, and frozen spin pairs, can account for the main features of the available data and connects observed temperature dependencies of T_1 and the spin susceptibility.

INTRODUCTION

The magnetic properties of disordered systems undergoing metal-insulator $(M-I)$ transitions have been the subject of considerable interest in recent years. Si:P in particular has been studied¹ using magnetic susceptibility and magnetic resonance methods over a range of applied magnetic fields and temperatures. The primary goal of that work was an increased understanding of the electronic structure and the electron spin dynamics of M-I materials in the critical region near the transition.

Paalanen, Ruckenstein, and Thomas² (PRT) have measured the ²⁹Si NMR spin-lattice relaxation rate $1/T_1$ in Si:P for values of the donor-electron concentration n_n near the critical value n_c as a function of temperature $(T < 200$ mK) and magnetic field $(B_0 < 2$ T). The relaxation rate was found to increase significantly with decreasing magnetic field for both insulating $(n_p = 0.09n_c)$ and just metallic $(n_p = 1.03n_c)$ samples. A dependence on temperature was also observed. PRT suggest that either of two classes of models which invoke strong electronspin effects might be used to explain these results. One involves localized spins and the other a metallic approach with spin correlations in an electron fluid. They favored the second approach which implies a drastic slowing of spin diffusion compared to charge diffusion. The field dependence of the relaxation is explained in terms of a distribution of correlation times for spin fluctuations which are linked to strong Coulomb correlations in the disordered electron fluid. Gan and Lee³ have put forward an alternative theory which accounts for the field dependence on the basis of coupled pairs of local electronic moments as a source of nuclear relaxation. No decisive test of the correctness of the various theoretical models has been possible with the available data.

We have carried out further 29 Si relaxation measurements on both uncompensated (Si:P) and compensated [Si:(P,B)] samples as a function of concentration and applied magnetic field at temperatures in the 1.3—1.5-K range. In addition, calculations based on a local-moment model have been carried out which allow for spindiffusion effects in the ^{29}Si spin system. Our results support the local-moment description and also suggest that exchange mediated spin-spin couplings between the electron spins are important in determining the spincorrelation function over a wide range of impurity concentration. The evidence points away from the applicability of models based on a homogeneous, strongly interacting electron gas. The model we propose can explain in a semiquantitative way the dependence of $1/T_1$ on n_p , B, and T over a wide range of conditions in the insulating phase and offers insight into the behavior in the just-metallic region as well.

EXPERIMENTAL DETAILS AND RESULTS

Measurements were made using pulsed NMR methods at a frequency of 8.45 MHz in a field of ¹ T. Low-field relaxation times were obtained using a field cycling procedure in which the magnetic field was adiabatically reduced to a chosen value in a time short compared to the low field T_1 ⁴ After allowing relaxation to proceed for a measured time interval, the field was adiabatically increased to 1 T where the 29 Si magnetization was measured. With the available magnet the shortest relaxation times which could be reliably determined were about 100 s. The samples consisted of \sim 150- μ m grains obtained by crushing a disk of material that had been characterized using room-temperature resistivity measurements together with the Mousty⁵ scale and, in the case of the compensated samples, additional neutron activation and Hall coefficient measurements.⁶

The present results are shown in Fig. ¹ together with a curve based on data obtained previously on an uncompensated sample $(n_p/n_c=0.68)$ by Jerome, Ryter, and Winter JRW).⁷ Two data points obtained by Jerome and Winter⁸ on a dilute, uncompensated sample are also plotted. Ikehata, Sasaki, and Kobayashi⁹ have also obtained field-dependent T_1 results for metallic Si:P ($n_p/n_c > 3$) at 0.55 K, but these are not reproduced here. As can be seen from Fig. I, results for all samples with

FIG. 1. Field dependence of the relaxation rate $1/T_1$ in Si:P and Si:(P,B) at low temperatures, for various values of the electron concentration *n* below and above the critical value n_c . Data for two samples are taken from Refs. 7 and 8, as labeled. The other curves give our data for four samples.

 $0.22 < n_n / n_c < 1.5$ show a strong increase in relaxation rate as the field is lowered. The increase is similar in form to that found by Paalanen et $al.$ ² in Si:P at lower temperatures. For $n_p / n_c \gg 1$, the field dependence disappears⁹ and samples obey the Korringa relation which applies to metals.

THE NUCLEAR-RELAXATION MODEL

The data of Fig. ¹ strongly suggest that a common, underlying mechanism is responsible for the field dependence of T_1 in all samples. Of particular interest is the fact that the curve for the sample with $n_p/n_c = 0.03$ mimics those with values of n_p near or just above n_c . The spin susceptibility for the sample with $n_p/n_c = 0.03$ lies close to the Curie-law prediction. ¹⁰ The spins may be regarded as weakly interacting and in a first approximation as isolated and fixed in position. The model we shall describe is cast in terms most applicable to this sample. After developing the model we then examine the implications of pushing its applicability into the range with n_p near n_c . Of further interest is the similarity in the behavior of the compensated and uncompensated materials. This similarity may be interpreted as providing support for the localized-moment model of nuclear relaxation rather than the Fermi-liquid approach, since compensation is expected to enhance the Anderson character of the transition and to change the properties of the electron fluid from those found in uncompensated material.

 29 Si may be directly coupled to paramagnetic centers by the hyperfine coupling \overline{H}_{hf} or by the dipolar coupling H_D . While one can show that the hyperfine coupling in Si:P exceeds the dipolar coupling at distances up to 70 \AA , it is nevertheless likely that the dipolar coupling is primarily responsible for relaxation of nearby 2^{5} Si nuclei at least for $n_p/n_c < 1$. This situation arises because H_D contains spin operators of the form S_zI^{\pm} which allow nuclear (*I*) flips unaccompanied by electron (*S*) flips whil H_{hf} contain spin operators $S^{\pm}I^{\mp}$ which produce simul taneous spin reversal. In the latter case energy conservation requires that there be closely spaced electron energy levels with energy difference $\Delta E \sim \hbar \omega_s$. Such levels will not, in general, be available with localized spins. Gan and Lee³ have proposed a model in which states closely spaced in energy arise through accidental neardegeneracy of singlet and triplet levels of an exchangecoupled pair of electrons. For their model, a broad distribution of values of J_{ij} , the exchange coupling between electron spins i and j , is assumed. They did not take the need to use nuclear-spin diffusion in the inhomogeneous system into account, however. It is well known that localized paramagnetic impurities in insulating materials often produce relaxation of the surrounding nuclear spins by means of an indirect mechanism involving nuclearby means of an indirect mechanism involving nuclear
spin diffusion.^{11,12} We believe Gan and Lee have overes timated the effectiveness of their mechanism. It appears likely to us that the electron-nuclear dipolar coupling will be of dominant importance in determining the field dependence of T_1 , at least for $n_p < n_c$.

Two limiting cases in which nuclear-spin diffusion plays a role in relaxation have been distinguished; one in which the spin-diffusion rate is the bottleneck for relaxation ("diffusion limited"), and one in which relaxation to the paramagnetic impurities is the bottleneck ("rapid diffusion"). The criterion to distinguish between the two cases has been given by Rorschach¹³ in the form of a dimensionless ratio $\delta = \beta^2 / 2b^2$ where $\beta = (C/D)^{1/4}$ is the radius outside of which spin diffusion is more efficient than direct relaxation, and b is the diffusion barrier inside which spin diffusion is blocked by large magnetic-field gradients produced by the impurity spin. For diffusion limited relaxation, $\delta > 1$, while for the rapid diffusion case, δ < 1. Treating dipolar coupling between paramagnetic centers and relaxing nuclei as the dominant relaxation mechanism gives

$$
C = 2/5\gamma_S^2 \gamma_I^2 \hbar^2 S(S+1) f(\omega_I) , \qquad (1)
$$

where the spectral function $f(\omega_I)$ is usually taken as $f(\omega_I) = \tau/(1+\omega_I^2 \tau^2)$ with τ the electron-spin correlation time. The spin diffusion coefficient in a regular lattice is $D \approx \frac{1}{30} \sqrt{M_2 a^2}$ with M_2 the dipolar second moment and a the nuclear spin spacing. For the ²⁹Si system in crystal line silicon, the value \overline{M}_2 is about 5.6 × 10⁵ (rad s⁻¹)². Taking $a \approx 0.55n_I^{-1/3} = 4.15$ Å (where n_I is the concentration of nuclear spins) gives $D \simeq 4 \times 10^{-14}$ cm² s⁻¹. Because of the random distribution of the $29\$ Si spins in the lattice this value for D should not be regarded as known to be better than an order of magnitude. In a random system the overall diffusivity may be strongly influenced by weak links whose effects are most difficult to estimate.

In estimating the radius of the diffusion barrier b we have considered the gradient in the local field produced by the S spins. This gradient is predominantly due to the spatial variation of the hyperfine coupling field
 $H_{\text{hf}} = (8\pi/3)(\mu_e) | \psi |^2$. Kohn-Luttinger¹⁴ wave functions were used together with the blocking condition a $(dH_{hf}/dr)_{r=b} \approx 1/\gamma_I T₂$ to obtain values for b in terms of $\langle \mu_e \rangle$. For $\mu_e B / kT < 1$, *b* is weakly field-dependent, ranging as $ln(B)$, while for $\mu_E B / kT > 1$, b reaches a constant maximum value of roughly 50 A.

At 1.3 K it appears likely that a transition in relaxation characteristic takes place moving from rapid diffusion in relatively high fields $(>0.1$ T) to the diffusion-limited case in lower fields $(0.05 T)$. This transition complicates the analysis of the data obtained at this temperature. For the PRT results obtained at 13.5 mK it is likely that rapid diffusion applies over the entire range of fields used because of the large value of $\langle \mu_e \rangle$.

For spin-diffusion mediated relaxation due to isolated paramagnetic impurities the general expression for the relaxation rate is 13,4

$$
1/T_1 = \frac{8\pi}{3} n\beta Dg(\delta) , \qquad (2)
$$

where $g(\delta)$ is given in terms of modified Bessel function as $g(\delta) = I_{3/4}(\delta)/I_{-3/4}(\delta)$ and *n* is the concentration of relaxing S spin centers. As we shall see, n is less than n_p . In the rapid diffusion limit Eq. (2) gives $1/T_1 = (4\pi/$ $3)$ nb⁻³C while in the diffusion-limited case the expression becomes $1/T_1 = (8\pi/3)nC^{1/4}D^{3/4}$. We take Eq. (2) as the basis for our calculations of $1/T_1$ for the ²⁹Si system.

We note that Ref. 4 describes a previous investigation of nuclear relaxation of the ${}^{63}Cu$ spin system in the dilute alloy, Cu:Mn. There are a number of general similarities between our Si:P system near the M-I transition and the Cu:Mn system. However, the details of the field dependence of $1/T_1$ for the host system (⁶³Cu and ²⁹Si, respectively) differ substantially because of the different nature of the paramagnetic species and differences in details of the nucleus-electron spin coupling.

DISCUSSION

For a strongly interacting spin system Eq. (2) must be used with care. Instead of assuming $n = n_P$ (uncomper sated case) or $n = n_p - n_B$ (compensated case), we must allow for the fact that a fraction of the spins are "frozen" in singlet states under the influence of the exchange coupling. Bhatt and Lee¹⁵ have successfully used a renormalization procedure based on this idea to calculate the magnetic susceptibility of insulating Si:P at low temperatures. In the same spirit we assume that spins for which the dominant pairwise exchange coupling $J \gg kT$ will, with high probability, be in the singlet state and will not contribute to nuclear relaxation. The fraction of spins $\eta = n / n_{\rm P}$ available for relaxation at a given temperature may be estimated using the susceptibility data of Andres et al .¹⁰ together with Curie-law predictions which they show on their plots. We have extracted values of η from the equation $\chi(T) = \eta \chi(T)_{\text{Curie}}$. For $n_p/n_c = 0.03$ values of η range from 0.8 at 1.3 K to 0.5 at 10 mK, while for $n_p/n_c \approx 1$, η varies from 0.1 to 0.01 over the same temperature interval. Table I shows some values of η derived from Ref. 10.

The assumption that $X(T)$ and T_1 ($n_p \le n_c$) are both controlled by a fraction of the localized spins may be tested using an empirical expression obtained by Zadrozny, Sachdev, and Paalanen¹⁶ $1/T_1 = K(T^\beta/B)$ (B > 0.4 T; $0.1 < n_p/n_c < 1$). This form with an exponent value β =0. 3 has been found to give a reasonable fit to available high-field ²⁹Si T_1 data. (This regime corresponds to the fast diffusion limit.) Andres et al .¹⁰ have shown that the magnetic susceptibility varies as $\chi(T) \propto T^{\nu}$ over a fairly wide temperature range (20 mK – 5 K). For $n_p/n_c < 1$ the exponent v lies in the range $-0.5(n_p/n_c \approx 1)$ to $-0.8(n_p/n_c = 0.1)$. If we write $\chi(T) \propto (n_p/T)$ and $1/T_1 \propto (\eta n_p) f(\omega_l)$, then we find values of β ranging from 0.5 to 0.2, in reasonable agreement with the empirical value of Zadrozny *et al.* (note that $\beta = v + 1$). This result provides support for the proposed relaxation mechanism involving localized moments, providing that $f(\omega_I)$ is not temperature dependent. Evidence that this temperature independence is correct is provided below.

The field dependence of T_1 given by Eq. (2) may be expected to arise primarily from the spectral function $f(\omega_t)$ contained in the factor C [see Eq. (1)], but al-

'This value uncertain because of unknown diamagnetic contribution from mobile electrons.

lowance must also be made for the expected field dependence of the diffusion barrier b . [At 1.3 K the condition γ_e $\hbar B \leq 2kT$ is satisfied for $B \leq 2.0$ T. In this field range one expects the form $b \propto \ln(B)$ to hold, as discussed in the second paragraph preceding Eq. (2)]. In calculating $f(\omega_I)$, it is necessary to estimate the S spin correlation time τ . Both electron spin-lattice and spin-spin relaxation processes must be considered. Spin-spin interactions within the exchange coupled reservoir (excluding the frozen singlet spins) can give rise to energy conserving flip-flop processes. Fairly isolated spins, spin pairs in the triplet state, and other small clusters of spins can participate in such processes through transitions amongst the Zeeman-split levels induced by $S_i^{\pm} S_j^{\mp}$ operators.

Assuming that spin-lattice and spin-spin processes are independent leads to a spectral density function which is the convolution of the separate spectral densities. If for simplicity exponential correlation functions are chosen, we have $1/\tau = 1/\tau_s + 1/\tau_L$ where τ_s and τ_L are, respectively, the spin-spin and spin-lattice relaxation times. For $n_p/n_c = 0.03$ we estimate $\tau_L \simeq 10^{-1}$ s at 1.3 K for $B \le 0.3$ \overline{T} , using the values given by Feher and Gere.¹⁷ It is likely that τ_s is much shorter than this. The exchange coupling between spins with a spacing of 115 A, which is the most probable spacing at this concentration, is 4×10^8 Hz. Inhomogeneous broadening processes such as unresolved hyperfine structure may inhibit spin diffusion within the electron system to some extent but it is likely that at least in dilute Si:P we will have the condition $\tau_s \ll \tau_L$ at 1.3 K and in low fields ($B \le 0.3$ T). The electron-spin system may be described by a spin temperature, and the nuclear spins achieve equilibrium with this thermal reservoir. The heat-capacity ratio $C_S/C_I = \gamma_S^2 n_S / \gamma_I^2 n_I \approx 10^7 n_S / n_I$. Thus, even at the lowest P concentrations the heat capacity of the electron reservoir is much greater than that of the ^{29}Si nuclear system. We therefore assume $\tau = \tau_S$ except perhaps at the highest fields and temperature used.

Figure 2(a) shows a plot of $log_{10}(1/T_1)$ versus $log_{10}B$ for the present data obtained on the dilute sample at 1.3 K and also for that of Jerome, Ryter, and Winter⁷ at very nearly the same temperature. The similarity in the shapes of the two experimental curves at 1.3 K is striking. Figure 2(b) shows similar plots for the PRT data at 13.5 mK for just-metallic and just-insulating samples.

In attempting theoretical fits to the data the form chosen for the spectral function $f(\omega_I)$ is of crucial importance. There is no justification for assuming that the Debye spectral function $\tau_s/(1+\omega_I^2\tau_s^2)$ (exponentially decaying correlation function with a single correlation time) should correctly describe the electron spin fluctuation spectrum if, as we believe, the spectrum is generated by exchange flips with ^a wide distribution in values of J. We take the distribution function to be $J(R)$ =K exp(-2R/ a_H^*), where a_H^* = 17 Å.¹⁰ The distribution in values of J should generate a corresponding distribution $P(\tau_s)$ of values of τ_s . If we assume that nearest neighbor interactions dominate in determining τ_s then one can show, using the nearest-neighbor distribution function, $P(R) = 4\pi R^2 n \exp[-(4\pi/3)R^3 n]$, that for localized spins $P(\tau_s)$ is proportional to $1/\tau_s$ over the range

of τ_s values that are of interest to us. [This form for $P(\tau)$ was introduced by PRT in their treatment of relaxation by mobile electrons, but the physics behind it is different in our model which assumes that localized spins are the source of relaxation.] If we combine this distribution with the Debye $f(\omega_I)$ and integrate over a range of τ_S values up to some upper limit τ_{max} we obtain $f(\omega_I) \propto (1/\omega_I) \tan^{-1}(\omega_I \tau_{\text{max}})$. It is likely that the nearest-neighbor model is oversimplified and that cluster-cluster interactions are important, particularly for $n_{n} \sim n_{c}$. Nevertheless, in order to make progress we assume that the arctan form for $f(\omega_I)$ with the proportionality constant of order unity represents a reasonable approximation to the actual spectral function.

Equation (2), using C given by Eq. (1) and the calculated values of b and D , permits us to attempt theoretical fits to the Si:P results. We take η , the fraction of spins involved in relaxation, as an adjustable parameter. The quality of fit is not very sensitive to the value τ_{max} . For consistency, we use a value of 10^{-5} s for τ_{max} , as suggested by the PRT measurements of $T_{1\rho}$ even though there is some uncertainty about the appropriateness of this value. The ESR linewidth measurements of Paalanen, Sachdev and Bhatt¹⁸ show that the electron spin lifetime $\tau \equiv 1/\gamma_e \Delta H_{1/2}$ has a value of the order of 10^{-7} s. How ever, since this time may represent an ESR dephasing time rather than a spin orientation lifetime, it is not at all clear that it should be identified with τ_s in the spectral function of Eq. (1).

At 1.3 K the fits depend in a crucial way on the value of nb^{-3} in the high-field region ($B > 0.1$ T) and on the value of $nD^{3/4}$ in the low-field region ($B < 0.1$ T) where we expect diffusion-limited effects to be important. At 13.5 mK we expect the rapid-diffusion limit to hold for all B values used, with b at a constant value. The theoretical fits to the data are shown in Fig. 2. The curves are numbered as follows: (1) $n = 1.1 \times 10^{17}$ cm⁻³, $T = 1.3$ K (present data); (2) $n = 2.5 \times 10^{18}$ cm⁻³. $T=1.28$ K (JRW); (3) $n = 3.33 \times 10^{18}$ cm⁻³, 13.5 mK (PRT); (4) $n = 3.81 \times 10^{18}$ cm⁻³, 13.5 mK (PRT). The values of η used to fit the vertical scale of the theoretical curves are shown in Table I, along with the values of η which we have extracted from Figs. 4 and 5 of Ref. 10, and from Refs. 18 and 19.

In fitting the 13.5 mK PRT data we are pushing the present expressions into a temperature range in which it is far from clear that they should hold. In this very low temperature range, γ_{ρ} $\hbar B > 2kT$ even at $B = 0.02$ T, and significant polarization of the electron-spin system is present when $B > 0.1$ T. If a polarization term $(1 - P_0^2)^{1/2}$, with P_0 having its Brillouin function value, tanh (γ_{ρ} *KB/2kT*), is introduced into Eq. (2), then our model predicts a significantly lower relaxation rate than those measured by PRT.

Sarachik et al .²⁰ have discussed the saturation properties of the magnetization in Si:P using a generalized scaling approach. Their analysis of magnetization data shows that the saturation effects in the exchange-coupled electron system of Si:P are less pronounced than the unmodified Brillouin function gives, but still sufficient that we would expect a significant dimunition of relaxa-

FIG. 2. log_{10} - log_{10} plot of the field dependence of experimental and calculated relaxation rates at 1.3 K (a) and 13.5 mK (b) for concentrations near the $M-I$ transition. The plotted points are experimental data taken from the indicated references. The solid lines are fitted curves based on Eq. (2). The dotted lines show the effect of adding a field dependence of the diffusion barrier radius b (see text).

tion effectiveness at the 13.5 mK temperature.

The nature of relaxation effects due to paramagnetic centers at very low temperatures has received recent attention in a paper by Waugh and Slichter.²¹ They proposed a spin "wobble" mechanism to account for the observation of surprisingly strong nuclear-spin relaxation in $CaF₂$. While our exchange-coupled system will certainly have a different Fourier spectrum than the system of isolated impurities in a material such as $CaF₂$, the Waugh and Slichter paper does serve as a warning that at very low temperatures, where saturation of the electron spin magnetization weakens the relaxation processes we have considered, processes whose nature is less obvious may take over as primary relaxation agents.

The shapes of the solid curves in Fig. 2 match the experimental data rather well. It will be noted that for both sets of 1.3-K data [Fig. 2(a)] keeping b constant at 50 \AA gives better agreement with the data (solid curves} than including a lnB field dependence of b (dotted curves). This result suggests that at this temperature b saturates at lower fields than expected. A possible underlying reason for this saturation is the fact that the systems are not "dilute" even for the $n_p = 1.1 \times 10^{17}$ cm⁻³ sample. (The $\ln B$ field dependence of b is based on the field distribution from an isolated impurity.)

Although there are uncertainties in values of a number of component quantities in our model, including b and D , we are nevertheless encouraged that all values of η derived from our fits (Table I) match the values extracted from the susceptibility measurements of Refs. 10, 18, and 19 within a factor of 2, both at 1.3 K and 13.5 mK. This parallel behavior is consistent with our earlier discussion in which the T_1 and χ temperature exponents were related.

For the two samples used by PRT, for which plots are given in Fig. 2(b), our model gives $\eta_4/\eta_3 = 0.2$. This ratio is consistent with the natural assumption that a significant drop in the number of localized spins occurs as the M-I transition is traversed from the insulated to metallic side. Using the values of η taken from the susceptibility data and shown in Table I give a ratio of values of η of 0.3 for a pair of samples slightly more separated in electron density than the PRT samples, with phosphorus concentrations of 3.8×10^{18} and 2.9×10^{18} cm⁻³, respectively.

We do not offer an explanation of the observed decrease in the value of I/T_1 in going from Si:P to samples of compensated Si:(P,B) at the same unpaired electron concentration and temperature. It could mean that there is a larger fraction of frozen singlet state spins in the compensated material compared to the uncompensated samples, because of larger values of J_{ij} in the compensate ed material. Serious consideration of this difference might best await completion of analysis of measurements of electron-spin susceptibility in $Si:(P,B)$.²²

In summary, the results we have presented suggest tha the field-dependent relaxation of ^{29}Si nuclei in Si:P and $Si: (P,B)$ in the insulating phase and apparently in the just-metallic phase is due to the localized moments. Using spin-difFusion theory it is possible to give a reasonably quantitative explanation of the observations. The spectrum of the electron spin fluctuations responsible for nuclear relaxation appears to be very similar in all samples and probably originates in the spin-spin exchange coupling. The $1/T_1$ versus B curves provide some information on this spectrum. In developing a theoretical model, use has been made of the Bhatt and Lee approach to the calculation of the magnetic susceptibility of insulating Si:P. This implies a link between the ²⁹Si T_1 behavior and that of χ in the concentration range in which local moments dominate in determining the magnetic properties. A relationship between the temperature exponents can be shown to follow from these ideas, and it is supported by available experimental evidence. Our model seems consistent with conclusions recently drawn by Alloul and Dellouve²³ from a study of the ${}^{31}P$ NMR in Si:P.

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- 'Permanent address: Department of Physics, University of the Witwatersrand, ¹ Jan Smuts Avenue, Johannesburg, South Africa.
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