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ESR linewidth behavior for barely metallic n-type silicon

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Measurements of the ESR linewidth of uncompensated Si:As close to n_c for the metal-insulator transition have been made at 9.4 GHz in the temperature range 4.2–1.4 K. The linewidth exhibits a minimum value $\Delta H_{\min}$ which occurs at $n = n_c$ and an excess component $[\Delta H_{\text{ex}} = \Delta H - \Delta H_{\min}]$ which exhibits scaling behavior of the form $B(n/n_c - 1)^p$ for $n > n_c$. The scaling behavior, established for both Si:As and Si:P (from earlier data), can be explained as a simple extension of the Elliot-Yafet mechanism for conduction-electron spin-resonance linewidths. The strong donor dependence of B is satisfactorily explained with impurity spin-orbit interaction parameters obtained from Orbach spin-lattice relaxation studies.

Since the discovery[1,2] that the electron-spin resonance (ESR) linewidth exhibits a minimum value $\Delta H_{\min}$ in Si:P near $n \sim 3\times10^{18}$ cm^{-3} [close to n_c for the metal-insulator (M-I) transition] there has been substantial interest in the density and temperature dependences of the linewidth[3–6] and spin susceptibility[4,6] on both the insulating and metallic sides of the M-I transition. Much of this early work has been reviewed by Holcomb,[7] particularly with respect to understanding why spin delocalization first occurs at a lower donor density than charge delocalization. Transport measurements[8] have not only established the scaling behavior of σ_{dc}, but have demonstrated the dominance of disorder-enhanced electron-electron interactions[9] for uncompensated Si:P. This led to the proposal[10] that uncompensated Si:P, behaving as a strongly correlated liquid near n_c, should exhibit enhanced spin fluctuations. Experimental studies by Paalanen and coworkers[11,12] in the millikelvin range utilizing NMR[11] and ESR[12] have yielded information pertaining to spin fluctuations. The results have produced evidence on the slowing of spin diffusion near n_c as $T \rightarrow 0$. What has not yet been recognized is that the excess ESR linewidth $[\Delta H_{\text{ex}} = \Delta H_{\text{tot}} - \Delta H_{\min}]$, which is proportional to the charge-diffusion coefficient D, exhibits scaling behavior for barely metallic samples. The scaling of D is in turn related to the scaling of σ_{dc} by the Einstein relationship. In addition, we demonstrate that the result $\Delta H_{\text{ex}}(n) \propto D(n)$ represents a natural generalization of the Elliott[13]-Yafet[14] (EY) mechanism for conduction-electron spin-resonance (CESR) linewidths near the M-I transition. Below we report new results for the ESR linewidth of Si:As which establish for the first time the scaling behavior of ΔH_{ex} for barely metallic samples. The data from earlier Si:P studies,[2,5] when replotted, also seem to show scaling behavior of ΔH_{ex} for $n > n_c$.

The theoretical model most frequently employed to explain CESR linewidths has been the EY model whereby $\Delta H_{\text{ex}} \sim 2/\gamma T_1 \simeq (2/\gamma\tau)(\Delta g/g)^2$ with τ the conductivity collision time and Δg the g shift. This mechanism has been successful in explaining CESR linewidths in many cases. As emphasized by Pifer,[5] however, the EY mechanism has not been able to explain the very strong donor dependence of ΔH_{ex} for $n > n_c$ if one employs the donor-dependent g shifts measured by Feher[15] in the dilute limit ($N_D \ll n_c$). These dilute-limit g shifts arise primarily from the effective-mass potential and yield a rather small donor dependence for P, As, and Sb donors that scales with the donor binding energy rather than with the strength of the central-cell impurity spin-orbit interaction strength. CESR studies of impurity-doped Li and Na (small-Z hosts) by Asik, Ball, and Slichter[16] have clearly established the importance of an impurity-dominated spin-orbit interaction for $1/T_1$ and ΔH_{ex}. The situation is similar in n-type Si for $n > n_c$, except that the pure host is an insulator at low temperatures. Below we note that the impurity spin-orbit interaction strengths necessary to explain the strong donor dependence of ΔH_{ex} can be approximately obtained from Orbach spin-lattice relaxation (SLR) results[17] and theory[18] for dilute n-type Si. The impurity spin-orbit interaction, which makes no contribution at all to dilute limit donor ($1sA_1$ state) g shifts measured by Feher, can satisfactorily explain the strong donor dependence of ΔH_{ex} for barely metallic samples just above n_c. This clearly demonstrates that the SLR rate is governed by charge diffusion and the impurity spin-orbit interaction.

The measurements reported below were made with an X-band ESR spectrometer at 9.4 GHz utilizing a TE_{102} mode cavity featuring a tilting sample holder that permitted the thin single-crystal Si:As sample to be accurately positioned at the electrical center (null), thus optimizing the cavity quality factor. The samples were carefully etched with CP-4 etchant solution and the thickness d was kept comparable to the skin depth δ and the line-shape asymmetry factor[5,19] A/B was kept less than 1.8. Calculated corrections were applied to obtain the peak-to-peak absorption derivative linewidth $\Delta H_{\text{p.p.}}$ in the limit $A/B \rightarrow 1$. The line-shape asymmetry results and the resulting microwave conductivity σ $(N, \nu = 9.4\ \text{GHz}, T)$ will be reported elsewhere.

The peak-to-peak linewidth $\Delta H_{\text{p.p.}}$ versus temperature for six Si:As samples near n_c is shown between 4.2 and 1.4 K in Fig. 1. There is virtually no temperature dependence of $\Delta H_{\text{p.p.}}$ in this temperature range although the linewidth is known to broaden at both higher[2,3,5,6] and lower[12] temperatures. The linewidth enhancement observed at much

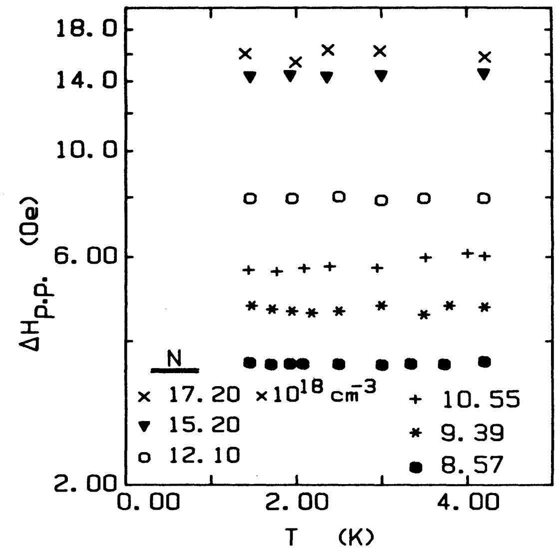

FIG. 1. Absorption derivative peak-to-peak linewidth vs temperature. Values are corrected for $A/B \rightarrow 1$. Errors are comparable to the size of the data points.

lower temperatures by Paalanen, Sachdev, Bhatt, and Ruckenstein[12] correlated well with an enhanced susceptibility as $T \rightarrow 0$. The enhancement is bigger as n approaches n_c+, which is consistent with local moments for barely metallic samples, although the actual mechanism for the low-temperature enhancement of $\Delta H_{\mathrm{p.p.}}$ and χ_s is not yet well understood. Here we concentrate on the donor-density dependence of $\Delta H_{\mathrm{p.p.}}$ in the broad temperature range where $\delta(\Delta H_{\mathrm{p.p.}})/\delta T \rightarrow 0$.

In Fig. 2 $\Delta H_{\mathrm{p.p.}}(T_{\mathrm{min}})$ is shown versus N_D where $n_c = 8.6 \pm 0.05 \times 10^{18}/\mathrm{cm}^3$. $\Delta H_{\mathrm{p.p.}}$ clearly shows a minimum very close to n_c. This was also the case for Si:P, although the minimum is a function of temperature as shown by Maekawa and Kinoshita.[2] There are small differences between the concentration values of $\Delta H_{\mathrm{p.p.,min}}$ for Si:P from different research groups, most likely from slightly different concentration scales. The Si:As results reported here come from the same wafers that bar samples were obtained for dc conductivity measurements[20,21] and disks for Hall measurements.[22] Within experimental errors ($\pm 1\%$) the minimum in $\Delta H_{\mathrm{p.p.}}$ occurs at $N_D \simeq n_c$.

Except for the rounding of $\Delta H_{\mathrm{p.p.}}(n)$ near the minimum at $n \simeq n_c$ the linewidth for $n > n_c$ can be well fit by $\Delta H_{\mathrm{p.p.}}(n,T) = A(n,T) + B(n/n_c - 1)^p$ where the first term $A(n,T) = M_2/\langle \omega(n,T) \rangle$ represents exchange or motional narrowing (T_2 process) from the insulating side of n_c (see discussion in Ref. 6). The second moment M_2 is given by $M_2 \sim \frac{4}{3} I(I+1)(A_0/2)^2$ resulting from the donor hyperfine interaction $A_0 \mathbf{S} \cdot \mathbf{I}$. The data[2,6] at a fixed temperature yield $A(n,T) \sim a(T)(n/n_c)^{-q}$ with $q \sim 1.6$ for both Si:P and Si:As for $0.8 < n/n_c < 1$ (here $n = N_D$). It is not known whether $A(n,T)$ retains this form near $n = n_c$ and for $n > n_c$, but we will utilize the expression for the present analysis. The minimum linewidth $\Delta H_{\mathrm{p.p.,min}} = a(T)$ and the excess linewidth $\Delta H_{\mathrm{p.p.,ex}} = \Delta H_{\mathrm{p.p.}}(n) - a(T)$. $\Delta H_{\mathrm{p.p.,ex}}$ versus reduced density $n/n_c - 1$ is shown in Fig. 3 for both Si:As and Si:P (from Refs. 2,5). The results yield $p \sim 1.0$ for both donor species, which is approximately twice the conductivity exponent μ for σ_{dc}. The parameters obtained from fitting the data $n/n_c > 1.05$ to the above expression for values of $q = 0$ $[A(n > n_c, T) = a(T)]$, $q = 1.6$, and 3 are shown in Table I. The exponent p is close to 1.0 for small q, but decreases as q increases and can in fact approach $\frac{2}{3}$ for $q \sim 10$. The ratio $B(\mathrm{As})/B(\mathrm{P})$ is large and can be accounted for by the EY mechanism if one utilizes the impurity spin-orbit interaction to obtain $\Delta g/g$. Pifer[5] reported that $\Delta H_{\mathrm{p.p.}} \propto n^{0.6}$ for Si:P for $n > 2n_c$, but also noted that as $n \rightarrow n_c+$ $\Delta H_{\mathrm{p.p.}}(n)$ fell faster than $n^{0.6}$. The basic result from Fig.

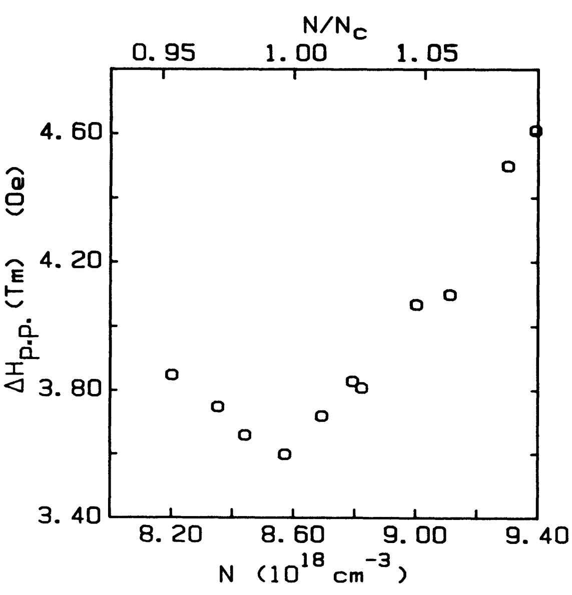

FIG. 2. $\Delta H_{\mathrm{p.p.}}(T_m \simeq 1.4\ \mathrm{K})$ vs donor concentration, close to N_c. The value of $\Delta H_{\mathrm{p.p.,min}}$ is 3.58 ± 0.05 Oe.

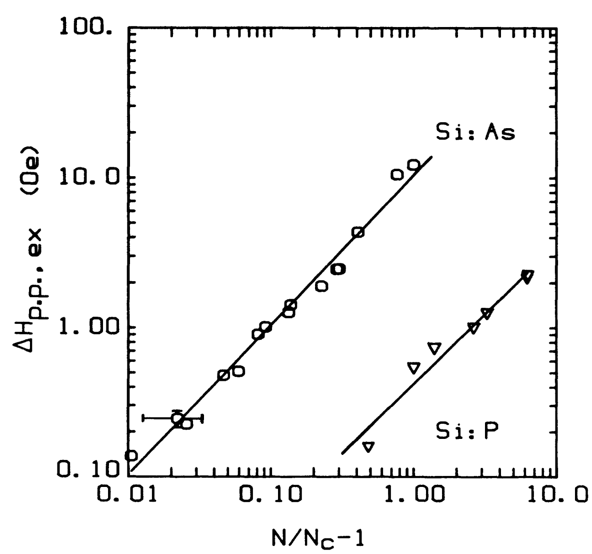

FIG. 3. Scaling behavior of $\Delta H_{\mathrm{p.p.,ex}}$ for both Si:As (this work) and Si:P (Refs. 2 and 5).

TABLE I. Experimental linewidth parameters.

Donor	q	a (Oe)	B (Oe)	p
Si:As	0	3.58 ± 0.05	10.3 ± 0.5	0.99 ± 0.05
	1	3.58 ± 0.05	12.7 ± 0.5	0.97 ± 0.05
	1.6	3.58 ± 0.05	13.6 ± 0.5	0.95 ± 0.05
	3.0	3.58 ± 0.05	15.1 ± 0.5	0.91 ± 0.05
Si:P	0	0.52 ± 0.05	0.50 ± 0.1	1.1 ± 0.1
	1.6	0.52 ± 0.05	0.76 ± 0.1	0.9 ± 0.1

3 and Table I is that $\Delta H_{\text{p.p.}}(n)$ contains a scaling component for both P- and As-doped Si although the exponent p, depending on the value of q for $n>n_c$, is most probably larger than the conductivity exponent. Finally, we note that the assumed form for $\Delta H_{\text{p.p.}}(n)$ yields a cusp at n_c, rather than a smooth minimum. Rounding of the cusp may occur from (1) inhomogeneous doping over the sample, (2) variable range-hopping conduction for $(1-n/n_c)\ll 1$, and (3) from possible other T_2 processes (the EY mechanism yields $1/T_1\rightarrow 0$ as $n\rightarrow n_c+$ at $T=0$).

The usual Elliott[13]-Yafet[14] theory for $n>n_c$ yields $\Delta H_{\text{p.p.}}\propto(1/\gamma\tau)(\Delta g/g)^2$ does not yield scaling behavior but can predict Pifer's $n^{0.6}$ dependence since $1/\tau\propto n^{2/3}$ on elementary grounds. A simple phenomenological model, representing an extension of EY theory, can yield $\Delta H_{\text{p.p.,ex}}\propto D(n)$ [or σ_{dc} employing $\sigma_{\text{dc}}(n)=e^2dN/d\mu D(n)$] where $D(n)$ is not the bare diffusion coefficient D_0, but is the scaling diffusion coefficient with the property $D(T=0,n)\rightarrow 0$ as $n\rightarrow n_c+$.

Let us introduce a spin-flip length $L_{\text{s.o.}}=(D\tau_{\text{s.o.}})^{1/2}$ where $1/\tau_{\text{s.o.}}$ is the spin-flip rate from spin-orbit scattering and the SLR rate $1/T_1=1/\tau_{\text{s.o.}}$. The D in $L_{\text{s.o.}}$ is the charge-diffusion coefficient, not the spin-diffusion coefficient, because for $n>n_c$ charge diffusion is dominant and the charges carry the spins. The number M of collisions (elastic) an electron undergoes before its spin is flipped is given by $M=(g/\Delta g)^2$. Hence $L_{\text{s.o.}}=l\sqrt{M}=l(g/\Delta g)$ where l is the mean free path. The linewidth contribution due to $\tau_{\text{s.o.}}=T_1$ is given for a Lorentzian line shape, by $\Delta H_{\text{p.p.}}=(2/\sqrt{3})(1/\gamma\tau_{\text{s.o.}})$, or

$$\Delta H_{\text{p.p.,ex}}=(2/\sqrt{3}\gamma)(\Delta g/g)^2[D(n)/l^2]. \quad (1)$$

For D the bare diffusion coefficient $D_0=v_F^2\tau/3=l^2/3\tau$ Eq. (1) yields the EY result. However, for $D=D_0(n/n_c-1)^\mu$ one obtains scaling where D has the same critical exponent as σ_{dc} assuming $dN/D\mu$ does not exhibit critical behavior. Equation (1) with $D(n)$ scaling to zero as $n\rightarrow n_c+$ represents an extension of the EY theory to the vicinity of the M-I transition.

The strong donor dependence of ΔH_{ex} (at fixed n/n_c) originates from $(\Delta g/g)^2/\tau_e$ where τ_e is the elastic collision time [$\tau_e\propto n^{-2/3}$]. Pifer[5] has suggested that the strong donor dependence of ΔH_{ex} originates from the impurity spin-orbit interaction. In the dilute limit of isolated donors this interaction accounts for the splitting of the $1s$-T_2 states into an orbital singlet and doublet split by $\Delta_{\text{s.o.}}$. The $1s-A_1$ state, which is spherical for small r inside the central cell, is unaffected by the impurity spin-orbit interaction [$\langle 1s-T_2|\mathbf{L}|1s-A_1\rangle=0$]. As n approaches n_c and for $n>n_c$ a random system of donors has complex overlapping electron wave functions that are no longer spherical about donor sites and the wave function overlaps many donor sites. Alternatively, there is a strong hybridization of the $1s-A_1$ and $1s-T_2$ impurity bands and the impurity spin-orbit interaction can now play an important role. An estimate of $(\Delta g/g)_{i,\text{s.o.}}$ for n near n_c can be inferred from the Orbach SLR rate parameters[17,18] obtained in the dilute limit. This yields $(\Delta g/g)_{i,\text{s.o.}}\sim\Delta_{\text{s.o.}}/\Delta E$ where $\Delta E=E_{1s\text{-}A_1}-E_{1s\text{-}T_2}$. Ochiai and Matsuura[6] have reported Orbach SLR even for $n>n_c$, suggesting that both the $1s-A_1$ and $1s-T_2$ impurity "bands" remain below the conduction-band edge, although they report concentration-dependent values of ΔE that are much smaller than for isolated donors. Assuming the ratio $(\Delta_{\text{s.o.}}/\Delta E)$ is not a strong function of n (both $\Delta_{\text{s.o.}}$ and ΔE decrease with increasing n) then one can employ the dilute-limit ratio[18] to calculate $\Delta H_{\text{p.p.,ex}}$, which is given by

$$\Delta H_{\text{p.p.,ex}}(n)=(2/3\sqrt{3})(1/\gamma\tau_e)(\Delta_{\text{s.o.}}/\Delta E)^2(n/n_c-1)^\mu. \quad (2)$$

Calculated values of $\Delta H_{\text{p.p.,ex}}(n=2n_c)$ are shown in Table II along with the parameters τ_e, $\Delta_{\text{s.o.}}$, ΔE, and the g shifts measured by Feher. τ_e is calculated either from

$$\tau_c=\hbar/2E_F(n=n_c)$$

or from

$$l_e=(v_F\tau_e)_c\simeq 0.55d_c \quad (d_c=N_c^{-1/3})$$

and the results agree to within 10%. $\Delta_{\text{s.o.}}$ and ΔE are the dilute limit values obtained from Ref. 18. The calculated values are larger than the corresponding experimental values of B shown in Table I. However, the calculated ratio $\Delta H_{\text{p.p.,ex}}(\text{As})/\Delta H_{\text{p.p.,ex}}(\text{P})$ is in good agreement with the experimental ratio $B(\text{As})/B(\text{P})\sim 17$. Pifer[5] has noted it was not possible to see the Si:Sb resonance signal, presumably because of the very broad linewidth resulting from the very much larger value of $(\Delta_{\text{s.o.}}/\Delta E)$ for Sb, as

TABLE II. Donor-dependent linewidth parameters.

Donor	$(g_D-g_{\text{CE}})^{\text{a}}$ $\times10^4$	$\tau_c\times10^{14}$ (sec)	$\Delta_{\text{s.o.}}{}^{\text{b}}$ (meV)	$\Delta E_{A_1-T_2}{}^{\text{b}}$ (meV)	$\Delta H_{\text{p.p.,ex}}(n=2n_c)$ (Oe)
P	−2.5	2.5	0.022	11.6	2.6
As	−3.8	1.6	0.13	21.1	45
Sb	−1.7	2.9	0.30	9.55	640

[a]Values from Ref. 15. [b]Values from Ref. 18.

shown in Table II. Any attempt to explain the linewidth data with the EY mechanism and the dilute limit g shifts[15] $(g_D - g_{CE})$ (see Table II) is totally untenable since it would yield the narrowest line for Si:Sb and the calculated linewidths would be much too small.

An anisotropic Zeeman-interaction intervalley scattering mechanism has been calculated by Chazalviel[23] and applied to n-type Ge where there is a very large g anisotropy ($g_l = 0.87$, $g_t = 1.92$). Chazalviel obtains, neglecting angular-dependence factors,

$$1/T_1 \simeq \tfrac{2}{9}[(g_l - g_t)^2/g^2]^2 \omega_0^2 \tau_{IV} \quad (3)$$

for $\omega_0 \tau_{IV} \ll 1$, where $\omega_0 = \gamma H_0$ is the Larmor frequency and τ_{IV} is the intervalley scattering rate. As determined by Wilson and Feher[24] $(g_l - g_t) \simeq 10^{-3}$ for Si and this mechanism leads to a totally negligible contribution to the linewidth for n-type Si. This mechanism could not yield the correct donor dependence of $\Delta H_{\text{p.p.}}$ for n-type Si in any case.

There is one other experimental feature that supports the role of the impurity spin-orbit interaction, namely the approximate linear (with n) decrease in the g value for Si:P reported by Quirt and Marko[4] for $n > 3 \times 10^{19}$ at 1.1 K and also by Kodera[1] at 77 K. The conduction electrons encounter more and more impurities, each of which gives an extra negative g shift of order $g_{CE}(\Delta_{\text{s.o.}}/\Delta E)$. This leads to the result

$$g(n) = g_{CE}(1 - nV_0 |\Delta_{\text{s.o.}}/\Delta E|), \quad (4)$$

where V_0 is a characteristic volume (per donor). For V_0 of order $(a_D^*)^3$ where a_D^* is the dilute limit donor Bohr radius and $(\Delta_{\text{s.o.}}/\Delta E)$ for Si:P from Table II one can explain the magnitude of the decreasing g value for $n > 3 \times 10^{19}$ quite satisfactorily. The approximate linear decrease of $g(n)$ with n suggests that $(\Delta_{\text{s.o.}}/\Delta E)$ is changing slowly with n. The g-shift data for Si:As is qualitatively similar to that for Si:P but is not accurate enough to quantitatively compare with the Si:P results.

The minimum linewidth $\Delta H_{\text{p.p.,min}} = a(T)$ also exhibits a strong donor dependence which has usually been interpreted[6] as resulting from exchange or motional narrowing. For the second moment ratio $M_2(\text{As})/M_2(\text{P}) \simeq 5[A_0(\text{As})/A_0(\text{P})]^2 \sim 14$ since $I = \frac{3}{2}$ for As and $\frac{1}{2}$ for P. Much of the ratio $a(T,\text{As})/a(T,\text{P})$ results from the M_2 ratio but one needs $\langle\omega(n = n_c, T, \text{As})\rangle \sim 2\langle\omega(n = n_c, T, \text{P})\rangle$ to explain the experimental ratio. Of importance here is the fact that the broadening on the insulating side as n decreases results from a different mechanism than the broadening on the metallic side.

In summary, it has been demonstrated that the excess ESR linewidth shows scaling behavior for barely metallic samples similar to the scaling of the dc conductivity although the critical exponents differ. The strong donor dependence results from the impurity spin-orbit interaction, the strength of which is estimated from dilute limit Orbach SLR rate parameters.

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