Observation of the conduction-electron spin resonance from metallic antimony-doped silicon

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Electron spin resonance has not previously been detected for barely metallic Sb-doped silicon. We report preliminary measurements at 9.4 GHz in the temperature range $1.4<T<4.2$ K for two Sb-doped silicon samples with concentrations close to the critical density n_c for the metal-insulator transition. The peak-to-peak linewidths were 21 and 45 Oe for the samples at n_c and $1.28n_c$, respectively. The results support Pifer's assertion that the conduction-electron spin resonance linewidth is determined by the impurity spin-orbit interaction. [S0163-1829(98)02123-7]

Conduction-electron spin resonance (CESR) has been observed at low temperatures ($T\leqslant 4.2$ K) for heavily doped Si:P (Refs. 1–7) and Si:As,[5,6,8] but has not been reported, to our knowledge, for metallic Si:Sb. Pifer[5] was unable to see CESR in Si:Sb, which he attributed to the broad linewidth expected because of the dominance of the impurity spin-orbit (SO) interaction in determining the linewidth. The interpretation of the CESR linewidth and its dependence on donor density is of interest in providing a different viewpoint about the transport mechanisms of n-type Si for n just above n_c, where n_c is the critical density for the onset of metallic behavior at $T=0$. There have been transport results for Si:Sb (Refs. 9 and 10) that show some unusual features and theorists have suggested[11,12] the impurity SO interaction might explain some of the features of the transport results for Si:Sb. Below, we report preliminary CESR results for two Si:Sb samples, one very close to the critical density n_c for Si:Sb and the second 28% above n_c. The results show larger linewidths than does Si:As and provide additional experimental support for showing the impurity SO interaction determines the peak-to-peak linewidth $\Delta H_{pp}(n)\propto 1/T_1=1/\tau_{SO}$ for $n>n_c$, where T_1 is the longitudinal spin-relaxation (SR) time and τ_{SO} is the SR time due to the SO interaction. Just as in the dilute limit for shallow donors, the results demonstrate the increasing strength of the impurity SO interaction with increasing Z of the substitutional donor. In addition, the results show $\tau_{SO}\gg\tau_e$ where $1/\tau_e$ is the elastic collision rate. The results do not support the notion of anomalous transport for Si:Sb for $n>n_c$ and suggest the universality class for Si:Sb should be the same as that for Si:P and Si:As. Although the preliminary results discussed below are not as detailed as earlier studies, they represent the only CESR results for Si:Sb in the metallic regime and provide additional experimental evidence supporting Pifer's notion that the impurity SO interaction is responsible for the CESR linewidth for Si:As and Si:Sb.

The measurements were made with an X-band ESR spectrometer at 9.4 GHz featuring a TE_{102} resonant cavity with a tiltable sample holder[13] to optimize the cavity Q. A power of approximately 1 mW was incident on the cavity yielding a microwave magnetic field $B_1\sim 8$ mG, which was well below saturation. The weak broad signals for Si:Sb required extensive signal averaging with a 1024-channel Nicolet signal averager. Although reference samples (dilute Si:As) were used to accurately set the microwave phase for χ'' in our Si:As study,[8] that was not possible here because of interference between the Si:As hyperfine spectrum and the much broader and weaker CESR signal of the Si:Sb sample. The field modulation amplitude H_m at the sample was increased by employing a lower modulation frequency of 35 Hz. Previously four to eight sweeps were employed[8] for the broader Si:As CESR lines; however here 256 sweeps (2.5 min each) were necessary to obtain satisfactory signal-to-noise ratio. As a result, it was not possible to carefully explore the temperature dependence of the linewidth. The samples were provided by the General Electric R&D Center (batch #1975) and were nominally uncompensated. They were etched with a CP4 etch to remove surface-state resonances and to minimize the asymmetric Dysonian line shape. The final thicknesses were $t=0.063$ and 0.065 mm. Unlike the Si:As study, where resistivity measurements were made on four bar samples between room temperature (RT) and 4.2 K adjacent to the thin rectangular slab used for ESR measurements, resistivity measurements were only made at RT. The donor density is therefore not as accurately determined as for the Si:As results.

Figure 1(a) shows the absorption derivative $d\chi''/dH$ for a $2.98\times 10^{18}/\mathrm{cm}^3$ Si:Sb sample ($\rho_{RT}=0.0137\ \Omega$ cm) at $T\sim 1.92$ K for a 100 Oe sweep. The peak-to-peak field modulation H_m was 3.6 Oe. The Dysonian line shape asymmetry ratio $A/B\sim 1.1\pm 0.05$ and the linewidth $\Delta H_{pp}\sim 21\pm 0.5$ Oe. The value $H_m/\Delta H_{pp}\sim 0.17$ was small enough to ensure any modulation-induced broadening is less than 1%. The asymmetry correction to ΔH_{pp} for this sample should be small. Data (not shown) for the same sample taken at $T=1.46$ K show a slightly different $\Delta H_{pp}\sim 21.6\pm 0.5$ Oe, but this is within the estimated error. Based on an estimate of the conductivity $\sigma(T\sim 1.9$ K) of 15 S/cm from Ref. 9 this leads to a skin depth $\delta\sim 0.14$ mm leading to $t/\delta\sim 0.45$, which translates to an $A/B\sim 1.08$ in reasonable agreement with the spectrum in Fig. 1(a). The g value was not measured with precision, but based on the nominal cavity frequency of 9.42 GHz was within 0.25% of the g value $g=1.9987$, as found for Si:As metallic samples. Figure 1(b) shows a 500 Oe sweep spectrum for the same sample at $T\sim 1.45$ K, which indicates a weaker broad signal on the low-field side of the 21 Oe width line. The center of this asymmetrical line is estimated to be between 40 and 60 Oe below the center of

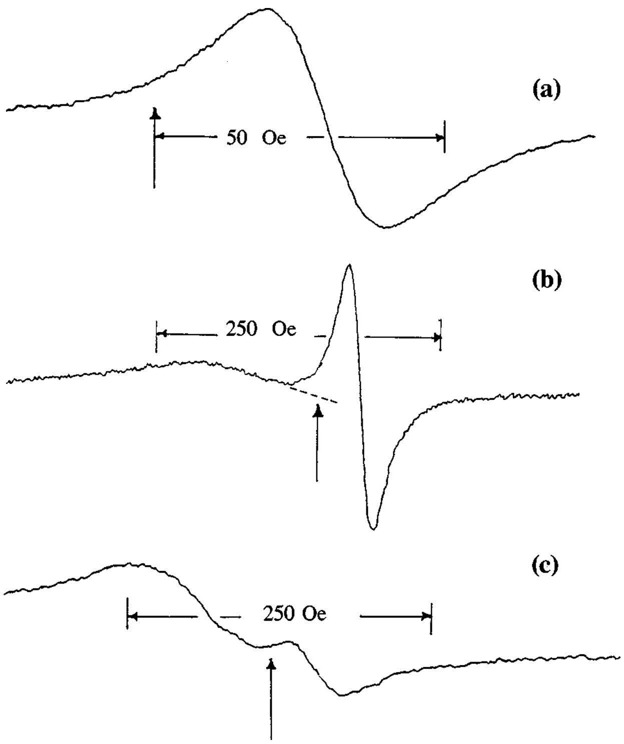

FIG. 1. Three absorption derivative spectra for two Si:Sb samples. The vertical arrows indicate a magnetic field of 3330 Oe. Each spectrum represents 256 sweeps. (a) A $2.98\times10^{18}/\mathrm{cm}^3$ Si:Sb sample at $T=1.92$ K. The sweep was 100 Oe. The peak-to-peak linewidth was determined to be 21 ± 0.5 Oe. (b) A 500 Oe sweep for the $2.98\times10^{18}/\mathrm{cm}^3$ sample showing a background signal centered approximately 40–60 Oe below the center of the Si:Sb CESR line. (c) The absorption derivative for a $3.8\times10^{18}/\mathrm{cm}^3$ Si:Sb sample at $T=1.93$ K for a 500 Oe sweep. The magnetic-field modulation was 2.22 times that used for (a) and (b). The peak-to-peak linewidth is 45 ± 7 Oe for the Si:Sb CESR line. The broad background signal from the Cu cavity has a width 167 ± 15 Oe.

the sharper line corresponding to a g value between 2.023 and 2.035. The halfwidth is between 80 and 100 Oe. The g value, the asymmetry, and the large linewidth suggest this signal arises from the Cu cavity. Schultz and Latham[14] have reported $g\sim2.033$ for high-purity Cu.

In Fig. 1(c) the absorption derivative is shown for a $3.8\times10^{18}/\mathrm{cm}^3$ Si:Sb sample ($\rho_{\mathrm{RT}}=0.0118\ \Omega$ cm) at $T\sim1.93$ K for a 500 Oe sweep and field modulation $H_m\sim8.0$ Oe, more than double the H_m used for Figs. 1(a) and 1(b). The spectrum now consists of two overlapping resonances, one broad and the smaller one a factor of about 4 narrower. The much broader line has a center very close to that for the weak background line in Fig. 1(b). The amplitude is consistent with the 2.22-fold increase in H_m and the g value is nearly the same as for the broad line in Fig. 1(b). In addition, there is a second narrower line of linewidth about 45 Oe with a g value close to 2.00; however the uncertainties in both quantities are larger because of the overlap of the two lines and also because of the large uncertainty in the baseline. The center of the broader line is approximately 50 Oe below the center of the narrower line, which is consistent with the results in Figs. 1(a) and 1(b). The broad line ($\Delta H_{pp}=167\pm15$ Oe), which contains most of the integrated intensity, is identified with the Cu background line from the cavity, while the narrower line is attributed to the CESR of Si:Sb.

The g values of the Si:Sb CESR signals are consistent to within 0.25% of the value $g=1.998\,75\pm0.0001$ obtained by Feher[15] for free carriers in Si:P, in addition to being in agreement with earlier CESR results for Si:As.[8] Because of the large linewidths, line-shape asymmetry, and overlap with the background signal no effort was made to more accurately determine the g values for these two samples. The $3.8\times10^{18}/\mathrm{cm}^3$ sample was also measured with only 128 sweeps at 4.2 K and the linewidths were the same to within the experimental errors. The overall behavior is the same as that for Si:As, but with larger values of the linewidth.

Table I shows that the linewidth at $n\sim n_c$ varies by a factor of about 50 from Si:P to Si:Sb, whereas at $n\sim1.28n_c$ the donor dependence is a factor above 80. The change in donor dependence (particularly striking from P to As) with just a 28% increase in donor density provides evidence that there is a different mechanism for the SR rate $1/T_1$ and linewidth for more metallic samples than for the linewidth at $n=n_c$ and for barely insulating samples. Right at n_c the linewidth can be explained by exchange and/or motional narrowing, which has been discussed by Anderson and Weiss.[16] The result for ΔH_{pp} in this case is

$$\Delta H_{pp}\sim\gamma[M_2/\langle\omega_{\mathrm{ex}}\rangle], \tag{1}$$

where M_2, the second moment or mean-squared spread of the spectrum about its center, has been given by Meier, Parks, and Hale[17] as $[(\delta H)^2+4/3I(I+1)(A_{\mathrm{hpf}}/2)^2]$ and γ is 1.76×10^7 rad/sec. $\langle\omega_{\mathrm{ex}}\rangle$ is the exchange and/or motional narrowing frequency. The first term in M_2 is the linewidth $\delta H$of the individual hyperfine lines of the resolved hyperfine spectrum in the dilute limit due to the ^{29}Si nuclei. δH is of order 3 Oe. The second term results form the donor hyperfine interaction with the donor nucleus of spin I. For Si:Sb, there are two isotopes with $I=\frac{5}{2}$ and $I=\frac{7}{2}$ for ^{121}Sb and ^{123}Sb, respectively. Thus M_2 is the weighted average $M_2=0.573M_2(^{121}\mathrm{Sb})+0.427M_2(^{123}\mathrm{Sb})$. The exchange (or motional) frequencies, calculated using Eq. (1) are shown in Table I and show a dependence that is qualitatively proportional to the donor binding energies. There have been numerous efforts to explain the temperature dependence of ΔH_{pp}

TABLE I. Linewidth parameters for barely metallic n-type silicon.

Dopant	$\Delta H_{pp}(n=n_c)$ (Oe)	M_2 (Oe^2)	$\langle\omega_{\mathrm{ex}}\rangle\times10^{-10}$ (rad/sec)	$\Delta H_{pp}(n\sim1.28n_c)$ (Oe)	$B(n=2n_c)$ (Oe)
Si:P	0.4	450	1.98	0.52 (Ref.5)	0.76
Si:As	3.4	6 310	3.53	5.6	13.6
Si:Sb	21	13 295	1.11	45	90 (est.)

for insulating samples in terms of a temperature-dependent exchange or motional narrowing $\omega_m = \omega_0 + \omega_h(T)$. Ochiai and Matsuura's results[6] suggest $\omega_h \propto T^{1/2}$. However, there has been no general agreement on the mechanism for this temperature dependence. Sachdev[18] has suggested that electron interaction effects, which also contribute to the spin susceptibility $\chi(T)$, provide the temperature dependence of ΔH_{pp} in the immediate vicinity of n_c. Meier, Parks, and Hale[17] have concluded the experimental results for Ge:As are not consistent with a conventional hopping contribution to ω_h and this conclusion may also be true for n-type Si. However, the temperature dependence of $\Delta H_{pp}(n,T)$ for $n < n_c$ is not relevant to the present discussion. The results suggests a much stronger density dependence for Si:Sb than for Si:P. Although we believe the $2.98\times10^{18}/\mathrm{cm}^3$ Si:Sb is very close to n_c (within $\pm1.5\%$), a 3% error in n_c toward the metallic side could increase the linewidth by 4 Oe, suggesting $\Delta H_{pp}(n=n_c)$ might be too large by this amount. This in turn would have the effect of increasing $\langle\omega_{\mathrm{ex}}\rangle$ by 23%, thus bringing the value closer to that for Si:P.

The last column in Table I indicates the magnitude of the mechanism for $n > n_c$ given by Zarifis and Castner[8] as $\Delta H_{pp,\mathrm{ex}} = \Delta H_{pp}(n) - \Delta H_{pp}(n=n_c) = B(n/n_c - 1)^p$, where B is the excess linewidth at $n = 2n_c$. B varies by roughly a factor of 100 from Si:P to Si:Sb, which is a factor of 2 greater than the variation of $\Delta H_{pp}(n=n_c)$. It has been demonstrated[8] that this strong donor dependence can be explained by the impurity SO interaction that splits the $1s$-T_2 states and is well documented from the Orbach spin-lattice-relaxation rate documented in the dilute limit ($N_d \leq 0.01n_c$). This $1/T_1$ process for barely metallic Si arises from the admixture of the $1s$-T_2 band into the ground state $1s$-A_1 band by the Anderson random potential.

One can make a rough estimate of both τ_{SO} and τ_e for the $1.28n_c$ Si:Sb sample using the linewidth shown in Table I and the scaling result $1/\tau_e(n) \sim 1/\tau_e(2n_c)(n/n_c - 1)^p$ using the same value of p (0.95) found for Si:As.[8] This estimate yields $\tau_{\mathrm{SO}} \sim 2.5\times10^{-9}$ sec and $\tau_e \sim 1.3\times10^{-13}$ sec, or $\tau_{\mathrm{SO}}/\tau_e \sim 1.9\times10^4$. Using the characteristic length $l \sim [3D\tau]^{1/2}$ one finds $l_{\mathrm{SO}}/l_e \sim 140$. Hence, even for Si:Sb where the impurity SO interaction is much larger than for Si:P, one still expects the corrections[11,12] to scaling theory from the impurity SO interaction to be small. Although these CESR results are for higher temperatures than some low-temperature studies[7,19,20] of Si:P, they still are in the same regime $kT/\hbar > g\mu_B H/\hbar \gg 1/\tau_{\mathrm{SO}}$. This suggests the physics should be the same. The shortest time scale is the elastic collision time τ_e on the metallic side of the transition.

In summary, the observation of the CESR linewidth for $n > n_c$ for Si:Sb is consistent with previous suggestions[5,8] that the CESR linewidth results form the impurity SO interaction associated with the $1s$-T_2 states. The linewidth very close to n_c results from the exchange-motional narrowing mechanism and provides an exchange-motional frequency close to that for Si:P. The results also suggest that corrections to scaling theory from the impurity SO interaction will be small and that Si:Sb as a metal-insulator transition system should be in the same universality class as Si:P and Si:As.

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