

## Spin relaxation of conduction electrons in *n*-type indium antimonide at low temperature

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We present a theory of the spin relaxation time of the conduction electrons in indium antimonide at liquid-helium temperature; earlier work on the subject is reviewed and discussed. The theory is compared with our measurements and the experimental data available in the literature: The relevant mechanism in highly doped samples ( $n \gtrsim 5 \times 10^{14} \text{ cm}^{-3}$ ) is shown to be scattering by ionized impurities. The spin-flip matrix element arises from the admixture of different spin states in the Bloch functions of the conduction band (Elliott process). Good agreement with experiment is obtained with no adjustable parameter. In the less-doped samples ( $n \lesssim 5 \times 10^{14} \text{ cm}^{-3}$ ) the currently invoked relaxation mechanism (modulation of the  $g$  factor or of the hyperfine interactions by the motion of the electrons) is shown to be ineffective. The impurity Elliott process is reconsidered; the effect of disorder is included by using a model of quasimacroscopic fluctuations of the electron density, which qualitatively accounts for the observed linewidths.

### I. INTRODUCTION

The spin resonance of conduction electrons in indium antimonide has been systematically investigated at low temperature by Isaacson.<sup>1</sup> The linewidth was measured as a function of electron concentration  $n$  for various samples from  $n \approx 3 \times 10^{13} \text{ cm}^{-3}$  up to  $n \approx 10^{16} \text{ cm}^{-3}$ . Gershenson, Pevin, and Fogelson reported similar experiments and also measured the linewidth as a function of temperature.<sup>2</sup> Here, our purpose will be to explain the low-temperature linewidths. At 1.3 °K the phonon processes do not contribute and the linewidth arises from impurity collisions. This process was mentioned by Elliott<sup>3</sup> and Yafet<sup>4</sup> but has often been overlooked in the literature<sup>2,5</sup>; no detailed calculation has been published until now. The equations used by some authors will be criticized; our theory will be compared with the experimental results of Refs. 1 and 2, and with our own linewidth and electron-mobility measurements.

### II. THEORY

#### A. Principle of the low-temperature spin-relaxation mechanism

The low-temperature spin-relaxation time arises from the spin-flip transition probability  $T_{\vec{k}\sigma \rightarrow \vec{k}'\sigma'}$  of an electron colliding with an impurity potential  $U$ . When calculated within the limits of first-order perturbation theory, this transition probability involves only the matrix element  $\langle \vec{k}\sigma | U | \vec{k}'\sigma' \rangle$ . This may be compared with the non-spin-flip matrix element  $\langle \vec{k}\sigma | U | \vec{k}\sigma \rangle$  which determines the momentum-relaxation time  $\tau$ .

Two contributions can lead to a nonvanishing spin-flip matrix element: (a) the impurity po-

tential  $U$  is the sum of two terms,  $U = V + \lambda$ ;  $V$  is a spin-independent potential, and  $\lambda$  is the spin-orbit part associated with  $V$ . This contribution has been considered for metals by Asik, Ball, and Slichter.<sup>6</sup> The corresponding spin-flip matrix element is of order  $\lambda a^3$  where  $\lambda$  is a typical spin-orbit energy, and  $a$  an atomic length. (b) As was first pointed out by Elliott,<sup>3</sup> a second contribution is due to the spin-orbit interaction in the host crystal, which results in a mixing of spin states in the eigenfunctions of the crystal Hamiltonian. The spin-flip matrix element is then of order  $V a^*$ , where  $a^*$  is some characteristic length of the range of  $V$ ; in a semiconductor,  $a^*$  is of the order of the first Bohr radius  $a_0^*$  of a neutral donor, which is much larger than an atomic length. For this reason, the Elliott process is expected to be much more efficient than the Asik process in most semiconductors, and especially in InSb, where  $a_0^* = 680 \text{ \AA}$ . In the following we shall be concerned only with the Elliott process, and thus we drop the spin-orbit part  $\lambda(\vec{r})$  of the impurity potential.

#### B. Detailed calculation of the impurity Elliott process

In order to compute the spin-flip matrix element between states  $|\vec{k}\rangle$  and  $|\vec{k}'\rangle$ , we need the expressions of the wave functions of the conduction band in a III-V semiconductor. These functions are of the Bloch type  $\psi_{\vec{k}\sigma} = u_{\vec{k}\sigma} e^{i\vec{k}\cdot\vec{r}}$ . The gap is direct and located at the center of the first Brillouin zone; at this point, the conduction-band Bloch function is  $s$ -like ( $u_0 = s$ ) and the valence-band ones ( $X, Y, Z$ ) are  $p$ -like. The valence band degeneracy is partially lifted by spin-orbit interaction. We call  $E_g$  the band gap and  $\Delta$  the spin-orbit splitting of the valence band.

Since we are concerned with small electron con-

centrations, the Fermi energy  $E_F$  remains much smaller than  $E_g$ , and we are mainly interested in the expressions of  $u_{\vec{k}\sigma}^{\uparrow}$  near the  $\vec{k}$  origin. These expressions may be derived, by  $\vec{k} \cdot \vec{p}$  perturbation theory, from the  $\vec{k}=0$  wave functions and energy levels. If one takes into account only the lower conduction band and the three upper valence bands, then the calculation may be carried out exactly. This is a very good approximation in InSb, because the energy gap is much smaller than the other interband spacings. The result has been given by Zawadzki.<sup>7</sup> Within our approximation  $E_F \ll E_g$ , it reduces to

$$|u_{\vec{k}\sigma}^{\uparrow}\rangle = |s\sigma\rangle + i \left( \frac{E_F}{E_g} \frac{3E_g + 2\Delta}{3(E_g + \Delta)} \right)^{1/2} \times \left( \frac{\vec{k} \cdot \vec{R}\sigma}{k_F} + \frac{2i\Delta}{2\Delta + 3E_g} \frac{\vec{S} \cdot (\vec{k} \times \vec{R}\sigma)}{k_F} \right), \quad (1)$$

where  $\vec{R}$  is the vector of components  $|X\rangle$ ,  $|Y\rangle$ ,  $|Z\rangle$ , and  $k_F$  is the Fermi wave vector. The admixture of spin states is brought by the spin operator  $\vec{S}$  in the last term and is of order  $(\Delta/E_g)(E_F/E_g)^{1/2}$ .

Once the expression for  $u_{\vec{k}\sigma}^{\uparrow}$  is known, we can compute the spin-flip matrix element  $\langle \vec{k}\sigma | V | \vec{k}'\sigma' \rangle$ . If we assume that  $V$  is slowly varying on the scale of a unit cell, and since  $u_{\vec{k}\sigma}^{\uparrow}$  has the lattice periodicity, the matrix element can be factorized

$$\begin{aligned} \langle \vec{k}\uparrow | V | \vec{k}'\downarrow \rangle &\approx V_{\vec{k}\vec{k}'} \langle u_{\vec{k}\uparrow}^{\uparrow} | u_{\vec{k}'\downarrow}^{\uparrow} \rangle \\ &\approx V_{\vec{k}\vec{k}'} \frac{E_F}{E_g} \frac{\Delta(\Delta + 2E_g)}{(\Delta + E_g)(2\Delta + 3E_g)} \\ &\quad \times \frac{k_x k'_x - k_y k'_y}{k_F}. \end{aligned} \quad (2)$$

Here the spin has been taken as quantized along the  $\vec{z}$  axis, and  $V_{\vec{k}\vec{k}'}$  is the matrix element of  $V$  between plane-wave states of wave vectors  $\vec{k}$  and  $\vec{k}'$ :

$$V_{\vec{k}\vec{k}'} = \iiint e^{i(\vec{k}' - \vec{k}) \cdot \vec{r}} V(\vec{r}) d^3r.$$

For the same slowly varying potential  $V$ , the non-spin-flip matrix element can be expressed as

$$\langle \vec{k}\sigma | V | \vec{k}'\sigma \rangle \approx V_{\vec{k}\vec{k}'} \langle u_{\vec{k}\sigma}^{\uparrow} | u_{\vec{k}'\sigma}^{\uparrow} \rangle \approx V_{\vec{k}\vec{k}'}. \quad (3)$$

The spin-flip and non-spin-flip transition probabilities  $T_{\vec{k}\uparrow \rightarrow \vec{k}'\downarrow}$  and  $T_{\vec{k}\sigma \rightarrow \vec{k}'\sigma}$  are then readily obtained from first-order time-dependent perturbation theory:

$$\begin{aligned} T_{\vec{k}\uparrow \rightarrow \vec{k}'\downarrow} &= \frac{2\pi}{\hbar} \delta(E_{\vec{k}'\downarrow} - E_{\vec{k}\uparrow}) V_{\vec{k}\vec{k}'}^2 \\ &\quad \times \left( \frac{E_F}{E_g} \right)^2 \left( \frac{\Delta(\Delta + 2E_g)}{(\Delta + E_g)(2\Delta + 3E_g)} \right)^2 \\ &\quad \times \frac{k_x^2 + k'_x{}^2 - 2k_x k'_x \cos\theta_{\vec{k}\vec{k}'}}{k_F^2}, \end{aligned}$$

$$T_{\vec{k}\sigma \rightarrow \vec{k}'\sigma} = \frac{2\pi}{\hbar} \delta(E_{\vec{k}'\sigma} - E_{\vec{k}\sigma}) V_{\vec{k}\vec{k}'}^2.$$

The spin relaxation time can then be computed together with the momentum relaxation time  $\tau$ , with the equations<sup>4, 8, 9</sup>

$$\frac{1}{T_1} = \frac{1}{T_2} = \iiint \frac{\delta(k - k_F) d^3k}{4\pi k_F^2} \times \left( \iiint \frac{d^3k'}{(2\pi)^3} 2T_{\vec{k}\uparrow \rightarrow \vec{k}'\downarrow} \right), \quad (4)$$

$$\frac{1}{\tau} = \iiint \frac{d^3k'}{(2\pi)^3} (1 - \cos\theta_{\vec{k}\vec{k}'}) T_{\vec{k}\sigma \rightarrow \vec{k}'\sigma}. \quad (5)$$

Our equation for the spin relaxation time refers to zero external magnetic field and weak polarization conditions, namely, the Fermi-Dirac distribution function  $f$  is assumed to be almost the same for the up and down electron subbands, i.e.,  $|f(E\uparrow) - f(E\downarrow)| \ll 1$ ; specifically, the polarization must be much smaller than  $k_B T / E_F$ . These conditions are experimentally fulfilled, since the resonance is usually performed at X-band frequency:  $g\mu_B B = \hbar\omega \ll k_B T$ , which is indeed the case for our measurements.

The exact calculation is performed by taking for  $V$  a screened Coulomb potential, which is adequate for InSb since the dominant scattering centers are ionized donors

$$V = (1/4\pi\epsilon_0)(e^2/r)e^{-k_s r}.$$

Rationalized units are used;  $\epsilon$  is the dielectric constant of the material and  $k_s$  the screening vector; according to Debye-Hückel theory in the case of Fermi statistics, we have  $k_s^2 = 3n/2\epsilon\epsilon_0 E_F$ . By substituting the matrix element  $\langle \vec{k}\sigma | V | \vec{k}'\sigma \rangle$  in the transition probability  $T_{\vec{k}\sigma \rightarrow \vec{k}'\sigma}$ , we obtain for the momentum relaxation time the well-known result of Brooks and Herring<sup>10</sup>

$$\frac{\hbar}{\tau} = \frac{\hbar^2}{m^* a_0^{*2}} \frac{N_i}{k_F^3} 2\pi \left( \ln(1+b) - \frac{b}{1+b} \right), \quad (6)$$

where  $b = 4k_F^2/k_s^2$  and  $N_i$  is the impurity density. The quantity  $\hbar^2/m^* a_0^{*2}$  is twice the effective Rydberg constant of the medium.

Similarly, by substitution of the spin-flip matrix element in the spin-flip transition probability and Eq. (4), we obtain the expression of the spin relaxation time

$$\frac{\hbar}{T_1} = \frac{\hbar}{T_2} = \frac{\hbar^2}{m^* a_0^{*2}} \frac{N_i}{k_F^3} \times \frac{2}{3} \left( \frac{\gamma E_F}{E_g} \right)^2 2\pi \left( \frac{b+2}{b} \ln(1+b) - 2 \right), \quad (7)$$

where  $\gamma$  is a function of  $\Delta/E_g$ ,

$$\gamma = 2\Delta(\Delta + 2E_g) / (2\Delta + 3E_g)(\Delta + E_g). \quad (8)$$

The  $\Delta$  factor in  $\gamma$  may be stressed by introducing the effective Landé factor  $g^*$  and the effective mass  $m^*$ :

$$\gamma = (2 - g^*)(m^*/m)(\Delta + 2E_g) / (\Delta + E_g). \quad (9)$$

In the limiting case  $b \rightarrow 0$ , our equation for  $1/T_2$  reduces to the result of Lewiner *et al.*<sup>11</sup> In the limiting case  $b \rightarrow \infty$ , we would expect to find the same result as Abakumov and Yassievich<sup>12</sup>; their relaxation rate appears, in fact, smaller than ours, by a factor  $\frac{3}{4}$  whose origin remains mysterious. Moreover we think that their equation is valid only for  $\Delta \gg E_g$ , since they implicitly make this assumption in the derivation of their Eq. (5).<sup>13</sup>

We may express the spin relaxation time as a function of the momentum relaxation time:

$$\frac{1}{T_1} = \frac{1}{T_2} = \frac{1}{\tau} \times \frac{2}{3} \left( \frac{\gamma E_F}{E_g} \right)^2 \frac{b+2}{b} \times \frac{\ln(1+b) - 2b/(b+2)}{\ln(1+b) - b/(1+b)}. \quad (10)$$

Since the first Born approximation is good only under the condition  $b \gg 1$ , the result becomes

$$\frac{1}{T_1} = \frac{1}{T_2} \approx \frac{1}{\tau} \times \frac{2}{3} \left( \frac{\gamma E_F}{E_g} \right)^2. \quad (11)$$

Because of the factor  $\gamma^2$ , the spin relaxation rate vanishes in the limit  $\Delta/E_g = 0$ ; this is to be expected, since mixing of spin states in the Bloch functions arises from spin-orbit interaction in the host crystal.

Our calculations have been carried out assuming Fermi-Dirac statistics. Although it is not relevant to the present experiments, appropriate averaging could be performed to obtain the corresponding results in the case of classical statistics. Assuming  $b \gg 1$ ;  $\tau$  is proportional to  $E^{3/2}$ ; for classical statistics, the  $E_F^{3/2}$  factor becomes<sup>9</sup>

$$\int_0^\infty E^{3/2} E^{3/2} e^{-E/k_B T} dE / \int_0^\infty E^{3/2} e^{-E/k_B T} dE = \frac{\Gamma(\frac{5}{2} + \frac{3}{2})}{\Gamma(\frac{5}{2})} (k_B T)^{3/2} = \frac{8}{\sqrt{\pi}} (k_B T)^{3/2}. \quad (12)$$

Similarly, the  $E^{1/2}$  dependence of  $1/T_1$  becomes

$$\int_0^\infty E^{1/2} E^{1/2} e^{-E/k_B T} dE / \int_0^\infty E^{1/2} e^{-E/k_B T} dE = \frac{\Gamma(\frac{3}{2} + \frac{1}{2})}{\Gamma(\frac{3}{2})} (k_B T)^{1/2} = \frac{2}{\sqrt{\pi}} (k_B T)^{1/2}. \quad (13)$$

Thus for classical statistics, Eq. (11) is replaced by

$$\frac{1}{T_1} = \frac{1}{T_2} \approx \frac{1}{\tau} \frac{32}{3\pi} \left( \frac{\gamma k_B T}{E_g} \right)^2. \quad (14)$$

Additional disagreement, of a factor of two, with Abakumov and Yassievich<sup>12</sup> for Eqs. (11) and (14) might arise from a different weighting factor in the averaging procedure for the definition of  $\tau$ .

At this point we can attempt a comparison of our theory with the experimental data in *n*-type InSb. Since our calculation proceeds from first-order perturbation theory, a good agreement is expected only for highly doped samples, where the first Born approximation is known to be valid.<sup>15</sup> The case of the less-doped samples is far more difficult and will be considered later.

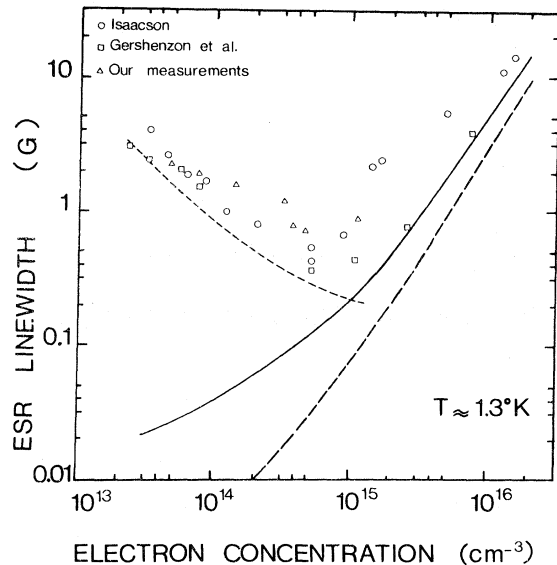


FIG. 1. Concentration dependence of the ESR linewidth (peak to peak of the derivative of the absorption curve). Dashed curve represents the results of our first-principles calculation [Eq. (17)] corrected for the electron-electron interactions. Solid curve corresponds to the semiempirical Eq. (11). Dotted curve in the lower-concentration range has been drawn according to Eq. (17), with an adjusted value  $\langle E_F^2 \rangle^{1/2} = 30^\circ \text{K}$ . Experimental points of various authors are reported on the same scale. Temperature is  $T = (1.3 \pm 0.1)^\circ \text{K}$ ; the results of Gershenson *et al.* were extrapolated from variable temperature measurements above  $1.7^\circ \text{K}$ . Linewidth is very sensitive to strains and surface defects so that the relevant points for comparison with theory are the lowest ones.

### III. COMPARISON WITH EXPERIMENT: HIGHER-CONCENTRATION RANGE

The linewidth, as measured by various authors, is plotted in Fig. 1 as a function of electron concentration  $n$ . As can be seen, there is a large dispersion of the experimental points. We do not think that this arises from a dispersion of the impurity content  $N_i$  due to the various origins of the samples. For higher-doped samples we can trust the impurity content  $N_i$  to be equal to the measured electron concentration  $n$ . Now in the case of the less-doped samples, where the compensation is important,  $N_i$  is the sum of the donor and acceptor contents  $N_D + N_A$ . But experience has shown<sup>15-17</sup> that for ingots of different origins  $N_i = N_D + N_A$  has an almost constant value, approximately  $4 \times 10^{14} \text{ cm}^{-3}$ . Thus, the samples obtained by different authors from an "undoped" commercial ingot are expected to exhibit similar characteristics, whatever be the subsequent doping method. We therefore believe that the dispersion of the experimental linewidths for different authors, or from one sample to the other, is due to the experimental difficulty of avoiding strains and surface defects, both of which can considerably increase the linewidth. The relevant experimental values for a comparison with our theory are thus the lowest ones.

Our samples were doped by neutron irradiation, according to the procedure described by Clark and Isaacson.<sup>18</sup> The electron and impurity content were deduced from classical transport measurements between 1.3 °K and 150 °K. The impurity content  $N_i$  as deduced from the 40 °K mobility measurements is of the form  $N_i \approx n + 4 \times 10^{14} \text{ cm}^{-3}$ . The momentum relaxation rate at 1.3 °K, as deduced from our mobility measurements, and in accordance with the literature,<sup>15</sup> is plotted in Fig. 2.

The theoretical curve on the same scale has been deduced from the Brooks-Herring formula [Eq. (6)], corrected for the electron-electron interactions which increase the momentum relaxation rate by a factor 1.58.<sup>19</sup> The critical electron concentration for the failure of the first Born approximation is clearly  $n \approx 5 \times 10^{14} \text{ cm}^{-3}$ . Below this concentration, the experimental points markedly depart from the theoretical curve. Moreover this concentration corresponds to a change of behavior of the linewidth.

Let us first focus our attention on the higher concentration range  $n \gtrsim 5 \times 10^{14} \text{ cm}^{-3}$ . In order to compare our theory with the experimental results, we can either compute  $1/\tau$  and  $1/T_2$  from first principles with Eqs. (6) and (7), or use the experimental value of  $1/\tau$  and compute  $1/T_2$  from  $1/T_2 = (1/\tau) \times \frac{2}{3} (\gamma E_F / E_g)^2$  [Eq. (11)]. This semiempirical

method is expected to give better results, since most of the difficulties are bypassed and the uncertainties are confined to a single dimensionless scaling factor. In the higher-concentration range, the two methods lead to the same result within a factor of three, provided the electron-electron interactions are taken into account in the first-principles calculation; this increases  $1/T_2$  [Eq. (7)] by a factor 1.58, in the same manner as  $1/\tau$ .<sup>19</sup> The theoretical value of the linewidth  $\Delta B$  can then be deduced using  $\Delta B = (2\hbar/\sqrt{3} g^* \mu_B) 1/T_2$ , where we take for  $\Delta B$  the derivative peak to peak width and we assume no inhomogeneous broadening of the line. The agreement with the measured values of  $\Delta B$  is good. The concentration dependence of the linewidth is of the type  $\Delta B \propto n^{1/3}$ .

### IV. DISCUSSION OF EXPERIMENTAL RESULTS IN THE LOWER-CONCENTRATION RANGE

In the lower-concentration range  $n \lesssim 5 \times 10^{14} \text{ cm}^{-3}$ , the linewidths as computed with the above equations become much smaller than the measured values. Moreover the observed linewidth varies as  $n^{-1}$ , in contrast with the high-concentration behavior. We first review and criticize the interpretations proposed in the literature to account for this concentration dependence; as they appear unsatisfying, we propose another explanation, based on an extension of the previous calculation.

The observed concentration dependence of the linewidth  $\Delta B \propto 1/n$  first suggested a relaxation mechanism of the type<sup>20</sup>

$$1/T_2 = \Omega^2 \tau_c . \quad (15)$$

The correlation time  $\tau_c$  associated with the inter-impurity motion, increases with decreasing concentration, which would give the right qualitative dependence. The magnetic energy term  $\Omega$  of Eq. (15), motion narrowed by  $\tau_c$ , should correspond to some magnetic interaction felt by an electron when located on an impurity center: shift of the  $g$ -value, or hyperfine interaction with donor nuclear spin or with the neighboring In and Sb nuclear spins.

The authors in Ref. 2 have proposed for  $\Omega$  a random distribution of  $g$  factors. This seems unlikely, since in this case the linewidth should increase with the resonance frequency, in disagreement with the experimental results of Ref. 1.

Another possibility for the origin of  $\Omega$  could be the hyperfine interaction with nuclear spins, as in phosphorus-doped silicon in the intermediate-temperature range.<sup>21</sup> This was suggested by the authors in Refs. 1 and 2. A quantitative discussion of this process is given in the Appendix, but the resulting linewidth is several orders of magnitude too small to account for the experimental results. We are thus left with the previous impurity Elliott

process; our treatment of Sec. II can be reconsidered and adapted to the lower concentration range, where the approximations  $b \gg 1$  and  $n \approx N_i$  are no longer valid.

#### V. IMPURITY ELLIOTT PROCESS IN THE LOWER-CONCENTRATION RANGE

For  $n \lesssim 5 \times 10^{14} \text{ cm}^{-3}$ , the parameter  $b$  becomes small and the first Born approximation fails. This failure is observable in the mobility behavior, since the calculated values of  $\tau$  become several times larger than the experimental ones. Higher Born approximations and multiple-scattering corrections for  $\tau$  have been successfully considered by Moore in a similar case.<sup>22</sup> This increases the momentum relaxation rate  $1/\tau$  by a factor of order  $1/b$ , which improves somewhat the agreement with experiment.

For the spin relaxation rate  $1/T_2$ , we have carried our previous calculation of Sec. II one order further in the Born expansion. An enhancement factor appears at low  $b$  but it is rather smaller than for  $1/\tau$ . This can be understood if a partial-wave analysis is used to solve the scattering problem. This has been done by Krieger and Strauss.<sup>23</sup> They find that most of the enhancement factor for  $1/\tau$  arises from an increase of the  $s$  phase shift ( $l=0$ ); the other phase shifts remain small and are correctly given (within 15%) by the first Born approximation. On the other hand, for the spin relaxation rate, the formulation of Nozières and Lewiner<sup>24</sup> shows that the spin-flip matrix element may be viewed as the matrix element between plane-wave states of an effective spin-orbit interaction, and thus vanishes for an  $s$  state. Thus the increase of the  $s$  phase shift, which is the principal source of the enhancement factor for  $1/\tau$ , is ineffective for  $1/T_2$ .

This argument falls down if multiple-scattering corrections are taken into account: In this case the whole scattering potential no longer has spherical symmetry, and a partial-wave analysis is irrelevant. The Born expansion is the only firm ground for this problem, but the corresponding terms are very difficult to handle. In fact, this is a problem of disordered systems. We thus content ourselves with a somewhat qualitative argument, given as follows: In low-doped InSb, the impurity content  $N_i = N_D + N_A$  is much greater than the electron concentration  $n = N_D - N_A$  and the interimpurity spacings become smaller than the Debye screening length. In such conditions, the random character of the impurity distribution leads to quasimacroscopic fluctuations of the self-consistent potential seen by an electron.

If the scale of the fluctuations is assumed to be

much greater than the electron mean free path, then a local Fermi energy  $E_F(\vec{r})$  can be defined from the local electron density  $n(\vec{r})$ . In spite of these local fluctuations, the resonance line still appears as a single homogeneous line, since, in any case, the scale of the fluctuations is much smaller than the spin diffusion length  $[(DT_1)^{1/2} \approx 1 \mu\text{m}]$ . The observed linewidth is then given by the average spin-relaxation rate

$$\left\langle \frac{1}{T_2} \right\rangle = \iiint \frac{1}{T_2(\vec{r})} n(\vec{r}) d^3r / \iiint n(\vec{r}) d^3r. \quad (16)$$

Since  $1/T_2$  is a fast rising function of  $n$ , this averaging procedure strongly favors the higher-concentration regions, which explains qualitatively the high value of the observed linewidth.

In fact, the scale of the fluctuations  $l$  and the electron mean free path  $\lambda$  are of the same order of magnitude ( $\lambda \approx l \approx a_0^* \approx 10^3 \text{ \AA}$ ) and most probably the momentum relaxation rate does not exhibit strong spatial fluctuations. Then, if the above argument holds, and if the semiempirical Eq. (11) is used for  $1/T_2$ ,

$$1/T_2(\vec{r}) = (1/\tau) \times \frac{2}{3} (\gamma E_F(\vec{r})/E_g)^2$$

and

$$\langle 1/T_2 \rangle = (1/\tau) \times \frac{2}{3} \gamma^2 \langle E_F^2 \rangle / E_g^2. \quad (17)$$

This equation fits the experimental points in the lower-concentration range with an almost constant value  $\langle E_F^2 \rangle^{1/2} \approx 30 \text{ }^\circ\text{K}$  (see Fig. 1). This value is consistent with the  $g$ -vs- $T$  measurements of Kaplan and Konopka,<sup>25</sup> from which a quite similar value can be obtained.<sup>26</sup> Although our argument is rather qualitative, we obtain the right order of magnitude,

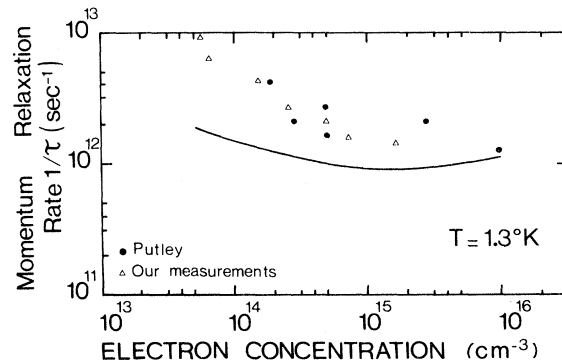


FIG. 2. Concentration dependence of the momentum-relaxation rate. Curve is theoretical and corresponds to the Brooks-Herring equation corrected for the electron-electron interactions. Our experimental points are reported on the same scale, together with the data of Putley (see Ref. 15). Latter have been extrapolated to the temperature  $T = 1.3 \text{ }^\circ\text{K}$ .

and thus we think that this local Elliott process is the clue to the observed linewidth in the lower-concentration range. The less ambiguous test of this mechanism would be the investigation of purer uncompensated samples, where the linewidth is expected to be smaller.

## VI. CONCLUSION

A detailed analysis of the spin-flip Elliott scattering of conduction electrons by charged impurities has been given. The results for the ESR linewidth in indium antimonide are in good agreement with experiment in the higher-concentration range ( $n \geq 5 \times 10^{14} \text{ cm}^{-3}$ ). In the lower-concentration range, the usually invoked relaxation process (modulation by the interimpurity motion of the  $g$  factor or of the hyperfine interaction) has been considered quantitatively and shown to give a relaxation rate too small by several orders of magnitude. Most probably the linewidth in the lower-concentration range is still accounted for by the Elliott process, provided allowance is made for the local fluctuations of the electron density.

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## APPENDIX: CONTRIBUTION OF HYPERFINE INTERACTIONS TO THE LINewidth (LOWER-CONCENTRATION RANGE)

Our purpose is to evaluate the order of magnitude of a process of the type  $1/T_2 = \Omega^2 \tau_c$ , where  $\tau_c$  is the correlation time associated with the interimpurity motion, and  $\Omega$  is the hyperfine interaction felt by an electron when located on a neutral donor.

At the concentration of  $10^{14} \text{ cm}^{-3}$ , where the Fermi energy  $E_F$  is of the same order of magnitude as  $k_B T$  and  $E_i$  (ionization energy of isolated donors), the correlation time  $\tau_c$  must be of order  $\tau_c \approx \hbar/E_F \approx 10^{-12} \text{ sec}$ . When the electron concentration becomes lower than  $10^{14} \text{ cm}^{-3}$ ,  $\tau_c$  increases above this value.

The spin relaxation rate for  $n = 10^{14} \text{ cm}^{-3}$  is  $1/T_2 = 4 \times 10^8 \text{ sec}^{-1}$ ; if it were ascribed to the present mechanism, this would require an interaction  $\Omega \approx 2 \times 10^{10} \text{ sec}^{-1}$ .

In order to see whether such a large value of  $\Omega$  is attainable, we now look for the hyperfine interaction of a donor electron with the nuclear spins in InSb.

We consider first the hyperfine interaction with the donor nucleus. The donors are most probably Sn or Te atoms; in both cases the hyperfine inter-

action in the isolated atom is the order of  $10^{11} \text{ sec}^{-1}$ ,<sup>27</sup> but the rather weak natural abundance of the magnetic isotopes of these nuclear species (16% and 8%, respectively) leads to a final hyperfine interaction  $\Omega \approx 3 \times 10^{10} \text{ sec}^{-1}$ , which is the right order of magnitude. However, this estimate requires the assumption that the donor wave function  $\psi$  near the impurity nucleus is the same as in the isolated impurity atom. This is completely impossible, since the effective-mass wave function of a neutral donor spreads over a great number of unit cells ( $a_0^* = 680 \text{ \AA}$ ), and even if central-cell corrections were important, such a value of  $\psi$  near the impurity nucleus is far from being attainable. Therefore the hyperfine interaction with the donor nuclei cannot be reasonably regarded as the cause of the observed linewidth.

A more probable origin for  $\Omega$  might be the hyperfine interaction with the In and Sb nuclei, whose magnetic moment and natural abundance are particularly large. We then consider the hyperfine interaction of a donor electron with a nuclear spin  $\vec{I}_i$  of the crystal<sup>20</sup>:

$$\hbar\Omega_i = \left[ \frac{4}{3} \mu_0 \mu_B (\mu_i/I_i) m_i \right] |F(\vec{r}_i)|^2 \eta_i, \quad (18)$$

where  $\mu_i$ ,  $m_i$ ,  $\vec{r}_i$  are, respectively, the nuclear magnetic moment, the  $z$  component of the nuclear spin, and the position of the nucleus.  $F(\vec{r})$  is the electron-envelope wave function.  $\eta_i$  represents the bunching of the crystal eigenfunctions at the nuclei. According to the day-shift data of Guéron<sup>28</sup> in InSb, we have  $\eta_i = 1.1 \times 10^4$  for Sb nuclei and  $\eta_i = 0.635 \times 10^4$  for In nuclei. For an electron in a donor state, we can sum the contributions of all the In and Sb nuclei; since the directions of two nuclear spins are uncorrelated, the result is

$$\Omega = \left( \sum \Omega_i^2 \right)^{1/2}. \quad (19)$$

If the hydrogenoid form is assumed for the wave-function envelope  $F(\vec{r})$ , we find

$$\hbar\Omega = \frac{8\mu_0\mu_B}{3(2\pi)^{1/2}} \frac{1}{(a_0^*a_i)^{3/2}} \times \left[ \left( \frac{\mu_{\text{In}}}{I_{\text{In}}} \right)^2 \langle m_{\text{In}}^2 \rangle \eta_{\text{In}}^2 + \left( \frac{\mu_{\text{Sb}}}{I_{\text{Sb}}} \right)^2 \langle m_{\text{Sb}}^2 \rangle \eta_{\text{Sb}}^2 \right]^{1/2}, \quad (20)$$

where  $\mu_B$  is the Bohr magneton,  $a_i$  is the lattice constant, and  $a_0^*$  is the effective Bohr radius. With  $a_i = 6.48 \text{ \AA}$  and  $a_0^* = 680 \text{ \AA}$ , the result is  $\Omega = 0.7 \times 10^8 \text{ sec}^{-1}$ , which is two orders of magnitude smaller than the required value.

The physical significance of this result is as follows: If the wave function were confined to a unit cell, i.e., a pair of In and Sb atoms, the hyperfine interaction  $\Omega_{\text{uc}}$  would be of order  $3 \times 10^{11}$

sec<sup>-1</sup>. When the wave function spreads over a number  $N$  of unit cells, the hyperfine interaction of the electron with a given pair of In and Sb nuclear spins is smaller than  $\Omega_{uc}$  by a factor  $N$ , due to the normalization factor of the wave function. As the electron can see  $N$  such pairs, the hyperfine interaction is increased again, but only by a factor  $\sqrt{N}$ , since the directions of the nuclear spins are uncorrelated. The final result is  $\Omega = \Omega_{uc} / \sqrt{N}$ . For the hydrogenoid donor wave function,  $N$  is of order  $10^7$ , which reduces the hyperfine interaction to the previously calculated value  $\Omega \approx 10^8$  sec<sup>-1</sup>.

This result might be in error if central-cell corrections to the donor ground state were important, namely if a small part  $\alpha$  of the donor wave function were localized in a small volume  $v$  around the impurity ion ( $\alpha = \iiint_v |\psi(\vec{r})|^2 d^3r$ ); in this case

if the volume  $v$  contained  $n$  unit cells, the hyperfine interaction would be  $\Omega \approx \Omega_{uc} \alpha / \sqrt{n}$ , which might be greater than  $10^8$  sec<sup>-1</sup>. However taking for  $n$  a few units, a value of  $10^{-1}$  for  $\alpha$  would still be required to reach the needed value  $\Omega \approx 2 \times 10^{10}$  sec<sup>-1</sup>. Although we lack knowledge about central-cell corrections for donors in InSb, a rough evaluation of  $\alpha$  can be made by writing the energy shift of the donor ground state due to the localized part of the donor potential. If kinetic-energy terms are neglected, this gives:  $\Delta E \approx \alpha E_{at}$ , where  $E_{at}$  is some atomic energy. The shift  $\Delta E$  of the donor ground state is certainly less than  $10^{-3}$  eV, otherwise this would affect the transport properties. Thus the most we could hope for  $\alpha$  is  $\alpha \approx (10^{-3} \text{ eV}) / (1 \text{ eV}) = 10^{-3}$ . The required value  $\alpha \approx 10^{-1}$  is thus unattainable and the contribution to the linewidth of the hyperfine interactions is negligible.<sup>29</sup>

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<sup>4</sup>Y. Yafet, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic, New York, 1963), Vol. 14, pp. 1–98.

<sup>5</sup>K. Sugihara, J. Phys. Soc. Jpn. **28**, 402 (1970).

<sup>6</sup>J. R. Asik, M. A. Ball, and C. P. Slichter, Phys. Rev. **181**, 645 (1969).

<sup>7</sup>W. Zawadzki and W. Szymańska, Phys. Status Solidi B **45**, 415 (1971).

<sup>8</sup>In the equation for the spin-relaxation rate, the summation over all the directions of  $\vec{k}$  occurs because the expression in the square bracket depends on the relative direction of  $\vec{k}$  and the axis of spin quantization. An alternative procedure would be to hold  $\vec{k}$  fixed and average over the spin direction. This would naturally lead to the same result.

<sup>9</sup>J. M. Ziman, *Electrons and Phonons* (Clarendon, Oxford, 1960), Chap. VII.

<sup>10</sup>See, for example, P. P. Debye and E. M. Conwell, Phys. Rev. **93**, 693 (1954).

<sup>11</sup>C. Lewiner, O. Betbeder-Matibet, and P. Nozières, J. Phys. Chem. Solids **34**, 765 (1973).

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<sup>13</sup>Equation (4) of Ref. 14 probably refers to strong shielding conditions ( $b \ll 1$ ); this may be inferred from the  $E_F^3$  dependence of the spin-relaxation rate in the case of Fermi statistics. However we could not identify it with our equation because of the occurrence of the dielectric constant ( $\kappa$  in their notation) in the so-called interaction constant  $C_{si}$ . Taking for  $\kappa$  the value in InSb ( $\kappa = 17.5$ ),

the two equations agree within a factor of 2, which seems fortuitous.

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<sup>20</sup>A. Abragam, *The Principles of Nuclear Magnetism* (Clarendon, Oxford, 1961), Chap. VIII.

<sup>21</sup>D. J. Lépine, Phys. Rev. B **2**, 2429 (1970).

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<sup>23</sup>J. B. Krieger and S. Strauss, Phys. Rev. **169**, 674 (1968).

<sup>24</sup>P. Nozières and C. Lewiner, J. Phys. (Paris) **34**, 901 (1973).

<sup>25</sup>D. Kaplan and J. Konopka, in *Proceedings of the Ninth International Conference on the Physics of Semiconductors, Moscow, 1968* (Nauka, Leningrad, 1968), p. 1146.

<sup>26</sup>Near the bottom of the conduction band in InSb, the  $g$  factor increases linearly with energy. For a given sample the  $g$ -vs- $T$  curve should then exhibit a linear dependence for  $k_B T \gg E_F$ , and a nearly constant behavior for  $k_B T \ll E_F$ ; the change of slope should occur at the temperature  $T = \frac{2}{3}(E_F/k_B)$ . In fact, for a sample in the lower-concentration range there is still a linear dependence in the region  $k_B T \ll E_F$ ; this is consistent with the picture of a fluctuating Fermi energy. The curve from Kaplan and Konopka (see Ref. 25) exhibits a change of slope near  $T_c \approx 15$  °K. This value corresponds roughly to the standard deviation of the probability distribution of  $E_F$ , say  $\bar{E}_F/k_B \approx 3T_c/2 \approx 23$  °K.

<sup>27</sup>C. F. Fisher, *Some Hartree-Fock Results For The Atoms Helium To Radon*, edited by the University of Waterloo (Waterloo, Ontario, 1968).

<sup>28</sup>M. Guéron, Phys. Rev. **135**, A200 (1964).

<sup>29</sup>Another origin for  $\Omega$  might be the presence of magnetic impurities inside the spreading region of a donor wave function. The resulting relaxation rate would be improved by a factor of order  $(\mu_B/\mu_i)^2 \approx 10^6$ , but because of the very small concentration of such impurities,

$10^{-9}$ – $10^{-7}$  [see F. Bridges and W. G. Clark, Phys. Rev. 182, 463 (1969)], it finally turns out to be several orders of magnitude smaller than the effect of the hyperfine interaction with In and Sb nuclei.