

## Temperature dependence of the electron Landé $g$ factor in InSb and GaAs

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We report Larmor precession in bulk InSb observed in the time domain from 77 to 300 K. The optically oriented polarization precesses coherently even at 300 K. The inferred Zeeman spin splitting is strongly nonparabolic, and the electron  $g$  factor ( $g^*$ ) is in good agreement with  $\mathbf{k}\cdot\mathbf{p}$  theory (provided we take only the dilational contribution to the change in energy gap with temperature). We also show here that correct application of the 14-band  $\mathbf{k}\cdot\mathbf{p}$  model agrees with apparently anomalous trends previously reported for GaAs and confirm that the most widely quoted formula for  $g^*$  in GaAs is incomplete.

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InSb is an interesting semiconductor from the point of view of tests of semiconductor band structure calculations because the heavy constituent atoms produce large relativistic effects such as spin-orbit coupling (responsible for the large, negative gyromagnetic ratio). InSb is also a candidate material for room-temperature spintronic devices such as the Das-Datta spin transistor, which relies on a coherent spin population manipulated by the Rashba effect, and thus, detailed investigation of the spin-electronic structure in this material at room temperature is of high topical interest. In the present work, we report the experimental measurement of the  $g$  factor in InSb at temperatures up to 300 K and the theoretical evaluation of  $g^*(T)$  for both InSb and GaAs.

Although it has been claimed that measurements of the  $g$  factor of GaAs at 300 K are inconsistent with  $\mathbf{k}\cdot\mathbf{p}$  perturbation theory,<sup>1-4</sup> this theory has been successfully used for decades to calculate the band structure in bulk semiconductors and heterostructures,<sup>5-14</sup> and, in particular, the conduction band effective mass and  $g$  factor. We show here that provided we include only the dilational change of the energy gap with temperature,<sup>8,13</sup> we obtain reasonable agreement between experiment and theory for the high-temperature  $g$  factor in both InSb and GaAs, and there is no anomaly. The higher band  $\mathbf{k}\cdot\mathbf{p}$  parameters have only a very small effect on the electron  $g$  factor for InSb, but they are very important for GaAs; in particular, we confirm that it is essential to include the effects of the interband spin-orbit coupling parameter, which was previously ignored in the Hermann and Weisbuch formula for  $g^*$  (Ref. 9) used in Refs. 1 and 2.

In an externally applied magnetic field, the electron energy is given by

$$E_{\pm,n,k_B}(B) = \frac{\hbar^2 k_B^2}{2m^*} + \left( n + \frac{1}{2} \right) \frac{\hbar e B}{m^*} \pm \frac{1}{2} g^* \mu_B B, \quad (1)$$

measured from the  $\Gamma_6$  conduction band edge, where  $\mu_B$  is the Bohr magneton, the quantum numbers  $\pm$  refer to the spin,  $n$  to the Landau level index, and  $k_B$  to the component of momentum parallel to the magnetic field  $B$ . In the parabolic

approximation, the effective mass and  $g$  factor,  $m^*$  and  $g^*$ , are constants and independent of  $n$ ,  $k_B$ , and  $B$ . The nonparabolicity of GaAs is usually taken to be small because the dependence of the effective mass on electron energy is small.<sup>7</sup> However, the  $g$  value, although small in magnitude, is significantly nonparabolic and changes from  $-0.44$  at the band edge for low temperature to zero at an excess energy of 80 meV.

We used a midinfrared circularly polarized pump-probe transient absorption technique, which we have previously used for investigation of spin relaxation in narrow-gap semiconductors.<sup>15,16</sup> In this experiment, a subpicosecond laser pulse excites electrons which quickly thermalize during the pulse, filling up the conduction band, and the resulting change in absorption (a dynamic Moss-Burstein shift) is measured by the probe pulse. For InSb, both the light hole and electron effective masses are very much less than the heavy hole mass (by a factor of  $\sim 30$ ), so that it is a good approximation to assume that the valence band is flat. The laser induced transitions involve electron states whose kinetic energy is simply given by the laser photon energy minus the optical energy gap. Bleaching of the probe absorption, which is easy to achieve due to the small density of states, therefore occurs when the conduction quasi-Fermi energy is also equal to the difference in the laser and band gap energies. By inclusion of a small magnetic field in the Voigt geometry, the optically oriented spins perform Larmor precession and a sinusoidal modulation of the transmission change is produced.<sup>1-4,15,17</sup> The experimental sensitivity on determination of the precession frequency, and hence  $g^*$ , was limited by the electron spin lifetime which limits the number of cycles for available magnetic fields. The InSb sample used for our study was undoped and 5  $\mu\text{m}$  thick, grown by molecular-beam epitaxy on a semi-insulating GaAs substrate. The mobility and carrier concentration ( $n$  type) measured by the Hall effect at 300 K (77 K) were  $6.37 \text{ m}^2/\text{V s}$  ( $3 \text{ m}^2/\text{V s}$ ) and  $1.4 \times 10^{15} \text{ cm}^{-3}$  ( $2.8 \times 10^{15} \text{ cm}^{-3}$ ).

Typical transients for different excitation wavelengths at 77 K with an applied magnetic field of 0.288 T are shown in

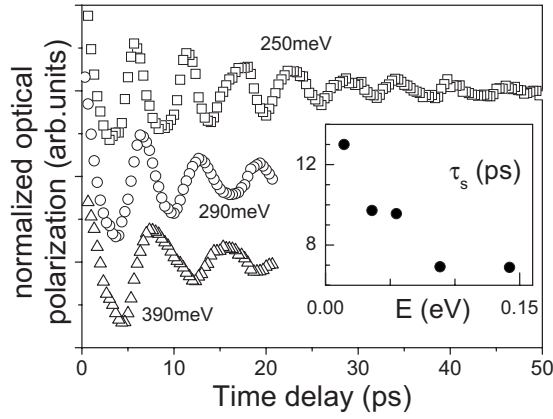


FIG. 1. Measured spin precession as a function of time for InSb taken using the circularly polarized pump-probe technique at 77 K and in an applied Voigt geometry magnetic field of 0.288 T. The optical excitation was of different wavelengths or photon energies:  $5 \mu\text{m} \equiv 250 \text{ meV}$  (circles),  $4.3 \mu\text{m} \equiv 290 \text{ meV}$  (triangles), and  $3.2 \mu\text{m} \equiv 390 \text{ meV}$  (squares). The spin lifetime taken from the decay of the envelope as a function of the excess energy above the optical gap is shown in the inset.

Fig. 1. The shapes of the transients are well described by a simple exponentially decaying sinusoid; the measured polarization precesses coherently at angular frequency  $g^* \mu_B B / \hbar$  while decaying with the spin lifetime  $\tau_s$ . The measured period of the oscillation becomes longer for excitation energies above the band gap illustrating the reduction of the absolute value of the effective  $g$  factor with the electron energy due to nonparabolicity. The macroscopic polarization decays because spins of different quantum numbers  $n$  or  $k_B$  precess at different rates due to the nonparabolicity of  $g^*$ , so the coherence is lost by dephasing. Increasing the pump photon energy increases the electron quasi-Fermi energy and therefore reduces the coherence time, as shown in the inset of Fig. 1. The effective  $g$  factor obtained from the transients for representative temperatures is plotted in Fig. 2 against excess optical energy above the band gap. The experiment is insensitive to the sign of  $g^*$ , and we assume that it is negative.

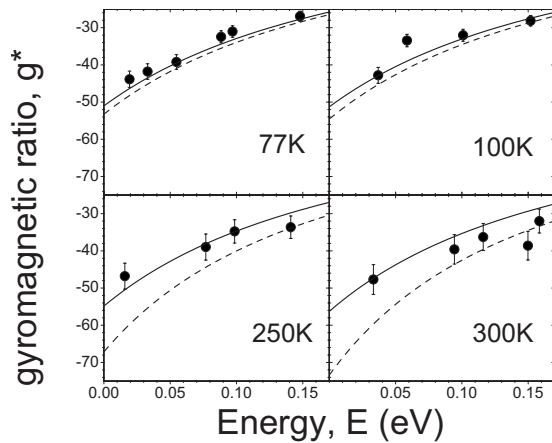


FIG. 2. Dependence of the  $g$  factor (symbols) on photon energy at different temperatures for InSb. The theoretical dependence from Eq. (3) is shown as a solid line (dilatational change of band gap) and dashed line (full optical band gap).

Modeling of InSb using  $\mathbf{k}\cdot\mathbf{p}$  theory requires only eight bands<sup>6,10</sup> (i.e., four spin-split bands: the three  $p$ -like  $\Gamma_7$  and  $\Gamma_8$  valence bands—heavy hole, light hole, and spin-orbit split-off bands—and the lowest  $\Gamma_6$   $s$ -like conduction band) because the energy separation between the  $\Gamma_6$  band and the next conduction level is much greater than the fundamental gap. Accurate modeling of GaAs on the other hand requires 14 bands (i.e., also including the higher  $p$ -like  $\Gamma_7$  and  $\Gamma_8$  conduction bands). The band-edge  $g$  factor from the 14-band model may be written analytically,<sup>7</sup>

$$\frac{g^*}{2} = 1 - \frac{E_{P0}}{3} \left( \frac{1}{G_0} - \frac{1}{E_0} \right) + \frac{E_{P1}}{3} \left( \frac{1}{G_1} - \frac{1}{E_1} \right) - \frac{2}{9} \bar{\Delta} \sqrt{E_{P0} E_{P1}} \left( \frac{2}{E_1 G_0} + \frac{1}{E_0 G_1} \right) + C', \quad (2)$$

where  $E_{P0}$  and  $E_{P1}$  are the squared interband momentum matrix elements,  $E_0$ ,  $G_0$ ,  $E_1$ , and  $G_1$  are the energies of the  $\Gamma_8$  and  $\Gamma_7$  valences and  $\Gamma_7$  and  $\Gamma_8$  conduction bands, respectively, measured relative to the  $\Gamma_6$  conduction band edge. The valence and higher conduction band spin-orbit splittings are  $\Delta_0 = G_0 - E_0$  and  $\Delta_1 = G_1 - E_1$ , and  $\bar{\Delta}$  is the spin-orbit coupling energy between the higher conduction band and the valence band. Equation (2) reduces to the expression derived by Hermann and Weisbuch<sup>9</sup> and used in Refs. 1 and 2 if one puts  $\bar{\Delta} = 0$ . [N.B., additional small corrections due to  $\bar{\Delta}$  to the energies  $E_0$ , etc., in the full formula from Ref. 7 have negligible effect and have been ignored in Eq. (2)]. For narrow-gap semiconductors (NGSSs), where  $E_0$  is small compared with the other gaps, it is necessary to retain only the second term on the right hand side of Eq. (2). It is plain that for InSb, accurate knowledge of fewer parameters is required.

For negligible higher band interactions, the energy dependence of the  $g$  factor for NGSSs may be written analytically<sup>10</sup> if the spin splitting is small compared with  $E - E_0$  (which it is for all reasonable magnetic fields),

$$\frac{g^*(E)}{2} = 1 - \frac{E_{P0}}{3} \left( \frac{1}{\Delta_0 + E_0 - E} - \frac{1}{E_0 - E} \right). \quad (3)$$

In the case of InSb at low temperature where<sup>11</sup>  $E_0 = -0.2352 \text{ eV}$ ,  $E_{P0} = 23.1 \text{ eV}$ , and  $\Delta_0 = -0.803$ , this leads to  $g^*(E=0, T=0) = -51$  (with negligible correction if we include the higher band terms), and the low-temperature slope  $dg^*/dE$  is approximately  $260 \text{ eV}^{-1}$  at the band edge. The use of the full  $8 \times 8$  Pidgeon-Brown matrix diagonalization<sup>6</sup> instead of Eq. (3) results not only in an increase in the slope  $dg^*/dE$  but also in an increase in the nonparabolicity so that the correction to  $g^*(E)$  is small. The model and the parameters agree well with experiment for low-temperature spin resonance.<sup>12</sup>

In order to model the temperature dependence of  $g^*$  from Fig. 2 (or of  $m^*$ ), the temperature dependent band gap is required. The temperature dependence of the optical band gap is well known. However, there are two contributions to the temperature change of the optical gap, which are firstly due to lattice dilation arising from the fact that the ionic potential is anharmonic,  $(\partial E_0 / \partial T)_P$ , and secondly the vibrational part is  $(\partial E_0 / \partial T)_V$ . It is well known that the effective

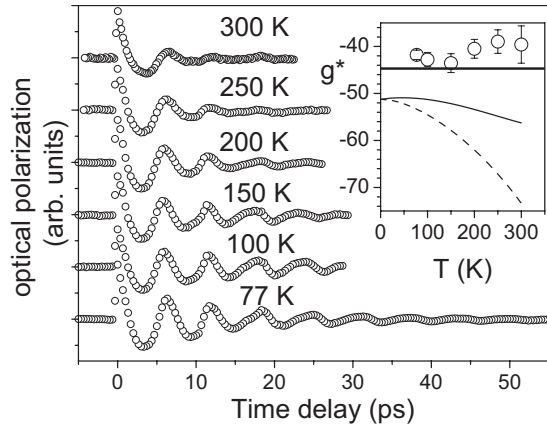


FIG. 3. (a) As in Fig. 1 but for different temperatures at a fixed excitation wavelength of  $4.7 \mu\text{m} \equiv 260 \text{ meV}$ . (b) The gyromagnetic ratio inferred from the transients of the main figure (circles). The thick solid line is calculated from Eq. (3), which shows that for fixed photon energy,  $g = -45$  is a constant. The rise in the data with temperature is due to the effects of higher bands. The thin lines are the band-edge  $g$  values calculated using the dilational (solid) and optical (dashed) band gap changes with temperature.

mass follows the dilational contribution only, since the vibrations occur on a much slower time scale than the time the electron takes to sample the interband interactions that determine the mass, which are at optical frequency. This has been used successfully to explain both temperature and pressure dependences of the effective mass for a variety of materials including InSb and GaAs.<sup>8,13</sup> Following Refs. 8, 13, and 18, we calculate the dilational change in the energy gap using InSb expansion coefficient measurements from Ref. 19. An approximate parametrization of the dilational change in  $E_0$  over the range of 0–300 K for InSb leads to a  $E_0^{\text{dil}} = -0.2352 + 5.17 \times 10^{-7}T^2 - 9.25 \times 10^{-9}T^3 + 3.81 \times 10^{-11}T^4 - 5.12 \times 10^{-14}T^5$ , where  $T$  is in kelvin. Simple consideration of the momentum operator suggests that the interband momentum matrix element should also scale (inversely) with the lattice dilation, so its square,  $E_{p0}$ , changes no more than 0.1% from 0 to 300 K. The spin-orbit splitting  $\Delta_0$  measured by modulated reflectance is temperature insensitive for the range from 5 to 300 K for InSb.<sup>14</sup> We therefore assume that other than the fundamental band gap, all band parameters are independent of temperature.

As can be seen from Fig. 2, Eq. (3) is quite consistent with the experimental data *without any fitting parameters*. Clearly, the data of Fig. 2 at high temperature (particularly, at low energies) is in better agreement with the theory including only the dilational band gap, in accord with the previous work on effective mass. The clinching point is that at the lowest energies, the *measured*  $g$  factor tends to a value of around  $-56$  at 300 K, corresponding to the dilational band gap of  $-0.217 \text{ eV}$ , and not to a value of  $-73$ , corresponding to the optical band gap of  $-0.174 \text{ eV}$ . We note that incomplete bleaching, if occurring, would be especially apparent at high energy and high temperatures, and the experimental points would fall to the right of the line. There may be some evidence of this at 300 K above 0.1 eV, but for lower energy or lower temperature, no significant effect of reducing the laser intensity was seen.

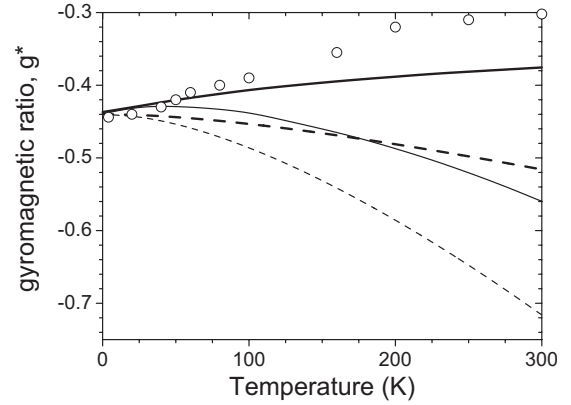


FIG. 4. Experimental and theoretical temperature dependences of  $g$  factor for GaAs. Data are taken from Ref. 1 (circles). The dashed lines are the band-edge  $g$  value of Eq. (2). The solid lines were calculated using the full 14-band  $\mathbf{k} \cdot \mathbf{p}$  matrix method (Ref. 7) at  $E = kT$  above the conduction band edge, appropriate for photoluminescence. The thin lines include the temperature variation of  $E_0$  and  $E_1$  using the optical gap and the higher band energy both from Ref. 20, whereas the thick lines use the same  $E_1$  variation but only the dilational part of the fundamental band gap change from Ref. 18.

According to Eq. (3), the  $g$  factor does not depend on temperature as long as the excitation photon energy is kept constant ( $h\nu = E - E_0 = \text{const}$ ), and any departures from this give a direct measure of the effect of higher bands. We have also performed such an experiment where we keep the photon energy constant and vary the temperature. The resulting transients are presented in Fig. 3, and we see a small reduction in the magnitude of the  $g$  factor associated with the higher band interactions. Figure 3 also shows the prediction for the band-edge  $g$  factor using the dilational change.

Having shown that the temperature dependence of the  $g$  factor for InSb, obtained from our own transient absorption measurements, can be satisfactorily explained by  $\mathbf{k} \cdot \mathbf{p}$  perturbation theory (provided that one includes only the dilational contribution to the change of energy gap), we may consider the situation for GaAs. For GaAs, the low-temperature parameters<sup>7</sup> are  $E_0 = -1.519 \text{ eV}$ ,  $\Delta_0 = -0.341 \text{ eV}$ ,  $E_{p0} = 27.86 \text{ eV}$ ,  $E_1 = 2.969 \text{ eV}$ ,  $\Delta_1 = 0.171 \text{ eV}$ ,  $E_{p1} = 2.36 \text{ eV}$ ,  $\bar{\Delta} = -0.061 \text{ eV}$ , and  $C' = -0.0215$ , leading to  $g^*(T=0) = -0.44$  from Eq. (2). Because of the near cancellation of the first and second terms on the right hand side in Eq. (2), the fourth term amounts to 30% of  $g^*$  for GaAs and must not be neglected, even though  $\bar{\Delta}$  is small. The third and fifth terms contribute only 5% each. Incomplete use of the Hermann and Weisbuch formula<sup>9</sup> for  $g^*$  requires modification of the other parameters to compensate, as was done in Refs. 1 and 2. In contrast, the widely used Hermann and Weisbuch formula for effective mass is perfectly adequate for GaAs because in the full  $m^*$  expression<sup>7</sup> corresponding to Eq. (2), the (second)  $E_0$  term dominates while the (fourth)  $\bar{\Delta}$  term is negligible.

We now turn our attention to interpreting the time-resolved photoluminescence results of Oestreich and co-workers for GaAs {Refs. 1 and 2} reproduced in Fig. 4. We should emphasize that the experiment of Refs. 1 and 2 is

different from our transient absorption experiment in that they pump at low laser intensity in order to avoid state filling and, consequently, they measure electrons with average energy approximately  $\langle E \rangle \sim kT$ . We have made a full 14-band  $\mathbf{k} \cdot \mathbf{p}$  calculation of  $g^*$  at  $E=kT$  shown in Fig. 4, following the procedure outlined in Ref. 7, i.e., not making use of arbitrary approximations. In brief, one diagonalizes two matrices, one for each conduction band spin state, which include exactly the interactions between the seven states of the same Landau quantum number and also the nearest adjacent interacting Landau levels coupled by the terms proportional to  $E_Q = 15.56$  eV (the momentum matrix element between the higher conduction and valence bands).<sup>7</sup> We define  $g^*(E) = (E_+ - E_-) / \mu_B B$  from Eq. (1), where  $E = (E_+ + E_-) / 2$ . As for InSb above, we assumed that  $E_{p0}$ ,  $E_{p1}$ , and  $E_Q$  are all constants (i.e., negligible dilational changes only). Also, as for InSb, reflectance measurements<sup>20</sup> have shown that the spin-orbit splitting  $\Delta_0$  is nearly constant (and very close to the value used above<sup>7</sup>). We therefore also assumed that the other spin orbit couplings  $\Delta_1$  and  $\bar{\Delta}$  are also constant. As already mentioned, the  $E_1$  (third) term in Eq. (2) is small, so using the optical or dilational change of that gap, or leaving it constant, makes negligible difference. The fundamental band gap is therefore the only temperature dependent parameter, and the dilational contribution to the change in  $E_0$  with  $T$  for

GaAs was taken from Ref. 18. We see from Fig. 4 that if we use the optical band gap, then  $g^*$  ( $E=kT$ ) should increase in magnitude (thick dashed line), whereas if we assume that only the dilational gap change is applied, the magnitude decreases (thick solid line) and we get a reasonable fit to the data. Averaging  $g^*$  over the electron distribution can produce much better agreement but is magnetic field dependent and therefore ambiguous.

In summary, good agreement is obtained between the experimental  $g$  factor and the predictions of  $\mathbf{k} \cdot \mathbf{p}$  theory for InSb up to 300 K provided we include only the dilational change in the band gap, confirming the prediction of the classic papers by Ehrenreich.<sup>8</sup> In addition, we have shown that the temperature dependence of  $g^*$  measured by other workers for GaAs (Refs. 1–4) is not, in fact, anomalous; on the contrary, good agreement is obtained with the 14-band model without adjustable parameters, providing, again, we use only the dilational change with temperature in the fundamental band gap. We reemphasize that the effect of  $\bar{\Delta}$  must not be neglected for  $g^*$ .

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