

Temperature dependence of the electron spin g factor in GaAs

W. Zawadzki*, P. Pfeffer
*Institute of Physics, Polish Academy of Sciences
 Al.Lotników 32/46, 02-668 Warsaw, Poland*

R. Bratschitsch**, Z. Chen, S. T. Cundiff
*JILA, National Institute of Standards and Technology and University of Colorado,
 Boulder, Colorado 80309-0440, USA*

B. N. Murdin
*Advanced Technology Institute, University of Surrey,
 Guildford GU2 7XH, United Kingdom*

C. R. Pidgeon
*Department of Physics, Heriot-Watt University,
 Edinburgh EH14 4AS, United Kingdom*

(Dated: September 15, 2008)

The temperature dependence of the electron spin g factor in GaAs is investigated experimentally and theoretically. Experimentally, the g factor was measured using time-resolved Faraday rotation due to Larmor precession of electron spins in the temperature range between 4.5 K and 190 K. The experiment shows an almost linear increase of the g value with the temperature. This result is in good agreement with other measurements based on photoluminescence quantum beats and time-resolved Kerr rotation up to room temperature. The experimental data are described theoretically taking into account a diminishing fundamental energy gap in GaAs due to lattice thermal dilatation and nonparabolicity of the conduction band calculated using a five-level $\mathbf{k}\cdot\mathbf{p}$ model. At higher temperatures electrons populate higher Landau levels and the average g factor is obtained from a summation over many levels. A very good description of the experimental data is obtained indicating that the observed increase of the spin g factor with the temperature is predominantly due to band's nonparabolicity.

PACS numbers: 72.25.Fe 71.70.Ej 72.25.Rb

I. INTRODUCTION

The temperature dependence of the spin g value of electrons in GaAs has been a subject of controversy since 1995, when it was shown that the experimental data, exhibiting an increase of the value of g as a function of temperature, are in a qualitative disagreement with the $\mathbf{k}\cdot\mathbf{p}$ theory, if one takes into account an experimental change of the fundamental energy gap [1]. The seeming qualitative disagreement between the experiment and the theory was confirmed by subsequent publications [2, 3]. The temperature dependence of the spin properties of electrons in GaAs is not only of academic interest since for possible spintronic applications the behavior of g near room temperature is clearly of great importance. It was recently shown that one can reach at least a qualitatively

correct description of the experimental g values if one includes in the $\mathbf{k}\cdot\mathbf{p}$ theory not the complete temperature change of the fundamental energy gap but the change due to lattice dilatation alone [4]. A similar result was obtained for the g value of electrons in InSb indicating that the better description was not fortuitous.

The experiments [1-3] and their analysis presented in Ref. [4] have two shortcomings. On the experimental side, the g value data was obtained by time-resolved photoluminescence alone, while it would be desirable to verify these data by some other method. On the theoretical side, the analysis in Ref. [4] is somewhat tentative as it does not account for the fact that at higher temperatures electrons populate many Landau levels, so that the measured spin g value represents an average over many transitions. Recognizing the importance of the subject and its controversial character the present work tries to overcome the above shortcomings in both experimental and theoretical aspects. First, we present new data on the temperature dependence of the electron g value in GaAs obtained with the time-resolved Faraday rotation and compare them with the photoluminescence data of

* e-mail address: zawad@ifpan.edu.pl

** Present address: University of Konstanz and Center for Applied Photonics, D-78457 Konstanz, Germany

Refs. [1-3]. Second, we complete the theoretical description by including the electron statistics which results in the necessity of summation over many Landau levels (LLs). We show that the complete theory considerably improves the description of experimental data.

The paper is organized as follows. We first describe new experiments on the temperature dependence of the electron g value in GaAs. Next, we describe the theoretical procedure used to evaluate the temperature dependence of the g value and compare the theory with all existing experimental data. We discuss our results and conclude the paper by a summary.

II. EXPERIMENT AND THEORY

The spin g factor of electrons in a nominally undoped $3 \mu\text{m}$ thick layer of GaAs was determined using an ultrafast degenerate setup for measuring the time-resolved Faraday rotation [5-7]. Spin polarized electrons were optically excited by a circularly polarized pump pulse which was spectrally tuned to the band gap of GaAs at all temperatures. After the excitation the electron spins begin to precess in a magnetic field applied parallel to the sample surface in the Voigt geometry. By measuring the Faraday rotation of a linearly polarized probe beam transmitted through the sample one is able to monitor the electron spin dynamics. The Larmor spin precession results in an oscillating signal that decays exponentially at a rate $1/T_2^*$, where T_2^* is the ensemble spin dephasing time (Fig. 1). The g factor is determined using the equation of Larmor precession: $g^* = \hbar\omega_L/\mu_B B$, where ω_L is the measured Larmor precession frequency, μ_B is the Bohr magneton, and B is the applied magnetic field. The ultrafast femtosecond laser pulses ($\Delta\lambda = 15.5 \text{ nm}$, $P_{\text{pump}} = 10 \text{ mW}$, $P_{\text{probe}} = 1 \text{ mW}$) were generated by a mode-locked Ti:sapphire laser oscillator operating at 76 MHz repetition frequency. The samples were held at low temperatures using an optical cryostat and a magnetic field was generated by a split-coil superconducting magnet. Magnetic fields up to 6 T have been applied.

GaAs is a medium-gap material, so that a three-level $\mathbf{k}\cdot\mathbf{p}$ description, successfully used for narrow gap semiconductors [8, 9], is not adequate for treating its band structure. The reason is that in GaAs the fundamental gap E_0 between the Γ_6^c and Γ_8^v levels is about 1.5 eV, i.e. it is not much smaller than the gap E_1 between the Γ_6^c level and the upper Γ_7^c conduction level (which is about 3 eV). It has been convincingly demonstrated that an adequate way to treat the conduction band of GaAs is to use a five-level model (5LM, which is equivalent to 14 bands including spin) in the $\mathbf{k}\cdot\mathbf{p}$ description (see Refs. [10, 11] and the references therein). In particular, for the description of electron spin g value in GaAs-based heterostructures the usefulness of the 5LM was demonstrated quite recently [12]. Since the 5LM for electrons in the presence of a magnetic field and its use for magneto-optical properties of GaAs was described in some details before [10,

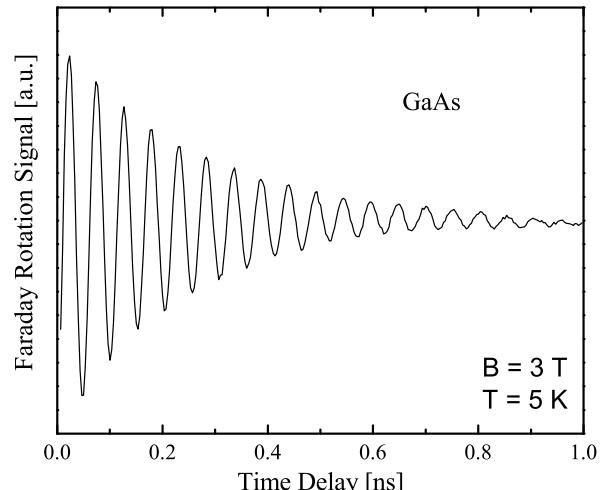


FIG. 1: Time-resolved Faraday rotation signal of nominally undoped bulk GaAs, recorded at a magnetic field of $B = 3 \text{ T}$.

[11], we only mention here the main elements of this approach. Thus the model includes exactly the Γ_7^v , Γ_8^v , Γ_6^c , Γ_7^c and Γ_8^c levels at the center of the Brillouin zone and the resulting $\mathbf{k}\cdot\mathbf{p}$ matrix has dimensions 14×14 . There exist three nonvanishing interband matrix elements of momentum: P_0 , P_1 and Q_2 , and one interband element of the spin-orbit interaction Δ . If one takes $Q = 0$ and $k_z = 0$ (where k_z is the momentum along the magnetic field direction) the 14×14 initial matrix factorizes into two 7×7 matrices for the spin-up and spin-down states. These matrices are soluble with the envelope functions in the form of harmonic oscillator functions and the eigenenergy problem for different Landau levels (LLs) n reduces to a diagonalization of 7×7 determinants. However, if the Q element is included (it comes from an inversion asymmetry of the GaAs crystal) the initial 14×14 matrix does not factorize and it is not soluble in terms of a single column of harmonic oscillator functions. Physically, this means that the resulting energy bands are not spherical. Since the nonsphericity of the conduction band in GaAs is small, one can solve for the eigenenergies looking for the envelope functions in terms of sums of harmonic oscillator functions. This leads to number determinants composed of the fundamental 7×7 blocks on the diagonal coupled by nondiagonal parts involving the Q elements. The eigenenergies are computed truncating the resulting big determinants. In our computations we used typically 21×21 determinants. All calculations were performed taking the magnetic field \mathbf{B} parallel to [001] direction and putting $k_z = 0$.

Now we turn to the temperature dependence of the fundamental gap E_0 . It was argued a long time ago [13] and confirmed by the behavior of the effective mass [14, 15] that the temperature dependence of the band parameters is governed by a dilatational change of the energy gap E_0 and not by its total (i.e. measured) change. The other contribution to the change of the gap is due to phonons.

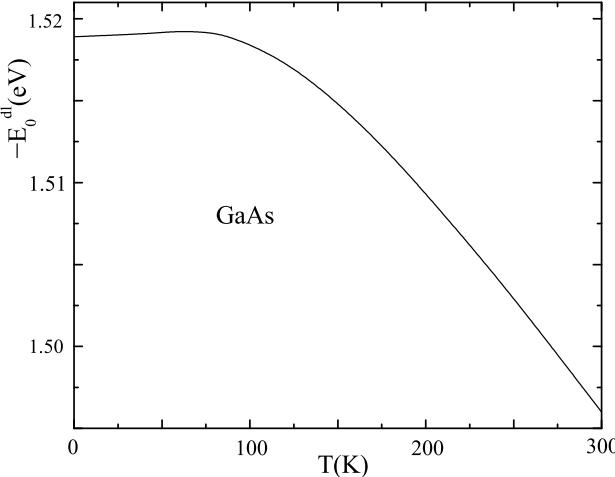


FIG. 2: Calculated change of the fundamental energy gap in GaAs due to thermal dilatation versus temperature (after [16]).

However, the phonon vibrations occur on a much slower time scale than the time the electron needs to sample the interband interactions determining the effective mass and the g factor, which are at optical frequencies. The dilatational change of the gap is given by [16]

$$\Delta E_0^{dl}(T) = -3D \left(\frac{\partial E}{\partial P} \right)_T \int_0^T \alpha_{th}(T') dT' , \quad (1)$$

where D is the bulk modulus, dE/dP is the pressure-induced band-gap shift, and $\alpha_{th}(T)$ is the linear thermal expansion coefficient (see also [17]). The quantities B and $\partial E/\partial P$ are readily measurable, the quantity $\alpha_{th}(T)$ was measured for GaAs by Novikova [18] and Soma et al [19]. Using the values of $\alpha_{th}(T)$ one calculates $\Delta E_0^{dl}(T)$. This was done by Lourenco et al [16], we replot their results in Fig. 2. It should be mentioned that $\alpha_{th}(T)$ goes through a minimum near $T \approx 50$ K, this results in a plateau and a flat maximum of $E_0^{dl}(T)$ seen in Fig. 2. The dilatational change ΔE_0^{dl} of the fundamental gap between 0 K and 300 K is about 23 meV (see Fig. 2), while the total change ΔE_0^{tot} is about 93 meV (cf. Refs. [21, 22]). These numbers are important when one tries to understand why putting the total change $\Delta E_0^{tot}(T)$ or the dilatational change $\Delta E_0^{dl}(T)$ into the calculations of g value, one obtains very different results in the two cases.

Finally, we consider average values of the spin g factor measured as a function of temperature. The measurements are usually done in relatively pure samples having low free electron densities. The electrons are excited across the gap into the conduction band and into both spin states. The spin states are equally populated, but the circularly polarized light produces a well defined coherence between them. The excited electrons quickly thermalize and are distributed among LLs ac-

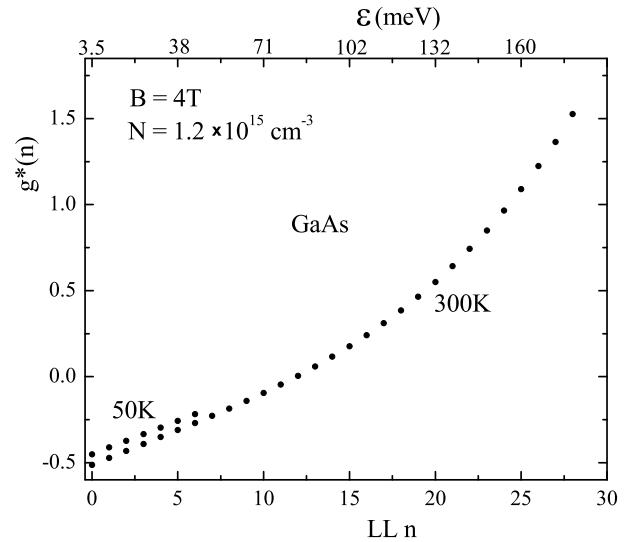


FIG. 3: Spin g values for consecutive Landau levels n , calculated for $T = 50$ K and $T = 300$ K, a magnetic field $B = 4$ T and the indicated electron density N . The indicated points correspond to LLs that give non-negligible contributions to the average g value. Approximate electron energies corresponding to LLs marked on the lower abscissa are shown on the upper abscissa.

cording to the lattice temperature without losing their spin or phase. Then they interfere and quantum beats in the photoluminescence or other effects are observed from many LLs. According to this picture the observed signal represents an average over the populated LLs in which the electron thermal distribution over LLs determines their contribution to the average g value. This explains why various experiments performed on weakly doped samples give basically the same results depending only on the temperature.

We assume that the electron energies are

$$E_{nk_z}^\pm = \mathcal{E}_n^\pm + \frac{\hbar^2 k_z^2}{2m_0^*} , \quad (2)$$

where n is the LL number, \pm signs correspond to the two spin states, k_z is the wavevector along the direction of \mathbf{B} , and m_0^* is the effective mass at the band edge. The energies \mathcal{E}_n^\pm contain the intricacies of the band structure mentioned above, but we first assume a simple parabolic dependence on k_z to make the averaging tractable. The spin g value is defined as (in formulas we use g^* symbol)

$$g^* = (E_{nk_z}^+ - E_{nk_z}^-)/\mu_B B . \quad (3)$$

An averaging procedure involves summation over n and integrations over k_x and k_z . A simple calculation gives the average value of g^* in the form

$$\overline{g^*}(T) = \frac{A}{C} , \quad (4)$$

where

$$A = \sum_{n=0}^{\infty} \int_{\mathcal{E}_n^i}^{\infty} \frac{g_n^*(\mathcal{E}) f(\mathcal{E}, \zeta)}{(\mathcal{E} - \mathcal{E}_n^i)^{1/2}} d\mathcal{E} , \quad (5)$$

and

$$C = \sum_{n=0}^{\infty} \int_{\mathcal{E}_n^i}^{\infty} \frac{f(\mathcal{E}, \zeta)}{(\mathcal{E} - \mathcal{E}_n^i)^{1/2}} d\mathcal{E} , \quad (6)$$

in which the summation is over the LLs, $f(\mathcal{E}, \zeta)$ is the Fermi-Dirac distribution function, and the square roots come from the integrations over k_z . The integrations begin from the lower states \mathcal{E}_n^i for each n , which can be either \mathcal{E}_n^+ or \mathcal{E}_n^- .

The average g value, as given by Eq. (4), is affected by the temperature in two opposite ways. According to the five-level model the spin g value at the conduction band edge is [10],

$$\begin{aligned} g_0^* = & 2 + \frac{2}{3} \left[E_{P_0} \left(\frac{1}{E_0} - \frac{1}{G_0} \right) + E_{P_1} \left(\frac{1}{G_1} - \frac{1}{E_1} \right) \right] + \\ & - \frac{4\bar{\Delta}\sqrt{E_{P_0}E_{P_1}}}{9} \left(\frac{2}{E_1 G_0} + \frac{1}{E_0 G_1} \right) + 2C' , \end{aligned} \quad (7)$$

where $E_{P_0} = 2m_0 P_0^2 / \hbar^2$, $E_{P_1} = 2m_0 P_1^2 / \hbar^2$, $G_0 = E_0 + \Delta_0$ and $G_1 = E_1 + \Delta_1$. The spin-orbit energies Δ_0 and Δ_1 relate to (Γ_7^v, Γ_8^v) and (Γ_7^c, Γ_8^c) levels, respectively, $\bar{\Delta}$ is the interband matrix element of the spin orbit interaction between the (Γ_7^v, Γ_8^v) and (Γ_7^c, Γ_8^c) multiplets (see [10, 22]), and C' is due to far-band contributions. For $\bar{\Delta} = 0$ Eq. (7) reduces to the formula given first by Hermann and Weisbuch [23].

It follows from Eq. (7) that, as the temperature T increases and the absolute value of the fundamental gap E_0 decreases (see Fig. 2), the spin g_0^* value at the band edge decreases. On the other hand, with increasing T the electrons populate higher LLs and band's nonparabolicity comes more and more into play. The latter is known to make the g value less negative (see Refs. [10, 24], and Fig. 3). Thus, as T increases, the average g^* decreases or increases depending on the relative strength of the two effects. It is now clear why putting into calculations only the dilatational part of the gap variation makes the first effect weaker, i.e. it favors increase of the g values. We emphasize that we do not use in our calculations Eq. (7), it is quoted only to make clear the dependence of g_0^* on E_0 . Calculating the electron energies away from the band edge we deal with the effects of band's nonparabolicity and inversion asymmetry, as explained above. We use the following band parameters of GaAs at $T = 0$: $E_{P_0} = 27.865\text{eV}$, $E_{P_1} = 2.361\text{eV}$, $E_0 = -1.519\text{ eV}$, $G_0 = -1.86\text{ eV}$, $E_1 = 2.969\text{ eV}$, $G_1 = 3.14\text{ eV}$, $C = -2.3107$, $C' = -0.0375$. The zero of energy is chosen at the conduction band edge. The above values are the same as those established by an overall fit of various experiments on GaAs

(see Ref. [10]) with the exception of the far-band contribution C' , as discussed below. It is assumed that only E_0 depends on the temperature, see discussion below.

In Fig. 3 we plot the g values for consecutive LLs at two temperatures, as determined from definition (3), in which the energies are calculated for $B = 4\text{ T}$ using the procedure described above. It is seen that, for a given temperature, as the energy increases with the growing Landau number n , the g factor increases due to band's nonparabolicity. On the other hand, for a given LL the g factor decreases with increasing temperature. The number of points for a given temperature indicates how many LLs are involved in the averaging, for still higher LLs the occupation by electrons is so small that their contribution is negligible. It should be noted that the g factors shown in Fig. 3 do not saturate at high energies at the free electron value of $+2$, as predicted by simple versions of the three-level or five-level $\mathbf{k}\cdot\mathbf{p}$ models (see e.g. [24]). The reason is that our present theory includes the bulk inversion asymmetry (also called the Dresselhaus effect), which is manifested by the appearance of the matrix element Q . Also, the increase of g factor with the energy, as shown in Fig. 3, is stronger than linear, whereas the one shown in Fig. 8 of Ref. [10] is weaker than linear. This is caused by the fact that in Fig. 3 the energy increases by going to high LLs (at constant B), whereas in Ref. [10] the energy is increased by going to high magnetic fields (at $n = 0$).

III. RESULTS AND DISCUSSION

Fig. 4 shows the temperature dependent g values of electrons extracted from time-resolved Faraday rotation measurements performed at $B = 4\text{ T}$ on undoped bulk GaAs (full squares). In addition, data taken from the literature and our calculations are shown. As to the experiments, the previously published data obtained with the use of time-resolved photoluminescence [1-3] and time-resolved Kerr rotation [25] are compared with our present results obtained by the time-resolved Faraday rotation. All these time-resolved experiments measure the phase coherence of spin states excited by circularly polarized light in the Voigt geometry and differ mostly in its detection. It is seen that the obtained data agree well with each other and all show an almost linear increase of g with the temperature. However, there is some discrepancy between various measured g values at $T = 0$, cf. [23, 26, 27]. Since all the g values presented in Fig. 4 were measured at magnetic fields B of a few Tesla, they should be *higher* than the band edge value g_0^* since they are affected by band's nonparabolicity. In this situation we take the value of the far-band contributions C' , as given above, to give our measured value of $g^* = -0.452$ at $T = 0$ and $B = 4\text{ T}$. This corresponds to the band edge value $g_0^* = -0.472$. Preference to a different experimental band edge g value would simply require a slightly different C' .

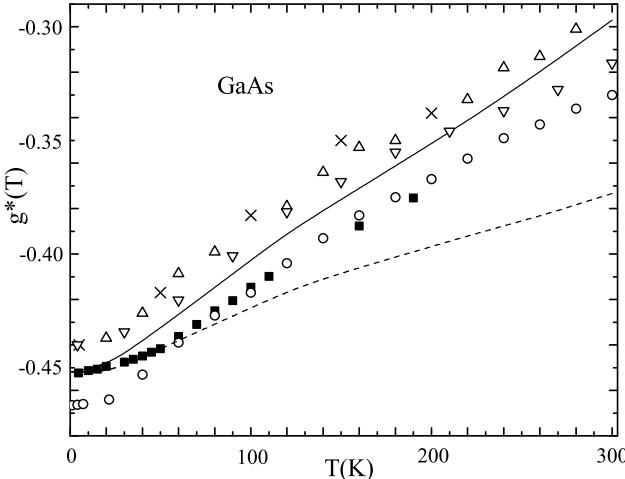


FIG. 4: Measured and calculated spin g values of electrons in GaAs versus temperature. Experimental data: full squares - our results, crosses - Ref. [1], triangles - Ref [2], circles - Ref. [3], reversed triangles - Ref. [25]. Theory: dashed line - calculation neglecting k_z -dependence of g_n^* ; solid line - calculation assuming interpolated k_z -dependence of g_n^* , see text.

As far as the theory is concerned, we show two calculations. The dashed line indicates an average value of g computed with the help of Eqs. (4), (5) and (6), in which the E_n^\pm energies are calculated for $B = 4$ T using the procedure outlined above. It is seen that, as the temperature increases and more LLs become populated, the band nonparabolicity effect is stronger than the dilatational decrease of the E_0 gap, and the average $g^*(T)$ increases. Still, the theoretical increase does not quite follow the experimental values for $T > 60$ K. The reason is that our simplified formula (2) assumes the spin splitting to be independent of k_z . This means that, as we average over the energy, the g value "jumps" from one LL to the next, as shown in Fig. 3. In reality, however, the g value depends also on k_z . As follows from the simplified three-level $\mathbf{k} \cdot \mathbf{p}$ model, the spin g value depends in fact on k_z similarly to its dependence on the orbital energy $\hbar\omega_c(n + 1/2)$. In other words, the decisive quantity is the total electron energy (see [8, 24, 28]). For the second calculation we *assume* that this is the case also in the complete five-level $\mathbf{k} \cdot \mathbf{p}$ model and we interpolate the g value to vary linearly with the energy (related to k_z) between consecutive Landau levels. If this is done, the averaging procedure gives the g values indicated in Fig. 4 by the solid line. It is seen that now the theory is in an excellent agreement with the experimental data. The moral of the story is that the almost linear increase of the average spin g value with the temperature in GaAs is caused by the dominating effect of band's nonparabolicity over the dilatational decrease of the fundamental gap. It should be emphasized that both our experimental data as well as our two theoretical curves approach the absolute zero of temperature at almost vanishing slope. It

is important that the improved experiment and theory exhibit this property.

Coming to the discussion of our results, one should keep in mind that our theoretical description, although representing a considerable improvement over the procedure of Ref. [4], is still approximate. The reason is that we made an assumption about the k_z dependence of the g factor. This assumption is reasonable and it allows us to calculate the \mathcal{E}_n^\pm energies and carry out the averaging procedure over LLs in a relatively simple way. Still, in principle one should include the k_z terms from the beginning in the computation procedure, calculate the k_z dependence of different LLs and carry out a numerical summation over k_z values. This, however, makes the problem difficult for two reasons. First, if the k_z terms are included, one must take much larger truncated determinants to obtain convergent values of $\mathcal{E}_n^\pm(k_z)$ energies. Second, when performing the averaging procedure one must integrate numerically over k_z for each LL separately since in principle each LL has a different k_z dependence. Our simplifying assumption allowed us to get around the above difficulties.

As to the work of other authors, Oestreich and Ruhle [1] put into the theoretical description the total temperature change of the energy gap but neglected band's nonparabolicity. Such a description predicted a decrease of the g factor with temperature which contradicted the experimental results. Oestreich et al [2] and Huebner et al [3] included in their description band's nonparabolicity but neglected important features of the band structure: the interband matrix element of momentum Q and of the spin orbit interaction $\overline{\Delta}$ between the (Γ_7^v, Γ_8^v) and (Γ_7^c, Γ_8^c) multiplets. Both these quantities have an important influence on the g value, which we illustrate numerically. Using our band parameters and $\overline{\Delta} \neq 0$ the band edge g value is $g_0^* = -0.472$, while for the same parameters and $\overline{\Delta} = 0$ there is $g_0^* = -0.346$ (see Eq. (7)). The element Q does not enter the conduction band edge quantities, but at $T = 300$ K and $B = 4$ T our calculated average value of g including Q is $\overline{g^*(T)} = -0.297$, while for $Q = 0$ the calculated average is $\overline{g^*} = -0.396$. Also, Refs. [2], [3] did not include the Landau quantization of the conduction band, so that the calculated average g value did not depend on the magnetic field intensity. Finally, and this is the decisive point, it was assumed that the g value depends on the temperature via the total (i.e. measured) change of the fundamental gap, whereas one should take the change due to thermal lattice dilatation alone. When this is done, there is no need to introduce a temperature variation of the interband matrix element of momentum P_0 , as was done in Ref. [3]. Consideration of the momentum operator and of the wave functions indicates that the interband momentum matrix element should also scale with the lattice dilatation, its square should change no more than 0.1 % in the temperature range from 0 to 300 K, so it can be neglected [4].

The latest analysis by Litvinenko et al [4], including the matrix elements $\overline{\Delta}$ and Q and the band nonparabolic-

ity on one side and taking only the dilatation contribution to the change of the energy gap on the other, was able to account for the first time for the experimental increase of the spin g factor with the temperature. However, this analysis assumed no k_z dependence of the Landau levels and, consistently, took the average electron energy for the Boltzmann statistics to be equal to kT . This corresponds roughly to our dashed curve in Fig. 4, which assumes no dependence of g on k_z . If one includes the k_z dependence of the energy, the average nondegenerate electron energy is $\bar{\mathcal{E}} = (3/2)kT$ which, upon using Fig. 3 for $g_n(\mathcal{E})$, corresponds quite well to the solid line in Fig. 4. In other words, if one applies a proper averaging procedure to the g value, the resulting $\bar{g}^*(T)$ corresponds quite well to averaging the electron energy $\bar{\mathcal{E}}$ and taking $g^*(\bar{\mathcal{E}})$. One should add that, taking the thermal change of gap due to the dilatation alone, Litvinenko et al [4] were able to account correctly for the temperature dependence of the electron spin splitting in InSb. All this, together with the published analysis of $m^*(T)$ in GaAs [14, 15], strongly confirms that both orbital and spin quantizations of the electron spectrum in a magnetic field are governed by the change of the energy gap due to lattice dilatation alone.

IV. SUMMARY

We investigated experimentally and theoretically the controversial problem of temperature dependence of the

spin g factor in GaAs. The time-resolved Faraday rotation technique was used to show that, in agreement with other data, the g factor increases almost linearly with T up to room temperature. This increase is successfully described by the five-level $\mathbf{k}\cdot\mathbf{p}$ model of the band structure for GaAs. As the temperature increases there occur two effects having an opposite influence on the g value. On one hand, a decrease of the fundamental energy gap caused by the thermal lattice dilatation leads to a decrease of g . On the other hand, at higher temperatures electrons populate higher Landau levels which, due to band's nonparabolicity, leads to an increase of the average $\bar{g}(T)$ value. A very good agreement of our theory with the experimental increase of $\bar{g}(T)$ indicates that the band nonparabolicity effect dominates. Our interpretation confirms the validity of the complete five-level $\mathbf{k}\cdot\mathbf{p}$ model for the conduction band of GaAs.

Acknowledgments

W. Z. and P. P. acknowledge a financial support of Polish Ministry of Science and Higher Education through Laboratory of Physical Foundations of Information Processing.

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