Temperature Dependence of the Electron Landé g Factor in GaAs

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The temperature dependent frequency of quantum beats of free electron Larmor precession in bulk GaAs yields the temperature variation from 5 to 200 K of the Landé g factor with high accuracy. The Landé g factor increases from -0.44 to -0.38 to -0.35 as the temperature increases from 5 to 100 to 150 K. The experimental results are in the opposite direction than prediction by $\vec{k} \cdot \vec{p}$ theory manifesting the need for appreciable, temperature dependent corrections of this band model.

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A recently introduced experimental technique enables us to measure with high accuracy the electron spin splitting by spin quantum beats [1]. We use this method to determine for the first time the temperature dependence of the electron Landé g factor of GaAs. A comparison of the experimental results with predictions of $\vec{k} \cdot \vec{p}$ perturbation theory yields obvious discrepancies.

The semiempirical $\vec{k} \cdot \vec{p}$ theory was developed to predict electronic properties such as the effective mass m^* [2] and the Landé g factor g^* [3] in the vicinity of the extremal points of semiconductor band structures using the knowledge of other experimentally determined semiconductor parameters as, e.g., band gap and spin-orbit splitting. The theory is of basic importance in solid state physics and is successfully used to estimate easily the pressure and alloy dependence of fundamental band parameters. Initially a three-band model was developed which takes into account the spin-orbit interaction of the valence band and its influence on the properties of the lowest conduction-band electrons. However, it turned out that this model was too coarse of an approximation. Corrections due to spin-orbit interaction of the conduction-band electrons had to be included, and a five-band model was developed [4,5]. This five-band model proved very useful in the prediction of low temperature properties of various semiconductors and, in particular, their alloys [6]. The question remains whether this theory is also able to describe the temperature dependence of an entity, as, e.g., the g factor. The value of g^* is different from the free electron Landé factor due to the presence of spin-orbit interaction, and this entity describes the fundamental property as the magnitude of the Zeeman splitting of electronic states in magnetic fields. The calculation of g^* in the semiconductor GaAs can be carried out with high accuracy, since (i) the interband matrix elements of the lowest conduction band are expected to be almost temperature independent, and (ii) the temperature variation of the energy gaps are known with high accuracies [7, 8].

The experimental determination of the temperature dependence of g^* was, however, up to now not possible. First, methods such as conventional electron spin resonance are difficult, even at low temperatures, in semiconductors with low doping levels [9], and high doping introduces difficulties in the interpretation of the data. Second, optical Zeeman spectroscopy on free and bound excitons [10] is not possible at elevated temperatures. Third, electron spin resonance using optical excitation has a poor signal to noise ratio, making a measurement of the temperature dependence at least rather difficult if not impossible [6].

Here we use a very precise measurement of g^* with time-resolved photoluminescence. The experiment relies on the detection of the quantum beats of electron Larmor precession. The oscillation frequency of the spin quantum beats directly reveals the electron spin splitting and therefore the electron Landé g factor.

The undoped bulk GaAs sample is mounted in the Voigt configuration in a He gas flow cryostat in a superconducting magnet. The magnetic field is in the x direction, observation and growth directions are in the z direction. We excite with ps pulses from a mode locked Ti:sapphire laser with a repetition rate of 80 MHz. The excitation density is about 10^{16} cm^{-3}. As a consequence, the electron gas is properly described by a Maxwell-Boltzmann distribution function. The photoluminescence (PL) is dispersed in a 0.32 m spectrometer and detected with a spectral and temporal resolution of 0.5 nm and 10 ps, respectively, by a streak camera with two-dimensional readout.

Figure 1 shows the temporal evolution of the circularly analyzed PL for four different temperatures at a magnetic field of 5 T. The PL is detected at the direct band gap with opposite handedness of circular polarization compared to the exciting laser pulses. Excitation with σ^+ polarized light creates more electrons with spin orientation $s_z = +1/2$ than with $s_z = -1/2$ [11]. The holes lose their angular momentum in a magnetic field rapidly because of the strong spin-orbit coupling in GaAs, whereas the electrons have a significant longer spin relaxation time [12]. The $s_z = +1/2$ electrons emit σ^- polarized light when they recombine with heavy holes. The magnetic field in the x direction leads to a Larmor precession of the electron spins around the axis of the magnetic field. This Larmor precession

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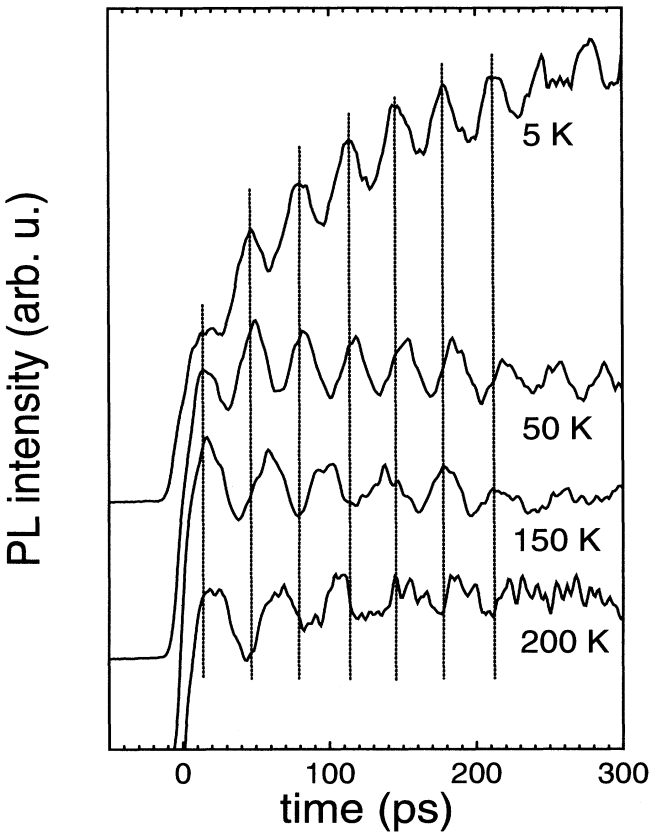

FIG. 1. Circularly polarized detected photoluminescence at four different temperatures after circularly polarized excitation in the continuum at a magnetic field of 5 T. The different oscillation periods of the photoluminescence directly demonstrate the temperature dependence of the electron g factor. The dotted lines are guides for the eye to illustrate the phase shifting.

causes oscillations in the average spin polarization in the z direction $\langle s_z \rangle$, which modulates the circular polarization of the luminescence. The time evolution of the spin part of the electronic wave function is

$$s_z^+(t) = s_z^+(0)\cos(\omega_L t/2) + s_z^-(0)\sin(\omega_L t/2), \quad (1)$$

where

$$\omega_L = g_e \mu_B B/\hbar \quad (2)$$

is the Larmor angular frequency [1].

The spatial part of the wave function suffers a very fast phase relaxation; i.e., the phase is equally randomized for both spins, and, therefore, the spin part of the wave function can be written separately. Spin relaxation damps the oscillations. We determine spin relaxation times of about 500 ps at 5 K and 150 ps at 150 K. As expected, the spin relaxation time decreases with temperature [13,14]. A quantitative analysis of spin relaxation is, however, difficult since this time is also slightly sample dependent. Figure 1 clearly reveals that the oscillation frequency decreases with increasing temperature; i.e., the absolute value of g^* must decrease with temperature. Spin splitting is linear up to at least 7 T in this temperature range; i.e., no magnetic field dependence of g^* has to be incorporated at 5 T.

Figure 2 depicts the temperature variation of the experimental g^* together with the theoretical prediction by a five-band model according to Ref. [5],

$$g^* = 2 - \frac{2P^2}{3}\left(\frac{1}{E_0} - \frac{1}{E_0 + \Delta_0}\right) - \frac{2P'^2}{3} \times \left(\frac{-1}{E(\Gamma_8^c) - E_0} + \frac{1}{E(\Gamma_7^c) - E_0}\right) + 2C', \quad (3)$$

where $P^2 = 28.9$ eV describes the coupling between the conduction band Γ_6^c and the valence bands Γ_8^v and Γ_7^v, $P'^2 = 6$ eV the coupling between the conduction band Γ_6^c and the higher conduction bands Γ_7^c and Γ_8^c, E_0 the $\Gamma_8^v - \Gamma_6^c$ gap, Δ_0 the valence band spin-orbit splitting, and $C' = -0.02$; the origin of the energies is taken at Γ_8^v [5]. The discrepancy between theory and experiment is obvious.

We now discuss uncertainties in theory and experiment. The temperature variation $E_0(T)$ is known with high precision [8]. This temperature dependence enters most strongly in Eq. (3). The spin-orbit splitting is almost temperature independent, and its slight temperature dependence $\Delta_0(T)$ is well known [7]. The exact values of $E(\Gamma_8^c)$ and $E(\Gamma_7^c)$ are debated [7,15]; however, these

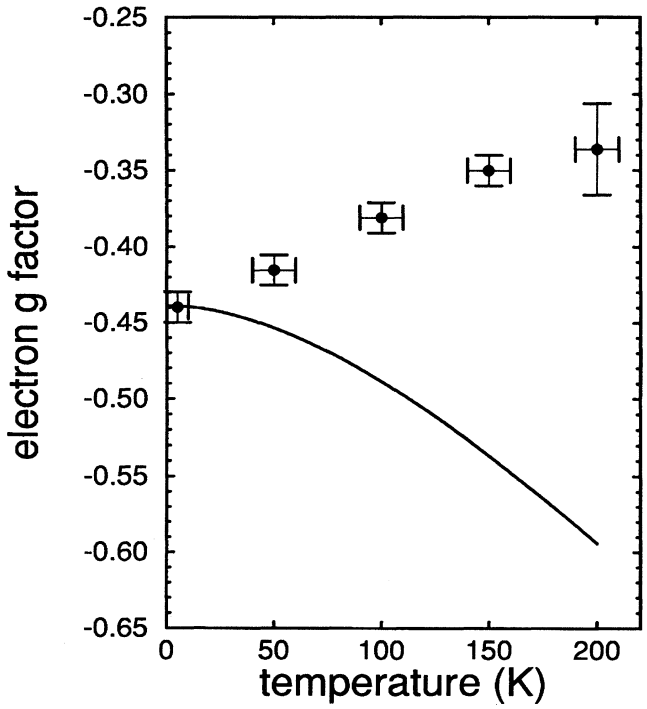

FIG. 2. Temperature dependence of the measured g^* (dots with error bars) and the calculated g^* after Ref. [5] if the temperature dependence of all bands is included (solid curve).

values enter only almost negligibly in the value of g^* obtained by Eq. (3). The temperature dependence of $E(\Gamma_8^c)$ is known [7], and the spin-orbit splitting $E(\Gamma_8^c) - E(\Gamma_7^c)$ once more should be almost independent of temperature. We took a variation of $E(\Gamma_8^c)(T) = 4.659\ \text{eV} - (4.0 \times 10^{-4}\ \text{eV K}^{-1}\ \text{T}^2)/(T + 241\ \text{K})$ according to Ref. [7] and a temperature independent $E(\Gamma_8^c) - E(\Gamma_7^c)$ of 0.171 eV [15]. The variation of P^2 and P'^2 is smaller than 0.2% for a variation of temperature from 5 to 200 K. The constant C' is also very small and should not change with temperature. To summarize, the main temperature variation of g^* according to Eq. (3) results from the variation of the fundamental energy gap. The other contributions can alter this variation only by less than 2%.

On the experimental side, first, GaAs is a particularly favorable system for our investigations: The small g^*, being detrimental for other experimental methods, enables us to measure small variations of g^* with extremely high accuracy. Second, quantum beat spectroscopy is free from influences of inhomogeneous broadening [16] and temperature dependent occupation of spin levels. Third, in our experimental arrangement no nuclear spin orientation [6,17] is produced by relaxation of electron to nuclear spin orientation. Fourth, surface effects can be neglected due to the large excitation penetration depth of 1 μm. The possibility to neglect surface effects is experimentally confirmed; when we use a 1 μm thick GaAs heterostructure instead of bulk GaAs we get the same results. We are able to detect the quantum beats with identical oscillation frequencies in all GaAs samples investigated. The different samples only show different electron spin relaxation times; i.e., the damping of the oscillations depends on specific sample preparation conditions. We determined at each temperature the frequency range for which a reasonable fit of the experimentally observed oscillations is possible. This frequency range determines the error bars in Fig. 2. Fifth, we are able to determine experimentally that g^* is independent of detection wavelength and of density for a nondegenerate electron gas; for example, at 150 K, g^* does not change for detection wavelengths between 835 and 825 nm, and for carrier densities between 10^{14} and 10^{16} cm^{-3}. Moreover, the low temperature g^* in our experiment is in excellent agreement with other experiments [6,10,18]. We are therefore convinced that our method is particularly well suited to yield reliable experimental results.

We performed similar experiments also in CdTe and InP. The results will be presented elsewhere, since a direct comparison with $\vec{k} \cdot \vec{p}$ theory is in these cases not straightforward. For CdTe, the temperature variation of several band parameters is not as well known as in GaAs, and for InP the calculated temperature variation of g^* is smaller than the experimental error.

We also performed spin quantum beat experiments at low temperatures in quantum wires [19]. Whereas the temperature dependence of the g factor disagrees with $\vec{k} \cdot \vec{p}$ theory, the anisotropy in a quantum confined structure is in reasonably good agreement. Figure 3 depicts the angular dependence of g^* in the quantum wire plane. The angles of 0°, 180°, and 360° correspond to a magnetic field —and therefore a g^*—along the wire direction; the angles of 90° and 270° correspond to B perpendicular to the wires. The solid line shows a theoretical estimate by $\vec{k} \cdot \vec{p}$ theory without adjustable parameters. The g factor in the quantum wire plane for an arbitrary angle ϕ becomes [20]

$$g^* = \{[g_\parallel^* B \cos(\phi)]^2 + [g_\perp^* B \sin(\phi)]^2\}^{1/2}, \qquad (4)$$

where $g_\perp^*$ and $g_\parallel^*$ denote the g factors according to Ref. [21] in the directions with and without confinement, respectively. Measurements of the g factor in quantum wells with different thicknesses at low temperatures show similar good agreements between $\vec{k} \cdot \vec{p}$ theory and experiment [22].

The apparent discrepancy between experiment and $\vec{k} \cdot \vec{p}$ theory at high temperatures is not easy to explain but temperature dependent effective mass measurements indicated already similar discrepancies [23,24]. However, the effective mass measurements were rather indirect including various corrections so that systematic errors obstructed a conclusive comparison. One possibility for the discrepancy is that in reality only part of the temperature variation of E_0 acts on the variation of g^*, namely, exclusively the part connected with the change in lattice constant alone and not with the part related to the electron-phonon interaction [25]. This would reduce the theoretical variation of g^* but not produce an opposite sign of this variation [26].

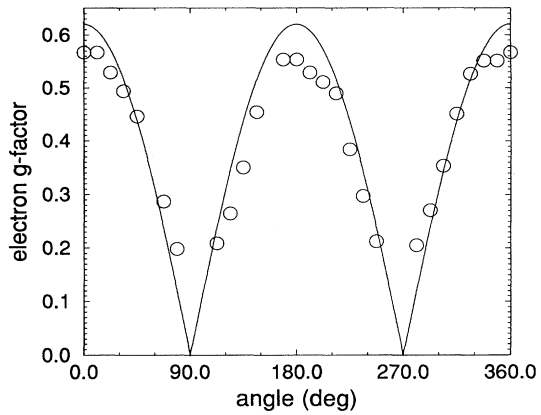

FIG. 3. Absolute value of the electron g factor in the quantum wire plane measured at 6 T. The open circles are experimental data. Perpendicular to the wires the PL oscillation period is longer than the electron spin dephasing time confirming that g^* is approximately 0. The solid line shows a theoretical estimate without any adjustable parameters according to Ref. [21].

In conclusion, we have presented novel experimental data on the temperature variation of the free electron Landé g factor in GaAs and compared it with a prediction by $\vec{k} \cdot \vec{p}$ theory. A striking discrepancy has become obvious which evidences the limits of the applicability of $\vec{k} \cdot \vec{p}$ theory even in its five-band approximation.

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