Temperature and density dependence of the electron Landé g factor in semiconductors

M. Oestreich
Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, 70569 Stuttgart, Federal Republic of Germany and Materials Department, University of California at Santa Barbara, Santa Barbara, California 93106

S. Hallstein
Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, 70569 Stuttgart, Federal Republic of Germany

A. P. Heberle
Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, 70569 Stuttgart, Federal Republic of Germany and Hitachi Cambridge Laboratory, Madingley Road, Cambridge CB3 OHE, United Kingdom

K. Eberl, E. Bauser, and W. W. Rühle
Max-Planck-Institut für Festkörperforschung, Heisenbergstrasse 1, 70569 Stuttgart, Federal Republic of Germany
(Received 25 September 1995)

The temperature and density dependence of spin quantum beats of electrons is measured by time-resolved photoluminescence spectroscopy and yields the electron Landé g factor in bulk GaAs, InP, and CdTe. In GaAs the g factor increases linearly from -0.44 at 4 K to -0.30 at 280 K; in InP the g factor is 1.20 at 4 K, exhibiting a very small temperature dependence up to 160 K, and in CdTe the g factor follows between $T=4$ K and 240 K the empirical equation $g=-1.653+4\times10^{-4}\ T+2.8\times10^{-6}\ T^2$. In GaAs we demonstrate the suppression of spin quantum beats due to Fermi blocking in a degenerate electron gas and measure an increase of the GaAs g factor from -0.44 at densities below $1\times10^{16}\,\mathrm{cm}^{-3}$ to -0.33 at $10^{17}\,\mathrm{cm}^{-3}$.

I. INTRODUCTION

The electron Landé g factor is one of the basic parameters in semiconductors that describes the magnitude of the Zeeman splitting of electronic states in magnetic fields. The g factor in semiconductors differs from the free electron g factor in vacuum, $g=2.0023$, due to the spin-orbit interaction. Depending on the semiconductor material, this interaction can change the effective g factor to very large positive as well as to very large negative numbers. Since various theoretical models predict the value of g, accurate experimental measurements of g provide a sensitive test of different band-structure calculations.[1–10]

A recently introduced experimental technique enables the measurement of the electron g factor g^* with high accuracy by spin quantum beats.[11] The technique proved to be feasible to measure various effects as the anisotropy of g^* in quantum wires,[12] the dependence of g^* on quantum well thickness,[13] and the temperature dependence of g^* in bulk GaAs.[14] The measurement of the temperature dependence with other techniques is not possible since methods such as, e.g., conventional electron-spin resonance and electron-spin resonance using optical excitation are difficult, in particular, at elevated temperatures. The temperature-dependent spin quantum beat experiments show interesting discrepancies between experiment and a well-accepted five-band $\boldsymbol{k}\cdot\boldsymbol{p}$ theory model.[14]

The $\boldsymbol{k}\cdot\boldsymbol{p}$ perturbation theory is a semiempirical theory that permits us to calculate the shape of the energy bands and the g factor in the vicinity of extremum points of energy bands from data of experimental energy gaps and matrix elements between various bands. In the late 1950s, a simple three-band model that included only the coupling between the states of the lowest conduction band and the two highest valence bands yielded good agreement with the experimental data of that time.[12] However, the measurement of the g factor in large band-gap semiconductors forced later the development of a more elaborate theory—the five-band $\boldsymbol{k}\cdot\boldsymbol{p}$ theory.[3,4] Three-band $\boldsymbol{k}\cdot\boldsymbol{p}$ theory is only suitable for narrow-gap semiconductors in which the $\boldsymbol{k}\cdot\boldsymbol{p}$ perturbation between the lowest conduction (Γ_6) level and the two nearest valence (Γ_8 and Γ_7) levels dominate; in large band-gap semiconductors like GaAs this is a rough approximation since the fundamental gap is not small compared to higher conduction bands. Five-band $\boldsymbol{k}\cdot\boldsymbol{p}$ theory fits with high precision the g factor and the effective mass of most common III-V compounds and alloys at low temperatures. However, the recently discovered discrepancy of the measured g factor at high temperatures and predictions by five-band $\boldsymbol{k}\cdot\boldsymbol{p}$ theory are not yet fully understood. Therefore, we present in this paper a detailed experimental study of the temperature-dependent free-electron g factor in GaAs, InP, and CdTe.

In addition, we describe the carrier density dependence of g^* in GaAs and demonstrate the suppression of spin quantum beats at high electron densities by Fermi blocking. We show that with increasing carrier density, the average electron energy increases due to band filling and the measured g factor increases as a function of energy tending towards the free-electron value $g=2$.[15,16]

II. EXPERIMENTAL ASPECTS

Electron spin quantum beats yield a method that allows the direct determination of g^* with high accuracy and at low

densities:[11] Energy levels separated by an energy difference ΔE can produce oscillations of the emitted light, when both energy states are excited coherently. A magnetic field $\boldsymbol{B}=Be_x$ applied on a semiconductor produces a splitting of the spin states $\chi_x^{\pm}$ by $\Delta E=\hbar\omega_L$ with ω_L being the Larmor angular frequency,

$$\omega_L=g_e^*\mu_B B/\hbar. \tag{1}$$

Excitation with circularly polarized light in the direction perpendicular to the direction of the magnetic field creates a carrier distribution with unequal occupation of the spin states χ_z^+ and χ_z^-, where z is the excitation and is opposite to the observation direction. The spin states $\chi_z^{\pm}$ can be described as a coherent superposition of $\chi_x^{\pm}$, provided both states are simultaneously excited by a short laser pulse. The spatial part of the wave function suffers a very fast phase relaxation. Therefore, the spin part can be written separately. The time evolution of the spin-state occupation in the z direction is then given by[11]

$$\begin{aligned}\chi_z(t)&=(\chi_x^- e^{+i(\omega_L/2)t}+\chi_x^+ e^{-i(\omega_L/2)t})/\sqrt{2}\\&=\chi_z^+\cos(\omega_L t/2)+i\chi_z^-\sin(\omega_L t/2).\end{aligned} \tag{2}$$

The spin orientation hence oscillates between $s_z=+1/2$ and $s_z=-1/2$. These oscillations can be understood in a semiclassical picture as the Larmor precession of the spins around the axis of the magnetic field (x axis). The photoluminescence emitted in the z direction after excitation by a short, circularly polarized laser pulse therefore shows a time-varying circular polarization.

In our experiment the luminescence is partially due to recombination of excitons. The magnetic field is, at least at higher fields (>1 T), strong enough to break up the exchange interaction of the order of 20 μeV in GaAs,[17] which couples the electron and hole spins. Therefore, two oscillation frequencies should be observable: one from oscillations of the electron spin and another one from oscillations of the hole spins. However, hole spin relaxation is in bulk extremely fast due to the degeneracy of light and heavy valence bands at $k=0$. Consequently, the oscillations of the hole spins are not observable in our experiment. We only see the oscillations of the electron spins. This assignment is confirmed by measurements in p-doped samples, which show the same oscillation frequencies for the band-to-acceptor luminescence as for the excitonic luminescence.[13] Additionally, the same oscillation frequency is observed at medium excitation densities, i.e., where excitons are completely screened and band-to-band recombination is observed. This method, therefore, although measuring the quantum beats from excitonic recombination, allows the direct determination of the free-electron g factor.

The undoped bulk GaAs, CdTe, and InP samples are mounted in 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 circularly polarized ps pulses from a mode-locked Ti:sapphire laser with a repetition rate of 80 MHz. The circular components of the photoluminescence (PL) are 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 (2D) readout.

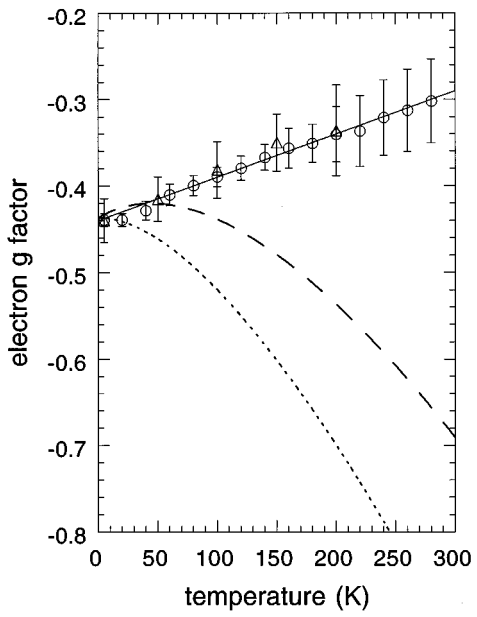

FIG. 1. Temperature dependence of the measured g^* in GaAs grown by molecular beam epitaxy (triangles with error bars) and grown by liquid phase epitaxy (circles with error bars) and an empirical fit with the equation $g^*(T)=-0.44+5\times10^{-4}\,T$ (solid line). The different samples show different electron-spin relaxation times, i.e., the damping of the oscillations depends on the 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. The error bars represent the absolute error and are therefore larger than the statistical scatter of the data. The dotted line is the calculated temperature dependence of g^* at the direct band gap and the dashed line the $\langle g^*\rangle$ calculated by Eq. (3).

III. EXPERIMENTAL RESULTS

A. Temperature dependence

Figure 1 shows the measured temperature dependence of the electron g factor in bulk GaAs. The g factor increases linearly from -0.44 (Ref. 18) at a lattice temperature of 4 K to about -0.3 at a lattice temperature of 280 K. The g factor is at carrier densities of 5×10^{15} cm^{-3} within our accuracy independent of the magnitude of the magnetic field up to 5 T for all temperatures. The g factor is also independent of sample quality. Figure 1 depicts the measured g factor in undoped molecular beam epitaxy GaAs (triangles) and high-quality liquid phase epitaxy GaAs (circles), but low-quality substrate GaAs with high defect concentration shows the same results. However, the sample quality determines the electron-spin dephasing time and, therefore, the measurement's accuracy and the maximum temperature where spin quantum beats can be observed with our experimental setup. The measured temperature dependence of bulk GaAs g^* contradicts predictions by $\boldsymbol{k}\cdot\boldsymbol{p}$ theory as shown in Ref. 14 for temperatures up to 200 K.

TABLE I. Temperature dependent band parameters in eV of GaAs and InP in the usual notation (see, e.g., Refs. 4 and 14).

	GaAs	InP
E_0	$(1.517-5.5)\times10^{-4}\ T^2/(T+225)$[a]	$(1.629-0.217)\times\{1+2/[\exp(697/T)-1]\}$[b]
Δ_0	$(1.851-3.5)\times10^{-4}\ T^2/(T+225)$[a]	$(1.579-0.061)\times\{1+2/[\exp(370/T)-1]\}-E_0$[b]
$E(\Gamma_8^c)$	$(4.659-4)\times10^{-4}\ T^2/(T+241)$[a]	$(5.112-4.25)\times10^{-4}\ T$[b]
$E(\Gamma_7^c)$	$E(\Gamma_8^c)-0.171$[c]	$(4.867-0.163)\times\{1+2/[\exp(775/T)-1]\}$[b]
P^2	29.3[d]	21.6[d]
P^2	6[e]	2.1[e]
C	-0.02[e,f]	-0.02[e,f]

[a]P. Lautenschlager, M. Garriga, S. Logothetidis, and M. Cardona, Phys. Rev. B **35**, 9174 (1987).
[b]P. Lautenschlager, M. Garriga, and M. Cardona, Phys. Rev. B **36**, 4813 (1987).
[c]D. E. Aspnes, C.G. Olson, and D.W. Lynch, Phys. Rev. B **12**, 2527 (1975).
[d]We use a slightly higher interband matrix element P^2 for GaAs and InP than Ref. 4. The adjustment is necessary because of the higher accuracy of the band-edge energies and—in the case of InP— of g^*. The adjustments are within the specified error of P^2 in Ref. 4.
[e]See Ref. 4.
[f]Not eV but dimensionless.

We want to point out that at finite temperatures, the electron distribution is described by a Maxwell-Boltzmann distribution for low electron densities and our experimental technique does not detect the g factor at the conduction-band minimum g_c^* but instead an energetically averaged g factor. The average g factor $\langle g^*\rangle$ results from the dependence of g^* on the electron excess energy[15] E weighted by the bulk density of states D^{3D} (where 3D is three-dimensional) and the Maxwell-Boltzmann distribution of the electrons:

$$\langle g^*\rangle = \frac{\int_0^\infty g^*(E) D^{3D}(E) \exp(-E/k_B T)\, dE}{\int_0^\infty D^{3D}(E) \exp(-E/k_B T)\, dE}, \tag{3}$$

where, according to Ref. 20,

$$g^*(E) = g_c^* + 6.3\times E \ \ (\text{eV}) \tag{4}$$

and

$$D^{3D}(E) = \frac{1}{2\pi^2}\left(\frac{2m}{\hbar^2}\right)^{3/2} E^{1/2}. \tag{5}$$

This assumes that each electron changes its energy due to electron-electron scattering much faster than the spin quantum beat oscillation period. Landau-level quantization is neglected since electron-electron scattering is, especially at high temperatures, faster than the cyclotron oscillation frequency. Excitonic effects are neglected since the exciton formation time and the carrier cooling at low temperatures is slow in bulk GaAs. The carriers are excited nonresonantly. Therefore, the lowest carrier temperature of the experiment is about 40 K (Ref. 21) and the comparison between measurement and calculation for lattice temperatures below 40 K deserves circumspection. The exact calculation of g^* at the direct band gap at a given temperature is described in Refs. 4 and 14 and the band parameters are summarized in Table I. The calculated temperature dependence of g^* at the direct band gap is depicted in Fig. 1 as a dotted line and $\langle g^*\rangle$ as a dashed line. All depicted experimental g factors are average g factors. The discrepancy between theory and experiment is obvious.

Figure 2 depicts the temperature dependence of g^* in InP. In contrast to GaAs, the temperature dependence of g^* calculated by $\boldsymbol{k}\cdot\boldsymbol{p}$ theory at the fundamental band gap in InP (dotted line) is small and agrees well with our experimental results (circles). The small variation of g^* in InP compared to the large variation in GaAs is reasonable since the g factor in InP is already closer to $g^* = +2$ of an electron in free space or at high energies.[16] The calculated $\langle g^*\rangle$ (dashed line) are slightly larger than the measured g^*; but the calculations of $\langle g^*\rangle$ overestimate the energy dependence since excitonic effects are neglected in the density of states $D^{3D}(E)$. The

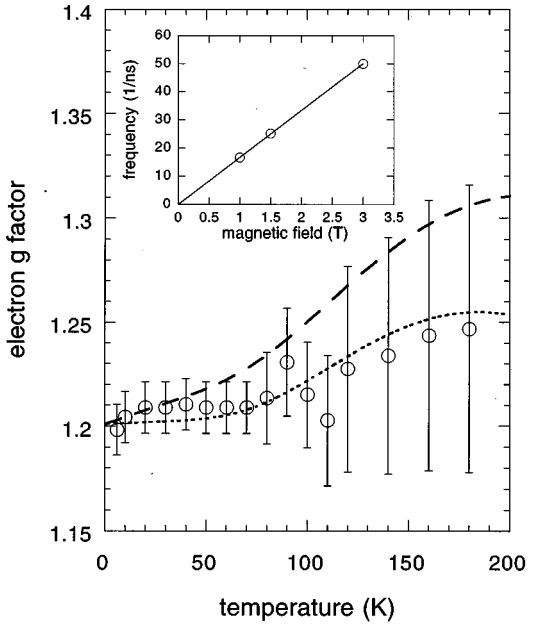

FIG. 2. Temperature dependence of the measured g^* (circles with error bars), the calculated g^* at the band edge after Refs. 4 and 14 (dotted line), and the calculated average g^* (dashed curve) in InP. Inset: Dependence of spin quantum beat frequency on the applied magnetic field at 5 K and a carrier density of 5×10^{15} cm^{-3}.

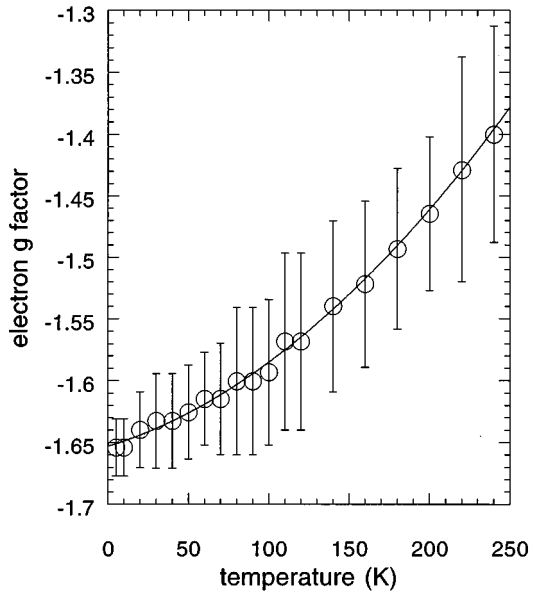

FIG. 3. Temperature dependence of the measured g^* (circles with error bars) and an empirical fit by the equation $g^*(T) = -1.653 + 4\times10^{-4}\ T + 2.8\times10^{-6z}\ T^2$ (solid curve).

inset of Fig. 2 demonstrates the linear relation between magnetic field and spin quantum beat frequency in InP at low magnetic fields and a carrier density of 5×10^{15} cm^{-3}. This linear behavior at low magnetic fields is observed at all measured temperatures in all measured semiconductors. At high magnetic fields, g^* depends on magnetic field as described in Refs. 6, 10, 13, and 20.

Figure 3 depicts $g^*(T)$ in CdTe. Since the temperature dependence of the higher conduction and valence bands is, to our knowledge, not exactly known, a comparison with $\boldsymbol{k}\cdot\boldsymbol{p}$ theory is not possible. The solid line in Fig. 3 shows an empirical fit with the equation $g^*(T) = -1.653 + 4\times10^{-4}\ \mathrm{T} + 2.8\times10^{-6}\ T^2$.

B. Density dependence in GaAs

Figure 4 shows the temporal evolution of the circularly analyzed bulk GaAs photoluminescence (PL) at two different excitation densities at a magnetic field of 3 T. The oscillations are detected on the high-energy side of the PL. They clearly show a faster oscillation period and, therefore, a larger absolute g factor for lower carrier densities. The high-density PL especially exhibits a decreasing oscillation period with time due to the temporal carrier density decrease.

Figure 5 shows the temporal high excitation PL of Fig. 4 at three different detection wavelengths: (a) above the electron Fermi energy, (b) approximately at the Fermi energy, and (c) at the direct band gap. The oscillation frequencies at the different detection energies (a) and (b) are the same because we again measure an average g factor as discussed in Sec. I A. Far below the Fermi level at the conduction-band minimum [Fig. 5(c)] the spin quantum beats are suppressed due to Fermi blocking. Fermi blocking of spin quantum beats occurs at high densities because spin-up and spin-down electron levels are, at the conduction-band minimum, both saturated and are therefore equally populated; the spin-up and

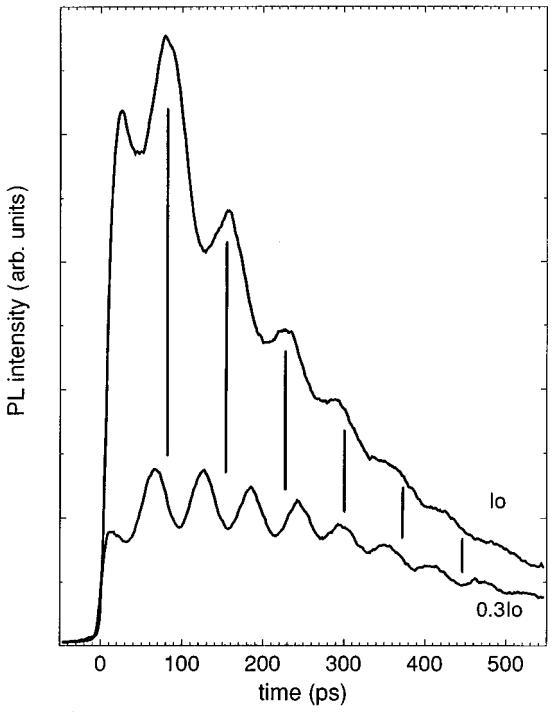

FIG. 4. Circularly polarized detected PL in bulk GaAs at two different excitation densities (I_0,0.3I_0) after circularly polarized excitation in the continuum at a magnetic field of 3 T. The different oscillation periods of the PL directly demonstrate the density dependence of the average g^*. The vertical solid lines are guides to the eye to illustrate the phase shifting. The excitation density at I_0 is 8×10^{16} cm^{-3}.

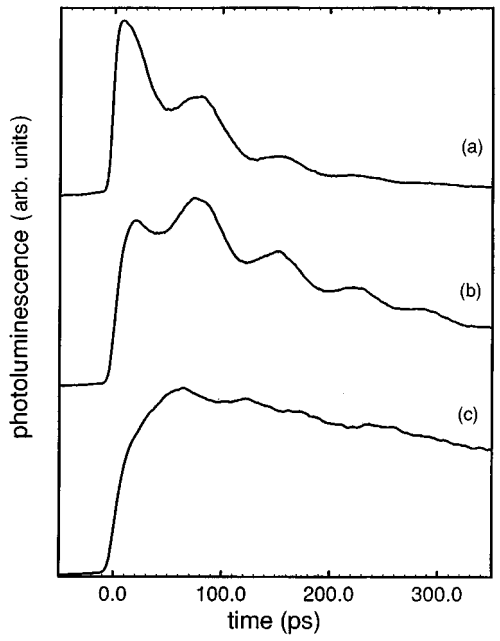

FIG. 5. Circularly polarized detected PL at an excitation density of 8×10^{16} cm^{-3} at three different detection energies: (a) above the Fermi level, (b) at the Fermi level, and (c) at the conduction-band minimum. Far below the Fermi level, spin quantum beats are strongly suppressed. We attribute the remaining weak oscillations at the band edge to regions with low electron densities due to density inhomogeneities.

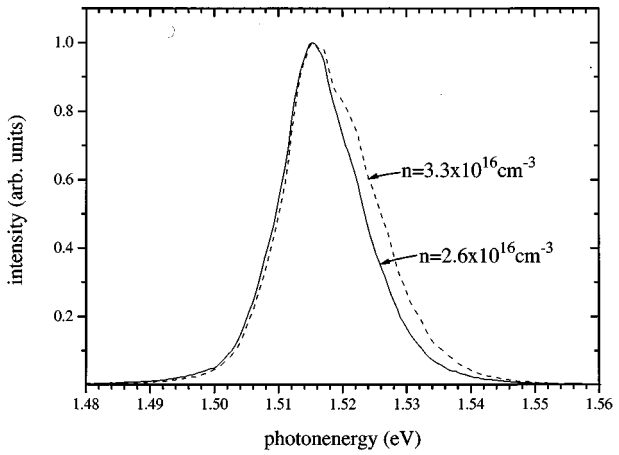

FIG. 6. Right and left circularly detected PL spectra. The spectra are measured one spin quantum beat oscillation period after excitation at a lattice temperature of 20 K and demonstrate two different Fermi-Dirac distributions for spin-up and spin-down electrons. A line-shape analysis yields the two different carrier densities.

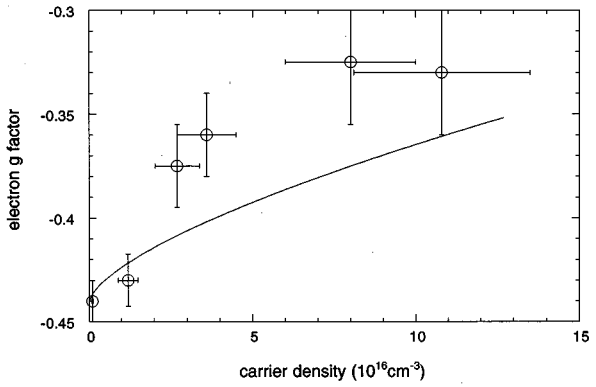

FIG. 7. Dependence of g^* on the excitation density. The open circles are measured and the solid line is the calculated g^* at the Fermi edge. The experimental densities are determined by a line-shape analysis of the transient spectra.

spin-down electron distributions are accurately described by Fermi-Dirac distributions with two different Fermi levels. The difference in spin-up and spin-down electron population yields two different PL spectra for opposite circularly polarized detection as shown in Fig. 6.

Figure 7 shows g^* in dependence on excitation density (circles with error bars) and calculations of g^* at the Fermi edge (solid line) assuming an energy dependent g factor $g^*(E) = -0.44 + 6.3 \times E$ (eV) (Refs. 10, 16, and 20) and a Fermi-Dirac distribution with $T = 0$ K.[22] At excitation densities above 10^{17} cm^{-3} no spin quantum beats are observable because spin dephasing is faster than one oscillation period. In a degenerate electron gas we mainly measure the g factor at the Fermi edge of the electron gas because the exchange of electrons at the Fermi edge with electrons at the fundamental band gap is weak at low temperatures. The discrepancy between measurement and theory indicates that theory underestimates the energy dependence of g^*.

IV. CONCLUSION

We have measured the temperature dependence of the electron Landé g factor in CdTe and InP and extended the temperature range in GaAs from 200 K to 280 K. The results in bulk GaAs verify the discrepancy between simple five-band $\boldsymbol{k}\cdot\boldsymbol{p}$ theory calculations and the experimental g factor. The theoretical energy dependence of g^* in a thermalized electron gas does not explain this discrepancy.

In InP, the temperature dependence of g^* is small because the low-temperature g^* is large compared to g^* in GaAs; and the agreement between theory and experiment is good. For CdTe, a comparison between theory and experiment is not possible because the temperature dependence of the conduction and valence bands is for this semiconductor not accurately known.

The density dependent measurements in GaAs show a small density dependence of g^* at densities below 1×10^{16} cm^{-3}, confirming earlier measurements,[14] and an enhanced g^* for higher densities—demonstrating the need for relatively low carrier densities for measurements of g^* at the Γ minimum. The fast spin dephasing time at densities above 10^{17} cm^{-3} prevents g-factor measurements by spin quantum beats at very high densities. The Fermi blocking of spin quantum beats demonstrates that two different Fermi distributions—one for spin up and one for spin down—can coexist in a degenerated electron gas.

Note added in proof. Measurements at a lattice temperature of 10 K and carrier densities below 10^{15} cm^{-3} yield a linear dependence of g^* in bulk GaAs on the magnetic field between 2 and 12 T. The change of g^* amounts to 0.005 per T. A linear extrapolation to 0 T yields a g^* of -0.47 instead of -0.44 at moderate densities and low magnetic fields. The spin quantum beats vanish at densities below 10^{14} cm^{-3}.

ACKNOWLEDGMENTS

We gratefully acknowledge the expert technical assistance of K. Rother and H. Klann, the critical and helpful discussions with U. Rössler, E. L. Ivchenko, and T. Ruf, and the financial support of the Bundesministerium für Bildung und Wissenschaft of the Federal Republic of Germany. M. O. expresses gratitude to the NATO Science Committee for financial support and to the DAAD. M.O. and K.H.S. would like to thank the Alexander von Humboldt-Stiftung for the financial support by Feodor-Lynen grants.

[1] L.M. Roth, B. Lax, and S. Zwerdling, Phys. Rev. **114**, 90 (1959).
[2] M. Cardona, J. Phys. Chem. Solids **24**, 104 (1963).
[3] C. Weisbuch and C. Hermann, Phys. Rev. B **15**, 816 (1977).
[4] C. Hermann and C. Weisbuch, Phys. Rev. B **15**, 823 (1977).
[5] M. Cardona, N.E. Christensen, and G. Fasol, Phys. Rev. B **38**, 1806 (1988).
[6] P. Pfeffer and W. Zawadzki, Phys. Rev. B **41**, 1561 (1990).
[7] M. Dobers, K. v. Klitizing, and G. Weimann, Phys. Rev. B **38**, 5453 (1988).
[8] E.L. Ivchenko and A.A. Kiselev, Fiz. Tekh. Poluprovodn. **26**, 1471 (1992) [Sov. Phys. Semicond. **26**, 827 (1992)].
[9] M.J. Snelling, G.P. Flinn, A.S. Plaut, R.T. Harley, A.C. Tropper, R. Eccleston, and C.C. Phillips, Phys. Rev. B **44**, 11 345 (1991).
[10] H. Mayer and U. Rössler, Phys. Rev. B **44**, 9048 (1991).
[11] A.P. Heberle, W.W. Rühle, and K. Ploog, Phys. Rev. Lett. **72**, 3887 (1994).
[12] M. Oestreich, A.P. Heberle, W.W. Rühle, R. Nötzel, and K. Ploog, Europhys. Lett. **31**, 399 (1995).
[13] R.M. Hannak, M. Oestreich, A.P. Heberle, W.W. Rühle, and K. Köhler, Solid State Commun. **93**, 313 (1995).
[14] M. Oestreich and W.W. Rühle, Phys. Rev. Lett. **74**, 2315 (1995).
[15] W. Zawadzki, Phys. Lett. **4**, 190 (1963).
[16] M.A. Hopkins, R.J. Nicholas, P. Pfeffer, W. Zawadzki, D. Gauthier, J.C. Portal, and M.A. DiForte-Poisson, Semicond. Sci. Technol. **2**, 568 (1987).
[17] W. Ekhard, K. Lösch, and D. Bimberg, Phys. Rev. B **20**, 3303 (1979).
[18] Our experiment does not yield the sign of g^*. However, many experiments and theories confirmed the negative value of g^* as first determined by White *et al.* (Ref. 19).
[19] A.M. White, I. Hinchliffe, P.J. Dean, and P.D. Greene, Solid State Commun. **10**, 497 (1972).
[20] M.J. Yang, R.J. Wagner, B.V. Shanabrook, J.R. Waterman, and W.J. Moore, Phys. Rev. B **47**, 6807 (1993).
[21] W.W. Rühle, K. Leo, and E. Bauser, Phys. Rev. B **40**, 1756 (1989).
[22] We can neglect the finite carrier temperature of the degenerated electron gas. With increasing carrier temperature electrons are transfered from below the Fermi energy to energies above the Fermi energy reducing the Fermi blocking. We measure an average g^*, which corresponds to g^* at the Fermi energy since $g^*(E)$ increases linearly with energy.