The sign of electron $g$-factor in GaAs$_{1-x}$N$_x$ measured by using the Hanle effect

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Received 10 July 2008, in final form 3 September 2008
Published 29 October 2008
Online at stacks.iop.org/SST/23/114008

Abstract

Positive signs of the effective $g$-factors for free electrons in the conduction band and electrons localized on deep paramagnetic centers have been measured in the nitrogen dilute alloy GaAs$_{0.979}$N$_{0.021}$ at room temperature. The $g$-factor signs have been determined from an asymmetry in the depolarization of edge photoluminescence in a transverse magnetic field (Hanle effect) at oblique incidence of the exciting radiation and oblique-angle detection of the luminescence. The tilted spin polarization of free electrons is induced under interband absorption of circularly polarized light, and the paramagnetic centers acquire spin polarization because of spin-dependent capture of free spin-polarized electrons by these centers. The measured Hanle curve is a superposition of two lines, narrow and broad, with the widths $\sim$400 G and $\sim$50000 G, arising due to the depolarization of localized and free electrons, respectively. The difference in the linewidths by two orders of magnitude strongly indicates much longer spin lifetime for the paramagnetic centers as compared with that for the free carriers. The magnitude and direction of the asymmetry in the measured Hanle curve have been found to depend on the partial contributions to the recombination radiation from the heavy- and light-hole subbands split by a uniaxial deformation of the GaAs$_{1-x}$N$_x$ film grown on a GaAs substrate. We have extended the theory of optical orientation in order to calculate the excitation spectrum of the photoelectron tilted-spin polarization and the circularly polarized luminescence spectrum taking into account that, in the strained samples under study, the light-hole subband lies above the heavy-hole one. The results have further been used to calculate the shape of Hanle curve as a function of the excitation and registration energies as well as the incidence and detection angles and to compare the theory with the experiment.

1. Introduction

Dilute III-N-V alloys with the group-V cations partially substituted by nitrogen, e.g., GaAsN and InGaAsN, have recently attracted particular attention owing to a number of their unusual properties. Thus, an increase of the nitrogen content in the alloy is accompanied by an anomalous reduction of the fundamental band gap, by more than 0.1 eV per 1% of N as the nitrogen concentration increases from zero up to 4% [1–4], as well as by a drastic increase of the electron effective mass and enhanced nonparabolicity of the $\Gamma_6$ conduction band [5–8] (for reviews, see also [9, 10]). These specific properties are related to the N-substitution-induced resonant electronic states within the conduction band continuum and an anticrossing of the localized nitrogen states with the extended conduction states of the semiconductor matrix [11–13]. Qualitatively, the available experimental data are consistent with the simple phenomenological band anticrossing (BAC) model which takes into account the anticrossing hybridization only with the levels of isolated (single) nitrogen atoms [11]. As a result, the two hybridized conduction bands, $E_+$ and $E_-$, are formed. As the nitrogen concentration increases, the lower band, $E_-$, shifts down in energy reducing the fundamental energy-band gap $E_g$. Simultaneously, the band-bottom
effective mass becomes heavier and the nonparabolicity coefficient rises considerably.

The band-structure modification involves a remarkable change in the gyromagnetic factor, \( g \), of electrons in the conduction band.\cite{14,15}. The change has two causes. Firstly, as in the case of conventional alloys \( \text{In}_x\text{Ga}_{1-x}\text{As} \)\cite{16,17}, the band-gap shrinkage leads to a monotonic downward shift of the effective \( g \)-factor below the value of the \( g \)-factor in the GaAs matrix \( g = -0.44 \) in GaAs at helium temperature\cite{14}. Secondly, the coupling with isolated nitrogen levels, with the Landé factor being positive and close to the electron \( g \)-factor in the vacuum \( g_0 = 2 \), tends to shift the \( g \) value upward, as was first found experimentally in InGaAsN when studying the dependence of the absorption coefficient on the magnetic field\cite{14}. The two effects have opposite signs and partially compensate each other. The BAC model predicts an initial growth in the dependence \( g(x) \)\cite{14}. The dependence is smooth and, as calculated for \( \text{GaAs}_{1-x}\text{N}_x \) in \cite{15}, has a maximum value \( g \approx 0 \) at \( x \approx 1\% \). At the same time, detailed measurement and comparison of the left- and right-circularly polarized photoluminescence spectra performed in \( \text{GaAs}_{1-x}\text{N}_x \) in a longitudinal magnetic field at helium temperature for \( x \leq 0.6\% \) have shown that \( g \) grows sharply with increasing \( x \), reverses its sign from negative to positive at \( x \approx 0.04\% \), reaches a value of 0.7 in the range \( x = 0.04\% \) and retains that value with small deviations as \( x \) increases up to 0.6\%\cite{15}. Such an abrupt dependence of \( g \) on \( x \) at small \( x \) has been interpreted in terms of a modified \( k \cdot p \) model\cite{13,15}. This model takes into account the hybridization of the host conduction band not only with the localized levels due to single N atoms but also with the levels which are formed by clusters of N atoms, pairs and triplets, and lie closer to the conduction-band bottom.

Data on the \( g \)-factor in \( \text{GaAs}_{1-x}\text{N}_x \) with high nitrogen concentration are highly desirable for the purpose of checking various theoretical models. However, to the best of our knowledge, such experimental data for \( x \approx 0.6\% \) are lacking.

In the present work the sign of the electron \( g \)-factor in the \( \text{GaAs}_{1-x}\text{N}_x \) conduction band has been measured at room temperature for \( x = 2.1\% \). To this end, we used the dependence of the sign of the asymmetry of the Hanle effect of optically spin-polarized electrons, measured under oblique excitation, on the \( g \)-factor sign. This method is based on the relationship between the direction of Larmor precession of electron spin in a magnetic field and the \( g \)-factor sign. As compared to other methods, this method is advantageous for studying crystals with high nitrogen content, particularly at high (up to room) temperatures when the photoluminescence occurs over a broad spectrum. The fundamentals of the method are presented in section 3, together with the discussion of peculiarities of the electron spin polarization and the oblique Hanle effect caused by the splitting of the \( \Gamma_8 \) valence band due to an uniaxial strain of the GaAsN films grown on GaAs substrates.

Section 4 provides the theory extended in order to calculate the magnitude and orientation of photoelectron spin polarization, the circular polarization of photoluminescence and the Hanle-curve shape as a function of the energy of absorbed and emitted photons in a uniaxially strained semiconductor with the top of the light-hole subband higher in energy than the top of the heavy-hole subband.

In the GaAsN alloys under study, the optical spin polarization of conduction electrons at room temperature has been found to be anomalously high\cite{4,18}. This is the result of spin-dependent recombination of polarized conduction electrons on deep paramagnetic centers which apparently appear during the introduction of nitrogen atoms into GaAs. The capture of free electrons is accompanied by efficient dynamic spin polarization of electrons bound on the centers. In turn, the polarized centers kinetically affect the polarization of free electrons so that a strongly coupled spin system of free and localized electrons is formed. A change in the polarization of localized electrons manifests itself in a remarkable change of the free-electron polarization. Thus, from the analysis of the curve of magnetic depolarization of the interband photoluminescence contributed by free particles, one can determine the \( g \)-factor signs of both free and bound electrons. In section 5, we present experimental results together with the results of numerical calculation taking into account the influence of spin-dependent recombination on the electron spin polarization.

2. Samples and experimental details

We studied the undoped 0.1 \( \mu \)m thick \( \text{GaAs}_{0.979}\text{N}_{0.021} \) layer grown by rf-plasma-assisted solid-source molecular-beam epitaxy at 350–450 °C on the semi-insulating \((0 0 1) \) GaAs substrate\cite{4}. The nitrogen content and crystallinity of the grown layer were examined by the x-ray diffraction technique. The as-grown structure was annealed for 5 min at 700 °C in a flow of arsenic in the growth chamber. Continuous-wave tunable Ti:sapphire laser was used for photoluminescence (PL) excitation. Spin polarization of electrons was created upon the interband absorption of circularly polarized light\cite{19}. It was monitored by measuring the degree of circular polarization of PL, defined as \( \rho = (I^+ - I^-)/(I^+ + I^-) \), where \( I^+ \) and \( I^- \) are the right (\( \sigma^+ \)) and left (\( \sigma^- \)) circularly polarized PL components. The value \( \rho \) and PL intensity in a wavelength range up to 1.4 \( \mu \)m were measured using a high-sensitivity polarization analyzer\cite{20} comprising a quartz polarization modulator\cite{21}, a lock-in two-channel photon counter and a photomultiplier with an InGaAsP photocathode. The measurements were carried out at 300 K.

Figure 1 shows the spectra of PL intensity (solid curve) and of PL circular polarization degree (circles) measured in the \( \text{GaAs}_{0.979}\text{N}_{0.021} \) layer under normal incidence of the pump beam and the detection of luminescence along the growth axis. The PL spectrum consists of two strongly overlapping inhomogeneously broadened bands where the low-energy PL band is negatively polarized (relative to the polarization of the exciting beam), whereas the high-energy band is polarized positively. As we have shown earlier\cite{4}, the two PL bands appear due to splitting of the light- and heavy-hole valence subbands induced by uniaxial compression along the growth axes of \( \text{GaAs}_{1-x}\text{N}_x \) film grown on the GaAs substrate. In its turn, this compression arises from large lattice mismatch.

\[ g \approx 0.7 \]
increase the free-electron polarization which can reach of the polarized conduction electrons. The polarized centers arise with the incorporation of nitrogen in GaAs and at strong pumping. The polarization in figure 1 reach 30–35%, which is near to the time, the absolute values both of negative and positive GaAsN is due to dynamic polarization of electrons bound on paramagnetic centers, increases with temperature and decreases electron polarization down to units and fractions of percentage at room temperature [19]. At the same time, the absolute values both of negative and positive polarization in figure 1 reach 30–35%, which is near to the maximal magnitude of 50% determined by selection rules. Such anomalous enhancement of free-electron polarization in GaAsN is due to dynamic polarization of electrons bound on paramagnetic centers [18, 23, 24]. The paramagnetic centers arise with the incorporation of nitrogen in GaAs and are polarized as a result of spin-dependent capture on them of the polarized conduction electrons. The polarized centers increase the free-electron polarization which can reach ≈100% at strong pumping.

3. The Hanle effect in GaAsN alloys

3.1. The electron Hanle effect under normal excitation and detection

The Hanle effect is a depolarization of photoluminescence by a magnetic field $B$ directed perpendicular to the continuous-wave pump beam [19]. The effect originates from Larmor precession of electron spins, which destroys their polarization. In the simplest case, the Hanle effect is described by Lorentzian $\rho(B)/\rho(B = 0) = 1/(1 + B^2/B_{1/2}^2)$ with half-width at half-maximum, $B_{1/2} = \hbar/g\mu_B T_s$, where $g$ is the electron Landé $g$-factor, $\mu_B$ is the Bohr magneton and $T_s$ is the electron spin-polarization lifetime. Figure 2 shows the Hanle curves measured in GaAs$_{0.979}$N$_{0.021}$ at excitation and detection along the normal to the sample. Excitation energy $h\omega_{exc} = 1.312$ eV, detection energy $h\omega_{det}$ is equal to $E_2 = 1.163$ eV and $E_1 = 1.117$ eV (b). Solid curves are superpositions of the calculated Hanle curves for free (dotted curves) and bound electrons (see the text for details). Inserts show the initial parts of the Hanle curves in the range of small magnetic field.

![Figure 1](image1.png)

**Figure 1.** Spectral dependences of the PL intensity (solid curve) and of the PL circular polarization degree (circles) for the GaAs$_{0.979}$N$_{0.021}$ layer under normal incidence (θ = 0) of the pump beam and detection of the PL. Excitation energy $h\omega_{exc} = 1.312$ eV. Arrows indicate the PL detection energies $E_1 = 1.117$ eV and $E_2 = 1.163$ eV at which the Hanle curves presented in figure 2 are measured.

![Figure 2](image2.png)

**Figure 2.** Experimental Hanle curves (open circles) measured in GaAs$_{0.979}$N$_{0.021}$ at excitation and detection along the normal to the sample. Excitation energy $h\omega_{exc} = 1.312$ eV, detection energy $h\omega_{det}$ is equal to $E_2 = 1.163$ eV (a) and $E_1 = 1.117$ eV (b). Solid curves are superpositions of the calculated Hanle curves for free (dotted curves) and bound electrons (see the text for details). Inserts show the initial parts of the Hanle curves in the range of small magnetic field.
two curves with strongly different half-widths, where the narrow curve describes the Hanle effect of bound electrons and the wide curve presents depolarization of free electrons. Solid curves in figure 2 are calculated using the equation \( \rho(B) = \rho_0/[1 + (B/B_1^{1/2})^2] + \rho_{\text{res}}/[1 + (B/B_1^{1/2})^2] + \rho_{\text{res}} \) at \( B_1/2 = 25000 \) G and \( B_1/2 = 185 \) G. It is seen that the calculated curves describe reasonably the measured ones in figure 2 for both positive and negative polarization. During the calculation the fitting parameters were \( \rho_0, \rho_{\text{res}}, \alpha \). At present, the origin of the constant polarization \( \rho_{\text{res}} \sim 4\% \) is not clear and additional studies should be done for its elucidation.

3.2. The electron Hanle effect under oblique excitation and detection

The Hanle effect can provide for measuring the sign of the Landé \( g \)-factor of electrons [25–27] since a rotation direction of mean electron spin in a magnetic field depends on the \( g \)-factor sign. The most suitable way to find the \( g \)-factor sign is to use a specular geometry of experiment. In this geometry, the exciting light falls on the sample obliquely, the luminescence is detected at an angle to the exciting beam while a magnetic field is perpendicular to both the excitation and detection directions, and lies in the crystal surface plane [26], as shown in figure 3.

In specular geometry, the dependence \( \rho(B) \) in the bulk zinc-blende semiconductor has the form [26]

\[
\rho(B) = \frac{\cos \alpha + \varphi \sin \alpha}{1 + \varphi^2},
\]

where \( \alpha \) is the angle between the directions of excitation and detection, \( \rho_0 \) is the degree of polarization for \( B = 0 \) and \( \alpha = 0, \varphi = \Omega T_e = g \mu_B B T_e/h \) is the angle through which the mean spin \( S \) of electrons with the \( g \)-factor equal to \( g \) turns during the electron-spin lifetime \( T_e = \tau_e/(\tau + \tau_e) \), \( \tau \) and \( \tau_e \) are the electron lifetime and spin relaxation time in the conduction band, \( \Omega = g \mu_B B/h \) is the Larmor frequency, and the Born magneton \( \mu_B > 0 \). Equation (1) was derived taking into account the fact that in a bulk unstrained GaAs-type crystal the mean electron spin in the moment of creation, \( S_0 \), is parallel to the exciting beam, and the degree of PL polarization is equal to the projection of \( S \) on the direction of detection: \( \rho = S n_1 \), where \( n_1 \) is the unit vector along the direction of registration [19, 28]. At \( \alpha = 0 \), the Hanle curve is a symmetrical function of the magnetic field as, for example, one can see in figure 2. For \( \alpha \neq 0 \), the \( \rho(B) \) plot is not symmetrical with respect to the \( B = 0 \) point. The value of \( \rho \) is larger for the magnetic field direction in which the electron spin precesses toward the luminescence observation axis. Note that for measuring the \( g \)-sign it is sufficient to record the asymmetry in the dependence of the absolute value of \( \rho \) on \( B \).

The above method acquires distinctive features [27] if the heavy- and light-hole subbands are split due to the uniaxial deformation in a bulk crystal or due to the quantum-size effect in a nanostructure.

First, in this case the direction of the mean spin of photogenerated electrons \( S_0 \) may not coincide with the direction of the exciting beam. Theory [28] (see also [29]) developed for uniaxially strained crystals yields the following expressions for the mean spins of the electrons created with \( k \approx 0 \) from states close to the tops of the light-hole (\( hh \rightarrow c \) transition) and the heavy-hole (\( hh \rightarrow c \) transition) subbands:

\[
S_{0hh} = -\frac{\nu(\nu_0)}{1 + (\nu_0)^2}, \quad S_{0lh} = \frac{3(\nu_0)^2 - 2\nu_0}{1 + (\nu_0)^2}.
\]

where \( \nu \) and \( \nu_0 \) are unit vectors along the deformation (growth) axis and the pump beam direction. As seen from equations (2), the spin \( S_{0hh} \) is parallel to the deformation axis for any light incidence angle, whereas the angle between \( S_{0lh} \) and \( \nu \) is twice the angle \( \theta \) between the excitation direction and the growth axis (figure 4). The light-hole and heavy-hole splitting can be neglited if the energy of the photogenerated holes exceeds substantially the splitting energy \( \Delta_c \) [28]. Such a situation can be realized when \( \hbar\omega_{\text{exc}} \sim E_g > \Delta_c \), where \( E_g \) is the forbidden gap of the deformed crystal. In this case, the electrons are excited from both valence subbands, and, as in a bulk unstrained crystal, their mean spin \( S_{0\Sigma} \) is directed along the excitation beam.

3 To be precise, \( \tan \xi = 2 \tan \theta \), where \( \xi \) is the angle between \( S_{0\Sigma} \) and \( \nu \). Since the angles \( \theta \) and \( \nu \) cannot exceed 16° due to the large refractive index, approximately 3.6, of GaAs and GaAsN, we can accept \( \xi \approx 2\theta \).
along the exciting beam: $S_{0}\uparrow\uparrow n_{0}$ (figure 4). Thus, with the increase of excitation energy from $\hbar\omega_{ex} \approx E_{g}$ toward $\hbar\omega_{ex} \gg E_{g} + \Delta_{c}$, the spin $S_{0}$ changes from $S_{0\uparrow}$ to $S_{0\uparrow}$ rotating counterclockwise through angle $(180^\circ - \theta)$.

Second, in the case of valence-subband splitting, the PL polarization is determined by projection of $S$ not on the direction of observation $n_{1}$, as this should be for a bulk unstrained semiconductor, but rather on the vector $S_{1}$, which depends on the angle between $n_{1}$ and $\nu$ vectors and on the actual recombination energy [19, 28]:

$$\rho = -4 S_{1} \cdot S_{1}. \quad (3)$$

For recombination with the tops ($k \approx 0$) of the light-hole and heavy-hole subbands, the vectors $S_{1\uparrow}$ and $S_{1\downarrow}$ are given by the same expressions (2) as $S_{lh\uparrow}$ and $S_{lh\downarrow}$, but with $n_{1}$ being replaced by $n_{1}$. We readily see that $S_{1\downarrow}$ is always parallel to the growth axis, while $S_{1\uparrow}$ is at an angle $\theta$ to this axis (figure 4). If the PL detection energy $\hbar\omega_{det} \approx E_{g}$, recombination with the light holes only is possible and, therefore, $S_{1} = S_{1\uparrow}$. When $\hbar\omega_{det} = E_{g} \gg \Delta_{c}$, the crystal valence-band splitting can be ignored and the effective direction of the PL detection, given by vector $S_{1\downarrow}$, coincides with the real one: $S_{1} \downarrow\uparrow n_{1}$. So, with increase of the detection energy, the vector $S_{1}$ changes its direction from $S_{1\downarrow}$ to ${-n_{1}}$, rotating clockwise through angle $(180^\circ - \theta)$.

Since both the value and the direction of the $S_{0}$ and $S_{1}$ vectors are energy dependent, the Hanle curves will be different for different excitation and detection energies. However, as shown in [27], they can still be described by equation (1), provided the angle $\alpha$ is replaced by the angle $(\gamma - \beta)$, where $\gamma$ and $\beta$ are the angles between the $z$-axis and vectors $S_{1}$ and $S_{0\uparrow}$, respectively, reckoned counterclockwise from the $z$-axis.

4. Theory

The above qualitative analysis of orientation of the vectors $S_{0}$ and $S_{1}$ and the angle between them is based on equations (2) valid for the $\Gamma$-point, $k = 0$. The values of $S_{0}$ and $S_{1}$ for arbitrary frequencies $\hbar\omega_{ex}$ and $\hbar\omega_{det}$ can be found taking into account the interband optical transitions with $k \neq 0$ where the heavy- and light-hole states are mixed. Such kinds of calculations have been performed for unstrained bulk zinc-blende-lattice semiconductors [29], strained bulk semiconductors with the top of the heavy-hole subband lying above that of the light-hole subband [30, 31], quantum-well structures in the approximation of infinitely-high barriers [32] and for finite barriers [33] (see also [34]), and semiconductor superlattices [35].

In this section we will follow the method of [30] developed in order to calculate the optical orientation of electron spins under interband absorption of circularly polarized light normally incident on a hexagonal crystal, e.g., CdS or CdSe. In fact, in [30] the quasicubic model of the valence band structure is used: the crystal splitting of the higher two valence subbands is obtained by applying an effective uniaxial strain to a crystal of cubic symmetry with the $\Gamma$$_{8}$ valence band. Here we generalize the approach of [30] to consider (i) the oblique incidence of the exciting light on the sample, and (ii) the opposite sign of the uniaxial strain resulting in the opposite sequence of the split subbands, now the light-hole subband lies above the heavy-hole subband.

The spin-polarized electrons are described by the spin-density matrix $f_{ss'}$, where $s, s' = \pm 1/2$ are the spin indices. This matrix can be found from the balance matrix equation

$$\begin{align*}
\begin{pmatrix}
\frac{\partial j_{\uparrow}}{\partial t} \\
\frac{\partial j_{\downarrow}}{\partial t}
\end{pmatrix}
\text{spin.rel.} + \begin{pmatrix}
\frac{\partial j_{\uparrow}}{\partial \tau}
\\
\frac{\partial j_{\downarrow}}{\partial \tau}
\end{pmatrix}
\text{Larmor} &= j,
\end{align*}
$$

(4)

where the first and second terms describe the spin relaxation and Larmor precession of the electron spins, and the right-hand term is the generation rate of the electron spin-density matrix. Four components of the latter matrix can be calculated by using the general equation

$$f_{ss'} \propto \sum_{kn} \delta[E_{k}(r) - E_{vn}(r) - \hbar\omega] \times \sum_{f} M_{cs,vnj}(r) M_{cs',vnj}^{*}(r). \quad (5)$$

Hereafter we omit common multipliers and use the following notation: $k$ is the electron wave vector, $E_{k}(r)$ and $E_{vn}(r)$ are the electron energies in the conduction band $c$ and the valence subband $vn(n = \pm)$ given by

$$E_{c}(r) = E_{0} + \frac{\hbar^{2} k^{2}}{2m_{c}}, \quad E_{vn}(r) = A k^{2} - \frac{\Delta_{c}}{2} \pm R,$$

(6)

$$R = \sqrt{\left(\frac{\Delta_{c}}{2}\right)^{2} + B^{2} k^{4} + \frac{A^{2}}{2} (3 k_{e}^{2} - k^{2})},$$

$z$ is the uniaxial-strain axis coinciding with the crystal growth axis, the index $j = 1, 2$ enumerates degenerate states in the $v_{n}$ and $v_{\nu}$ valence subbands; $m_{c}$ is the electron effective mass in the conduction band, $A$ and $B$ are the standard valence-band parameters entering the Luttinger Hamiltonian taken in the spherical approximation ($D = \sqrt{3}B$); $\Delta_{c}$ is the splitting of the $\Gamma$$_{8}$ valence band at $k = 0$ called the crystal splitting. In the following we assume a value of $\Delta_{c}$ to be positive in which case the indices $v_{n}$ and $v_{\nu}$ correspond to the light-hole and the heavy-hole subbands, respectively, and the parameter $E_{0}$ coincides with the fundamental band gap $E_{c}$. The opposite order of the light- and heavy-hole subbands considered in [30] is described by negative values of $\Delta_{c}$. The matrix elements for interband electron transitions are related to the interband matrix elements of the momentum operator $\hat{p}$ by

$$M_{cs,vnj}(r) \propto e \cdot p_{cs,vnj}(r),$$

where $e$ is the light polarization unit vector.

Under oblique incidence with the incidence plane containing the axis $x$ and $z$ (and perpendicular to the $y$ plane) the initial spin $S_{0}$ of the photogenerated electrons is related to $\hat{j}$ by

$$S_{z} = \frac{1}{2} \frac{\hat{j}_{x} - \hat{j}_{z} - \hat{j}_{x} + \hat{j}_{z}}{2}, \quad S_{x} = \frac{\text{Re} \left\{ \hat{j}_{x} \hat{j}_{y} \right\}}{\hat{j}_{x}^{2} + \hat{j}_{y}^{2} + \hat{j}_{z}^{2}}. \quad (7)$$

We omit further details and present the final result for the generation matrix (5) written as a sum of the partial contributions, $f_{ss'}^{\alpha} + f_{ss'}^{\beta}$, due to the optical transitions.
from the subbands $v_n$ with $n = \pm$. These contributions can eventually be presented in the form

\[ \tilde{f}^{(n)}_{st} = W_n \delta_{s\nu} + \rho^0 t_s t_p (R_n^\dagger \sin \theta \sigma_s + R_n \cos \theta \sigma_s), \]

where $\rho^0$ is the degree of circular polarization of the light wave in the vacuum, $\theta$ is the refraction angle, $t_s$ and $t_p$ are the amplitude transmission coefficients of the $s$- and $p$-polarized light, $\sigma_s$ and $\sigma_p$ are the Pauli spin $2 \times 2$ matrices.

The coefficients $W_n$, $R_n^+$ and $R_n^-$ can be readily calculated assuming

\[ \eta = \frac{|B|}{(h^2/2m_e)} \ll 1. \]

In GaAs the dimensionless parameter $\eta$ is about 0.23. For the light propagating in the direction $n = (\sin \theta, 0, \cos \theta)$ one has

\[ W_\pm = \sqrt{\chi_\pm} \left[ \frac{t_s^2 + t_p^2}{2} - \frac{1}{2} \frac{\Pi(\chi_\pm) F(\theta)}{\chi_\pm} \right]. \]

\[ W_\mp = \left\{ \begin{array}{ll} 0 & \text{for } 1 > \epsilon > 0, \\
\sqrt{\chi_\mp} \left[ \frac{t_s^2 + t_p^2}{2} + \frac{1}{2} \frac{\Pi(\chi_\mp) F(\theta)}{\chi_\mp} \right] & \text{for } \epsilon > 1. \end{array} \right. \]

Here

\[ \chi_+ = \eta \epsilon, \quad \chi_- = \left\{ \begin{array}{ll} 0 & \text{for } 1 > \epsilon > 0, \\
\eta (\epsilon - 1) & \text{for } \epsilon > 1. \end{array} \right. \]

\[ \epsilon = (\hbar \omega - E_g)/\Delta_c, \omega \text{ is the light frequency, and the function } \Pi(\chi) \text{ of a variable } X \text{ is defined by} \]

\[ \Pi(X) = \frac{1}{4} \left( \frac{3 + 2X - 4X^2}{\sqrt{6X}} \right) \arcsin \sqrt{\frac{6X}{1 + 4X^2 + 2X + 1 - 2X}}. \]

The photoelectron average spin components in the directions $z$ and $x$ are determined by

\[ S_z = \rho^0 t_s t_p \cos \theta \frac{R^+_z + R^-_z}{W_+ + W_-}, \]

\[ S_x = \rho^0 t_s t_p \sin \theta \frac{R^+_z + R^-_z}{W_+ + W_-}, \]

where

\[ R^+_z = -\frac{1}{2} \sqrt{\chi_+}[1 - 2\Pi(\chi_+)], \quad R^-_z = -\frac{1}{2} \sqrt{\chi_+}[1 + \Pi(\chi_+)], \]

\[ R^-_z = 0 \text{ if } 1 > \epsilon > 0, \quad \text{and} \]

\[ R^+_z = -\frac{1}{2} \sqrt{\chi_-}[1 - 2\Pi(\chi_-)], \quad R^-_z = -\frac{1}{2} \sqrt{\chi_-}[1 - \Pi(\chi_-)], \]

if $\epsilon > 1$.

The above equations correspond to positive $\Delta_c$. However, they are also valid for negative $\Delta_c$ if the function (9) is replaced by

\[ \Pi(X) = -\frac{1}{4} \left( 1 + 2X + \frac{3 - 2X - 4X^2}{\sqrt{6X}} \right) \right. \]

\[ \times \left. \ln \frac{1 + 2X + \sqrt{6X}}{\sqrt{1 + 4X^2 + 2X}}. \right. \]

The angle $\varphi$ between the vector of the average spin and the axis $z$ equals

\[ \varphi = \arctan \left( \frac{R^+_z}{R^-_z} \tan \theta \right) + (1 - \text{sign}(S_z)) \frac{\pi}{2} \]

\[ \approx \arctan \left( \frac{R^+_z}{R^-_z} \right) + (1 - \text{sign}(S_z)) \frac{\pi}{2}, \]

where $R^+_z = R^+_z + R^-_z, R^-_z = R^+_z + R^-_z$. Here we take into account that, in the medium with a big index of refraction, the refraction angle $\theta$ is small even for remarkable incidence angles.

Under photoexcitation near the fundamental edge, $\hbar \omega \approx E_g$, one has $\chi_- = 0$, $W_- = 0$ and the optical transitions are allowed only from the upper light-hole subband. For $0 < \epsilon = (\hbar \omega - E_g)/\Delta_c \ll 1$, the function $\Pi(\chi)$ in (9) tends to 1, and according to equation (10) the ratio $S_+/S_-$ is given by $-2 \tan \theta$ and $\varphi$ by $-\arctan 2 \tan \theta$ in agreement with the first equation (2). At the edge of the transitions from the heavy-hole subband, $\epsilon - 1 = (\hbar \omega - E_g - \Delta_c)/\Delta_c \ll 1$, the function$\Pi(\chi_-) \rightarrow 1$ and, therefore, $R^-_z \rightarrow 0$ and $R^+_z$ is negative. Therefore, for electrons excited from the top of the $v_--$ subband, $\varphi \rightarrow \pi$, in agreement with the second equation (2).

5. Calculation, experimental results and discussion

5.1. Specular configuration

It is convenient to use the reduced excitation and detection energies $\hbar \omega_{\text{exc}} = (\hbar \omega_{\text{exc}} - E_g)/\Delta_c$ and $\hbar \omega_{\text{det}} = (\hbar \omega_{\text{det}} - E_g)/\Delta_c$. Our measurements have been made for a large excitation energy $\hbar \omega_{\text{exc}} = 7.2$ permitting the crystal splitting of the valence subbands to be neglected. Below we present the results of calculation for that energy. They show that in this case $\beta \approx 180^\circ - \theta$, i.e. $S_0 = n_0$, and $|S_0| = 0.25 \cos \theta \approx 0.25$, which is in full agreement with the above qualitative analysis based on the expressions (2).

The dependences of the polarization $\rho(0) = \rho_0 \cos(\gamma - \beta)$ and of the angle $(\gamma - \beta)$ on the detection energy $\hbar \omega_{\text{det}}$ calculated for zero magnetic field and specular configuration are shown by solid lines in figures 5(a) and 5(c). One can see that for the detection energy $\hbar \omega_{\text{det}} = 0$, when recombination with the light holes with $k = 0$ only is possible and the vector $S_1$ is directed at the angle $\gamma = 180^\circ - 2\theta$ to the $z$-axis, the angle $(\gamma - \beta) = -\theta$ and polarization $\rho(0) \approx -0.5$. In this case, $|S_1| = 0.5$. When increasing $\hbar \omega_{\text{det}}$, the admixture of the heavy-hole states leads to the increase of $\rho(0)$ and $(\gamma - \beta)$. These changes of $\rho(0)$ and $(\gamma - \beta)$ are small when $\hbar \omega_{\text{det}}$ varies from 0 to 1. However, the $\rho(0)$ and $(\gamma - \beta)$ dependences have abrupt humps at $\hbar \omega_{\text{det}} = 1$, when the transitions from the heavy-hole subband are switched on, involving the creation of electrons with opposite signs. With further increase of $\hbar \omega_{\text{det}}$, the quantity $\rho(0)$ increases rapidly, then reverses its sign from negative to positive and approaches its maximal value $\rho(0) = 0.25$ at $\hbar \omega_{\text{det}} \approx 2$, while the angle $(\gamma - \beta)$ approaches the maximal value of $(180^\circ + 2\theta)$, corresponding to which are $\gamma = \theta$ and $|S_1| = 0.25$. Those values of $\rho(0)$ and $(\gamma - \beta)$ remain practically invariable with $\hbar \omega_{\text{det}}$ increasing still further.
shown for the normal incidence, absence of magnetic field. In (b) incidence angle of 48° detection.

... bisectrix between the exciting beam and the direction of the PL configuration the normal to the sample surface coincides with the figure 5). Therefore at such an energy of detection the angle changes drastically within the range 1|ε| = 0 only when the direction of PL detection 90° to +90°. With such definition of angle α, expression (1) for the Hanle effect takes the form

\[ \rho(B) = \rho_0 \frac{\cos \alpha + \varphi \sin \alpha}{1 + \varphi^2} \text{sign}(\epsilon_{\text{det}} - \epsilon_\perp), \]  

where \( \rho_0 = 4|S_0||S_1|T_s/\tau \).

We consider the coupled spin system of free and bound electrons in the model of spin-dependent recombination proposed by Weisbuch and Lampel [36], applied by Paget [37] and generalized in [18]. In this model the magnetic-field dependence of the PL polarization can be, with high accuracy, described by two Lorentzians:

\[ \rho(B) = \rho_0 \left[ \frac{\cos \alpha + \varphi \sin \alpha}{1 + \varphi^2} + \rho_{\text{res}} \frac{\cos \alpha + \varphi_c \sin \alpha}{1 + \varphi_c^2} \right] \times \text{sign}(\epsilon_{\text{det}} - \epsilon_\perp). \]  

5.1.1. High energy detection (α = 2θ). We realized the first case at \( \epsilon_{\text{det}} = \epsilon_\perp = 2.03 \) (here, just as in all the following measurements, \( \epsilon_{\text{exc}} = 7.2 \) and the refraction angle \( \theta = 12° \)). The experimental Hanle curve (open circles in figure 6) has a manifestly asymmetric shape in this case. The
maximum of its narrow part (figure 6(a)), which corresponds to depolarization of bound electrons, shifts to positive values of the magnetic field. For the employed specular geometry of experiment, to this corresponds the positive sign of $g_e$ [18] (note that the Hanle curve, measured in the same geometry in bulk GaAs, in which $g < 0$, has a maximum at $B < 0$). The maximum of the broad part of the experimental curve $\rho(B)$ determined by depolarization of free electrons is less pronounced, since it is superimposed by the depolarization curve of bound electrons, which has a greater-by-an-order amplitude in the zero magnetic field. The polarization of bound electrons, however, decreases rapidly as the magnetic field increases, and we can neglect its contribution at high-energy excitation and the electron recombination with the light holes when $\alpha \approx -\theta$, $\hbar \omega_{\text{exc}} = 1.312 \, \text{eV}$, $\hbar \omega_{\text{det}} = E_1 = 1.117 \, \text{eV}$, $\theta = 12^\circ$.

5.1.2. Low energy detection ($\alpha = -\theta$). The experimental dependence $\rho(B)$ for this case that has been realized at $\hbar \omega_{\text{det}} = 0.45$ is shown by open circles in figure 7. The negative sign of the measured luminescence polarization results from the fulfillment of the condition $\epsilon_{\text{det}} < \epsilon_{\perp}$. The solid curve in figure 7 is calculated using equation (13) for $\alpha = -\theta$. The maxima (in the absolute value) on the depolarization curves for the bound (figure 7(a)) and free (the dotted line in figure 7(b)) electrons are situated at $B < 0$, which also provides evidence that the signs of $g$ and $g_e$ are positive.

The measured positive sign of the free-electron $g$ factor can be compared with an estimation of the $g$ value following from the relationship between $g$ and the electron effective mass $m^*$ given by equation (5) in [15] and the measured value $m^* = 0.13-0.15 \, m_0$ ($m_0$ being the free-electron mass) for GaAs$_{0.979}$N$_{0.021}$ in the backscattering configuration where the directions of detection and excitation are antiparallel, $n_1 \downarrow \uparrow n_0$. To realize that possibility, the magnetic field $B$, lying in the sample surface plane, is directed perpendicular to the exciting beam, while the sample is turned around vector $B$ in such a way that there is nonzero angle $\theta$ between the normal to the sample (axis $\nu$) and the exciting beam (vector $n_0$), as shown in the inset in figure 8.

5.2. Backscattering configuration

Deformation-induced splitting of the light- and heavy-hole subbands allows the Hanle curve asymmetry to be observed even in the backscattering configuration where the directions of detection and excitation are antiparallel, $n_1 \downarrow \uparrow n_0$. To realize that possibility, the magnetic field $B$, lying in the sample surface plane, is directed perpendicular to the exciting beam, while the sample is turned around vector $B$ in such a way that there is nonzero angle $\theta$ between the normal to the sample (axis $\nu$) and the exciting beam (vector $n_0$), as shown in the inset in figure 8.

The dependence of angle $\alpha$ between vectors $S_0$ and $S_1$ on detection energy $\epsilon_{\text{det}}$ calculated for a backscattering
configuration and \( \varepsilon_{\text{exc}} = 7.2 \) is shown by the dashed line in figure 5(d). One can see that \( \alpha = +3\theta \) if \( \varepsilon_{\text{det}} < 1 \) and \( \alpha \approx 0 \) if \( \varepsilon_{\text{det}} > 2 \).

5.2.1. Low energy detection (\( \alpha = +3\theta \)). At \( \varepsilon_{\text{det}} < 1 \) angle \( \alpha \approx 3\theta \), which makes the asymmetry of the experimental Hanle curve (open circles in figure 8) more pronounced than in the case of the specular configuration when \( \alpha \approx -\theta \). The negative sign of polarization is due to the fact that \( \varepsilon_{\text{det}} < \varepsilon_{\perp} \approx 1.07 \). Solid and dotted curves in figure 8 are calculated from equation (13) and the second term of equation (13), respectively, for \( \alpha = 3\theta \). It is seen that the maxima of depolarization curves both for the bound (figure 8(a)) and the free electrons (the dotted line in figure 8(a)) are found in the region of positive values of the magnetic field. For a backscattering configuration this indicates that the signs of \( g \) and \( g_c \) are in full agreement with the result obtained with the use of specular configuration.

5.2.2. High energy detection (\( \alpha = 0 \)). At \( \varepsilon_{\text{det}} > 2 \) (when deformation splitting can be neglected) the angle \( \alpha \approx 0 \) and, according to equation (13), the Hanle curve must be symmetrical and insensitive to the sign of \( g \)-factor. Indeed, the experimental Hanle curve (open circles in figure 9), measured at \( \varepsilon_{\text{det}} = \varepsilon_2 = 2.03 \), is symmetric.

The experimental dependences \( \rho(B) \) presented above have been measured under high-energy excitation (\( \varepsilon_{\text{exc}} > 1 \)) and detection energy \( \varepsilon_{\text{det}} > 1 \) or \( \varepsilon_{\text{det}} < 1 \), when the angle \( \alpha \) remains practically invariable with varying \( \varepsilon_{\text{det}} \). Under these conditions the greatest absolute value of the angle \( \alpha \) and, respectively, the greatest asymmetry of the Hanle curve are observed in a specular configuration at \( \varepsilon_{\text{det}} > 1 \) (\( \alpha = 2\theta \)) and in a backscattering configuration at \( \varepsilon_{\text{det}} < 1 \) (\( \alpha = 3\theta \)). A greater asymmetry of the Hanle curve can be obtained in specular configuration under excitation and detection with only a light hole participating (\( \varepsilon_{\text{det}} < \varepsilon_{\text{exc}} < 1 \), when \( \alpha = -4\theta \) [27]. Note that the maximum asymmetry of the Hanle curve can be realized both in specular and backscattering configurations under high-energy excitation and detection energy \( \varepsilon_{\text{det}} = \varepsilon_{\perp} \), when in the absence of magnetic field the vectors \( \mathbf{S}_1 \) and \( \mathbf{S}_0 \) are perpendicular to each other (angle \( |\gamma - \beta| = 90^\circ \)) and \( \rho \propto \mathbf{S}_0 \mathbf{S}_1 = 0 \), while in a magnetic field \( \rho \propto \pm \mathbf{S}_0 || \mathbf{S}_1 | | \psi/(1 + \psi^2) \), where the sign \((\pm)\) before the right-hand side corresponds to the specular (backscattering) configuration. However, at \( \varepsilon_{\text{det}} = \varepsilon_{\perp} \) the modulus of \( \mathbf{S}_1 \) vector diminishes sharply and is near zero if the refraction angle does not exceed a few degrees (\( |S_1(\varepsilon_{\perp})| = 0 \) for \( \theta = 0 \)), amounting to \( \approx 0.1 \) only at angles \( \theta \) close to the maximum value (\( \theta_{\text{max}} = 16^\circ \) in GaAsN)

Experimental dependences \( \rho(B) \) measured in GaAsN consist of narrow and broad parts. The large amplitude of the narrow part bears witness to strong polarization of bound electrons, arising due to spin-dependent recombination. We have approximated the experimental curves \( \rho(B) \) by the sum of two Lorentzians, one of which describes the depolarization of bound electrons, and the other, that of free electrons. At the same time, we have shown in [18] that the polarizations of free and bound electrons are summed up additively \( \rho \propto S + S_c \) only in the limit of low polarization of bound electrons. If polarization of bound electrons is strong, it becomes necessary to solve a system of nonlinear equations, coupling polarizations of free and bound electrons [18]. As the preliminary analysis shows, in that case a substantially better fit of the experimental Hanle curve is obtained for the region of low magnetic fields, where the polarization of bound electrons dominates. The detailed theoretical description of the compound Hanle effect under an arbitrary polarization (including 100\% of bound electron) will be presented in a separate publication. Here we note that the shape of Hanle curves (in particular their asymmetry), obtained from the solution of a system of nonlinear equations does not differ qualitatively from the results of the approximation by two Lorentzians, which makes it possible to employ the latter for determination of the signs of \( g \) and \( g_c \).

6. Summary

In order to determine the sign of the electron \( g \)-factor in a semiconductor film, we have applied the method based on the analysis of the asymmetrical Hanle effect under oblique incidence of the exciting light onto the sample and detection of the photoluminescence at oblique emission. The relationship between the \( g \)-factor sign and direction of the asymmetrical shift of the Hanle curve is governed by the valence band structure, particularly, by its splitting due to an internal uniaxial strain in the film, and depends on the photoexcitation.

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4 See footnote 3.
and photodetection energies. In the present experimental and theoretical work we have extended the method to the case where the internal strain shifts the light-hole subband upward relative to the heavy-hole subband. The interband optical orientation of electron spins, circular polarization of photoluminescence, and shape of the Hanle curve have been calculated in dependence on the energies of excitation and detection as well as on the angles of incidence and secondary emission.

The experimental study has been performed on uniaxially compressed GaAs₁₋ₓNₓ epitaxial films. The observed Hanle effect is a result of the magnetic-field induced depolarization of the coupled system of spin-polarized free electrons and electrons bound on deep paramagnetic centers. The asymmetry of the resultant Hanle curve has been measured at room temperature under oblique incidence of the pump radiation and detection in the specular or backscattering configurations. Positive signs of the gyromagnetic g-factors of the free and bound electrons in the GaAs₀.₉₇₅N₀.₀₂₁ alloy have been deduced from comparison between experiment and theory.

Acknowledgments

We are grateful to the late B P Zakharchenya for stimulating interest in this work, and M M Glazov, K V Kavokin and V M Ustinov for fruitful discussions. Partial support by the Russian Foundation for Basic Research, grants of the Russian Academy of Sciences, and TARA project of the University of Tsukuba is acknowledged.

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