Optical orientation and the Hanle effect of neutral and negatively charged excitons in GaAs/$Al_xGa_{1-x}As$ quantum wells

R. I. Dzhioev,[1] V. L. Korenev,[1] B. P. Zakharchenya,[1] D. Gammon,[2] A. S. Bracker,[2] J. G. Tischler,[2] and D. S. Katzer[2]
[1]*A. F. Ioffe Physical Technical Institute, St. Petersburg, 194021 Russia*
[2]*Naval Research Laboratory, Washington DC 20375*

(Received 12 June 2002; revised manuscript received 23 August 2002; published 30 October 2002)

We report on the optical orientation of negatively charged excitons (trions) in [001] GaAs/$Al_xGa_{1-x}As$ quantum wells (QW's). The spin orientation of equilibrium electrons reveals itself in the polarized luminescence of both excitons and trions. The Hanle effect of the trions differs substantially from that of the neutral excitons. It is found experimentally that the electron-spin-relaxation time in a 14-nm QW exceeds 10 ns at $T=4.2$ K.

DOI: 10.1103/PhysRevB.66.153409 PACS number(s): 78.67.De, 78.67.−n

The potential for using the spin degree of freedom in functional devices (spintronics) has renewed interest in spin systems in semiconductors.[1] An important example is quantum information processing, where it is necessary to initialize, manipulate, and read out the state of quantum bits. The spin of an electron in a semiconductor heterostructure is a good candidate for a quantum bit,[2] because of its long spin-relaxation time.[3] At the same time there is a fundamental interest in quantum-confined systems and their excitations. In particular there has recently been interest in charged excitons (trions) in quantum wells and quantum dots.[4–7] The trion is interesting, in part, because it is an optically excited state of a single electron and could prove useful in the optical measurement of isolated electronic spins. For example, optically detected magnetic resonance has been used to measure the change in the equilibrium electron polarization in quantum wells with resonant microwave radiation through the change of luminescence polarization of excitons and trions.[8]

Here we show that spin polarized single electrons can be probed through the polarization of exciton and trion photoluminescence (PL) under optical orientation conditions. The PL polarization, however, arises not only from the spin polarization of the equilibrium electrons but also from that of the excitons and trions. Nevertheless, the polarization can be fully analyzed using the Hanle effect—the dependence of the PL polarization as a function of a transverse magnetic field (in the plane of the quantum well). The Hanle curves for exciton and trion PL lines under quasiresonant excitation show distinct shapes that can be understood through rate equations. This analysis allows us to resolve the contribution of the equilibrium electrons from that of the excited states (excitons and trions), providing good measures of the magnitude of the equilibrium electron polarization and spin relaxation. From these experiments we show that the spin-relaxation time of single electrons is longer than 10 ns in a 14-nm quantum-wwll, (QW), in agreement with the theory of spin relaxation by hyperfine interactions.

The optical orientation experiments were carried out on a sample with a semi-insulating GaAs substrate, on which a 500-nm-thick GaAs buffer and a 25-nm-thick AlAs barrier separating the substrate from the main structure were molecular beam epitaxy grown. Following this is a 100-nm-thick GaAs layer capped by a 25-nm-thick $Al_{0.3}Ga_{0.7}As$ barrier, above which a series of five quantum wells of different thickness separated by 25-nm barriers $Al_{0.3}Ga_{0.7}As$ were prepared. The sample, though nominally undoped, has a background doping level of order 10^{14} cm^{-3} *n* type from Hall measurements on test structures. The polarization of exciton and trion emission from a 14-nm QW has been studied. The sample was placed in a liquid-helium cryostat and pumped by a tunable Ti-sapphire laser with the circular polarization alternated in sign at a frequency of 26.61 kHz with a photoelastic quartz modulator. This permitted us to eliminate the effect of the lattice nuclear polarization on the optical orientation of the electrons.[1] The PL polarization was measured in the reflection geometry with a circular-polarization analyzer. The electronics provided measurement of the *effective* degree of circular polarization, $\rho=(I_+^+-I_+^-)/(I_+^++I_+^-)$, where I_+^+ and I_+^- are the intensities of the σ^+ PL component under the σ^+ and σ^- pumping, respectively.

The low-temperature PL spectrum of the 14-nm QW (Fig. 1) consists of a neutral exciton (1.534 eV) and a negatively charged trion (1.5327 eV) with an energy splitting corresponding to the trion binding energy $E_b=1.3$ meV.[7] The optical orientation of the exciton and trion is analyzed by the dependence of the degree of the circular polarization of the PL on a transverse magnetic field in the Voigt geometry (Hanle effect) for the X and X^- lines. Under *nonresonant* excitation of the QW, generation of free electrons and holes takes place. Figure 2 shows the magnetic depolarization of

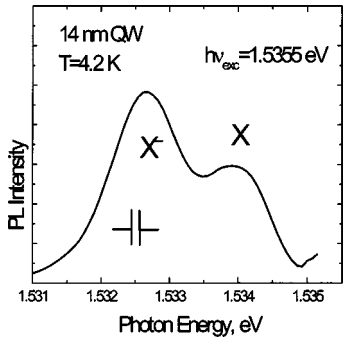

FIG. 1. Spectrum of the low-temperature ($T=4.2$ K) photoluminescence of a 14-nm quantum well under quasiresonant creation of excitons ($h\nu_{exc}=1.5355$ eV).

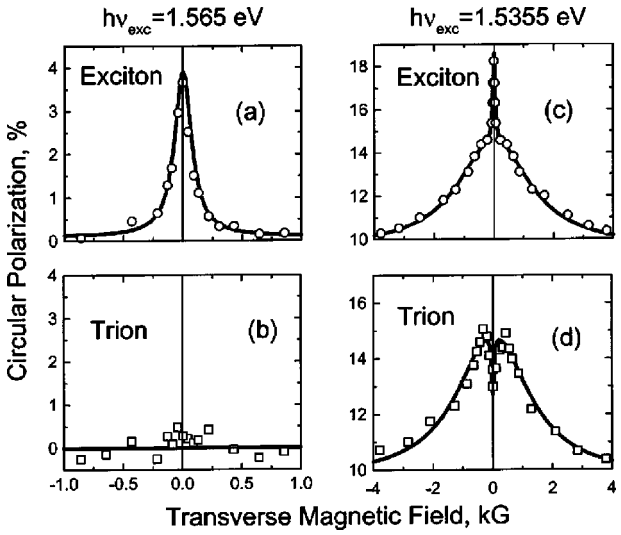

FIG. 2. Hanle effect for exciton [circles (a)] and trion [squares (b)] under nonresonant excitation of electron-hole pairs ($h\nu_{\mathrm{exc}}=1.565$ eV). The solid curve is a Lorentzian with $B_{1/2}=90\ G$. The Hanle effect on an exciton PL line [circles (c)] and negatively charged exciton [squares (d)] under quasiresonant creation of excitons ($h\nu_{\mathrm{exc}}=1.5355$ eV). Solid curves are calculated according to Eqs. (3)–(5).

exciton (a) and trion PL (b) under excitation with a laser energy $h\nu_{\mathrm{exc}}=1.565$ eV, inducing transitions from both heavy- and light-hole subbands. One can see that the polarization of exciton PL (circles) decreases to zero even at a low magnetic field (~90 G). The trion line is practically unpolarized (squares) in this field region (The absence of optical orientation of negatively charged excitons under nonresonant excitation was previously reported).[9]

In sharp contrast, the Hanle effect changes radically under *quasiresonant* creation of excitons in the QW. Figure 2(c) shows that depolarization of the exciton emission has two regions. In the low-field region (~30 G) the polarization decreases sharply from 18% to 15%. In the high-field region (~1.5 kG) it decreases more slowly to 10%. The Hanle effect of the trion [Fig. 2(d)] also has two regions, but the polarization grows initially (from 13% to 15%), and then it decreases as for the exciton.[10]

We start the discussion with the nonresonant excitation of electrons and holes [Figs. 2(a) and 2(b)]. This case is analogous to the usual optical orientation of electrons in bulk *n*-GaAs.[1] The right-hand polarized light creates the electrons with spin projection $|-\frac{1}{2}\rangle$ and heavy holes with an angular momentum projection $|+\frac{3}{2}\rangle$ onto the [001] axis, as well as electrons in the $|+\frac{1}{2}\rangle$ state and light holes with a momentum projection $|+\frac{1}{2}\rangle$. The transition probabilities from heavy- and light-hole subbands go as 3:1.[1] In wide quantum wells (>10 nm) free holes lose their spin quickly, whereas electrons conserve their polarization prior to recombination. At low temperatures electrons bind with holes into excitons in states with total momentum $|\pm 1\rangle=|\pm\frac{3}{2};\mp\frac{1}{2}\rangle$ (optically active or bright excitons) and $|\pm 2\rangle=|\pm\frac{3}{2},\pm\frac{1}{2}\rangle$ (optically forbidden or dark excitons). The degree of polarization of bright and dark excitons is determined by that of electrons at the moment of exciton formation. The magnetic field transverse to the [001] axis induces the Larmor precession of electron spin that decreases the electron polarization, and therefore, that of the excitons in steady state conditions.[11] This is in agreement with experiment [Fig. 2(a)], which shows the depolarization of the exciton emission.

The situation differs for trions. For example, a single electron with spin $|+\frac{1}{2}\rangle$ and a bright exciton with momentum $|+1\rangle=|+\frac{3}{2},-\frac{1}{2}\rangle$ form a trion in the ground state $|+\frac{3}{2}\rangle$ (a singlet with respect to the electron spins). However, the fast hole spin relaxation creates the same number of dark excitons with a momentum $|-2\rangle=|-\frac{3}{2},-\frac{1}{2}\rangle$. Their binding with $|+\frac{1}{2}\rangle$ electrons creates trions with a momentum $|-\frac{3}{2}\rangle$ with the same probability. Therefore, the polarization of the trion should be zero. In the same way, trion formation from single electrons with spin $|-\frac{1}{2}\rangle$ gives the same answer. This agrees well with experiment [Fig. 2(b)], which shows the absence of trion polarization under nonresonant excitation.

Under *quasiresonant* creation of excitons by circularly polarized light, the polarization behavior of both the exciton and trion changes substantially [Figs. 2(c) and 2(d)]. Light creates excitons rather than uncoupled electrons and holes. In this case the magnetic depolarization curve of the exciton [Fig. 2(c)] consists not only of the low-field ''electronic'' part presented in Fig. 2(a) but also a high-field region. The latter result suggests that resonantly excited excitons now contribute to the Hanle curves, but require a higher magnetic field to depolarize.[12] Interesting behavior is also found for the trion polarization. Comparison of Figs. 2(b) and 2(d) shows that an optical orientation of trions is now obtained under quasiresonant excitation conditions. These results suggest an incomplete spin relaxation of holes during the exciton and trion lifetimes. Additional evidence for hole spin orientation is the ''incomplete'' depolarization [Figs. 2(c) and 2(d)]–the PL polarization saturates at the 10% level instead of going to zero. The zero-field hole polarization is not affected by magnetic field because the transverse hole *g* factor is close to zero. Our conclusion about incomplete spin relaxation of holes agrees with Ref. 13 where hole spin relaxation was suppressed under resonant creation of excitons. Under these conditions bright excitons preferentially form the trion because their number exceeds the number of dark excitons.

The fact that the Hanle curve of the trion contains both ''electronic'' and ''excitonic'' regions is not surprising because the trions are formed from single electrons and excitons. It is more surprising that the single electron polarization contributes to the polarization of the exciton and trion (low-field region) in an opposite manner: exciton polarization decreases while trion polarization increases at low magnetic fields.

To understand the origin of the qualitative differences in line shapes of the Hanle curves under *quasiresonant* creation of excitons, let us consider the simplest model of optical orientation of excitons and trions. Under quasiresonant excitation bright excitons are mainly created, and, as we discussed above, the optical orientation of the trion unambiguously proves the suppression of hole spin relaxation. Therefore, trions are preferentially formed by bright excitons

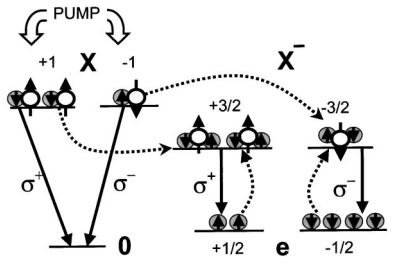

FIG. 3. An illustration of the optical pumping process under resonant excitation conditions. In this case the hole spin relaxation is slowed down, and bright states $|\pm 1\rangle$ are populated preferentially. The bright excitons $|+1\rangle$ ($|-1\rangle$) combine with the $|+\frac{1}{2}\rangle$ ($|-\frac{1}{2}\rangle$) ground state electrons to form the $|+\frac{3}{2}\rangle$ ($|-\frac{3}{2}\rangle$) trions. After the $\sigma^+(\sigma^-)$ radiative transitions of the $|+\frac{3}{2}\rangle$ ($|-\frac{3}{2}\rangle$) trions, the ground-state $|+\frac{1}{2}\rangle$ ($|-\frac{1}{2}\rangle$) electrons are left behind. The radiative transitions of $|+1\rangle$ ($|-1\rangle$) excitons into the ground state $|0\rangle$ with $\sigma^+(\sigma^-)$ polarizations are also shown.

that bind to an additional electron. Figure 3 reflects this process. To be more specific, bright excitons with a total momentum $|\pm 1\rangle$ may recombine radiatively, emitting circularly polarized $\sigma^{\pm}$ photons. Alternatively, they may bind with single electrons, forming singlet state trions with a total momentum $|\pm\frac{3}{2}\rangle$. The rate of this process is proportional to the number of single electrons with spin antiparallel to the spin of the electron within the exciton. Trions emit $\sigma^{\pm}$ light from the $|\pm\frac{3}{2}\rangle$ states, leaving behind the electron in the $|\pm\frac{1}{2}\rangle$ state. The processes enumerated above are described below by the rate equations for concentrations of bright excitons, $n_{\pm 1}$, and trions, $n_{\pm 3/2}$.

At zero magnetic field the kinetic equations for exciton concentrations $n_{\pm 1}$ are

$$\frac{dn_{\pm 1}}{dt} = g_{\pm 1} - \frac{n_{\pm 1}}{\tau_b} - \frac{n_{\pm}(1 \pm P_e)}{\tau_{bT}}. \tag{1}$$

The first term on the right hand side of Eq. (1) describes the generation of bright excitons into the $|\pm 1\rangle$ states with a rate $g_{\pm 1}$. The second term takes into account the radiative recombination with a characteristic time τ_b. The third term describes the spin-dependent binding of $|\pm 1\rangle$ excitons into trions with a total momentum $|\pm\frac{3}{2}\rangle = |\pm 1; \pm\frac{1}{2}\rangle$ during a time $\tau_{bT}/(1 \pm P_e)$. The single electron polarization $P_e = (n_{+1/2} - n_{-1/2})/(n_{+1/2} + n_{-1/2})$ is determined by electron concentrations $n_{\pm 1/2}$ with spins along (opposite to) the z axis.

The kinetic equations for trion concentrations $n_{\pm 3/2}$ are

$$\frac{dn_{\pm 3/2}}{dt} = \frac{n_{\pm 1}(1 \pm P_e)}{\tau_{bT}} - \frac{n_{\pm 3/2}}{\tau_T}. \tag{2}$$

The first term on the right-hand side of Eq. (2) describes the trion formation, and the second describes the trion recombination with a time τ_T. Note that in Eqs. (1) and (2) we have taken into account the spin-dependent formation rate of the trion. As a result, the steady-state polarizations of bright excitons (P_1) and trions (P_T) contain contributions from spin-polarized single electrons:

$$\text{(a)} \quad P_1 \equiv \frac{n_1 - n_{-1}}{n_1 + n_{-1}} \approx P_{\text{in}} - f P_e,$$

$$\text{(b)} \quad P_T \equiv \frac{n_{3/2} - n_{-3/2}}{n_{3/2} + n_{-3/2}} \approx P_1 + P_e. \tag{3}$$

Here the initial polarization of excitons $P_{\text{in}} = (g_1 - g_{-1})/(g_1 + g_{-1})$, the exciton lifetime $\tau = (1/\tau_b + 1/\tau_{bT})^{-1}$, and the factor $f = \tau/\tau_{bT} = \tau_b/(\tau_b + \tau_{bT})$ characterize the efficiency of trion formation with respect to the exciton radiative recombination. In these conditions the ratio of intensities of the exciton $I_X = (n_1 + n_{-1})/\tau_b$ and trion $I_T = (n_{3/2} + n_{-3/2})/\tau_T$ PL lines is determined solely by the factor f: $I_X/I_T = 1/f - 1$. Equation (3) is deduced under the condition $P_1 P_e \ll 1$. In the absence of single-electron polarization ($P_e = 0$) the degree of exciton polarization P_1 [Eq. 3(a)] is determined as usual[1] by direct optical pumping, and the polarizations of the excitons and trions coincide. Once electrons are polarized, an additional term appears ($-fP_e$), resulting from the spin-dependent formation of the polarized electron and exciton into the trion: the excitons having angular momentum along the electron polarization (P_e) form a trion more quickly than those with the opposite projection. This tends to enhance the exciton concentration with polarization that is opposite to the single electron polarization [because of the minus sign for fP_e in Eq. (3a)]. In contrast, trion formation is favored by parallel orientations of exciton and electron momentum projections [the positive sign of P_e in Eq. (3b)].

The spin polarization of equilibrium electrons is the result of various mechanisms. For example, there can be spin-flip scattering of excitons by single electrons, as well as the accumulation of electron mean spin as a result of the usual optical pumping process.[1] The excitation by σ^+ light creates excitons in a $|+1\rangle = |+\frac{3}{2}, -\frac{1}{2}\rangle$, state, where the hole polarization is positive when the electron polarization in the exciton is negative. The polarization of equilibrium (single) electrons will be negative for both of the mechanisms considered above. Therefore, the single electron polarization increases the zero-field exciton polarization and decreases the trion polarization [compare Eqs. (3a) and (3b)] in agreement with the experimental results [Figs. 2(c) and 2(d)].

A magnetic field in the Voigt geometry depolarizes both the single electrons and the electron in the exciton as a result of Larmor precession. Single electrons are depolarized at low fields. Therefore, the exciton polarization goes down while that of the trion goes up, in good agreement with experiment [Figs. 2(c) and 2(d)]. The effect of the external magnetic field on the mean spin of single electrons can be taken into account if we replace P_e in Eq. (3) by

$$P_e(B) = \frac{P_e}{1 + (B/B_{1/2})^2}, \tag{4}$$

where the halfwidth $B_{1/2} = \hbar/\mu_B g_e T_s$ of the Hanle curve determines the spin lifetime of the single electrons, T_s.[1] The in-plane electron g factor is $g_e = -0.26$ for the 14-nm QW.[14]

Larger magnetic fields ($\sim$1 kG) depolarize the excitons. As a result, both exciton and trion polarizations go down further. The Hanle effect of excitons has been considered theoretically[12] and the polarization of bright excitons is

$$P_1(B) = P^\infty + \frac{P_1 - P^\infty}{1 + (B/B^X_{1/2})^2}$$

$$\approx P^\infty + \frac{P_{\rm in} - P^\infty}{1 + (B/B^X_{1/2})^2} - \frac{f P_e}{1 + (B/B_{1/2})^2}. \qquad (5)$$

Here the polarization P^∞ takes into account the ''incomplete'' Hanle depolarization; $P_1 = P_{\rm in} - f P_e(B)$ is determined by Eq. (3a) where $P_e(B)$ describes the Larmor precession of single electron spin. To deduce Eq. (5) we took into account that the halfwidth of the magnetic depolarization curve for the excitons, $B^X_{1/2} \gg B_{1/2}$. It is determined by a number of parameters,[12] and will be considered as a fitting parameter of the simple model. The magnetic field dependence of the trion polarization is determined by Eq. (3b), where the dependencies, $P_1(B)$ and $P_e(B)$, are given by Eqs. (5) and (4), respectively. The degree of PL circular polarization of excitons, $\rho^X_c(B) = P_1(B)$, and trions, $\rho^T_c(B) = P_T(B)$, is measured experimentally.

The solid lines in Figs. 2(c) (exciton) and 2(d) (trion) are fits to Eqs. (3)–(5) with the parameters $B_{1/2} = 35$ G, $B^X_{1/2} = 1.45$ kG, $P_{\rm in} = 5\%$, $P^\infty = 10\%$, and $P_e = -6\%$. The parameter $f = \tau_b/(\tau_b + \tau_{bT}) = \frac{2}{3}$ implies that the trion formation time is two times shorter than the radiative lifetime, and that $I_X/I_T = 1/f - 1 = \frac{1}{2}$. This is in agreement with the PL spectrum, where the trion intensity is 1.5 times greater than the exciton intensity. The halfwidth $B_{1/2} = 35$ G gives a spin lifetime of $T_s = 10$ ns for the single electrons. This time is determined by the shortest of the lifetime and spin relaxation time,[1] and thus we can conclude that $\tau_s > 10$ ns.

The electron-spin relaxation likely arises from hyperfine interaction with nuclear spin. At low temperatures, the electrons are localized by potential fluctuations (the QW thickness and/or charged impurity concentration fluctuations). For a dilute and localized electron ensemble, the dominant spin relaxation mechanism is expected to be static fluctuations in the nuclear field distribution over different electrons. The characteristic value for this lifetime distribution is expected to be of order 5 ns,[15] in agreement with the result. It should be noted that a previous pump-probe study[16] of [001]-GaAs/Al_xGa_{1-x}As quantum wells revealed a relatively fast electron spin relaxation (~100 ps). The difference may be connected with a higher excitation density in the pump-probe experiment that introduces additional spin relaxation mechanisms (for example, electron-hole exchange and Dyakonov-Perel relaxation).[1] We have also measured the spin-relaxation time of electrons in quantum wells of different thicknesses. The τ_s values are of the order of nanoseconds for QW thicknesses in the range 4–14 nm, which again implies that spin relaxation arises from the hyperfine channel over this entire well width range. The details of these results will be published elsewhere.

In conclusion, we have presented an optical orientation study of neutral excitons, trions, and single electrons in an *n*-type GaAs QW. The polarization of single electrons is probed through the Hanle depolarization of the PL of the exciton and trion. Although the trion PL is unpolarized under nonresonant excitation, it becomes polarized under quasiresonant excitation and displays a sharp and distinctive Hanle lineshape that can be understood from a simple rate equation model. The linewidth of this feature gives a relatively long electron spin relaxation time of 10 ns in a 14-nm GaAs QW, that is in agreement with spin dephasing arising from hyperfine interactions.

We thank I.A. Merkulov for helpful discussions. Partial support of the Russian Basic Research Foundation (Grant Nos. 00-02-16991 and 01-02-17906), CRDF (Grant No. RP1-2252), Young Scientists Grant No. 36 of RAS, DARPA/SPINS, NSA/ARDA/ARO and ONR is gratefully acknowledged.

[1] *Optical Orientation. Modern Problems in Condensed Matter*, edited by F. Meier and B. P. Zakharchenya (North-Holland, Amsterdam, 1984).

[2] G. Burkard *et al.*, Phys. Rev. B **59**, 2070 (1999).

[3] R. I. Dzhioev *et al.*, Fiz. Tverd. Tela (St. Petersburg) **39**, 1975 (1997) [Phys. Solid State **39**, 1765 (1997)]; J. M. Kikkawa and D. D. Awschalom, Phys. Rev. Lett. **80**, 4313 (1998); Z. Wilamowski *et al.*, Physica B **284**, 1926 (2000); R. I. Dzhioev *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. [JETP Lett. **74**, 182 (2001)]; R. I. Dzhioev *et al.*, Phys. Rev. Lett. **88**, 256 801 (2002).

[4] K. Kheng *et al.*, Phys. Rev. Lett. **71**, 1752 (1993).

[5] G. Finkelstein *et al.*, Phys. Rev. Lett. **74**, 976 (1995).

[6] A. J. Shields *et al.*, Phys. Rev. B **51**, 18 049 (1995).

[7] J. G. Tischler *et al.*, Phys. Rev. B **66**, 081310 (2002).

[8] C. Y. Hu *et al.*, Phys. Rev. B **58**, R1766 (1998).

[9] O. V. Volkov *et al.*, Pis'ma Zh. Eksp. Teor. Fiz. **71**, 465 (2000) [JETP Lett. **71**, 465 (2000)].

[10] An M-shaped Hanle curve has been found earlier in bulk solid solution $Al_xGa_{1-x}As$ due to the Overhauser effect (Ref. 1), as well as in semimagnetic semiconductors [Yu. G. Kusrayev *et al.*, in *Proceedings of the 23rd International Conference on the Physics of Semiconductors, Berlin, Germany* (World Scientific, Singapore, 1996), p. 2459] that is due to a large number of Mn atoms. Here both the Overhauser effect and Mn atoms are absent.

[11] The measurement of the halfwidth of the Hanle curve in Fig. 2 enables us to determine a spin lifetime $T_s = 5$ ns (Ref. 1). The spin lifetime is determined by the shortest of the electron lifetimes and spin relaxation times. Such a long time (5 ns) cannot be due to excitons which have a lifetime one order of magnitude shorter.

[12] The heavy-hole exciton spin-level structure in a quantum well is close to that of $\Gamma_7 \times \Gamma_9$ exciton in a bulk hexagonal crystal where the Hanle effect has been studied theoretically: G. L. Bir and G. E. Pikus, Pis'ma Zh. Eksp. Teor. Fiz. **15**, 730 (1972) [JETP Lett. **15**, 516 (1972)]; Zh. Eksp. Teor. Fiz. **64**, 2210 (1973) [Sov. Phys. JETP **37**, 116 (1973)].

[13] T. Amand *et al.*, Phys. Rev. Lett. **78**, 1355 (1997); M. Dyakonov *et al.*, Phys. Rev. B **56**, 10412 (1997).

[14] E. L. Ivchenko and A. A. Kiselev, FTP **26** 1471 (1994).

[15] I. A. Merkulov *et al.*, Phys. Rev. B **65**, 205309 (2002).

[16] Y. Ohno *et al.*, Phys. Rev. Lett. **83**, 4196 (1999).