

Spin dynamics in the charged InP quantum dots

I. V. Ignatiev^{1,2,3}, I. Ya. Gerlovin^{3,4}, S. Yu. Verbin^{1,3}, I. A. Yugova,
and Y. Masumoto¹

1) Institute of Physics, University of Tsukuba, Tsukuba 305-8571, Japan

2) Venture Business Laboratory, University of Tsukuba, Tsukuba
305-8571, Japan

3) Institute of Physics, St.-Petersburg State University, St.-Petersburg,
198904, Russia

4) Vavilov State Optical Institute, St.-Petersburg, 190034, Russia

Abstract. Kinetics of polarized photoluminescence (PL) of the InP quantum dots with variable number of resident electrons is studied. A long-lived component of the degree of circular polarisation of the PL is found to be present for charged quantum dots and absent for neutral ones. Behaviour of the long-lived polarisation in longitudinal and transverse magnetic field is studied. Experimental data obtained allowed us to conclude that the long-lived polarisation is related to spin orientation in the quantum dots containing odd number of carriers. We found that the irreversible spin relaxation time in these dots substantially exceeds 1 ns.

1. Introduction

In recent years, a considerable attention has been focused on spin dynamics of carriers and excitons in semiconductor heterostructures. Particular interest was attracted to prospects of development of elements of spin memory and spin transistors based on these structures [1]. Quantum dots (QDs) structures have been suggested as the most promising for this purpose. It is supposed that localisation of carriers should eliminate the impact of spatial motion on their spin state, and, for this reason, can greatly reduce the spin relaxation rate. A recent theory [2] predicts very low spin scattering rates of the order of 10^3 - 10^4 s⁻¹ for semiconductor QDs. In a neutral QD, nonzero spin may belong only to an electron-hole pair. Therefore, the spin lifetime proves to be limited by the recombination process and does not exceed 1 ns. This limitation is removed in the charged QDs, where the majority (equilibrium) carriers are permanently present. From this point of view, spin dynamics in the charged QDs attracts considerable interest.

Our work is aimed at investigation of spin relaxation in controllably charged InP QDs. We have found that spin dynamics of the neutral and charged quantum dots is essentially different. The spin polarisation decay time in neutral dots is of the order of tens of picosecond, whereas, in the presence of resident charges, the spin lifetime can exceed 1 ns. In order to identify the mechanisms responsible for formation of the long-lived component of the spin polarisation, we studied spin dynamics of the charged QDs

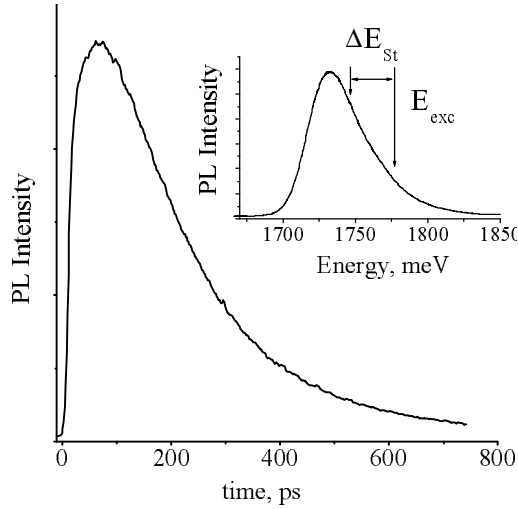


Fig. 1. A typical time dependence of the unpolarized PL of the InP QDs. In the inset it is shown the PL spectrum measured at nonresonance excitation. Arrows indicate spectral points of quasi-resonance excitation and detection of the PL, as well as the Stokes shift between them

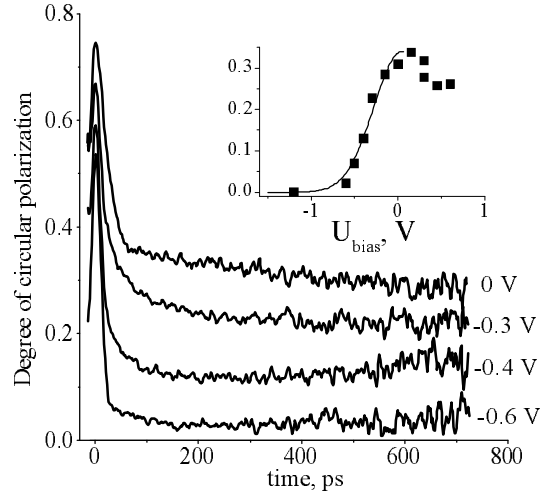


Fig. 2. Degree of circular polarisation of the PL at different U_{bias} shown against each curve. Inset: bias dependence of amplitude of the long-lived polarisation component (squares). Solid line is the fit by a Gaussian. $E_{\text{St}} = 14$ meV.

as a function of the magnitude and orientation of the external magnetic field. Here we present the main results of this study.

2. Experimental details

We studied the photoluminescence (PL) of a heterostructure with a single layer of InP QDs sandwiched between the $\text{In}_{0.5}\text{Ga}_{0.5}\text{P}$ barrier layers. The sample was grown by the gas source molecular beam epitaxy on an n^+ GaAs substrate. The areal density of the QDs was about 10^{10} cm^{-2} . The average base diameter of the QDs was about 40 nm and the height about 5 nm. In order to control the charge of the QDs, the sample was provided with a semitransparent indium tin oxide Schottky contact on the top surface and with an ohmic contact on the bottom. The thickness of the undoped epitaxial layers, to which the electric voltage was applied, was about 0.5 μm .

In kinetic studies, the photoluminescence was excited by a 3-ps pulses of a Ti:sapphire laser within the PL band (quasi-resonant excitation) as shown in the inset of Fig. 1. The PL kinetics was measured with a time resolution of 6 ps using a 0.25 m subtractive double monochromator and a streak camera. The measurements were made in a cryostat with a superconducting magnet. The design of the cryostat allowed us to excite the sample and to detect its emission either along the magnetic field direction (the Faraday configuration) or across the field (the Voigt configuration). The PL was detected in the backscattering geometry.

3. Circular polarisation in the absence of the magnetic field

A key result of the present work is detection of the long-lived component in the PL polarisation decay. Figure 2 demonstrates kinetics of the degree of circular polarisation of the PL measured at different biases.

The degree of polarisation was calculated in a standard manner:

$$\rho_{\text{circ}}=(I^+ - I)/(I^+ + I), \quad (1)$$

where I^+ and I are the PL intensities in the co- and counter-polarized light, respectively.

At zero and positive biases, the decay of the degree of polarisation contains a long lived polarisation component, which remains virtually constant during the PL lifetime of about 300 ps (see Fig. 1). The amplitude of this component, ρ_{slow} , lies in the range of tens of percent for small Stokes shift (about 15 meV). The value, ρ_{slow} , rapidly drops with increasing negative voltage and vanishes at $U_{\text{bias}} < -0.5$ V.

It has been shown in Refs. [3,4] that the InP QDs in the structure under study contain resident carriers (electrons) that arrived from the doped substrate. The negative voltage removes these electrons, so that the QDs become neutral at $U_{\text{bias}} < -0.5$ V. Therefore, one can conclude that the long-lived circular polarisation of the PL is observed only for the QDs containing resident electrons.

As has been shown in Ref. [5], the main reason for the fast decay of the degree of circular polarisation in the *neutral* InP QDs is related to the reversible dephasing resulted from a large spread of anisotropic exchange splittings in the ensemble of the QDs. The presence of the long-lived spin polarisation in the *negatively charged* QDs indicates that the anisotropic component of the exchange coupling, in these dots, is, to a considerable extent, suppressed.

The reason for this suppression in QDs containing *odd* numbers of carriers can be readily explained by the fact that such QDs have a half-integer spin and the anisotropic exchange does not split the spin states due to the Kramers theorem [4,6]. In the quantum dots with *even* number of carriers, such a mechanism cannot be realized, and we can suggest that these dots, like the neutral ones, should show a fast decay of the degree of polarisation. Indeed, as can be seen from Fig. 2, the decay of circular polarisation of the charged dots displays, with an approximately equal weight, both the fast and slow components. In the framework of the model proposed, we can suggest that the dots with even and odd number of carriers are responsible, respectively, for the fast and slow components.

The analogy, noted above, between spin dynamics of neutral dots and of charged dots with even number of carriers is, however, far from complete. The difference between them is revealed distinctly in kinetics of circular polarisation of the PL in the longitudinal magnetic field discussed in the next section.

4. Circular polarisation in the longitudinal magnetic field

A characteristic feature of the kinetics of the *charged* dots in the longitudinal magnetic field is its dependence on the magnetic field direction for a specified sign of circular polarisation of the exciting light. For one field direction, the amplitude of the slow component increases with increasing magnetic field strength, whereas for the opposite direction, it remains practically constant or even slightly decreases (Fig. 3). Note that the difference between the values of the degree of polarisation for two directions of the field increases in time with the characteristic constant τ of about 150 ps.

Since we suppose that PL of the QDs with *odd* number of carriers is completely polarised, the increase of the degree of polarisation in longitudinal magnetic field can be caused only by changes of spin states of the QDs with *even* number of carriers. These changes of the spin states can be naturally attributed to the temperature redistribution of populations of the fine-structure components split by the magnetic field. The measured value of τ should be considered, in this case, as the time of establishment of thermal

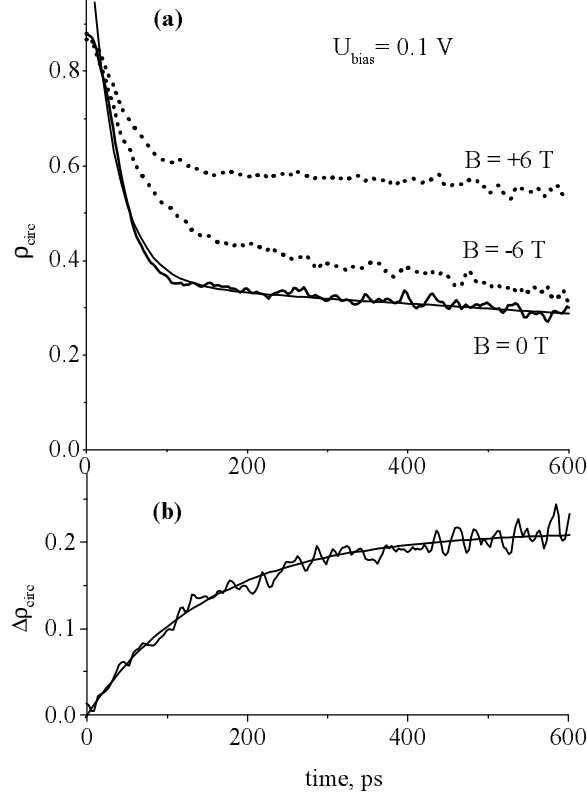


Fig. 3. (a) Degree of circular polarisation of the PL, ρ_{circ} , for *charged* InP quantum dots at zero magnetic field and $B = \pm 6$ T. Smooth solid curve is fit by two-exponential function: $\rho_{\text{circ}} = \rho_1 \exp(-t/\tau_1) + \rho_2 \exp(-t/\tau_2)$ with $\tau_1 = 33$ ps, $\tau_2 = 2900$ ps. (b) Difference of the circular polarisations, $\Delta\rho_{\text{circ}} = \rho_{\text{circ}}(+6\text{T}) - \rho_{\text{circ}}(-6\text{T})$. Smooth solid curve is the fit by function: $\Delta\rho_{\text{circ}} = \rho_1 (1 - \exp(-t/\tau))$ with $\tau = 150$ ps.

equilibrium in the spin system. By comparing the data of Figs. 2 and 3, one can readily notice that the rate of establishment of equilibrium is much smaller than the reversible dephasing rate in the dots with even number of carriers.

The polarisation dynamics, in these dots, cannot be elementary. Specifically, first, the dephasing rapidly destroys the light-induced polarisation and only after that the thermal equilibrium between the spin sublevels starts being established, which is observed experimentally (see Fig. 3). Under the temperature equilibrium, the PL polarisation is determined only by the order of arrangement of the Zeeman sublevels, i.e., by the magnetic field direction. Thus, to explain the experimental data, we have to assume that in the QDs with even number of electrons the irreversible relaxation processes occur that lead to establishment of thermal equilibrium in the spin subsystem during the time of the order of 150 ps.

As was already mentioned above, we ascribe the long-lived polarisation observed in the absence of the magnetic field, to the spin orientation in the QDs with *odd* number of carriers. The irreversible phase relaxation between spin sublevels of these dots should have led to a decay of the degree of circular polarisation with the same characteristic time $\tau = 150$ ps. However, as can be seen from Fig. 2, the slow polarisation component of PL of the charged dots virtually does not decay during the measuring time (1000 ps).

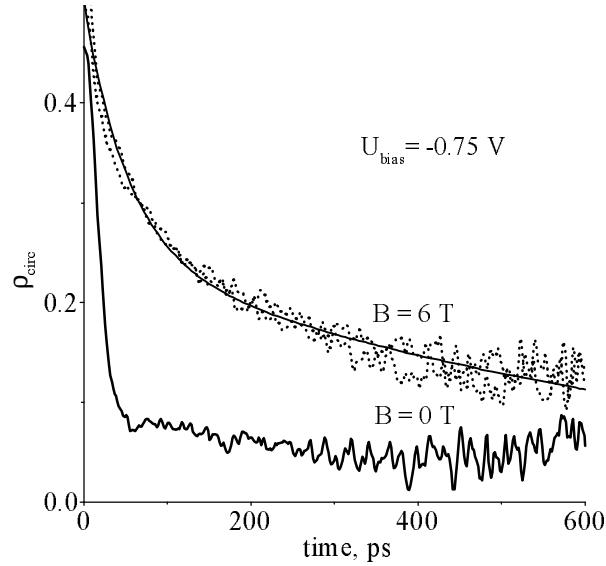


Fig. 4. Degree of circular polarisation of the PL, ρ_{circ} , for *neutral* InP quantum dots at zero magnetic field and $B = \pm 6$ T. Note that the polarisation kinetics at $B = +6$ T is not distinguished from that at $B = -6$ T. Smooth solid curve is the fit by two-exponential function with $\tau_1 = 50$ ps, $\tau_2 = 800$ ps.

We can conclude from this that the processes of irreversible dephasing in QDs with *odd* number of carriers are suppressed.

For the *neutral* QDs, the effect of longitudinal magnetic field on the PL kinetics is revealed in appearance of a slowly varying circularly polarised component, whose amplitude increases with the field strength (Fig. 4). An analysis shows that the slow component arises in magnetic fields for which the Zeeman splitting exceeds the anisotropic one. As a result, the spread of the exchange energy values does not cause the reversible dephasing of the spin states. The structure under study is characterised by a large spread of Zeeman splittings [7]. This is why the number of dots where the Zeeman splitting exceeds the anisotropic one increases with increasing field. This fact accounts for a relative growth of the slow component in the decay of the degree of polarisation.

It is important that the PL kinetics of neutral QDs, unlike that of the charged ones, does not show any appreciable dependence of the slow component amplitude on direction of the magnetic field (see Fig. 4). This means that the irreversible spin relaxation rate in neutral dots is low, and lifetime of the electron-hole pair appears to be too small for the temperature equilibrium between the Zeeman components to be established.

5. The kinetics in the transverse magnetic field

In the transverse magnetic field, the kinetics of the polarised PL of the charged and neutral dots proves to be strongly different also. As was shown previously [7], in the presence of a transverse magnetic field, circularly polarised PL of the neutral dots reveals pronounced oscillations. The decay time of the oscillations is about 100 ps, with the oscillation frequency linearly growing with the field strength. It was shown that these oscillations are related to quantum beats between the states of the bright and dark excitonic doublets, mixed by the transverse magnetic field. The Zeeman splitting of these components is governed by transverse component of the electron g -factor, which is equal, for the structure under study, to $g_{e,x} = 1.43$.

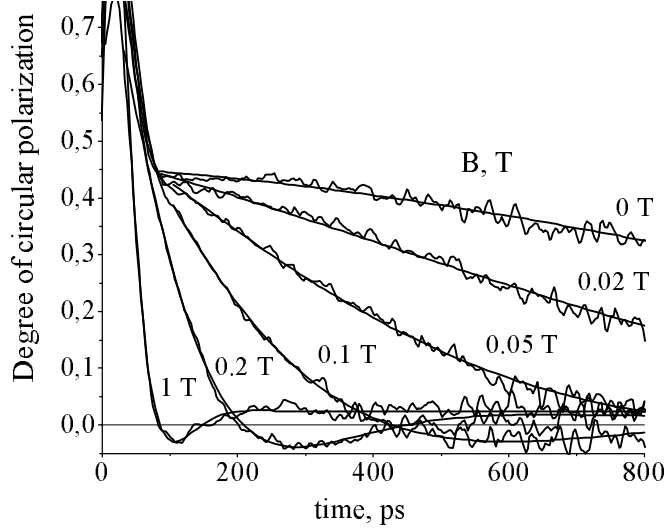


Fig. 5. Degree of circular polarisation for the charged QDs in transverse magnetic field indicated against each curve. $\Delta E_{St} = 15$ meV, $U_{bias} = 0$ V.

For the charged QDs, the kinetics of the degree of circular polarisation in a transverse magnetic field, is shown in Fig. 5. As it was pointed above, in the absence of the magnetic field, a slow polarisation component with the initial amplitude above 40% is observed. With the magnetic field on, the polarisation decay time sharply decreases, and at $B > 0.1$ T the polarisation decay becomes nonmonotonic. This behaviour may be regarded as related to the rapidly damping quantum beats.

The time dependence of the long-lived polarisation component can be well described by the expression:

$$\rho(t) = \rho_0 \exp(-t/\tau) \cos(\omega t). \quad (2)$$

The beat frequency ω determined in this way increases with the magnetic field and the decay time τ decreases.

The suppression of the circular polarisation in a transverse magnetic field is usually referred to as the Hanle effect [8]. This effect is related to precession of the spins of carriers around the magnetic field direction, which gives rise to quantum beats, as was repeatedly observed in quantum wells. In the case of free carriers, the electron and hole spins can precess independently [9,10]. The beat frequency is governed by the splitting of the electron and hole levels in the transverse magnetic field. For the hole spin precession, this frequency is small because the transverse component of the g -factor for the heavy hole is much smaller than for the electron, $g_{hh,x} \ll g_{e,x}$

The frequency of the beats corresponds to the g -factor of about 0.25 in the magnetic field $B = 1$ T. This value is much smaller than the transverse component of the electron g -factor ($g_{e,x} = 1.43$) and, at the same time, exceeds the transverse component of the hole g -factor ($g_{hh,x} \sim 0.1$), estimated for the neutral QDs in Ref. [7]. Nevertheless, we believe that the beats observed are related to the hole spin precession. This is supported by the fast decay of the beats that may be caused by a large spread of the hole g -factor. As was established in Ref. [7], the spread of the longitudinal component of the hole g -factor in the structure under study is large and virtually coincides with its mean value. The reasons for the discrepancy between the value of the g -factor given above ($g = 0.25$) and the value of transverse component of the hole g -factor in the neutral dots calls for further investigation.

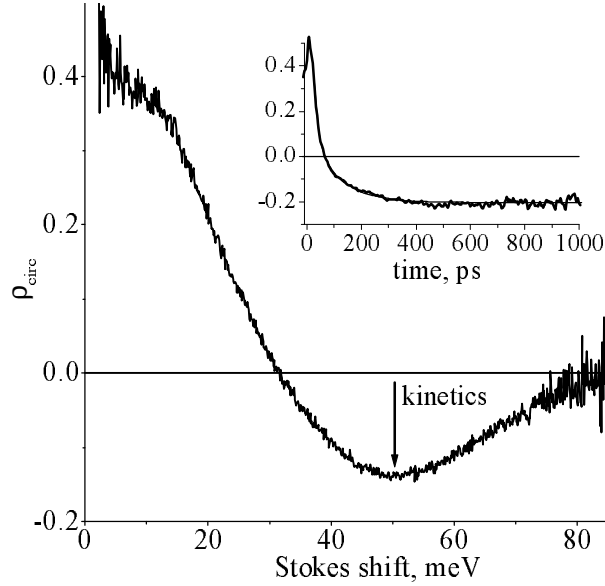


Fig. 6. Spectral dependence of degree of the circular polarisation for the charged InP QDs. Inset: PL kinetics measured at Stokes shift 50 meV. Smooth solid line is the fit by a two-exponential function with $\tau_1 = 30$ ps, $\tau_2 = 100$ ps.

6. Spectral dependence of kinetics of circular polarisation

A specific feature of the circularly polarised PL of the charged QDs is a change of the sign of the degree of polarisation with increasing Stokes shift as shown in Fig. 6. As is seen from the figure, for the Stokes shift exceeding 30 meV, the degree of circular polarisation becomes negative approaching, for the Stokes shift about 50 meV, 25 %. Inset in Fig. 6 shows kinetics of the degree of circular polarisation measured for this value of the Stokes shift. It is seen that, at the first moment after the excitation, the degree of polarisation is positive, like it is for small Stokes shifts. The initial region of the kinetics is characterised by fast decay with the time constant about 30 ps. After that, a slower process can be observed leading to establishment of negative polarisation. A characteristic time of this process lies in the range of 100 ps.

The negative circular polarisation was recently observed in PL of the negatively charged InAs QDs [11]. This means that such effect is common for the structures with charged QDs. The negative circular polarisation was also observed in structures with the GaAs quantum wells [12] where it was ascribed to excitation of the light-hole states. For our experiments, however, this explanation does not seem appropriate, because the energy gap between the heavy- and light-hole states in the InP quantum dots (units of meV) is much smaller than the values of the Stokes shift for which the negative polarisation is observed.

The model proposed in Ref. [11] explains the negative polarisation as a result of simultaneous flip of the electron and hole spins due to the anisotropic exchange interaction. Without entering into details of this model, it should be noted that it implies fast loss of the hole-spin orientation upon relaxation of the photoexcited hole to the ground state. For the electron-hole pair excited directly in a QD (which is exactly what occurs in our study), this process seems to be unlikely. It is possible that it is spin orientation of resident electrons that is of importance in our case. To verify this suggestion, however, additional experiments are needed.

7. Conclusion

In summary, the main result of this paper consists in detection of a nearly undamped component in kinetics of the degree of circular polarisation of PL of the InP QDs with resident electrons. We come to the conclusion that the long-lived polarisation is related to spin orientation in the QDs containing odd number of carriers. The spin orientation in the dots with even number of carriers is destroyed by the reversible dephasing, resulted from a spread of the anisotropic exchange splittings. In addition, we have found a dependence of kinetics of the degree of polarisation of the charged quantum dots on the magnetic field direction. It is concluded that this dependence is related to irreversible spin relaxation leading to temperature redistribution of populations of the Zeeman sublevels. It is shown that this relaxation can be realised only in the quantum dots with even number of carriers. The reason for the dependence of the irreversible spin relaxation on the number of carriers in the QDs calls for further studies.

Acknowledgments

Authors thank to Dr. T. Okuno and M. Ikezawa for help in performing of experiments, to Dr. K. V. Kavokin and Dr. V. K. Kalevich for a fruitful discussion, and to Dr. Zapasskii for a critical reading of the manuscript. This work is supported by program "Research for the Future" from JSPS-RFTF97P00106, by INTAS, grant No. 1B 2167, and by RFBR.

References

- [1] E. Pazy, I. D'Amico, P. Zanardi, and F. Rossi, *Phys. Rev. B* **64**, 195320 (2001).
- [2] A.V. Khaetskii and Yu.V. Nazarov, *Phys. Rev. B* **61**, 12639 (2000).
- [3] Y. Masumoto, I. V. Ignatiev, I. E. Kozin, V. G. Davydov, S. V. Nair, H.-W. Ren, J. -S. Lee, S. Sugou, *Jpn. J. Appl. Phys.* **40**, 1947 (2001).
- [4] I. E. Kozin, V. G. Davydov, I. V. Ignatiev, A. V. Kavokin, K. V. Kavokin, G. Malpuech, H. W. Ren, M. Sugisaki, S. Sugou, and Y. Masumoto, *Phys. Rev. B* **65**, 241312(R) (2002).
- [5] I.A. Yugova, I. Ya. Gerlovin, I.V. Ignatiev, S. Yu. Verbin, Y. Masumoto. In *Proceedings of the 14th Indium Phosphide and Related Materials Conference, Stockholm, Sweden, May 12-16 (2002)*, p. 71-74.
- [6] K. V. Kavokin, *Physica Status Solidi (to be published)*
- [7] A. Yugova, I. Ya. Gerlovin, V. G. Davydov, I. V. Ignatiev, I. E. Kozin, H.-W. Ren, M. Sugisaki, S. Sugou and Y. Masumoto, *Phys. Rev.*, submitted.
- [8] *Optical orientation*, edited by F. Meier and B. Zakharchenya, *Modern Problems in Condensed Matter Sciences Vol. 8* (North-Holland, Amsterdam, 1984).
- [9] A. P. Heberle, W. W. Ruhle, and K. Ploog, *Phys. Rev. Lett.* **72**, 3887 (1994).
- [10] X. Marie, T. Amand, P. Le Jeune, M. Paillard, P. Renucci, L. E. Golub, V. D. Dymnikov, and E. L. Ivchenko, *Phys. Rev. B* **60**, 5811 (1999).
- [11] S. Cortez, A. Jbeli, X. Marie, O. Krebs, R. Ferreira, T. Amand, P. Voisin, and J-M. Gerard, *Proceedings of the MSS10 Int. Conf. on Modulated Semiconductor Structures, 23-27 July 2001, Linz, Austria [to be published in Physica E (2002)]*.
- [12] R. I. Dzhiyev, B. P. Zakharchenya, V. L. Korenev, P. E. Pak, D. A. Vinokurov, O. V. Kovalenkov, and I. S. Tarasov, *Fiz. Tverd. Tela* **40**, 1745 (1998) [*Phys. Solid State* **40**, 1587 (1998)].