JETP Letters, Vol. 71, No. 3, 2000, pp. 117–122. Translated from Pis'ma v Zhurnal Éksperimental'noĭ i Teoreticheskoĭ Fiziki, Vol. 71, No. 3, 2000, pp. 174–181.

CONDENSED MATTER

Collective Behavior of Interwell Excitons in GaAs/AlGaAs Double Quantum Wells

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Received December 30, 1999

Abstract—Photoluminescence spectra of interwell excitons in double GaAs/AlGaAs quantum wells (*n–i–n* structures) have been investigated (an interwell exciton in these systems is an electron–hole pair spatially separated by a narrow AlAs barrier). Under resonance excitation by circularly polarized light, the luminescence line of interwell excitons exhibits a significant narrowing and a drastic increase in the degree of circular polarization of photoluminescence with increasing exciton concentration. It is found that the radiative recombination rate significantly increases under these conditions. This phenomenon is observed at temperatures lower than the critical point and can be interpreted in terms of the collective behavior of interwell excitons. © *2000 MAIK "Nauka/Interperiodica".*

PACS numbers: 73.20.Mf; 73.20.Dx; 78.55.Cr

1. Tunneling quantum systems (superlattices and double quantum wells) have long been the subjects of intensive studies [1–8]. In principle, electron and hole charge carriers can be spatially separated in these two-dimensional systems, which is the reason for this interest. In double quantum wells with an applied band-declining electric displacement, one can excite excitons with the electron and the hole located in different quantum wells separated by a tunnel-transparent barrier. Such excitons are named spatially indirect (I) or interwell excitons, as distinct from direct intrawell excitons, in which the electron and the hole are located in the same quantum well. Interwell excitons are long-lived as compared to intrawell excitons. Therefore, these excitons can easily be accumulated, and the gas of such excitons can be cooled down to rather low temperatures. Because the central symmetry is broken, interwell excitons have a dipole moment already in the ground state. Theory predicts various possible scenarios of the collective behavior in a system of spatially separated electrons and holes [1, 9–14]. Thus, in the recent work [14], it was shown that a liquid dielectric phase of such excitons could be a stable state of the electron–hole system at certain critical parameters (the dipole moment of interwell excitons, their density, and temperature) in spite of the dipole–dipole repulsion of interwell excitons. Previously, it was indicated [11] that a condensed dielectric excitonic phase can occur only in the case of lateral confinement (accidental or artificially prepared) in the quantum well plane. This confinement and the related external compression allow rather large densities to be achieved in the gas of interwell excitons.

A random potential due to diverse structural defects (residual charged and neutral impurities, fluctuations of the barrier width and the widths of the interwell excitons themselves, etc.) is always present in the real tunneling quantum systems based on semiconductor heterostructures. These fluctuations generate a random potential profile in quantum well planes. Therefore, photoexcited electrons and holes spatially separated between neighboring quantum wells, as well as excitons, become strongly localized in the regions of these fluctuations at sufficiently low temperatures. This localization effect in coupled quantum systems is, in particular, manifested in the lateral, thermally activated tunneling of charge carriers and is revealed in experiments on spectral narrowing of the luminescence line corresponding to interwell radiative recombination with increasing temperature [7].

This work is devoted to studying the photoluminescence of interwell excitons in a double quantum well with the barrier separating the quantum wells that contains four AlAs monolayers. When the barriers are so narrow, interwell excitons are rather strongly bound. In this case, if the linear scale of fluctuations $l > a_B$ (the exciton Bohr radius is about $a_B \leq 100$ Å) and the amplitude of fluctuations $\Delta > kT$, interwell excitons are to be localized at lateral fluctuations of the random potential without changing significantly their internal structure. It is known that the barrier width in structures with narrow AlAs barriers is characterized by large-scale fluctuations. Therefore, the lateral potential barrier will also be characterized by large-scale fluctuations. It is

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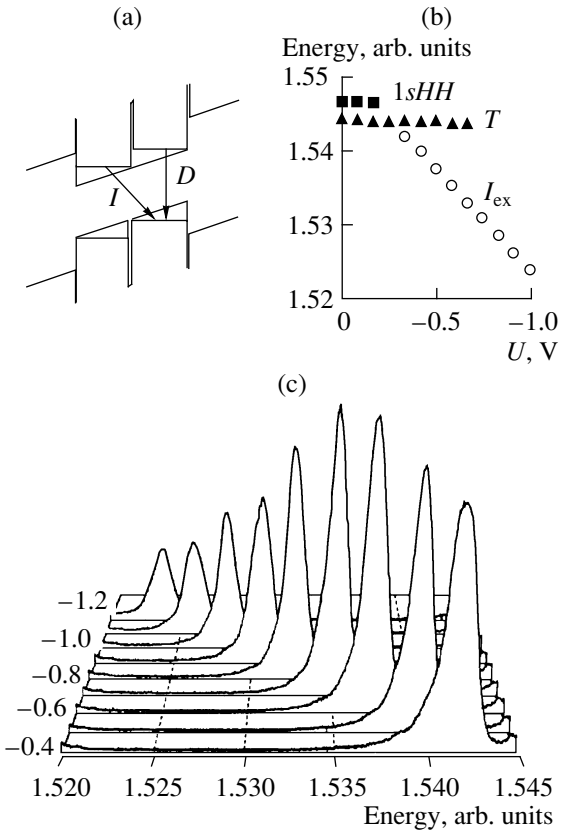

Fig. 1. (a) Schematic diagram of optical transitions; (b) spectral positions of lines of the direct exciton (1*sHH*), the excitonic complex (*T*), and interwell excitons (I_{ex}) as a function of the electric displacement; and (c) the behavior of luminescence spectra of interwell excitons as a function of the applied voltage (numbers on the right of the spectra correspond to the electric voltage in volts) at $T = 2$ K.

reasonable to expect that interwell excitons at sufficiently low temperatures will reside in such accidental lateral large-scale potential wells. It is interesting to know whether the system of excitons under conditions of such accidental lateral confinement will exhibit critical behavior when the density increases at low temperatures. In this work, we attempt to answer the above questions.

2. We investigated an *n–i–n* GaAs/AlGaAs heterostructure with a double quantum well GaAs/AlAs/GaAs (the width of GaAs wells is about 120 Å, and the width of the AlAs barrier is about 11 Å). The structure was grown using molecular-beam epitaxy on an *n*-type doped GaAs support (the concentration of the doping Si: impurity was 10^{18} cm^{-3}) with the (001) crystallographic orientation. First, a 0.5-μm buffer layer of Si-doped (10^{18} cm^{-3}) GaAs was grown on the support. Next, an insulating AlGaAs layer ($x = 0.33$) with a thickness of 0.15 μm was put in place. Next, GaAs/AlAs/GaAs double quantum wells were grown. After the double quantum well, an insulating AlGaAs layer with a thickness of 0.15 μm was put in place, followed by a 0.1-μm layer of Si-doped (10^{18} cm^{-3}) GaAs. The whole structure was covered by a 100-Å GaAs layer. Mesas of size 1×1 mm^2 were made on the structure by the lithographic method. Metallic contacts of an Au + Ge + Pt alloy were applied to the buffer layer and to the doped layer in the upper part of a mesa.

Luminescence spectra were investigated under conditions of resonance excitation of intrawell direct heavy-hole excitons with the use of a tunable Ti-sapphire laser. With the aim of optically orienting the angular momentum of the exciton, we used circularly polarized resonance excitation. The kinetics of luminescence spectra was studied under pulse excitation by a picosecond laser (wavelength 6200 Å, pulse duration 30 ps, and pulse frequency 0.8 MHz). The time evolution of spectra and the kinetics of intensities were measured using a system of time-correlated single-photon counting.

3.1. Figure 1c demonstrates the luminescence spectra of interwell excitons (I_{ex}) measured under resonance excitation of the 1*sHH* exciton and at various applied electric displacements. Two lines are observed in the region of intrawell luminescence at a zero electric displacement: the line 1*sHH* of the free exciton and the line *T* of the bound exciton. At a negative electric displacement, starting from –0.4 V, the spectra exhibit a line of interwell radiative recombination, which linearly shifts toward lower energies as the applied voltage increases (see Fig. 1b). In this case, only the line of the charged excitonic complex *T* remains in the spectrum of intrawell recombination [15]. At large negative displacements $U < -0.8$ V and stationary excitation, only the luminescence line of the interwell exciton remains detectable in the spectra, whereas the luminescence of direct intrawell excitons and excitonic complexes is several orders of magnitude weaker in intensity. The intensity of the interwell-exciton line behaves as a nonmonotonic function of the applied negative displacement. This line appears in the luminescence spectrum at such electric displacements when the Stark shift exceeds the difference of binding energies of the intrawell and interwell excitons, $eFz \geq E_D - E_I$. At $U \sim -0.7$ V, the intensity of this line is a maximum and then monotonically drops with increasing electric field (see Fig. 1c). This behavior can easily be understood with regard to the fact that, as the field increases, the effective dipole moment of the interwell exciton increases in the direction of structure growth (*z*-axis). In this case, the overlap between the wave functions of the electron and the hole in the exciton monotonically decreases.

3.2. The luminescence line of the interwell exciton at sufficiently low temperatures ($T = 2$ K) and small pumping intensities has a large width (FWHM = 4–5 meV), and the line shape is asymmetric with a rather extended long-wavelength tail and a relatively sharp violet edge (see Fig. 2). These features of the photoluminescence line of interwell excitons are the

consequences of their strong localization in the regions of fluctuations of the random potential. In this case, the line width reflects the statistical distribution of amplitudes of the random potential (it is implied that the pumping is so small, $n_{e-h} < 10^9$ cm^{-2}, that the occupation of accidental potential wells with linear scales $l < 1$ μm does not exceed one).

The luminescence line width and shape of interwell excitons significantly change with increasing power of the resonance excitation of direct 1*sHH* intrawell excitons by circularly polarized light (see Fig. 2). With increasing pumping, line *I* is narrowed down to 1.5 meV, that is, more than three times. The narrowing of the line is accompanied by a strong superlinear increase in its intensity (see the inset in Fig. 2). Only at high pumping intensities ($P > 6$ W/cm^2) does this line shift toward high energies and somewhat broaden. The line shift toward high energies indicates that the applied electric field is screened when the interwell-exciton density becomes sufficiently large. From here, the interwell-exciton density can be estimated from the above when the luminescence line width reaches its minimum. This estimation gives $n = 3 \times 10^{10}$ cm^{-2} for the concentration. We observed a significant narrowing of the luminescence line of interwell excitons at various applied negative displacement voltages from –0.5 V to –1.2 V. At large negative displacements, the narrowing of the luminescence line of interwell excitons occurred at considerably lower pumping intensities.

3.3. Of interest is the behavior of the degree of circular polarization γ of the interwell-exciton line with increasing power of resonance excitation (see the inset in Fig. 2). Direct, 1*sHH*, fully spin-oriented excitons ($J_h = +3/2$ and $S_z = -1/2$) were generated in our experiments on resonance excitation by circularly polarized light. Because of the tunneling of carriers, their binding into interwell excitons, spin–lattice relaxation, and strong spin–orbit interaction of holes, the spin memory of interwell excitons is partially lost but, nevertheless, remains high and comprises almost 15–20% at a low excitation density. When the power of resonance excitation increases so that the luminescence line of interwell excitons becomes strongly narrowed, the degree of circular polarization of the corresponding line increases more than two times. If it is suggested that the spin relaxation rate τ_s varies only slightly with increasing pumping intensity (most likely, it can only increase), then the increase in the degree of circular polarization can be naturally associated with the decrease in the lifetime τ_d of interwell excitons. This follows from the simple kinetic equation relating the degree of circular polarization with the lifetime and the spin relaxation time $\gamma = \gamma_0/(1 + \tau_d/\tau_s)$ [16]. A threshold increase in the circular polarization of the narrow photoluminescence line of interwell excitons with increasing pumping was observed at resonance excitation by linearly polarized light (polarization was parallel to the plane of layers). When the excitation densities were not high and interwell excitons were strongly localized in the regions of fluctuations of the random potential, the photoluminescence spectra remained fully depolarized under such conditions. The details of experiments with linearly polarized excitation will be published separately.

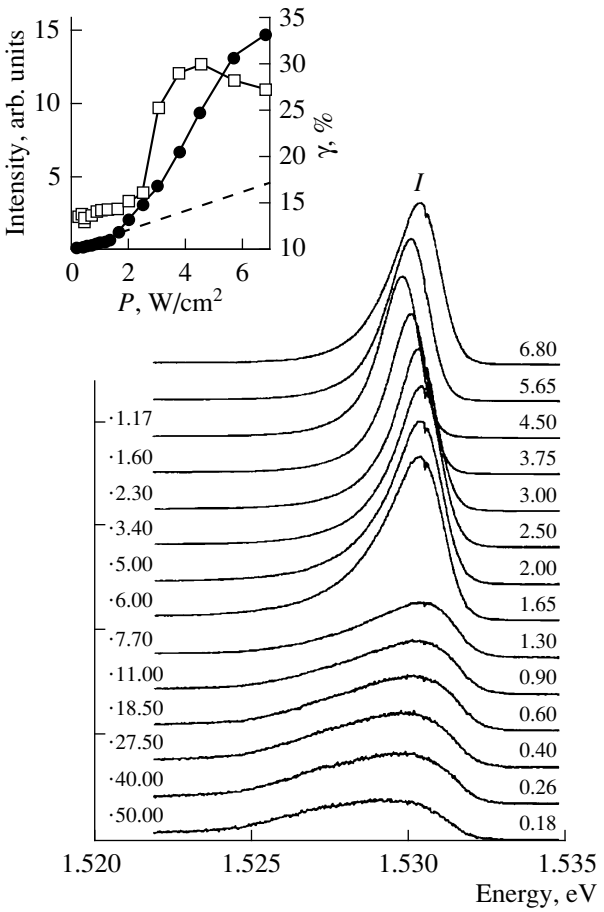

Fig. 2. Photoluminescence spectra of interwell excitons (line *I*) at various powers of the resonance excitation of the direct 1*sHH* exciton by circularly polarized light (σ^+), applied displacement –1 V, and $T = 2$ K. Numbers on the right of the spectra correspond to pumping intensities in W/cm^2, those on the left correspond to the scale factors in respect to the two upper spectra. The inset presents the line intensity of interwell excitons (round symbols, left scale) and its degree of circular polarization (square symbols, right scale) as functions of the power density. The dashed line is an extrapolation of the linear dependence of the intensity.

Thus, with increasing concentration of interwell excitons, the corresponding photoluminescence line is narrowed down and the degree of its polarization increases, which gives indirect evidence that the radiative decay time shortens. This phenomenon turned out to be very sensitive to temperature. At temperatures higher than critical values and fixed large pumping intensities, the line width of interwell excitons increases abruptly and the degree of circular polariza-

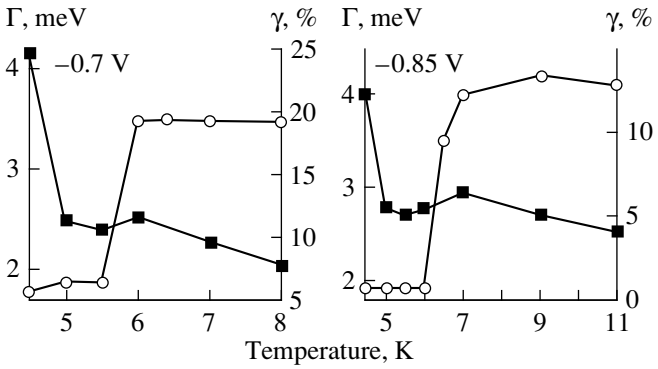

Fig. 3. Temperature dependence of the luminescence line width of interwell excitons Γ (round symbols, left scale) and its degree of circular polarization γ (square symbols, right scale) for electric displacements of –0.7 and –0.85 V.

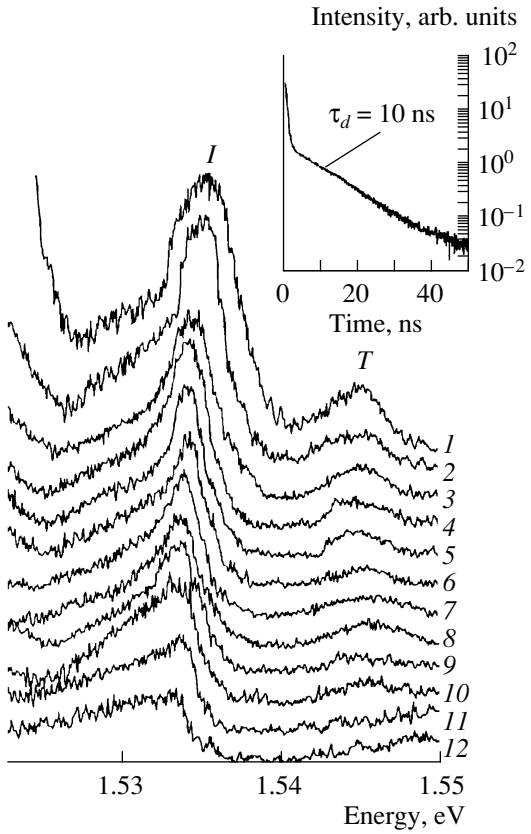

Fig. 4. Time evolution of spectra and the kinetics of the luminescence decay of interwell excitons (see inset) under conditions of pulse excitation at T = 5 K and displacement –0.7 V. Spectra *1–8* were measured with time delays of 2, 3, 4, 5, 6, 7, 8, and 10 ns and integration for 2 ns. Spectra *9–12* correspond to delays of 12, 16, 20, and 25 ns and integration for 4, 4, 5, and 6 ns, respectively. The intensity decay time of the narrow line of interwell excitons τ_d = 10 ns.

tion decreases down to previous values (see Fig. 3). In this case, the critical temperature at which such dramatic spectral changes were observed was $T_c \le 6$ K (ΔT = ±1 K).

3.4. We investigated the kinetics of photoluminescence spectra under conditions of pulse excitation with the use of a picosecond laser. Under this excitation, hot photoexcited electrons and holes are generated at the instant of laser pulse action in each quantum well with the same density and are not spatially separated. Figure 4 presents the time evolution of spectra under pulse excitation. The spectra were measured with different time delays in reference to the exciting laser pulse at T = 5 K and the applied voltage U = –0.7 V. At zero delays, only the region of direct intrawell luminescence is observed in the spectra. The photoluminescence spectrum of interwell excitons is formed at delays $\tau \ge 2$ ns. This delay is a consequence of carrier tunneling through the barrier accompanied by spatial separation of charge carriers among quantum wells; their intrawell energy relaxation; and, simultaneously, recombination. Figure 4 demonstrates that a sufficiently narrow line dominates at the violet edge of interwell photoluminescence spectra at small delays (<10 ns). With increasing delay, this line is significantly narrowed. Its width reaches 1.5 meV at a delay of 6 ns; that is, it decreases almost three times in comparison with its width at initial delays. The intensity of this line decays with time much faster than the structureless spectrum of interwell luminescence beneath it. At delays longer than 20 ns, this line is not resolved and blends into the structureless part of the spectrum, which remains almost unchanged in its shape and is observed at delays longer than 40 ns. This behavior is clearly evident in the kinetics of intensities measured directly at the spectral position of the narrow line (see the inset in Fig. 4). It is essential to note that the narrow band distinguished by the short-term kinetics of intensity decay under conditions of pulse excitation is observed, as well as in the case of stationary excitation, only at sufficiently low temperatures. Thus, in the case of delays of 12 ns, the narrow line starts to broaden and blends into the structureless background beneath it at $T \ge 6$ K. At shorter delays and, hence, at a higher density of interwell excitons, this line disappears in the spectrum at distinctly higher temperatures. For example, at a delay of 7 ns, the narrow line disappears in the spectra at $T \ge 10$ K. We observed the same behavior of the time-resolved spectra and kinetics of interwell luminescence at various applied voltages in the range from –0.4 to –0.9 V.

4. The totality of the experimental results presented above cannot be explained within the simple one-particle description of radiative annihilation of interwell excitons localized at fluctuations of the random potential. Thus, the appearance of the narrow line in the photoluminescence spectrum can be associated with delocalized interwell excitons, which appear above the percolation threshold. However, in this case, it remains completely inexplicable why this phenomenon is so sensitive to temperature and is not observed at $T > T_c$. The threshold increase of the degree of circular polarization and the superlinear growth of the intensity of

this line also remains unclear. At the same time, these results can be explained, at least, qualitatively as the consequence of the collective behavior of delocalized interwell excitons upon attaining the critical temperature and concentration. It can be suggested that the investigated structures with a narrow AlAs barrier are characterized by large-scale fluctuations of the potential, in particular, by those due to variations of the barrier width. Such fluctuations of the potential are poorly screened, as distinct from the random potential due to residual charged impurities, which also arises in the studied structures. Because of large-scale fluctuations of the potential profile in the plane of the quantum well, photoexcited interwell excitons can accumulate in macroscopically large areas with lateral confinement up to several tenths of a micron. It is in these areas that the main events associated with photoexcited interwell excitons proceed. These events can develop by independent scenarios, but collective interaction in the system of interwell excitons delocalized within macroscopically large lateral areas is suggested in the basis of each of them. Within one of the scenarios, it can be suggested that interwell excitons are condensed into a metallic *e–h* liquid with spatially separated electron–hole layers upon attaining the critical densities and temperature. If one associates the narrow line in the spectrum with the emerging liquid phase, then its density can be easily estimated from the above by the line width, which, in this case, must equal the sum of Fermi energies of electrons and holes. The density determined in this way equals 3×10^{10} cm^{-2}, and the dimensionless parameter r_s corresponding to this density equals $r_s = 1/\sqrt{\pi n a_B^2} = 4$. The condensed phase within this approach turns out to be too loose to shield completely the Coulomb interaction in interwell excitons; that is, excitons at these concentrations must retain their individuality. Therefore, the condensed phase cannot be metallic. This conclusion is confirmed by studies of photoluminescence spectra in a magnetic field with the Faraday geometry. It was found that the narrow line, similar to a free exciton, is split into a Zeeman doublet with the intensity ratio of components σ^+/σ^- corresponding to the temperature and the spin splitting value. That is, the individual properties of the exciton are retained in the condensed phase. The details of these experiments will be described in a separate publication.

We suggest that the effect of strong narrowing of the photoluminescence line of interwell excitons and the critical character of this phenomenon with respect to density and temperature can be associated with the condensation of interwell excitons into a dielectric liquid. It was shown in [14] that, at certain values of the dipole moment of interwell excitons, a sufficiently dense system of interwell excitons could condense into a dielectric liquid in spite of the dipole–dipole repulsion between such excitons. It was indicated in [11] that this condensation could most likely occur in the regions with lateral confinement. According to our experiments, under stationary excitation, the condensation occurs at $T < 6$ K and at an average exciton concentration of 3×10^{10} cm^{-2}.

The condensed part of excitons must be phased within the coherent length. Spatial coherence must occur at least at scales of the de Broglie wavelength of interwell excitons λ_{ex}, which equals $\lambda_{ex} = h/\sqrt{\pi m k T} = 1.5 \times 10^3$ Å at $T = 2$ K and exceeds the exciton Bohr radius by more than an order of magnitude. At the same conditions, the dimensionless parameter $r = n \times \lambda_{ex}^2 = 4$. The observed threshold increase of the circular polarization of interwell excitons gives indirect evidence of an increase of spatial coherence in the condensed phase. Furthermore, the radiative decay of phase-correlated interwell excitons in the condensate must be distinguished by considerably larger radiative probabilities, as compared to the photoluminescence of excitons beyond the condensate. This conclusion also agrees with the experiment.

Nevertheless, an essential question in justifying the assumption of the condensation of interwell excitons into a dielectric liquid phase remains open and requires an experimental answer. This question concerns the determination of the linear scales of spatial coherence. This problem can be solved experimentally, in particular, by measuring intensity correlations under conditions when the suggested exciton condensation occurs.

ACKNOWLEDGMENTS

The authors are grateful to V.F. Gantmakher, S.V. Iordanskiĭ, A.S. Ioselevich, V.D. Kulakovskiĭ, Yu.E. Lozovik, V.G. Lysenko, and R.A. Suris for interesting discussions. This work was in part supported by the Russian Foundation for Basic Research, project no. 98-02-16656, and by the interdepartmental program "Nanostructures."

REFERENCES

1. E. Yu. Lozovik and V. I. Yudson, Zh. Éksp. Teor. Fiz. **71**, 738 (1976) [Sov. Phys. JETP **44**, 389 (1976)].
2. T. Fukuzawa, E. E. Méndez, and J. M. Hong, Phys. Rev. Lett. **64**, 3066 (1990).
3. J. E. Golub, K. Kash, J. P. Harbison, and L. T. Flórez, Phys. Rev. B **41**, 8564 (1990).
4. J. A. Kash, M. Zachau, E. E. Méndez, *et al.*, Phys. Rev. Lett. **66**, 2247 (1991).
5. L. V. Butov, A. Zrenner, G. A. Abstreĭter, *et al.*, Phys. Rev. Lett. **73**, 304 (1994); L. V. Butov, in *Proceedings of the 23rd International Conference on Physics of Semiconductors, Berlin, 1996*.
6. V. B. Timofeev, A. I. Filin, A. V. Larionov, *et al.*, Europhys. Lett. **41**, 435 (1998).
7. V. B. Timofeev, A. V. Larionov, A. S. Ioselevich, *et al.*, JETP Lett. **67**, 613 (1998).

8. V. V. Krivolapchuk, E. S. Moskalenko, A. L. Zhmodikov, *et al.*, Solid State Commun. **111**, 49 (1999).
9. D. Yoshioka and A. H. MacDonald, J. Phys. Soc. Jpn. **59**, 4211 (1990).
10. X. M. Chen and J. J. Quinn, Phys. Rev. Lett. **67**, 895 (1991).
11. Xuejun Zhu, P. B. Littlewood, M. S. Hybertsen, and T. Rice, Phys. Rev. Lett. **74**, 1633 (1995).
12. J. Fernández-Rossier and C. Tejedor, Phys. Rev. Lett. **78**, 4809 (1997).
13. Lerwen Liu, L. Swierkowski, and D. Nelson, Physica B **249**–**251**, 594 (1998).
14. Yu. E. Lozovik and O. L. Berman, Zh. Éksp. Teor. Fiz. **111**, 1879 (1997) [JETP **84**, 1027 (1997)].
15. V. B. Timofeev, A. V. Larionov, M. Grassi Alessi, *et al.*, Phys. Rev. B **60**, 8897 (1999).
16. *Optical Orientation, Modern Problems in Condensed Matter Sciences*, Ed. by F. Meyer and B. P. Zhakharchenya (Elsevier, Amsterdam, 1984), Vol. 8.

Translated by A. Bagatur'yants