Charged excitonic complexes in GaAs/Al0.35Ga0.65As p-i-n double quantum wells

Timofeev, V. B.; Larionov, A. V.; Alessi, M. Grassi; Capizzi, M.; Frova, A; Hvam, Jørn Marcher

Published in:
Physical Review B Condensed Matter

Link to article, DOI:
10.1103/PhysRevB.60.8897

Publication date:
1999

Document Version
Publisher's PDF, also known as Version of record

Charged excitonic complexes in GaAs/$Al_{0.35}Ga_{0.65}As$ *p-i-n* double quantum wells

V. B. Timofeev and A. V. Larionov
Institute of Solid State Physics, Russian Academy of Science, 142432 Chernogolovka, Russia

M. Grassi Alessi, M. Capizzi, and A. Frova
Istituto Nazionale di Fisica della Materia, Università degli Studi di Roma ''La Sapienza,'' Piazzale Aldo Moro 2, 00185 Roma, Italy

J. M. Hvam
Research Center COM, The Technical University of Denmark, DK 2800 Lyngby, Denmark

(Received 7 May 1999)

Photoluminescence (PL) and PL excitation measurements (PLE) have been performed in GaAs/$Al_xGa_{1-x}As$ double quantum well (QW) structures under different applied electric fields. An emission due to charged excitons (trions) has been identified in the PL spectra ~ 3 meV below the heavy-hole exciton emission. These trions are localized by random potential fluctuations, at the interfaces or in the QW, as shown by the saturation of their emission intensity with respect to that of the heavy-hole excitons. Trions are positively charged, namely, they are made by two holes and one electron, as shown by (i) an analysis of the PL polarization for resonant excitation of the heavy- and the light-exciton ground state, and (ii) the analysis of the Zeeman effect for the trion PL band in the Faraday geometry, i.e., for a magnetic field normal to the QW's.
[S0163-1829(99)10235-2]

I. INTRODUCTION

The stability of charged excitons (trions) and their observation in semiconductors was discussed first by Lampert 41 years ago.[1] Stable trions can be either positively charged (two holes bound to one electron) or negatively charged (two electrons bound to one hole). Their formation requires the coexistence in the same system of excitons and carriers, either free or localized by random potential fluctuations. Moreover, carriers need to be diluted in order to minimize the effects of screening and of the exclusion principle, which reduce the stability of these excitonic complexes. In bulk, three-dimensional semiconductors the detection of trions is difficult since their binding energy is very small (a few percent of the exciton binding energy).[2] The first claim of a trion contribution to the photoluminescence (PL) line shape of a dense electron-hole plasma in bulk Ge and Si has been made in the late seventies.[3] After Stebe and Ainane have pointed out that in a two-dimensional system the binding energy of trions should increase significantly due to the effect of confinement,[4] many efforts have been spent to observe charged exciton complexes in quantum wells (QW's). In the last years, trions have been clearly identified in $CdTe/Cd_xZn_{1-x}Te$ QW's (Ref. 5) and in GaAs/$Al_xGa_{1-x}As$ heterostructures.[6–8]

In the present paper, we investigate the existence of trions in double symmetric GaAs/$Al_xGa_{1-x}As$ QW's where an external electric field is applied parallel to the growth direction. This system is a good candidate for the search of trions, as explained in the following. The applied bias introduces an asymmetry between the two wells, each one collecting preferentially one type of carrier. In both wells, the random potential fluctuations, at the interfaces or in the well, induce a localization of carriers into traps that may later bind excitons and give rise to charged three-particle exciton complexes, namely, to excitonic trions. As a matter of fact, in the low-temperature PL spectra of these *p-i-n* double quantum wells (DQW) we observe a band ~ 3 meV below the heavy-hole exciton (HHE) band. For an increasing excitation power, the intensity of this band increases superlinearly first, then it saturates with respect to the exciton emission. Measurements of excitation photoluminescence (PLE) for different polarization of the exciting and emitted light demonstrate that this excitonic complex is formed by two holes and one electron (positive trion) localized at random potential fluctuations. This attribution is supported also by PL measurements in presence of a magnetic field applied perpendicularly to the QWs.

II. EXPERIMENT

GaAs/$Al_xGa_{1-x}As$ ($x=0.35$) DQW structures have been grown by molecular-beam epitaxy on the (001) surface of a *n*-doped GaAs substrate (Si doping: 2×10^{18} cm^{-3}). The sequence of layers is the following: 0.5 μm *n*-type GaAs buffer layer (Si doping: 10^{18} cm^{-3}); 0.3-μm $Al_xGa_{1-x}As$ insulating barrier; 8-nm GaAs well; 5-nm $Al_xGa_{1-x}As$ barrier; 8-nm GaAs well; 0.3-μm $Al_xGa_{1-x}As$ insulating barrier; 0.1-μm *p*-type GaAs top contact layer (Be doping: 10^{18} cm^{-3}), and, finally, 5-nm GaAs cap layer. We have studied 0.1×0.1 cm^2 lithographically etched mesas made in these DQW's, *n* and *p* regions contacts being made by gold. In dark or at low-laser photoexcitation power, for an exciting photon energy lower than the $Al_xGa_{1-x}As$ barrier band gap (resonant excitation), the samples behave like typical *p-i-n* diodes, with a current smaller than 10 nA in the whole range of bias used (from -1 to $+0.8$ V). In the undoped region of the *p-i-n* structure, the residual impurity concentration is $\leqslant 10^{15}$ cm^{-3}. All experiments have been carried out with a Ne-He or a tunable Ti-sapphire laser. PL and PLE emitted

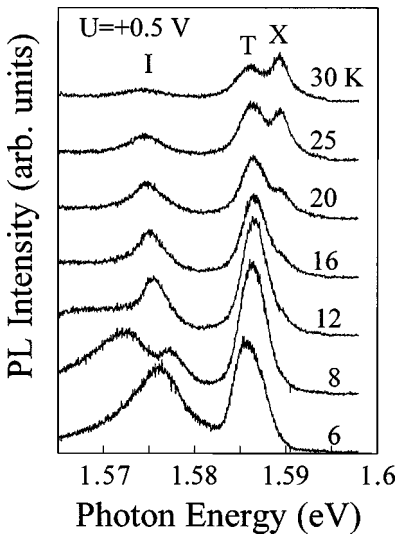

FIG. 1. Photoluminescence spectra of a GaAs/$Al_xGa_{1-x}As$ *p-i-n* DQW measured at different temperatures for a fixed forward bias ($U=+0.5$ V). The excitation power is equal to 1.5×10^{-3} W cm^{-2}, the excitation wavelength to 632.8 nm. The interwell exciton, *I*, the charged exciton complex, *T*, and the 1*sHH* intrawell exciton, *X*, are indicated.

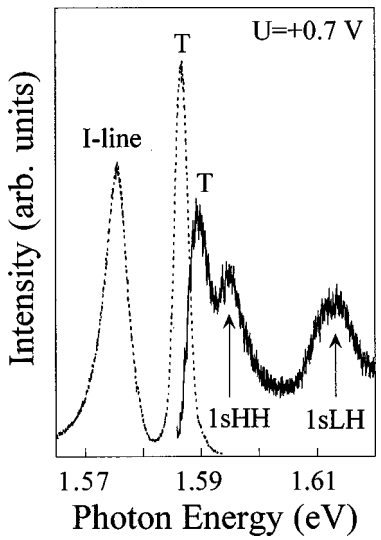

FIG. 2. Photoluminescence excitation spectrum (solid line) taken at $T=4.5$ K for a forward bias $U=+0.7$ V and an excitation power equal to 1.2×10^{-3} W cm^{-2} (detection energy 1.585 eV). A PL spectrum (dashed line) taken at $T=4.5$ K for resonant excitation of the intrawell 1*sHH*-exciton is also reported. The interwell exciton, *I*, the charged exciton complex *T*, and the 1*s* ground-state level of the heavy-hole (HH) and light-hole (LH) exciton are indicated.

light has been analyzed by a double monochromator, the signal being measured with a charge coupled device-camera or a cooled GaAs photomultiplier. The circular polarization of PL under resonant photoexcitation has been studied by means of linear polarizers followed by quarter-wave plates.

III. RESULTS AND DISCUSSION

Let us discuss first the radiative recombination of excitons in the regime of low-excitation power and low temperature, when the applied electrical bias is not screened by photoexcited carriers. PL spectra at fixed $+0.5$ V forward bias are reported in Fig. 1 for temperatures ranging from 6 to 30 K. Three bands are observed, *I*, *T*, and *X*. For increasing temperature, the intensity of the first two bands strongly decreases with respect to that of the *X* band, thus suggesting the connection of those two bands to bound states. When the applied bias is changed from $+1$ to -2 V, the peak energy of the *I* band shifts linearly to lower energy. Those of the *T* and *X* bands, however, do not change. This different behavior with the applied field indicates that the *T* and *X* bands are due to intrawell excitons (exciton complexes where electrons and holes are in the same well and therefore their energy levels are equally affected by the applied field). On the same grounds, the *I* band has been attributed already to emission from an interwell exciton, namely, an exciton complex where photoexcited electrons and holes are spatially separated between the two adjacent wells.[9,10] It has been shown that at low temperature, typically below 6–8 K, those interwell excitons are localized by potential fluctuations due to charged residual impurities in the insulating regions of the *p-i-n* structure.[9] A thermoactivated tunneling mechanism between electrons and holes, each one localized by potential fluctuations in the respective well, accounts then for peculiar features of this band, e.g., the decrease of the band full width at half maximum for increasing temperature.[9,10]

Let us now discuss the intrawell exciton bands, the object of the present investigation. The *X* band is due to the recombination of a HH-intrawell exciton (1*sHH* state), as supported by the PLE spectra reported in Fig. 2 where both heavy- and light-hole intrawell excitons are observed. The *T* band is 2.8–3.1 meV below the HHE band in the PL spectra (Fig. 1), 5.3 meV below the same band in the PLE spectra (Fig. 2). Both the *T* and HHE bands exhibit a large Stokes shift (*SS*), namely, their PL peak energies are lower than the absorption peak energies as measured in the PLE spectra. *SS* is around 2.9 meV for the *T* band, see Fig. 2, and 5.1 meV for the HHE band, see Figs. 1 and 2. These large values of *SS* are due to the competition between carrier localization at random potential minima, in the QW and at the interfaces, and carrier thermalization.[11]

PL spectra taken at $T=11$ K for different exciting power densities are reported in Fig. 3. The intensity of the *T* band first increases superlinearly with respect to that of the *X* band, then it saturates, as also shown in the inset. The superlinear regime indicates that more than two carriers are involved in the complex responsible for the *T*-band emission. The saturation at high-power density, on the other hand, indicates that the density of these complexes is much lower than that of the delocalized excitons responsible for the *X*-band emission. Recently, an emission band has been observed in the PL spectra of $In_xGa_{1-x}As$/GaAs QW's, whose power and temperature dependences were quite similar to those reported here for the *T* band, and has been attributed to charged excitons localized by potential fluctuations at the $In_xGa_{1-x}As$/GaAs interfaces.[12] This attribution holds also for the present *T* band, as confirmed by an analysis of the type and degree of polarization of the PL emission when either the heavy- or the light-hole excitons are resonantly excited (resonant PL, RPL) by σ^+ circularly polarized light. Moreover, this investigation allows us to establish that the *T*

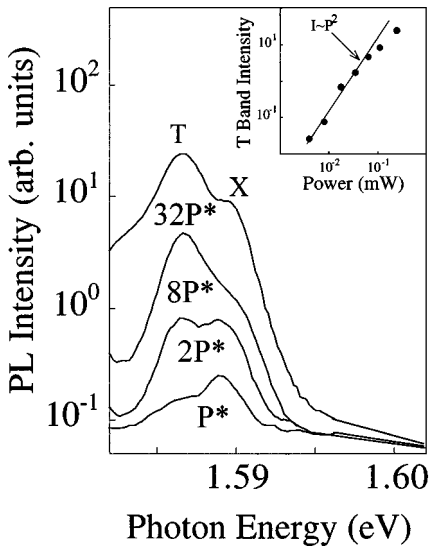

FIG. 3. Photoluminescence spectra at $T=11$ K for different exciting power density ($P^*=\mathrm{W\,cm^{-2}}$). The peak intensity of the T band as a function of the excitation power is reported in the inset (the full line corresponds to a quadratic dependence of intensity on power).

band is due to positively charged trions. RPL spectra taken at 4.5 K for different circular polarization σ^+ and σ^- of the emitted light are reported in Fig. 4 for resonant excitation of the $1sHH$ exciton state (a) and of the $1sLH$ (light-hole) exciton state (b). The spectra cover the energy range of the I and T bands. The corresponding estimate of the degree of polarization γ for the PL intensity, namely, the ratio between σ^+ and σ^- polarized emission, is reported in Fig. 5. The dependence of γ on temperature in the case of the T-band emission is finally shown in Fig. 6. When the $1sHH$ exciton state is excited resonantly with σ^+ circularly polarized light, the T band maintains at low temperature almost the same type and degree of polarization of the exciting light and γ is 0.82 at $T=2$ K; γ then decreases for increasing temperature until it vanishes above $T=25$ K, as shown in Fig. 6, because of the insurgence of spin-lattice relaxation processes. On the other hand, when the $1sLH$ exciton state is excited resonantly with circularly polarized light, the T band is weakly

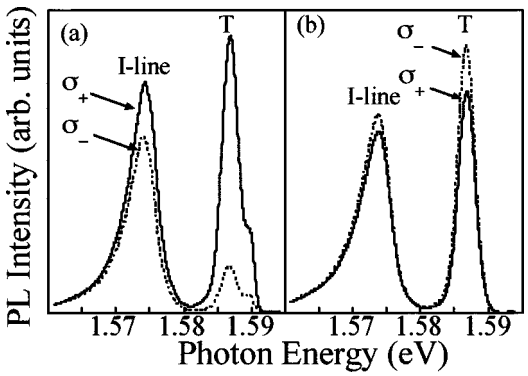

FIG. 4. Photoluminescence spectra at $T=4.5$ K for different polarization σ^+ and σ^- of the emitted light. The polarization of the exciting light is fixed (σ^+). (a) Resonant excitation of the 1s ground state of the heavy-hole exciton. (b) Resonant excitation of the 1s ground state of the light-hole exciton.

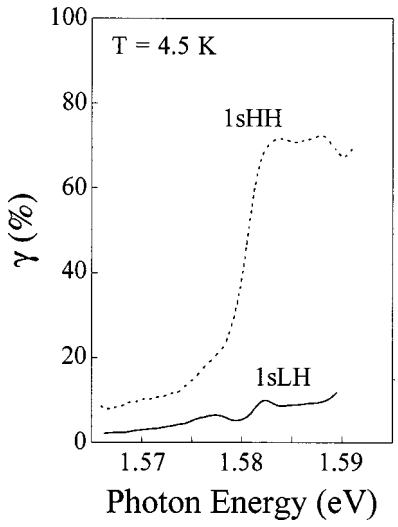

FIG. 5. Ratio $\gamma=|\sigma^+-\sigma^-|/|\sigma^+ + \sigma^-|$ between the photoluminescence intensities σ^+ and σ^- at $T=4.5$ K reported in Fig. 4 (the polarization of the exciting light is σ^+). The full (dashed) line refers to the case of resonant excitation of the 1s ground state of the light-hole (heavy-hole) exciton.

polarized ($\gamma=10\%$ at $T=4.5$ K) and its polarization is opposite with respect to that of the incident light.

These features indicate that the T band is due to emission from positively charged trions. When HHE are photoexcited by circularly polarized σ^+ light, the photogenerated HH has a momentum component $J_z=+3/2$, the electron has $J_z=-1/2$. The spin-orbit interaction gives rise to a relaxation of the HH momentum much faster than that of the electron.[13] When this photoexcited exciton binds to a localized HH to form a positively charged trion, the electron maintains its value of J_z $(-1/2)$, while the hole is fully relaxed. Emission from this trion, therefore, will be polarized as the exciting light, namely, σ^+. On the contrary, when LHE are photoexcited by circularly polarized σ^+ light, the electron should have $J_z=+1/2$ and the emission from trions is polarized

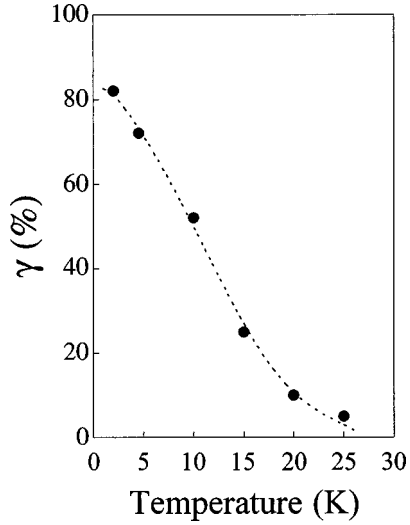

FIG. 6. The ratio $\gamma=|\sigma^+-\sigma^-|/|\sigma^+ + \sigma^-|$ between the photoluminescence intensities σ^+ and σ^-, evaluated at the T-band peak energy for the case of resonant excitation of the $1s$ HH exciton ground state, is reported as a function of temperature.

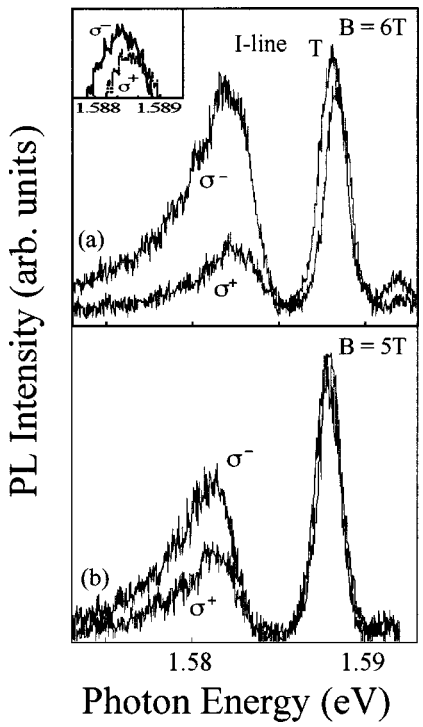

FIG. 7. Zeeman splittings at $T=2$ K for the interwell exciton, *I*, the positively charged exciton complex, *T*, and the intrawell 1*s*HH exciton as measured for a magnetic field *B* perpendicular to the DQW. (a) $B=6$ T; (b) $B=5$ T. In the inset, the splitting of the *T* band is shown on an expanded scale.

σ^-.[14] However, in this case the degree of polarization should be quite low because of mixing between the light and heavy holes, rather strong for the $1sLH$ exciton state. All these features are actually shown by the *T*-band emission, as seen in Fig. 4 and Fig. 5. If the same polarization analysis is performed for negative trions, one finds that the trion PL band should not be polarized at all when the HHE state is resonantly excited with circularly polarized light, contrary to experimental results.[15]

The present assignment of the *T* band to positive trions is further supported by PL measurements in the Faraday geometry, namely, in presence of an applied magnetic field perpendicular to the QW planes. PL spectra taken at $T=2$ K for σ^+ and σ^- polarization are reported for an applied magnetic field *B* of 6 and 5 T in Figs. 7(a) and 7(b), respectively. All the *I*, *T*, and *X* bands exhibit a diamagnetic shift and split into two bands at different energies for the two different circular polarizations of the light (Zeeman doublets), whose difference in energy increases with magnetic field. However, while the intensities of the two components of the doublet is quite different for the interwell *I* and intrawell *X* exciton bands, the intensities of those components for the trion *T* band are almost equal, at least for magnetic field up to 6 T. This behavior is a signature of positively charged trions. Indeed, in the case of a T^+ complex the splitting of the PL band is determined mainly by the splitting of the final state, namely, by the splitting of the recoil particle, a heavy hole. In the case of a T^- complex, instead, the Zeeman splitting of the corresponding PL doublet is determined by the splitting of the initial state. Therefore, the intensities of the two components σ^+ and σ^- of the doublet are expected to be quite similar for positive trions, essentially different for negative trions because of the different level populations of the initial states. The value of the Zeeman splitting has also allowed us to estimate the *g* factor of the heavy hole in the positively charged trion $[g(h_{3/2})=1.4]$.

Finally, the temperature dependence of the *T*-band intensity reported in Fig. 1 is consistent with the thermal ionization of a charged exciton complex into a free exciton and a hole. For increasing temperature, indeed, the intensity of the *X* band increases at the expenses of that of the *T* band. Also the observed activation temperature of about 25 K, see Fig. 1, is close to the binding energy of the second hole in the trion (around 3 meV, as measured by the energy separation between the peaks of the *X* and *T* bands measured in PL).

IV. CONCLUSIONS

Suitably biased double-quantum-well heterostructures give rise to separate confinement of photoexcited electrons and holes in the two adjacent wells. Photoluminescence, resonant photoluminescence and photoluminescence excitation measurements have shown that charged exciton complexes form in those systems, where they can be easily studied. In the presence of a forward bias, photoexcited holes preferentially accumulate in one of the quantum wells and, at low-exciting power, are almost all localized on random potential fluctuations. At low temperatures, excitons bind to those localized holes thus giving rise to charged exciton complexes, trions, whose thermal activation energy is on the order of 3 meV. The state of charge of the trions has been determined by an analysis of the PL polarization for resonant excitation of the heavy- and the light-exciton ground states. This attribution has been confirmed by an analysis of the Zeeman effect of the trion PL band in the Faraday geometry, i.e., for a magnetic field normal to the QW's.

ACKNOWLEDGMENTS

V.B.T. acknowledges financial support from the Landau-Centro Volta Network and the Russian Foundation of Fundamental Research. A.V.L. acknowledges financial support from the ''Nanostructure Program'' of the Russian Ministry of Science and Technology. The samples were grown by C. B. Soerensen, III-V Nanolab.

[1] M. A. Lampert, Phys. Rev. Lett. **1**, 450 (1958).
[2] G. Munschy and B. Stebe, Phys. Status Solidi B **64**, 213 (1974).
[3] G. A. Thomas and T. M. Rice, Solid State Commun. **23**, 359 (1977).
[4] B. Stebe and A. Ainane, Superlattices Microstruct. **23**, 545 (1989).
[5] K. Kheng, R. T. Cox, M. Y. d'Aubigné, F. Bassani, K. Saminadayar, and S. Tatarenko, Phys. Rev. Lett. **71**, 1752 (1993).
[6] G. Finkel'stein, H. Shtrikman, and I. Bar-Joseph, Phys. Rev. Lett. **74**, 976 (1995).
[7] H. Buhmann, L. Mansouri, J. Wang, P. H. Beton, N. Mori, L. Eaves, M. Henini, and M. Potemski, Phys. Rev. B **51**, 7969 (1995).
[8] A. J. Shields, M. Pepper, D. A. Ritchie, M. Y. Simmons, and G.

A. C. Jones, Phys. Rev. B **51**, 18 049 (1995).

[9] V. B. Timofeev, A. I. Filin, A. V. Larionov, J. Zeman, G. Martinez, J. M. Hvam, D. Birkedal, and C. B. Soerensen, Europhys. Lett. **41**, 535 (1998).

[10] V. B. Timofeev, A. V. Larionov, A. S. Ioselevich, J. Zeman, G. Martinez, J. M. Hvam, and C. B. Soerensen, Pisma Zh. Eksp. Teor. Fiz **67**, 580 (1998) [JETP Lett. **67**, 613 (1998)].

[11] E. Runge and R. Zimmermann, in *Festkörperprobleme/Advances in Solid State Physics*, edited by Bernhard Kramer (Friedr. Vieweg, Braunschweig/Weisbaden, Germany, 1999), Vol. 38, p. 251; R. Zimmermann and E. Runge, Phys. Status Solidi A **164**, 511 (1997).

[12] F. Martelli, A. Polimeni, A. Patanè, M. Capizzi, P. Borri, M. Gurioli, M. Colocci, A. Bosacchi, and S. Franchi, Phys. Rev. B **53**, 7421 (1996).

[13] See, e.g., *Optical Orientation, Modern Problems in Condensed Matter Sciences*, edited by F. Meyer and B. P. Zakharchenya (Elsevier, Amsterdam, 1984), Vol. 8.

[14] After photoexcitation, LH excitons quickly relax into HH excitons, which bind to localized heavy holes and give rise to charged trions. In the ground state of this complex, the two holes have opposite momenta ($\pm 3/2$), the electron keeps its momentum ($+1/2$). Therefore, the electron can recombine only with the ($J=+3/2$) heavy hole and give rise to an emission band polarized σ^-.

[15] The lack of a full polarization for the *T* band under resonant excitation of HHE at the lowest temperature could be attributed to a small contribution of negatively charged trions, localized in the neighbor QW, to the *T*-band emission. The binding energy of trions is very small and weakly depends on trion state of charge (Ref. 4), thus leading to an overlap between the emission of oppositely charged trions.