Intensity dependence of the Fermi edge singularity in photoluminescence from modulation-doped $Al_xGa_{1-x}As/In_yGa_{1-y}As/GaAs$ heterostructures

H. Kissel,* U. Müller, C. Walther, and W. T. Masselink
Department of Physics, Humboldt-Universität zu Berlin, Invalidenstrasse 110, D-10115 Berlin, Germany

Yu. I. Mazur, G. G. Tarasov, G. Yu. Rud'ko, M. Ya. Valakh, V. Malyarchuk, and Z. Ya. Zhuchenko
Institute of Semiconductor Physics, National Academy of Sciences, Prospect Nauki 45, 252650 Kiev, Ukraine
(Received 27 October 1999; revised manuscript received 13 December 1999)

A photoluminescence (PL) study of pseudomorphic modulation-doped $Al_xGa_{1-x}As/In_yGa_{1-y}As/GaAs$ heterostructures with high-electron density reveals a fundamental change in the PL spectrum under increased excitation density. The high-energy tail of the PL is transformed due to the mutual repulsion of the Fermi-edge singularity (FES) and the excitonic states. The details of this repulsion depend on the excitation density and temperature. At low temperature, the dependence of the existence of the FES feature on excitation density has been demonstrated for the first time. The appearance of the FES is accompanied by the formation of an abrupt high-energy edge and occurs at intensities far below those required for the appearance of the hybridized $n=2$ exciton. Strong screening of the $n=2$ exciton state by photoexcited carriers is observed. The PL behavior under excitation density and temperature variation near the Fermi edge is explained in terms of a strong effect of the carrier density on the appearance of the FES; the details of this effect have not yet been fully theoretically explored.

Nearly ideal conditions for the study of many-body effects, namely, the Fermi-edge singularity (FES), band-gap renormalization, or shake-up processes are provided by pseudomorphic modulation-doped $Al_xGa_{1-x}As/In_yGa_{1-y}As/GaAs$ heterostructures.[1–5] Due to the high density of the two-dimensional electron gas (2DEG) in these systems, the correlation effects are of great importance, resulting in the rearrangement of electron and hole subsystems in order to reduce the Coulomb interaction. It has been shown theoretically that two principal conditions must be satisfied in order to observe the FES : (i) the strong overlap of the wave functions of spatially separated electrons and holes to ensure the effective electron-hole correlation and (ii) an abrupt Fermi edge.[6–8] Recently, it has been demonstrated that these conditions are necessary but, likely, not sufficient for such an observation.[9] We report here principally a behavior of the FES providing strong support for this latter assertion.

Typically the FES is identified: (i) in photoluminescence (PL) as an enhancement of the oscillator strength for transitions close to the Fermi edge if disorder or hole localization spreads the hole wavefunction sufficiently in k-space to allow transitions otherwise forbidden due to momentum conservation; (ii) in absorption it appears as an excitonic enhancement near the Fermi level of a factor ~ 2 at 4 K.[10] FES related effects demonstrate a strong temperature, excitation density, and magnetic-field dependence. Chen *et al.*[11] observed for $Al_xGa_{1-x}As/In_yGa_{1-y}As/GaAs$ in perpendicular magnetic field B very strong periodic intensity oscillations in B^{-1} for the PL at low temperature caused by the resonant enhancement of the FES in the highest occupied Landau level. The electron scattering by holes becomes substantially enhanced due to resonance with excitons of the empty $n=2$ electron subband. The behavior of the FES and the $n=2$ exciton under near-resonance condition between the $n=2$ subband and the Fermi energy E_F has been recently studied in $Al_xGa_{1-x}As/In_yGa_{1-y}As/GaAs$ by means of PL and PL excitation spectra under temperature and excitation density variation.[12] It has been shown that the FES quenches rapidly with a temperature and/or excitation density increase, whereas the magnitude of the $n=2$ exciton feature grows accompanied by a blueshift of the energetic position. In all previous studies the FES feature either definitely exists or definitely does not exist at low temperature. If existing, its further evolution can be followed under temperature, magnetic field, or excitation density variation. We demonstrate here for the first time the low-temperature origination of the FES in the PL spectra, although it was absent initially.

We have grown pseudomorphic modulation-doped $Al_xGa_{1-x}As/In_yGa_{1-y}As/GaAs$ heterostructures to optimize the FES observation in PL. The typical epilayer sequence consists of a GaAs buffer layer ($d=80$ nm), an undoped $In_yGa_{1-y}As$ strained quantum well (QW) (2DEG channel) ($y=0.19$, QW thickness $D=15$ nm), an $Al_xGa_{1-x}As$ undoped spacer ($x=0.2$, $d=7.5$ nm), an $Al_xGa_{1-x}As$ heavily Si-doped supplier layer ($x=0.2$, $d=35$ nm, $N_D=2\times10^{18}$ cm^{-3}), and a Si-doped GaAs cap layer ($d=5$ nm, $N_D=2.5\times10^{18}$ cm^{-3}). The electron sheet density n_s of the 2D electrons, obtained from the Hall measurements down to 4.2 K is $N_s=1.18\times10^{12}$ cm^{-2}. Double-crystal x-ray diffraction and simulation of the double x-ray rocking curve were used to verify the samples' structural parameters. The x-ray patterns indicated a high quality of the structures under investigation. The PL was excited by the 514.5-nm line of a cw Ar^+ laser. The PL radiation was dispersed through a 3/4-m Czerny-Turner scanning spectrometer, with a spectral resolution better than 0.1 meV.

In order to be more precise in energy assignments, we calculate the subband structure for the $In_yGa_{1-y}As$ QW in

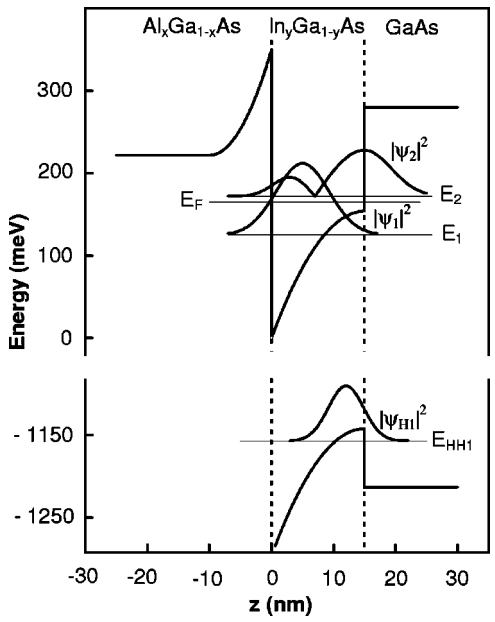

FIG. 1. Conduction- and valence-band profiles found from a coupled Schrödinger-Poisson analysis for $Al_xGa_{1-x}As/In_yGa_{1-y}As/GaAs$ heterostructure. Squared envelope functions for electrons and holes are also shown. Distance is measured from $Al_xGa_{1-x}As/In_yGa_{1-y}As$ interface position.

our heterostructures, self-consistently solving the coupled Schrödinger and Poisson equations. The Fermi level for a given temperature was calculated by iteration from the charge-neutrality condition. The subband energies and the energy eigenfunctions for the $In_{0.19}Ga_{0.81}As$ QW are shown in Fig. 1. The intersubband separation $\Delta = E_2 - E_1$ is found to be 48 meV, and E_F is about 5 meV smaller than the E_2 energy. As seen, the wave function of the $n=1$ electron state has its maximum near the $Al_xGa_{1-x}As/In_yGa_{1-y}As$ interface, while the $n=2$ electron state has its main peak near the $In_yGa_{1-y}As/GaAs$ interface, as does the hole wave function. The calculated wave functions allow one to compute the ratio of the electron-hole overlap integrals squared for the E_{21} and E_{11} transitions; this value is found to be 12.7. Based on the results derived from these calculations, one can calculate a single-peaked PL spectrum from our $Al_{0.2}Ga_{0.8}As/In_{0.19}Ga_{0.81}As/GaAs$ heterostructures.

Figure 2 shows a characteristic PL spectrum at 4.2 K [Fig. 2(a)] and the evolution of its high energy tail under different excitation densities [Fig. 2(b)]. The dominant feature (E_{11}-transition) at 1.284 eV [Fig. 2(a)] is due to the recombination of $n=1$ electrons with $n=1$ heavy holes and has a typical asymmetric line shape. The recombination reaches its maximum near the $n=1$ subband bottom ($\vec{k}=0$) and strongly decreases in intensity toward E_F. Due to the presence of impurities, the momentum conservation is broken and the transitions from electrons in states up to $k=k_F \sim 2\times10^6$ cm^{-1} become allowed. The shape of the spectral feature is well described in terms of a microscopic theory developed by Lyo and Jones[13] for steady-state PL at low temperatures in modulation-doped degenerate direct-band-gap semiconductor QW's. The following parameters were derived from the fit: $E_g = 1.278$ eV, $\Gamma = 13.8$ meV (Γ is the full width at half maximum), and $E_F = 44$ meV. The obtained E_F value coincides well with that found from the self-consistent solution of Schrödinger and Poisson equations ($E_F = 43$ meV). The low-energy PL threshold on the energy axis [see Fig. 2(a)] is slightly redshifted with respect to the E_{11} peak position. This is a consequence of 2D-carrier scattering by charge impurities strongly depending on the thickness of the spacer layer. These observations demonstrate that the PL profile is primarily determined by electron-hole recombination accompanied by impurity and disorder-assisted processes, resulting in broadening and blue-shifting of the PL maximum.

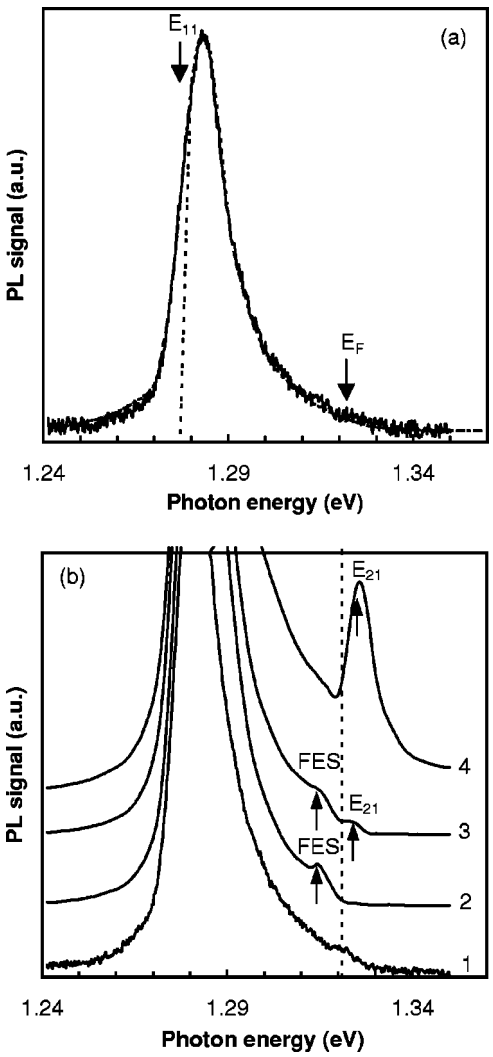

FIG. 2. PL spectrum from a $Al_{0.2}Ga_{0.8}As/In_{0.19}Ga_{0.81}As/GaAs$ heterostructure at 4.2 K under various excitation intensity: (a) PL spectrum at low excitation power of 0.0012 I_0, where I_0 is the maximum intensity used and approximately 20 W/cm^2. Dashed line presents the fit in terms of Lyo and Jones theory [11]. The low-energy tail is approximated by the Urbach form. Arrows stand for E_{11} and E_F energies. (b) Transformation of the high energy PL tail under intensity elevation. Curves 1 to 4 correspond to the intensities: 1 - 0.0025 I_0, 2 - 0.01 I_0, 3 - 0.1 I_0, 4 - I_0, respectively. Transition energies for the FES and the $n=2$ exciton are shown by arrows pointing up.

The FES is recognized by the transformation of the PL line shape in the close vicinity of E_F under excitation density increase [Fig. 2(b)]. Taking into account the close proximity of E_F and E_2 in our experiment, a strong interaction between

the elementary excitations near the Fermi level and the $n=2$ bare (atomic) exciton is expected. Several surprising new characteristics arise from this interaction. Figure 2(b) shows that the PL tail spreads far beyond the E_F boundary at the modest excitation level. At very low excitation, transitions associated with neither the FES nor the $n=2$ exciton are seen. Increasing the excitation density, luminescence from the FES clearly develops with a pronounced shift toward the energies below E_F. The steepness of the spectral drop fall after the FES feature abruptly grows and no PL signal is detected behind E_F. Further increase of the excitation density results in the appearance of a $n=2$ feature (E_{21}-transition) moving toward higher energy. Finally, the E_{11} and E_{21} peaks are present only at the highest intensities and the FES decays completely.

The optical oscillator strength for electrons at the Fermi level recombing with photo-generated holes becomes enhanced by over two orders of magnitude when the continuum of Fermi sea excitations is resonantly coupled with the exciton state.[7] This coupling is described in terms of Fano interference.[14] The interaction between an exciton and 2D electrons in asymmetrically-doped single $In_yGa_{1-y}As$ QW's was also discussed by Chen *et al.* in terms of a Fano resonance[11] without solving the many-body problem. A more detailed theoretical treatment including the band-gap renormalization, static screening, and dynamical response of the Fermi sea has been carried out by Hawrylak.[7] With inclusion of the hole selfenergy, the position of the actual emission line falls close to the bare excitonic transition, and in absorption two peaks arise, one related to the bound exciton state. For increasing carrier density the emission spectrum resembles the single-particle density of states in the conduction band with enhancement at the Fermi level, while the absorption spectrum evolves into a blueshifted FES. This predicted behavior is somewhat obscured both in our data and in those presented in Ref. 12.

Indeed, at low temperature and low pumping level, the high-energy tail of the PL is reasonably described in terms of the electron-heavy hole recombination contributed by impurity and disorder-assisted processes [Fig. 2(a)]. The FES enhancement, if exists, is of a negligible magnitude. Elevating the excitation density in addition to excitations of the Fermi sea, an increasing contribution from excitations including holes in the $n=1$ heavy-hole subband and electrons in the second subband arises. Due to strong resonance coupling of excitons with excitations of the Fermi sea, two hybridized states appear, separated by the region of destructive Fano interference. The state related to the Fermi edge is energetically shifted toward lower energy and the state related to hybridized excitons shifts to higher energy. At these relatively low-excitation levels, the states related to the second conduction subband exciton cannot be observed in PL because the $n=2$ electron level is empty according to Fermi statistics with the electron temperature measured to be approximately 9 K. The result is the characteristic FES enhancement of the PL with an abrupt fall-off on the high energy side due to the Fano interference. A further increase of excitation density leads to the population of the $n=2$ electron level and observation of the E_{21} transition, corresponding to the hybridized excitonic state [curve 3 in Fig. 2(b)]. At still higher excitation intensity, both the FES and excitonic emission are quenched and the free-2D electron-hole recombination dominates the high-energy PL tail [curve 4 in Fig. 2(b)].

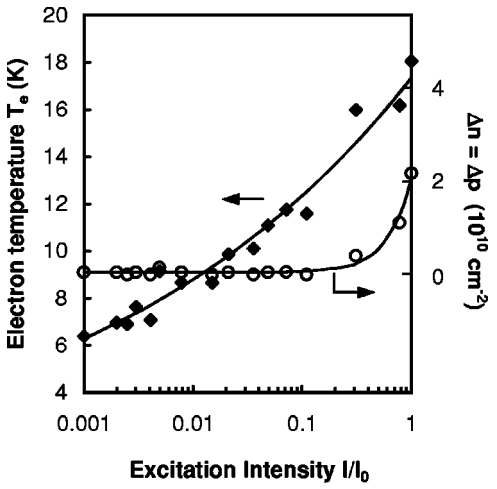

FIG. 3. Excess electron concentration compared to the thermal equilibrium value and electron temperature as functions of excitation intensity. The maximum intensity I_0 is approximately 20 W/cm^2.

The Fermi level and the electron density can be determined by fitting the detailed line shape of the photoluminescence to a theoretical model.[15] Figure 3 depicts the excess electron density due to optical excitation as determined from such a fit. It also shows the electron temperature as determined from the shape of the high-energy tail of the PL shown in Fig. 2. The data of Fig. 3 indicated that at the highest excitation density studied here, $I_0=20$ W/cm^2, is sufficient to result in population of the n=2 electronic subband in the $In_yGa_{1-y}As$ QW.

Figure 4 shows the development energies of the FES and $n=2$ related features as the intensity is varied. At the lowest excitation intensities, no PL that can be attributed to the FES is observed. At about 40 mW/cm^2, the FES appears near the Fermi energy, immediately shifting to pronounced lower energies as the excitation intensity is increased. This shifting

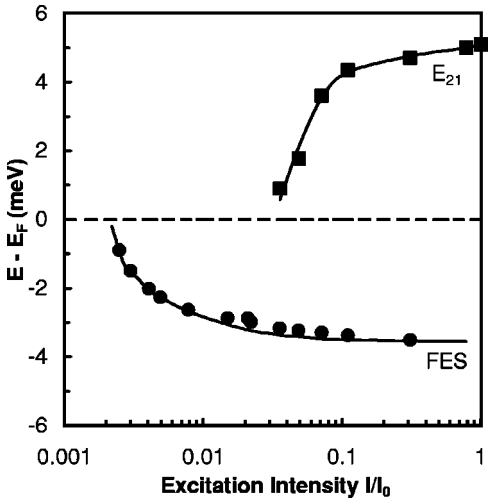

FIG. 4. Shift by energy for the FES and $n=2$ related features in the PL spectrum under excitation power increase. The maximum intensity I_0 is approximately 20 W/cm^2.

essentially saturates when the hybridized $n=2$ exciton state appears. The $n=2$ related feature undergoes a pronounced blueshift after its appearance with a tendency to saturation, reflecting the screening of the $n=2$ exciton by excess electrons. It is noteworthy that the spectral shifts within the E_F proximity are not accompanied by any noticeable shift of the E_{11} transition when the excitation level increases. The blueshift of the E_{21} peak is interpreted in terms of the transition from the hybridized exciton state to the bare (atomic) exciton,[10–12,16] if the screening of the $n=2$ exciton by excess electrons is included. Insensitivity of the transition E_{11} to the pumping density shows that the internal electrical field does not change with intensity, and therefore the blueshift saturation for the E_{21} transition is mainly connected with the exciton screening and the transition from the excitonic recombination to the free-electron-hole recombination. In fact, we show here that the carrier concentration strongly influences the character of both the FES and the $n=2$ exciton. This result is consistent with the conclusion derived from PL investigations of low-doped $In_xGa_{1-x}As/InP$ heterostructures.[9] Here, it was shown that in spite of the fact that both a close position of the Fermi edge to the empty $n=2$ electron subband and a hole localization by alloy fluctuation potential provide the optimal conditions for the occurrence of the FES, these conditions are not sufficient for the FES enhancement, but, rather, the carrier concentration also plays a role. We conclude that even modest variation of the concentration, caused by light illumination or heating, can substantially influence the FES development. Indeed, an elevation of the sample temperature from 4.2 to 15 K gives rise to a substantial broadening and subsequent smearing out the FES related feature in PL.

In summary, we observe a fundamental change in the PL spectrum as function of excitation density reflected in the high-energy tail due to the mutual repulsion of the FES and the excited-subband excitonic states. The details of this interaction depend critically on the excitation density and temperature. At low temperature, the dependence of the existence of the FES feature on excitation density has been observed for the first time in PL. The FES appearance is accompanied by the formation of an abrupt high-energy edge and occurs far lower in intensity than the luminescence associated with the hybridized $n=2$ exciton. Strong screening of the $n=2$ exciton state by photoexcited carriers is observed, resulting in 2D electron gas heavy-hole recombination for second electronic subband. A strong excitation power dependence of the 2D-related PL could be the result of the photoinduced change of the potential distribution across the QW structure. Such an effect has also been previously observed in $Al_xGa_{1-x}/GaAs$ quantum structures.[17,18] A strong band bending can arise from the neutralization of the ionized residual acceptors by photocreated holes in the $In_yGa_{1-y}As$ channel and the neutralization of the ionized donors in the barriers by photocreated electrons. The reduction of the band bending under illumination leads to a characteristic blueshift of the PL spectrum. We have not observed a noticeable blue-shift of the PL spectrum and, hence, these explanations must be ruled out. We believe that the behavior of the PL near the Fermi edge must be explained by a strong carrier density effect on the FES which has not yet been explored theoretically.

This work was supported in part by a NATO Linkage Grant.

*Electronic address: kissel@physik.hu-berlin.de

[1] C. Delalande, G. Bastard, J. Orgonasi, J. A. Brum, H. W. Liu, M. Voos, G. Weimann, and W. Schlapp, Phys. Rev. Lett. **59**, 2690 (1987).

[2] M. S. Skolnick, J. M. Rorison, K. J. Nash, D. J. Mowbray, P. R. Tapster, S. J. Bass, and A. D. Pitt, Phys. Rev. Lett. **58**, 2130 (1987).

[3] Y.-C. Chang and G. D. Sanders, Phys. Rev. B **32**, 5521 (1985).

[4] D. Heiman, B. B. Goldberg, A. Pinczuk, C. W. Tu, A. C. Gossard, and J. H. English, Phys. Rev. Lett. **61**, 605 (1988).

[5] J. Rubio, H. P. van der Meulen, J. M. Calleja, V. Haïrle, R. Bergmann, and F. Scholz, Phys. Rev. B **55**, 16 390 (1997).

[6] T. Uenoyama and L. J. Sham, Phys. Rev. B **39**, 11 044 (1989).

[7] P. Hawrylak, Phys. Rev. B **42**, 8986 (1990); **44**, 3821 (1991); **44**, 6262 (1991); **45**, 4237 (1992).

[8] H. P. van der Meulen, I. Santa-Olalla, J. Rubio, J. M. Calleja, K. J. Friedland, R. Hey, and K. Ploog, Phys. Rev. B **60**, 4897 (1999).

[9] I. A. Buyanova, T. Lundström, A. V. Buyanov, W. M. Chen, W. G. Bi, and C. W. Tu, Phys. Rev. B **55**, 7052 (1997).

[10] T. A. Fisher, P. E. Simmonds, M. S. Skolnick, A. D. Martin, and R. S. Smith, Phys. Rev. B **48**, 14 253 (1993).

[11] W. Chen, M. Fritze, W. Walecki, A. V. Nurmikko, D. Ackley, J. M. Hong, and L. L. Chang, Phys. Rev. B **45**, 8464 (1992).

[12] S. J. Xu, S. J. Chua, X. H. Tang, and X. H. Zhang, Phys. Rev. B **54**, 17 701 (1996).

[13] S. K. Lyo and E. D. Jones, Phys. Rev. B **38**, 4113 (1988).

[14] U. Fano, Phys. Rev. **124**, 1866 (1961).

[15] K. H. Herrmann, M. Happ, H. Kissel, K.-P. Möllmann, J. W. Tomm, C. R. Becker, M. M. Kraus, S. Yuan, and G. Landwehr, J. Appl. Phys. **73**, 3486 (1993).

[16] C. Colvard, N. Nouri, H. Lee, and D. Ackley, Phys. Rev. B **39**, 8033 (1989).

[17] I. V. Kukushkin, K. von Klitzing, K. Ploog, and V. B. Timofeev, Phys. Rev. B **40**, 7788 (1989).

[18] Y. R. Yuan, K. Mohammed, M. A. A. Pudensi, and J. L. Merz, Appl. Phys. Lett. **45**, 739 (1984).