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Role of hole localization in the optical singularities of a two-dimensional electron gas studied by time-resolved photoluminescence

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Abstract. The coexistence of localized and free holes in modulation doped InGaAs/InP quantum wells, which show Fermi edge singularities in the cw photoluminescence and excitation spectra, has been determined by picosecond time-resolved luminescence spectroscopy. A long-lived peak at the low-energy side of the emission spectrum is observed at 5 K and weak excitation. The peak, which vanishes on either increasing the temperature at ∼30 K or increasing the excitation density, is attributed to recombination of free electrons with localized holes. The simultaneous presence of free and localized holes explains the lineshapes observed in cw measurements and the presence of Fermi edge singularities.

1. Introduction

The presence of free carriers in a semiconductor quantum well (QW) modifies most notably its optical properties. A strong enhancement in the oscillator strength of optical transitions in the vicinity of the Fermi edge has been reported both in absorption and emission of modulation doped QWs [1–6]. This enhancement, known as Fermi edge singularity (FES), results, in n-type samples, from many-body interactions between a photoexcited hole and the sea of electrons in the QWs [7]. Due to the difficulties of controlling some crucial experimental parameters, such as the valence band structure, the intersubband coupling and the localization of the carriers by impurities or potential fluctuation, the physical mechanisms giving rise to the singularities are still not fully understood. Photocreated holes in semiconductors are free to move in the valence band and therefore its recoil reduces the FES. Furthermore, to observe the FES in emission, non-k-conserving transitions have to contribute to the photoluminescence (PL) spectrum [2]. Localization of the holes, for example by random alloy fluctuations, allows efficient recombination of electrons up to their Fermi energy and enhances the electron–hole interaction due to the fact that the hole becomes a practically infinite effective mass.

The temporal evolution of FES has been studied by time-resolved photoluminescence (TRPL) spectroscopy in a two-dimensional hole gas in GaAs QWs [8]. In this work, we present a study of the temperature and excitation-density dependence of the TRPL spectra in n-type modulation doped QWs, which show FES in the cw PL and excitation (PLE) spectra. Our results confirm the coexistence of localized and free valence band holes in these samples.

2. Experimental details

We have investigated three single $In_{0.53}Ga_{0.47}As/InP$ QWs grown by low-pressure metal-organic vapour phase epitaxy on InP substrates. Two 10 nm thick layers containing 1.7×10^{18} cm^{-3} S atoms are symmetrically located in the InP barriers around the 8 nm wide QWs. The different widths of the space layer separating the dopants from the well, 30, 15 and 5 nm, lead to sheet-carrier concentrations of 5×10^{11}, 8×10^{11} and 1.2×10^{12} cm^{-2}, and to Fermi energies of 27, 42 and 72 meV respectively. Typical electron mobilities at low temperature are $\sim 1.8 \times 10^5$ cm^2 V^{-1} s^{-1}. The electron

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densities and Fermi energies are obtained from Shubnikov–de Haas oscillations and from the width of the PL spectra respectively.

Photoluminescence and PLE spectra were taken in an He bath cryostat, exciting the samples with the light from a tungsten lamp passed through a grating monochromator. The emission was analysed in a 0.85 m focal length double spectrometer and detected by a liquid nitrogen cooled Ge detector. TRPL spectra were obtained using an up-conversion spectrometer (5 ps resolution), with the samples mounted in a cold finger cryostat. Pulses from a Styryl 8 dye laser, synchronously pumped by a mode-locked Nd:YAG laser, were used to excite the samples. We will concentrate here on the sample with the lowest carrier density.

3. Results and discussion

The cw PL (dotted curve) and excitation (•) spectra of the sample with an electron density $n = 5 \times 10^{11}$ cm^{-2} are shown in figure 1. The shift between the PL and PLE arises from the phase-space filling by the electrons, which blocks the absorption underneath the Fermi energy in the conduction band. The PL spectrum of a two-dimensional (2D) electron gas depends on the degree of hole localization: since the joint density of states is energy independent, the lineshape is determined by the energy distribution of the photocreated holes and the possible relaxation of the momentum conservation. For free holes with finite mass, the PL has a maximum at energies corresponding to transitions to the top of the valence band and a monotonic decrease at higher energies following a Boltzman distribution. On the other hand, the spectrum is square-like if the holes are completely localized and k is no longer conserved. Many-body interactions strongly modify the single particle picture: exchange and correlation introduce corrections to the self-energy of the carriers, and the electrons at the Fermi level respond collectively to the presence of a photocreated hole, giving rise to an FES [9]. The intensity of the FES increases with the hole mass as a result of the reduction of hole recoil [1].

The Fermi edge singularities are clearly observed as peaks at the high energy side of the PL and at the beginning of the pseudo-absorption spectrum in figure 1. Transitions to the second electron subband (E_1) can be recognized at an energy of ∼1 eV. The strong temperature dependence of the peak labelled FES in the PL is documented in the inset of the figure: on increasing the temperature from 5 K to 30 K the peak vanishes. This dependence confirms the assignment of this structure to a Fermi edge singularity [7]. The step-like onset of the low-temperature PL, reflecting the 2D density of states, indicates that localized holes are involved in the emission. This fact is also evidenced in the low-energy tail of the PL arising from phonon replica processes, which require hole localization to preserve k conservation [6, 10].

Figure 2 shows low-temperature TRPL spectra taken 50 ps after excitation at power densities ranging from 1 W cm^{-2} to 30 W cm^{-2}. The excitation energy was 1.68 eV, above the InP bandgap. The low energy peak

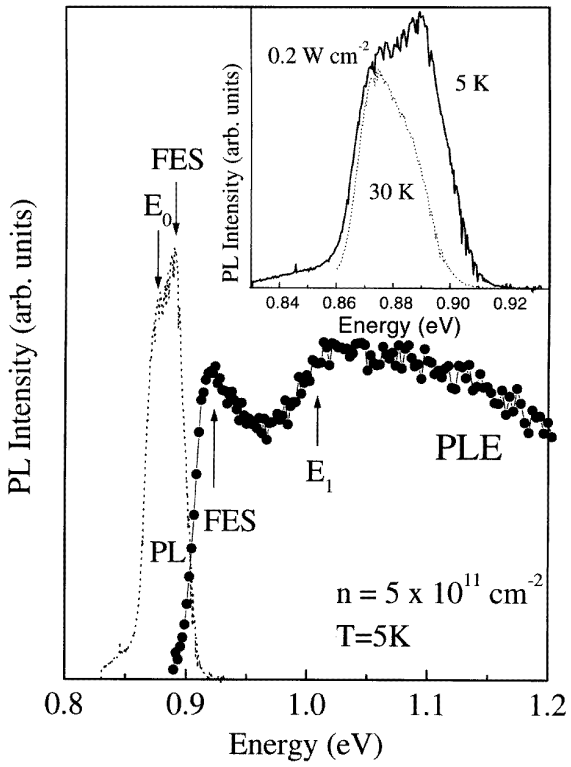

Figure 1. Low-temperature photoluminescence (PL, dotted curve) and excitation (PLE, •) spectra of an *n*-modulation doped ($n = 5 \times 10^{11}$ cm^{-2}) $In_{0.53}Ga_{0.47}As$/InP quantum well (8 nm width). E_0 and E_1 denote transitions from the bottom of the first and second electronic subbands respectively. Fermi edge singularities are labelled with FES. Inset: PL spectra at two temperatures.

(E_0) corresponds to recombination of holes with electrons in the Fermi sea of the first subband. The small blue shift of this peak with increasing power density denotes the displacement towards higher energies of the product of the density of states and the subband occupation with increasing band filling. Concurrently, the onset of the peak red-shifts as a consequence of band gap renormalization [11]. The second peak, E_1, assigned to recombination of electrons from the second subband, shows also a small blue shift with increasing power density. The lineshapes of these spectra are very similar to those obtained under cw illumination at moderate and high excitation densities [11]. The behaviour of the PL between 0.93 eV and 1 eV indicates that the carrier temperature increases with power, in agreement with previous reports [12, 13]. Only for power densities below 5 W cm^{-2} does the spectral range of the PL become comparable with that of the cw spectrum (inset). However, even at the lowest power used in our experiments, the carrier temperature is still too high to allow a direct observation of the FES in time-resolved measurements. The reduction of the FES at high densities of photoexcited carriers and/or high temperatures has been also observed in cw measurements [14].

The time evolution of the PL at two different detection energies is shown in figure 3 for a power density of 30 W cm^{-2}. The inset depicts the corresponding TRPL spectrum at 50 ps (the arrows mark the detection energies).

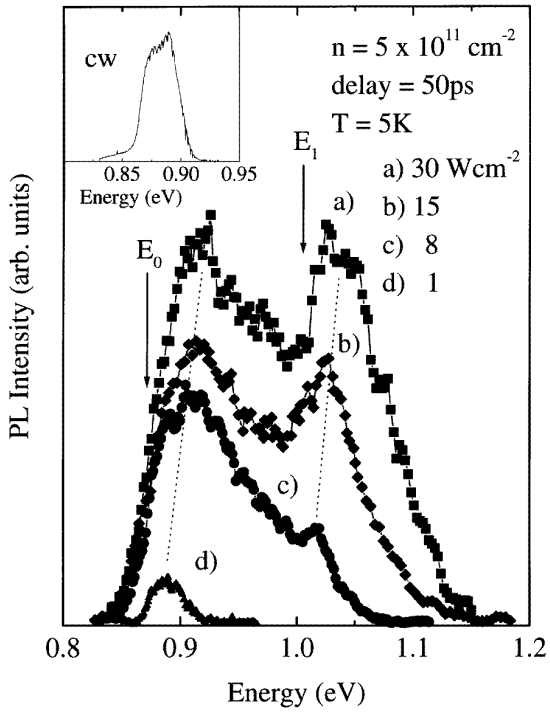

Figure 2. Time resolved PL spectra, corresponding to the sample of figure 1, taken 50 ps after excitation at 1.68 eV, for different excitation densities. The beginning of the PL from the first (E_0) and second (E_1) subbands is marked by arrows. Inset: cw PL for comparison.

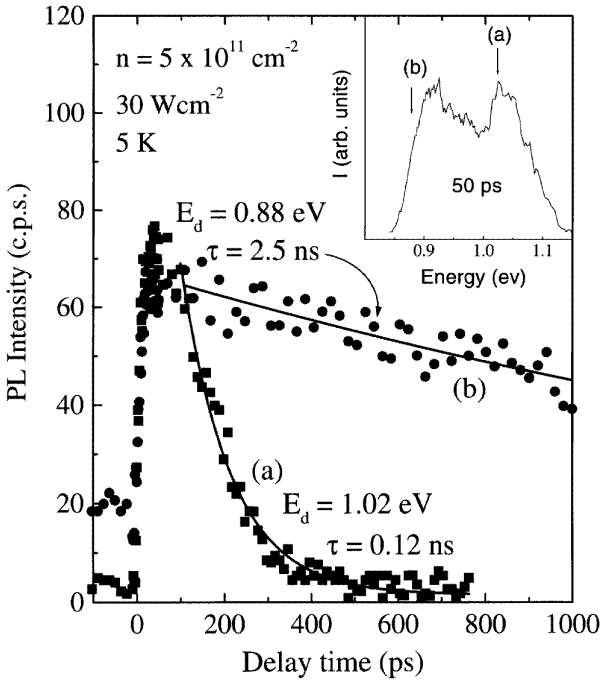

Figure 3. Time evolution of the PL for an excitation power density of 30 W cm^{-2} at two different detection energies: (a) E_d = 1.02 eV (■), (b) E_d = 0.88 eV (●). The curves show the best fit to an exponential decay. The inset shows the TRPL spectrum at 50 ps and 30 W cm^{-2}; the arrows mark the detection energies.

The rise-times are of the order of 10–20 ps. The second subband presents a fast decay with a time constant of 120 ps, comparable with that of the barrier PL (not shown). The decay time grows considerably for the first subband to 2.5 ns, detecting at 0.88 eV. This time is considerably longer than the one obtained for 8 nm wide undoped $In_{0.53}Ga_{0.47}As/InP$ QWs, which amounts to 0.9 ns [15]. The slow decay time of the first subband gives rise to the signal at negative times in trace (b), which arises from the emission excited by a previous pulse (12 ns in advance).

Lowering the excitation density to 4 W cm^{-2}, the contribution from the second subband disappears (see inset in figure 4). The high-energy side of the first-subband PL shows rise and decay times of ∼150 ps and 1.9 ns respectively, as shown in trace (a) of figure 4. Lowering the detection energy, the time evolution of the PL changes considerably: at 0.88 eV (figure 4, trace (c)), the rise and decay times increase to ∼1 ns and ∼6 ns, respectively†. Similar results to those presented in this figure are obtained for excitation below the bandgap of InP, which excludes a slow transfer of carriers from the barrier to the QW as the mechanism responsible for the increase in the rise and decay times. The long decay time indicates a contribution from localized holes, which have a large lifetime due to their small wavefunction overlap with free electrons. The origin of the localization could arise from potential fluctuations at the interfaces, which have been found to be responsible for the localization of excitons in InGaAs/GaAs QWs [16], or from alloy disorder in the InGaAs QW [10].

The contribution of the localized holes to the emission is more evident in the spectral lineshape of the PL. A new peak (L) becomes resolvable in the low-energy side of the spectrum at long times as can be seen clearly in figure 5, which compiles TRPL spectra of the first subband at different times. The spectrum taken 1 ns after excitation (●) has a maximum at ∼0.88 eV (peak L), while at short times (○) the maximum of the PL lies at ∼0.885 eV (peak labelled F). A lineshape analysis of the spectra gives an energy separation of the maxima of 9 meV and a ratio of the areas A_L/A_F which increases from 0.1 at 50 ps to 0.4 at 1 ns. The high temperature of the carriers at short times hinders the observation of peak L, which we attribute to recombination of 2D electrons with localized holes. Additional proof of the localization of the holes is obtained from the temperature dependence of the TRPL spectra, as shown in the inset of figure 5. Increasing the temperature from 5 K to 30 K is enough to free a considerable number of holes and therefore the peak L becomes less prominent in the PL spectra. Note that the behaviour of the TRPL spectra with increasing temperature is opposite to that of the cw PL, indicating that the temperature of the carriers is still too high in the time-resolved experiments to allow the observation of the FES. A related shift of the emission towards low energies with increasing time delay has been observed in the time-resolved photoluminescence of localized excitons in GaAs/GaAlAs QWs [17], but no additional peak has been resolved in the spectra in this case. Similarly to

† The decay time has been estimated from the signal at negative times in trace (c).

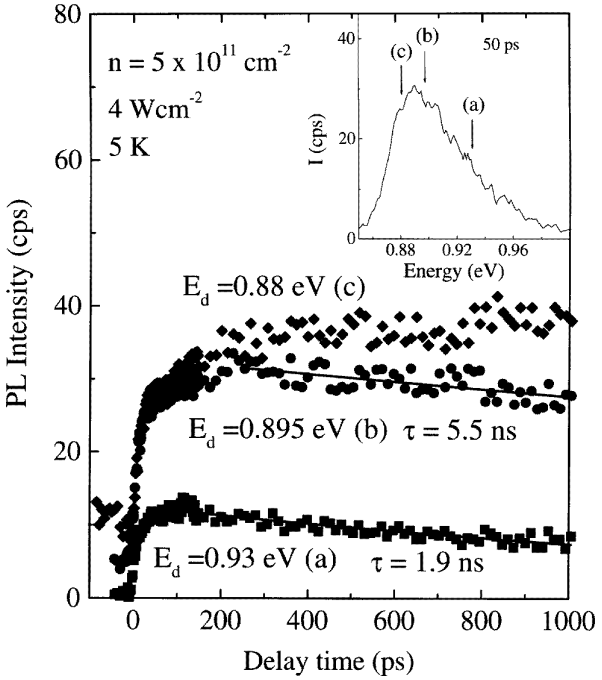

Figure 4. Time evolution of the PL for an excitation power density of 4 W cm^{-2} at three different detection energies: (a) E_d = 0.93 eV (■), (b) E_d = 0.895 eV (●), (c) E_d = 0.88 eV (♦). The curves show the best fit to an exponential decay. The inset shows the TRPL spectrum at 50 ps and 4 W cm^{-2}; the arrows mark the detection energies.

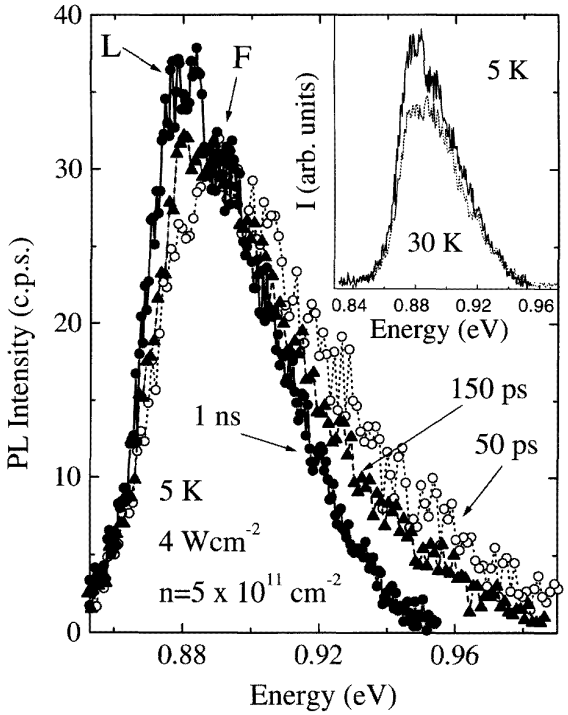

Figure 5. TRPL spectra at different time delays for low-power excitation. The inset shows the temperature dependence of the spectra at 1 ns.

the temperature effect, an increase of the excitation power density (≥8 W cm^{-2}) leads to the disappearance of peak L and the PL spectrum becomes temperature independent (up to ∼60 K). These two facts can be easily explained as a result of an increase in the carrier temperature with excitation density, which also reduces the effects of the lattice temperature.

The dependence of peak L on temperature and photoexcited carrier density resembles that of charged excitons [18–21], which show an intensity decrease relative to that of free excitons with increasing excitation density and temperature. However, in contrast with the dependence found for charged excitons [20, 21], our results are independent of excitation energy. The similarity between excitation below and above the bandgap of the InP excludes a transfer of carriers from the barriers to the QW as the mechanism responsible for the decrease of the intensity of peak L.

4. Conclusions

In summary, we have studied the dynamics of the optical singularities in the emission spectra of a two-dimensional gas of electrons by time-resolved photoluminescence spectroscopy. The temperature and excitation density dependence of the time-resolved spectra establish the coexistence of localized and free valence band holes in samples which present Fermi edge singularities in the cw spectra. Although we do not demonstrate explicitly the indispensability of hole localization to obtain Fermi edge singularities in the optical spectra, since these are not observed in the time-resolved measurements, our results strongly suggest that hole localization provides the mechanism to break the k-conservation needed to observe the singularities in emission. This study demonstrates the importance of the dynamics of the recombination processes in reaching a correct understanding of the role of different carriers types in highly correlated systems.

Acknowledgments

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References

[1] Mahan G D 1981 *Many Particle Physics* (New York: Plenum)
[2] Skolnick M S, Rorison J M, Nash K J, Mowbray D J, Tapster P R, Bass S J and Pitt A D 1987 *Phys. Rev. Lett.* **58** 2130
[3] Chen W, Fritze M, Walecki W, Nurmikko A V, Ackley D, Hong J M and Chang L L 1992 *Phys. Rev.* B **45** 8464
[4] Livescu G, Miller D A B, Chemla D S, Ramaswamy M, Chang T Y, Sauer N, Gossard A C and English J H 1988 *IEEE J. Quantum Electron.* **24** 1677
[5] Mélin T and Laruelle F 1996 *Phys. Rev. Lett.* **76** 4219
[6] Rubio J, Meulen H P, Calleja J M, Bergmann R and Scholz F 1997 *Phys. Rev.* B at press

[7] Schmitt-Rink S, Ell C and Haug H 1986 *Phys. Rev.* B **33** 1186
[8] Richards D, Wagner J, Schneider H, Hendorfer G, Maier M, Fischer A and Ploog K 1993 *Phys. Rev.* B **47** 9629
[9] Schmitt-Rink S, Chemla D S and Miller D A B 1989 *Adv. Phys.* **38** 89
[10] Skolnick M S, Nash K J, Saker M K and Bass S J 1994 *Phys. Rev.* B **50** 11 771
[11] Kulakoovskii V D, Lach E, Forchel A and Grützmacher D 1989 *Phys. Rev.* B **40** 8087
[12] Tatham M, Taylor R A, Ryan J F, Wang W I and Foxon C T 1988 *Solid State Electron.* **31** 459
[13] Tomita A, Shah J, Cunningham J E, Goodnick S M, Lugli P and Chuang S L 1993 *Phys. Rev.* B **48** 5708
[14] Kalt H, Leo K, Cingolani R and Ploog K 1989 *Phys. Rev.* B **40** 12 017
[15] Cebulla U, Bacher G, Forchel A, Mayer G and Tsang W T 1989 *Phys. Rev.* B **39** 6257
[16] Martelli F, Polimeni A, Patanè A, Capizzi M, Bori P, Gurioli M, Colocci M, Bosacchi A and Franchi S 1996 *Phys. Rev.* B **53** 7421
[17] Zhou P, Jiang H X, Bannwart R, Solin S A and Bai G 1989 *Phys. Rev.* B **40** 11 865
[18] Kheng K, Cox R T, d'Aubigne Y M, Bassani F, Saminadayar K and Tatarenko S 1993 *Phys. Rev. Lett.* **71** 1752
[19] Finkelstein G, Strikman H and Bar-Joseph I 1995 *Phys. Rev. Lett.* **74** 976
[20] Shields A J, Osborne J L, Simmons M Y, Pepper M and Ritchie D A 1995 *Phys. Rev.* B **52** 5523
[21] Shields A J, Foden C L, Pepper M, Ritchie D A, Grimshaw M P and Jones G A C 1994 *Superlatt. Microstruct.* **15** 355