Single carrier localization in $In_xGa_{1-x}As_{1-y}N_y$ investigated by magnetophotoluminescence

A. Polimeni[a)] and F. Masia
INFM-Dipartimento di Fisica, Universita' di Roma "La Sapienza," Piazzale A. Moro 2, 00185 Roma, Italy

A. Vinattieri
INFM-Dipartimento di Fisica and LENS, Universita' di Firenze, via Sansone 1, 50019 Sesto Fiorentino, Italy

G. Baldassarri Höger von Högersthal and M. Capizzi
INFM-Dipartimento di Fisica, Universita' di Roma "La Sapienza," Piazzale A. Moro 2, 00185 Roma, Italy

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We investigated the origin of radiative recombination in $In_xGa_{1-x}As_{1-y}N_y$/GaAs quantum wells by photoluminescence (PL) after picosecond excitation and under a magnetic field, B. Continuous wave and time-resolved PL show that at low temperature T localized states are mainly involved in the radiative recombination processes. Most importantly, the shift of the PL peak position induced by B depends dramatically on temperature, being higher at lower T. This result indicates that the PL emission at low temperature is determined by the recombination of loosely bound electron-hole pairs in which one carrier is localized by N-induced potential fluctuations, and the other carrier is delocalized. © *2004 American Institute of Physics.* [DOI: 10.1063/1.1688003]

Recombination from localized states generally dominates emission processes at low temperature in semiconductor alloys.[1–4] Local fluctuations in the composition lead to an exponential tail of localized states within the crystal forbidden gap.[1,5] The preferential occupancy of these low-energy states by carriers at low T is responsible for asymmetric photoluminescence (PL) spectra,[2,4,6–11] a blueshift of the PL peak energy as the excitation power increases,[7–10,12] a decreasing emission decay time of PL with increasing emission energy,[4,8–10,13,14] and an anomalous dependence of the PL maximum energy on temperature.[4,9,11,13] All these effects are particularly important in $In_xGa_{1-x}As_{1-y}N_y$, a material of relevance for optoelectronic and solar cell applications.[15] Usually, it is assumed that radiative recombination from states induced by composition disorder is excitonic in nature. Very recently, this assumption has been questioned for $GaAs_{1-y}N_y$, where the fast rise time ($\sim$25 ps) of the PL signal was used to establish that radiative recombination occurs between localized electrons and delocalized holes.[16]

Here, we employed time-resolved and magneto-PL spectroscopy at different temperatures to ascertain the nature of radiative recombination in $In_xGa_{1-x}As_{1-y}N_y$/GaAs quantum wells (QWs). Localized (delocalized) states dominate the PL spectra at temperatures lower (higher) than 100 K, as deduced from power, temperature, and decay time studies. We found that the PL rise time is independent of temperature and detection energy, thus not being conclusive about the origin of the states involved in the emission processes. The shift of the PL peak energy induced by a magnetic field (B up to 12 T) decreases sizably and changes its dependence on B from linear to quadratic when going from low to high T. These findings suggest that low temperature PL arises from the recombination between localized and delocalized carriers. Moreover, from an analysis of the magneto-PL data, we derived the in-plane effective mass m^* of the delocalized carrier. The values of m^* are in good agreement with those of the in-plane effective mass of heavy (light) holes in $In_xGa_{1-x}As$ (GaAs).

We investigated $In_xGa_{1-x}As_{1-y}N_y$/GaAs single QWs having x (y)=0.0 (0.016), 0.25 (0.011), 0.34 (0.007), 0.38 (0.052), and a QW thickness of L=9.0, 6.0, 7.0, 8.2 nm, respectively. All samples were grown by solid source molecular beam epitaxy. Continuous wave (cw) PL was excited by the 515 nm line of an Ar^+ laser or the 532 nm line of a vanadate-yttrium aluminum garnet (YAG) laser, dispersed by a double 3/4 m monochromator, and detected by a N-cooled Ge detector or by a N-cooled InGaAs linear array. The magnetic field (B=0–12 T) was applied parallel to the growth axis of the samples. Time-resolved (TR) PL measurements were performed by using a mode-locked Ti:sapphire laser, pumped by a cw Ar^+ laser, and providing 1.2 ps pulses with a repetition rate of 81 MHz. The typical average excitation density was 10 W/cm^2. The PL was collected, dispersed through a 25 cm flat field monochromator, and detected by a S1 streak camera with a resolution of 4 ps.

First, we describe the results of TR PL. Figure 1 shows the PL decay curves for different energies of a 6.0-nm-thick $In_{0.25}Ga_{0.75}As_{0.989}N_{0.011}$ QW at T=10 and 140 K. At T=10 K, the decay time τ_D decreases from 500 ps to 100 ps with increasing emission energy. This decrease is consistent with the presence of localized states, mainly on the low-energy side of the PL spectra, as reported elsewhere.[8–10,13,14] τ_D remains constant ($\sim$100 ps) with emission energy at T=140 K due to the ionization of carriers from localization centers. The rise time τ_R measures how quickly carriers relax into the states from which they recombine. In Ref. 16, the very small τ_R values ($\sim$25 ps) measured in $GaAs_{1-y}N_y$ epilayers were considered to be inconsistent with the formation

a)Electronic mail: polimeni@roma1.infn.it

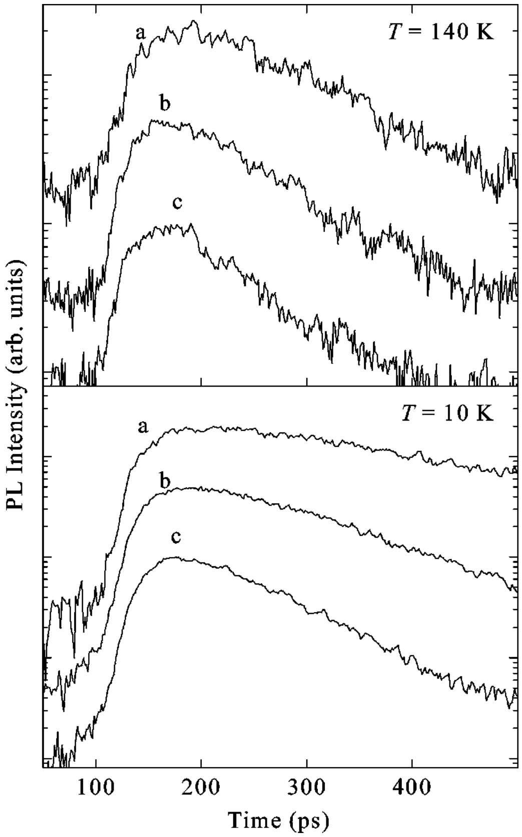

FIG. 1. (Lower panel) Decay curves of photoluminescence at $T=10$ K for an $In_{0.25}Ga_{0.75}As_{0.989}N_{0.011}$ QW recorded on the (a) low-energy side, (b) maximum, and (c) high-energy side of the cw PL spectrum. (Upper panel) The same as the lower panel, except for $T=140$ K.

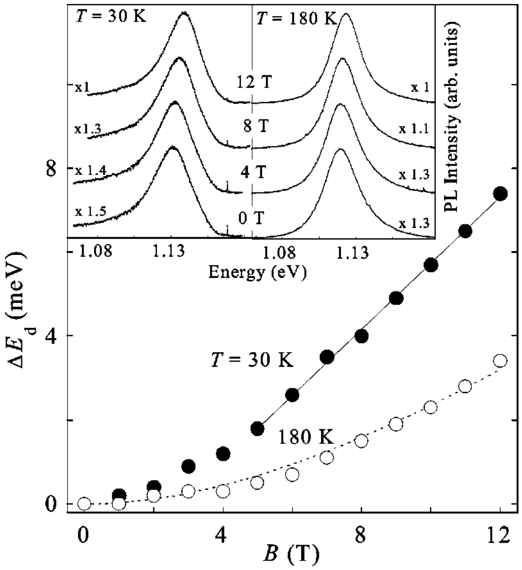

FIG. 2. Diamagnetic shift ΔE_d of the PL peak measured in an $In_{0.25}Ga_{0.75}As_{0.989}N_{0.011}$ QW at $T=30$ K (full dots) and $T=180$ K (open circles) vs magnetic field B. The dashed curve is a fit to the $T=180$ K data of a model for 2D Coulombic systems at arbitrary magnetic fields (Refs. 19 and 20). The continuous line is a fit to the $T=30$ K data ($B>4$ T) by $\Delta E_d=(e\hbar/2m_h^*)B$, where m_h^* is the in-plane hole effective mass. The inset shows the PL spectra at representative magnetic field values and at different temperatures. Multiplication factors are given. The laser power density is $P=15$ mW/cm^2 and 20 W/cm^2 for $T=30$ and 180 K, respectively.

and recombination of excitons. A new recombination mechanism was then proposed, where a delocalized hole annihilates with a localized electron.[16] In our samples, τ_R is equal to ~ 50 ps (estimated as the time elapsed between 10% and 90% of the PL signal at maximum), independent of T and emission energy. Moreover, on the basis of PL measurements at a different temperature and excitation power density (not shown here),[12] the radiative recombination in our $In_xGa_{1-x}As_{1-y}N_y$ samples is determined by localized states in a finite number for $T<100$ K, and by delocalized states for $T>100$ K. Following the arguments of Ref. 16, one would expect an increase in the rise time for $T\sim 100$ K. However, carrier relaxation becomes faster (namely, τ_R decreases) as T increases due to efficient carrier-phonon interaction.[17] No clear variation of τ_R with T has been found in our samples or in those of Ref. 16, whereas a decrease in τ_R has been reported between $T=4$ K ($\tau_R\sim 200$ ps) and room temperature ($\tau_R<50$ ps) in Ref. 13. As a consequence, the rise time alone cannot be considered an unambiguous test of the nature of the states involved in the recombination processes.

The degree of localization and/or confinement of carriers in semiconductor heterostructures can be investigated through the dependence of carrier energy levels on B as measured, for instance, by magneto-PL.[18] Preliminary measurements show that the shift of the PL peak energy with B in N-containing samples depends on temperature.[19] Here, we extend those measurements to different $In_xGa_{1-x}As_{1-y}N_y$ QWs. The inset in Fig. 2 shows for representative B values the PL spectra recorded on the same QW used for TR PL (very similar results have been obtained in all N-containing samples). At $T=30$ K, the PL line shape shows a long low-energy tail characteristic of localized state recombination, which is absent at $T=180$ K, where emission is dominated by free excitons. The diamagnetic shift ΔE_d of the PL peak energy is shown in Fig. 2 as a function of B for the two measurement temperatures. Since the highest value of the magnetic energy in these samples is comparable with the exciton binding energy, the high T data have been fitted for simplicity purposes by using a model developed by MacDonald and Ritchie (dashed line in Fig. 2).[20] This model has been derived for two-dimensional (2D) Coulombic systems at arbitrary magnetic fields, and the exciton effective mass is the only fitting parameter.[19] The model does not fit the low T dependence of ΔE_d on B, which is linear for $B>4$ T. It should be noted that at any value of B, ΔE_d is higher at low T than at high T (a factor two for $B=12$ T). This indicates that at low temperature the recombining electron-hole pair is more loosely bound than an *exciton*, either localized or free. Indeed, the interaction of these electron-hole pairs at low T with a magnetic field results in a perturbation stronger than Coulomb attraction and, therefore, in a greater diamagnetic shift with respect to the exciton case. Before continuing, we point out that (i) ΔE_d is independent of T at high temperature where free excitons only contribute to PL,[19] (ii) ΔE_d measured at low T decreases when very high power densities are employed (namely, when free excitons start contributing to the PL signal), and (iii) these effects are absent in the N-free $In_xGa_{1-x}As$ QWs studied for comparison purposes.

On the basis of these observations, the PL emission at low temperature can be attributed to recombination of a localized with a delocalized carrier.[21] As for the charge of the

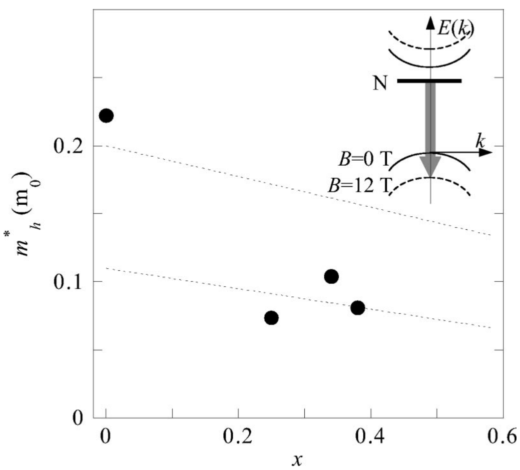

FIG. 3. Dependence of the in-plane hole effective mass in $In_xGa_{1-x}As_{1-y}N_y$ as a function of the In concentration x. The dashed lines are the values of the in-plane light (upper line) and heavy (lower line) hole, as estimated by using the Luttinger parameters.[26] The inset depicts the recombination at low temperature occurring in $In_xGa_{1-x}As_{1-y}N_y$ in a reciprocal space scheme at $k\sim 0$ for $B=0$ T (continuous parabolas) and $B=12$ T (dashed parabolas). N indicates the localized level related to nitrogen.

localized carriers, one can invoke the model first proposed by Hopfield, Thomas, and Lynch,[22] who suggested that N in GaP is an isoelectronic *electron* trap. Since N in GaAs shares several common features with GaP:N, we argue that N in $In_xGa_{1-x}As$ also behaves as an isoelectronic *electron* trap. Consequently, the potential minima due to N compositional disorder capture electrons with which *free* holes can recombine, similar to a free-hole to neutral-donor recombination (this is shown schematically in the inset of Fig. 3). Under this hypothesis, the electron is strongly localized and the shift of the PL peak with B can be ascribed entirely to the free hole, namely $\Delta E_d=(e\hbar/2m_h^*)B$, where m_h^* is the hole in-plane effective mass. The continuous line in Fig. 2 is a fit of this formula to the $T=30$ K data with $m_h^*=0.074\,m_0$ (m_0 is the electron mass in vacuum).[23] A similar approach has been used for deriving the electron-effective mass from the B-induced shift of free-electron to neutral-acceptor recombinations.[24,25] Figure 3 shows the m_h^* values derived in $In_xGa_{1-x}As_{1-y}N_y$ QWs as a function of the In concentration. Since N incorporation affects mainly the conduction band states,[15] we compare the m_h^* values derived here with those of the heavy and light holes of the N-free $In_xGa_{1-x}As$ host. In fact, due to the different type of strain, compressive in $In_xGa_{1-x}As_{1-y}N_y$ and tensile in $GaAs_{1-y}N_y$, in-plane heavy and light holes should be considered in the former and latter case. The dashed lines in Fig. 3 are the $In_xGa_{1-x}As$ in-plane hole masses, as estimated through the Luttinger parameters.[26] The good agreement of the experimental data with the hole curves supports our hypothesis about the hole nature of the delocalized carrier.

In conclusion, cw and TR PL at different temperatures show that in $In_xGa_{1-x}As_{1-y}N_y$, radiative recombination at low T involves localized states. However, no clue about the excitonic or otherwise nature of these states can be derived from PL only. Magneto-PL measurements allow probing the localization degree of recombining carriers at different T's and indicate that the low-T PL emission is due to *free* holes recombining with electrons *localized* in N-rich regions.

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