Photoluminescence kinetics of indirect excitons in GaAs/Al_xGa_{1-x}As coupled quantum wells

L. V. Butov
Institute of Solid State Physics, Russian Academy of Sciences, 142432 Chernogolovka, Moscow District, Russia

A. Imamoglu
Department of Electrical and Computer Engineering, University of California, Santa Barbara, Santa Barbara, California 93106

A. V. Mintsev
Institute of Solid State Physics, Russian Academy of Sciences, 142432 Chernogolovka, Moscow District, Russia

K. L. Campman and A. C. Gossard
Department of Electrical and Computer Engineering, University of California, Santa Barbara, Santa Barbara, California 93106

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Photoluminescence (PL) kinetics of long-lifetime indirect excitons in a GaAs/Al_xGa_{1-x}As coupled quantum well characterized by a small in-plane random potential was studied at temperatures $1.5 \leqslant T \leqslant 15$ K for a wide range of exciton densities. Strong deviations of the indirect exciton PL kinetics from monoexponential PL rise/decay were observed at low temperatures and high exciton densities. In particular, right after the excitation is switched off, the spectrally integrated indirect exciton PL intensity *increased* sharply. Simultaneously, the indirect exciton energy distribution was observed to narrow significantly. The observed increase in intensity is attributed to the sharp increase of occupation of the optically active exciton states. The energy distribution narrowing is explained in terms of the phonon mediated exciton energy relaxation in momentum space and in the in-plane random potential. [S0163-1829(99)00104-6]

A number of collective phenomena, in particular exciton condensation and superfluidity, have been predicted for a low-temperature two-dimensional (2D) exciton system in semiconductor quantum well structures.[1] The principal limitation for the realization of the low-temperature exciton system is the electron hole recombination, which limits the exciton lifetime and results in the high effective exciton temperature. The latter is determined by the ratio between the exciton energy relaxation rate and the exciton recombination rate. The indirect (interwell) excitons in coupled quantum wells (CQW's) are characterized by a long recombination lifetime; therefore, CQW's present an opportunity for experimental study of a high-density low-temperature exciton system.

In the general case of nonresonant excitation, the occupation of low-energy exciton states that are responsible for the formation of collective states[2,3] is determined by the exciton energy relaxation and can be revealed in the exciton photoluminescence (PL) kinetics. In the present paper we study the PL kinetics of long-lifetime indirect excitons in a GaAs/Al_xGa_{1-x}As CQW characterized by a small in-plane random potential. The earlier studies of the PL kinetics of indirect excitons in GaAs/Al_xGa_{1-x}As CQW's have demonstrated a long indirect exciton recombination time,[4–7] revealed a metastable exciton energy distribution,[8] and allowed for the measurement of the exciton energy relaxation in the hopping regime.[9]

The studied electric field tunable n^+-i-n^+GaAs/Al_xGa_{1-x}As CQW structure has been grown by molecular-beam epitaxy. The i region consists of two 8-nm GaAs quantum wells (QW's) separated by the 4-nm $Al_{0.33}Ga_{0.67}As$ barrier and surrounded by two 200-nm $Al_{0.33}Ga_{0.67}As$ barrier layers. The n^+ layers are Si doped GaAs with $N_{Si}=5\times10^{17}$ cm^{-3}. A schematic band diagram of GaAs/Al_xGa_{1-x}As CQW's is shown in Fig. 1. The electric field in the z direction was monitored by the external gate voltage V_g. Carriers were photoexcited by a pulsed semiconductor laser ($\hbar\omega=1.85$ eV) with a pulse duration of 50 ns. The edge sharpness of the pulses including the system resolution was 0.2 ns. The PL kinetics was measured by a time correlated photon counting system.

The gate voltage dependence of the time integrated PL spectrum and the exciton PL kinetics is shown in Fig. 1. At $V_g \lesssim 0.2$ V the ground state is direct exciton, which is seen from the gate-voltage-independent exciton energy and short PL decay time. The double structure of the direct exciton line results presumably from the difference in the widths of two QW's. At $V_g \gtrsim 0.2$ V the indirect exciton is lower in energy than the direct exciton and its energy reduces approximately linearly with V_g [Fig. 1(a)]. The observed transition from the direct to the indirect exciton ground state with increasing V_g is typical of CQW's with a wide interwell barrier (see Ref. 10 and references therein).

Figure 1(b) shows the indirect exciton PL kinetics measured at the maximum of the time integrated indirect exciton spectrum. The main feature of the indirect exciton PL kinetics is a strong and sharp increase of the indirect exciton PL intensity just after the excitation is switched off: In about 1 ns the PL intensity is increased by a factor of 2 in magnitude. This after-excitation PL intensity jump (PL jump) was observed for all studied excitation pulse widths (from 3 to 500 ns) and excitation energies (from 1.598 to 1.94 eV). The PL jump was observed only for indirect excitons with a long decay time [Fig. 1(b)] and was found to be a general effect

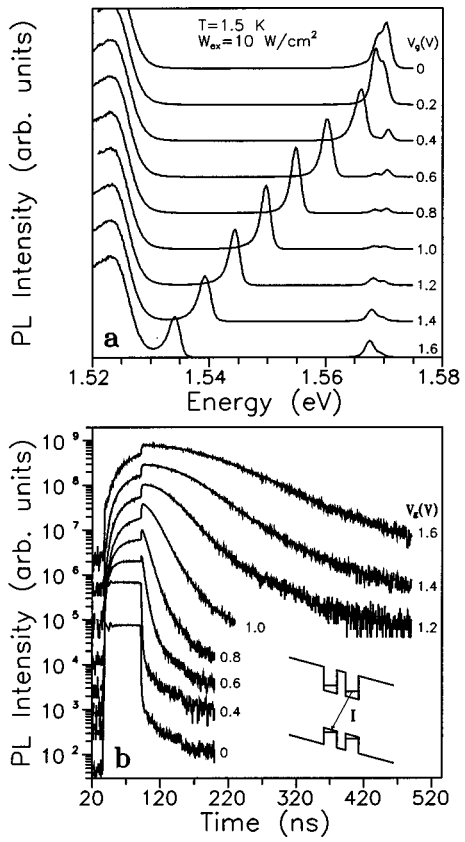

FIG. 1. (a) Gate voltage dependence of the time integrated PL spectrum and (b) the PL kinetics measured at the line maximum of direct (at $V_g=0$ and 0.2 V) and indirect (at $V_g=0.4-1.6$ V) excitons at $T=1.5$ K and $W_{ex}=10$ W/cm^2. The broad line at the lowest energies corresponds to the n^+-GaAs emission. The excitation pulse has a steplike shape ~ 50 ns wide that is reflected by the time dependence of the direct exciton PL at $V_g=0$. Inset: schematic band diagram of the GaAs/Al_xGa_{1-x}As CQW.

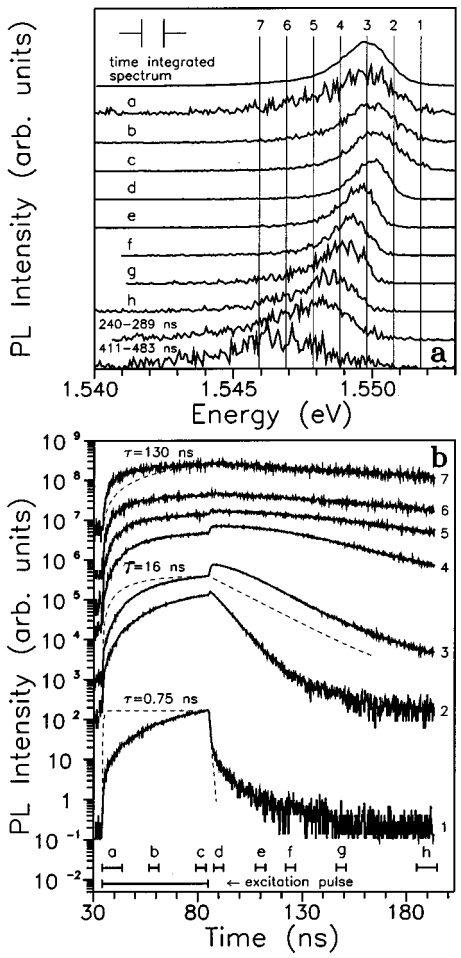

FIG. 2. (a) Time delay dependence of the indirect exciton PL spectrum at $V_g=1$ V, $T=1.5$ K, and $W_{ex}=10$ W/cm^2. The spectra are integrated in the time intervals shown in (b). The excitation pulse has a steplike shape ~ 50 ns wide with the end between the intervals *c* and *d*. The spectrum intensities are equalized for clarity. (b) Energy dependence of the indirect exciton PL kinetics at the same V_g, T, and W_{ex}. The PL kinetics are measured at energies shown in (a). For comparison, dashed curves represent the monoexponential PL rise/decay with the time constants corresponding to the fastest decay rate for decays 1, 3, and 7.

for indirect excitons in various CQW structures: We have observed the PL jump also in $In_xGa_{1-x}As/GaAs$ CQW's, and Γ-X_z AlAs/GaAs CQW's. The necessary condition for the observation of the PL jump is a small linewidth of indirect exciton PL, which indicates a small disorder in the structure; no PL jump was observed in GaAs/$Al_xGa_{1-x}As$, $In_xGa_{1-x}As/GaAs$, and AlAs/GaAs CQW's with an indirect exciton PL linewidth at cw excitation larger than ~ 3 meV. The largest PL jump was observed in the CQW with the narrowest indirect exciton PL line and this CQW is considered in the present paper.

The indirect exciton PL kinetics after the PL jump also strongly deviates from the monoexponential PL decay: The PL decay rate first strongly increases and then reduces with the delay time [Fig. 1(b)]. This behavior is also characteristic of high-quality samples only; in samples with large disorder the decay rate reduces monotonically with the delay time.

Figure 2(a) shows the time delay dependence of the indirect exciton PL spectrum. This dependence presents the time evolution of the exciton (electron-hole pair) energy distribution with a weight proportional to the radiative recombination probability. During the excitation pulse (time intervals *a*, *b*, and *c* the indirect PL line is relatively broad with a large exciton occupation of high-energy states (between lines 1 and 2). Just after the excitation is switched off (time interval *d*) the high-energy tail of the indirect exciton PL line strongly reduces, i.e., the exciton energy distribution sharply narrows. The narrowing of the indirect exciton PL line is accompanied by the increase of its spectrally integrated intensity by about 1.4 times (the intensity maximum is increased by 1.9 times). With a further time delay the indirect line monotonically broadens and shifts to lower energies.

Another aspect of the spectrum time delay dependence [Fig. 2(a)] is the energy dependence of the PL kinetics [Fig. 2(b)]. The PL rise time monotonically decreases with reducing detection energy; at high energies (decays 1–3) the PL rise is slower than the monoexponential rise shown by the dashed curves, while at low energies (curves 6 and 7) it is faster. The PL jump is maximum at energy corresponding to the maximum of the time integrated PL spectrum (decay 3) and is not observed at the tails of the spectrum (this is valid also for higher temperatures and lower excitation densities studied). The PL decay time monotonically increases with reducing detection energy; at lowest energies (decay 7) the

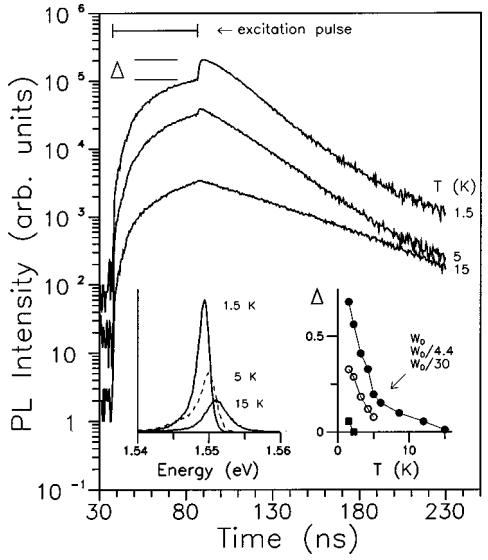

FIG. 3. Temperature dependence of the indirect exciton PL kinetics measured at the maximum of the time integrated spectrum at $V_g = 1$ V and $W_{ex} = W_0 = 10$ W/cm^2. The excitation pulse has a steplike shape ~ 50 ns wide. Left inset: the corresponding time integrated indirect exciton spectra. Right inset: temperature dependence of the indirect exciton PL intensity increase after excitation switching off, $\Delta = \ln(I_{PL-max}/I_{PL-pulse-end})$, for different W_{ex}.

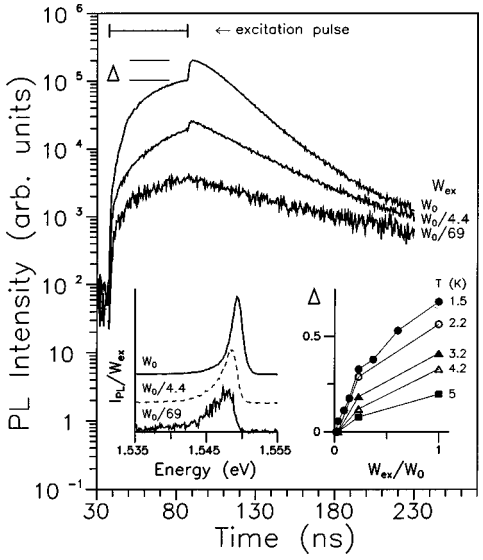

FIG. 4. Excitation density dependence of the indirect exciton PL kinetics measured at the maximum of the time integrated spectrum at $V_g = 1$ V and $T = 1.5$ K. $W_0 = 10$ W/cm^2. The excitation pulse has a steplike shape ~ 50 ns wide. Left inset: the corresponding time integrated indirect exciton spectra. Right inset: excitation density dependence of the indirect exciton PL intensity increase after excitation switching off, Δ, for different T.

PL decay is close to the monoexponential decay with a long time constant.

With increasing temperature the indirect exciton PL line shifts to higher energies, broadens, and reduces in intensity and the PL jump disappears (Fig. 3). At high temperatures the PL decay is close to the monoexponential decay with a long time constant.

When the excitation density is reduced, the indirect exciton line shifts to lower energies and broadens, and the PL-jump disappears (Fig. 4). At lowest excitation densities the PL decay is close to the monoexponential decay with a long time constant.

The observed indirect exciton PL kinetics is discussed below. The integrated exciton PL intensity remains almost constant with V_g variation, while the decay time varies by several orders of magnitude (Fig. 1). This indicates that radiative recombination is more efficient than nonradiative recombination. For delocalized 2D excitons only the states with small center-of-mass momenta $k \leqslant k_0 \approx E_g/\hbar c$ can decay radiatively (where E_g is the energy gap and c is the speed of light in the medium). The measured radiative decay rate is proportional to the fraction of excitons with $k \leqslant k_0$.[11–15] The exciton PL kinetics is determined by the kinetics of occupation of the optically active exciton states. The occupation of these states is increased by the energy relaxation of photoexcited high-energy excitons via phonon emission and decreased by the exciton recombination and phonon absorption. The PL jump denotes the sharp increase of occupation of the optically active exciton states just after excitation is switched off; the sharp increase of occupation of the low-energy exciton states ($E \leqslant E_0 = \hbar^2 k_0^2/2m \approx 1$ K) and the sharp narrowing of the exciton energy distribution indicate the sharp reduction of the effective exciton temperature.

The PL jump indicates that the kinetics of exciton-phonon system strongly depends on the presence or absence of the excitation. Photogenerated electron-hole pairs first relax predominantly by emitting multiple LO phonons (in an approximately picosecond time scale) and eventually form high center-of-mass momentum excitons. These excitons then relax down in energy by LA-phonon emission and recombine when reaching the low-energy states with $E \leqslant E_0$. Since LO phonons decay into multiple LA phonons in $5-10$ psec, a large LA-phonon population builds up during pumping, in spite of the fact that 3D phonons propagate out of the QW plane in a 10-psec time scale. The high LA-phonon occupancy in turn keeps the exciton temperature relatively high during the pumping cycle. After the excitation is switched off, the LA-phonon population decays sharply, which in turn results in the reduction of the effective exciton temperature as the excitons cool down by emitting phonons into the cold lattice. This process typically has a nanosecond time scale, consistent with our observation for the rise time of the PL jump.[16] Since the recombination time scale is much longer, fast exciton cooling sharply increases the number of optically active excitons with $E \leqslant E_0$. With increasing bath temperature the occupation of the low-energy optically active states reduces, which results in the observed disappearance of the PL jump and increase of the PL decay time (Fig. 3).

The exciton PL kinetics is strongly influenced by the in-plane random potential. (i) The potential results in the mixing of exciton states with different momenta and therefore washing out of the border between optically active and inactive exciton states.[16] (ii) The excitons migrate in the well plane in search of the lower-energy sites with emission of acoustic phonons; this migration results in the slow exciton energy relaxation.[17] (iii) The oscillator strength of optically active 2D excitons is increased with an increase of the lateral size of exciton center-of-mass wave function (and saturates when the coherence length reaches the inverse wave vector of the emitted light). For excitons localized in the potential

site the oscillator strength is proportional to the localization area[11–15] and the mean localization length is reduced with reducing energy in random potential.[18] (iv) The potential influences the exciton statistics: The 0D exciton state in a potential site has finite degeneracy (neglecting the exciton-exciton interaction, the degeneracy is $\sim S/a_B^2$, where S is the localization area and a_B is the exciton Bohr radius). (v) A strong potential can break the exciton resulting in the separate localization of the electron and hole in different in-plane potential sites.

The PL jump is not observed in CQW samples with a large random potential because the large potential washes out the border between the optically active and inactive exciton states and therefore a possible sharp increase of the low-energy state occupation would not result in the PL intensity increase. Similarly, the PL jump is not observed at low excitation density (Fig. 4) as at low exciton density the excitons are strongly localized in deep potential minima. The increase of the exciton localization with decreasing exciton density is also revealed in the broadening and shifting to lower energies of the indirect exciton line (Fig. 4).

The observed energy dependence of the exciton PL kinetics (Fig. 2) is understood as follows. The PL decay is faster at higher energies because of a possibility of exciton energy relaxation both in momentum space and in the in-plane random potential and because of the stronger exciton localization for the lower-energy exciton states resulting in a smaller oscillator strength. Note that at largest delays and lowest energies the PL decay could be dominated by slow recombination of electrons and holes separately localized in different potential sites. The PL jump is not observed for high-energy states due to their fast decay rate and is not observed for low-energy states because they are strongly localized. The exciton energy relaxation from the potential higher-energy sites increases the occupation of lower-energy states and therefore also contributes to the PL jump. The observed slow PL kinetics just after the PL jump is due to the photoexcited high-energy exciton relaxation in momentum space and in the in-plane random potential. Note that the fast rise time for the low-energy exciton states [decay 7 in Fig. 2(b)] indicates a low degeneracy of these states, which is due to the small localization area.

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[1] Yu. E. Lozovik and V. I. Yudson, Zh. Eksp. Teor. Fiz. **71**, 738 (1976) [Sov. Phys. JETP **44**, 389 (1976)].
[2] V. N. Popov, Theor. Math. Phys. **11**, 565 (1972); P. N. Brusov and V. N. Popov, *Superfluidity and Collective Properties of Quantum Liquids* (Nauka, Moscow, 1988), Chap. 6.
[3] D. S. Fisher and P. C. Hohenberg, Phys. Rev. B **37**, 4936 (1988).
[4] S. Charbonneau, M. L. W. Thewalt, E. S. Koteles, and B. Elman, Phys. Rev. B **38**, 6287 (1988).
[5] C. C. Phillips, R. Eccleston, and S. R. Andrews, Phys. Rev. B **40**, 9760 (1989).
[6] J. E. Golub, K. Kash, J. P. Harbison, and L. T. Florez, Phys. Rev. B **41**, 8564 (1990).
[7] A. Alexandrou, J. A. Kash, E. E. Mendez, M. Zachau, J. M. Hong, T. Fukuzawa, and Y. Hase, Phys. Rev. B **42**, 9225 (1990).
[8] J. E. Golub, K. Kash, J. P. Harbison, and L. T. Florez, Phys. Rev. B. **45**, 9477 (1992).
[9] J. E. Golub, S. D. Baranovskii, and P. Thomas, Phys. Rev. Lett. **78**, 4261 (1997).
[10] L. V. Butov, A. Zrenner, G. Abstreiter, A. V. Petinova, and K. Eberl, Phys. Rev. B **52**, 12 153 (1995).
[11] J. Feldmann, G. Peter, E. O. Göbel, P. Dawson, K. Moore, C. Foxon, and R. J. Elliott, Phys. Rev. Lett. **59**, 2337 (1987).
[12] E. Hanamura, Phys. Rev. B **38**, 1228 (1988).
[13] L. C. Andreani, F. Tassone, and F. Bassani, Solid State Commun. **77**, 641 (1991).
[14] B. Deveaud, F. Clerot, N. Roy, K. Satzke, B. Sermage, and D. S. Katzer, Phys. Rev. Lett. **67**, 2355 (1991).
[15] D. S. Citrin, Phys. Rev. B **47**, 3832 (1993).
[16] W. Zhao, P. Stenius, and A. Imamoglu, Phys. Rev. B **56**, 5306 (1997).
[17] T. Takagahara, Phys. Rev. B **31**, 6552 (1985).
[18] Zh. S. Gevorkyan and Yu. E. Lozovik, Fiz. Tverd. Tela (Leningrad) **27**, 1800 [Sov. Phys. Solid State **27**, 1079 (1985)].