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Nonlinear Photoluminescence Kinetics of Indirect Excitons in Coupled Quantum Wells

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We report the strongly nonlinear dependence of indirect exciton photoluminescence (PL) kinetics on excitation density. In the indirect regime characterized by a slow exciton recombination, at high excitation densities, low magnetic fields, low temperatures, and for samples with small in-plane disorder, we observe that the indirect exciton PL intensity increases sharply within a few nanoseconds after the excitation is switched off. This PL intensity jump is also accompanied by a subsequent PL decay rate that increases with excitation density. The effects are observed only when the estimated occupancies of the lowest energy exciton states exceed unity.

The system of indirect (interwell) excitons in coupled quantum wells (CQWs) is characterized by much longer indirect exciton recombination time compared to single-layer exciton systems. Therefore, CQWs provide a unique opportunity for studying low temperature 2D exciton systems. In particular, the long lifetime of indirect excitons promotes the exciton accumulation in the low energy exciton states and allows the system to cool down to temperatures where the exciton gas becomes degenerate at densities well below the Mott density [1].

We report on the study of photoluminescence (PL) kinetics of indirect excitons in GaAs/AlGaAs CQW at low temperatures $50\,\mathrm{mK} \leq T \leq 15\,\mathrm{K}$ and high magnetic fields $0\,\mathrm{T} \leq B \leq 16\,\mathrm{T}$. The electric-field-tunable n^+–i–n^+ GaAs/AlGaAs CQW structure was grown by MBE. The *i*-region consists of two 8 nm GaAs QWs separated by a 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 $N_{Si} = 5 \times 10^{17}\,\mathrm{cm}^{-3}$. The electric field in the *z*-direction is monitored by the external gate voltage V_g applied between n^+-layers. The small disorder in the CQW is indicated by the indirect exciton PL linewidth of about 1 meV. Carriers were photoexcited by a pulsed semiconductor laser ($\hbar\omega = 1.85\,\mathrm{eV}$, the pulse duration was about 50 ns, the edge sharpness including the system resolution was $\approx$0.2 ns, and the repetition frequency was 1 MHz). The PL measurements were performed in a He^3/He^4 dilution refrigerator by means of an optical fiber with diameter 0.1 mm positioned 0.3 mm above the mesa.

A typical V_g dependence of the ground state PL line positions at low excitation densities is shown in the inset of Fig. 1. At finite V_g the ground state is indirect exciton constructed from electron (e) and hole (h) in different layers (the indirect regime).

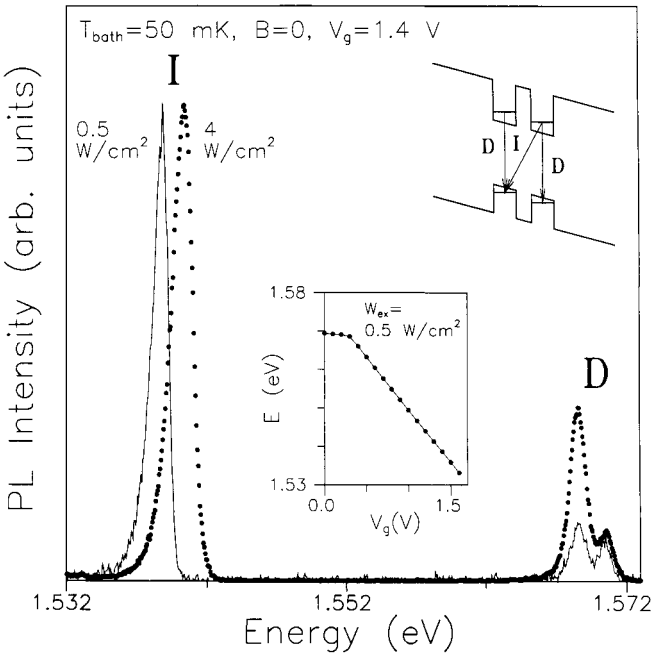

Fig. 1. PL spectra in the indirect regime. Top inset: band diagram of the GaAs/AlGaAs CQW structure under applied gate voltage; the direct (D) and indirect (I) exciton transitions are indicated. Bottom inset: the ground state PL line energy as a function of gate voltage

Figure 1 shows that, unlike the direct exciton, the indirect exciton energy increases with excitation density. This observation is consistent with the theoretically predicted enhancement of the indirect exciton energy with e–h density: it can be understood in terms of the net repulsive interaction between indirect excitons which are dipole oriented in the z-direction for low exciton densities, and in terms of the energy shift originated from the electric field between the separated e and h layers for high e–h densities [2]. In the latter case the energy shift can be roughly estimated using the plate capacitor formula $\delta E = 4\pi n_{\mathrm{eh}} e^2 d/\varepsilon$, which allows the estimation of the exciton density $\approx 9 \times 10^9$ cm^{-2} at $W_{\mathrm{ex}} = 4$ W/cm^2 for the data of Fig. 1.

The indirect exciton PL kinetics is shown in Fig. 2. At high excitation densities, low temperatures, and low magnetic fields the indirect exciton PL kinetics strongly differs from the monoexponential PL rise/decay: right after the excitation is switched off, the indirect exciton PL intensity first jumps up and then decays with a rate that changes non-monotonously with time. On the contrary, at low excitation densities, high magnetic fields, and high temperatures the indirect exciton PL kinetics are close to monoexponential with long time constants (Fig. 2).

The measurements on a set of CQW samples have shown that the nonlinearities in the indirect exciton PL kinetics are reduced with increasing in-plane disorder. In particular, in a GaAs/AlGaAs CQW sample with a large in-plane disorder (indirect exciton PL linewidth of 6.5 meV) no PL enhancement is observed after excitation is switched off; the PL kinetics in this case is close to monoexponential and is practically independent of the excitation density at the same excitation densities studied [3].

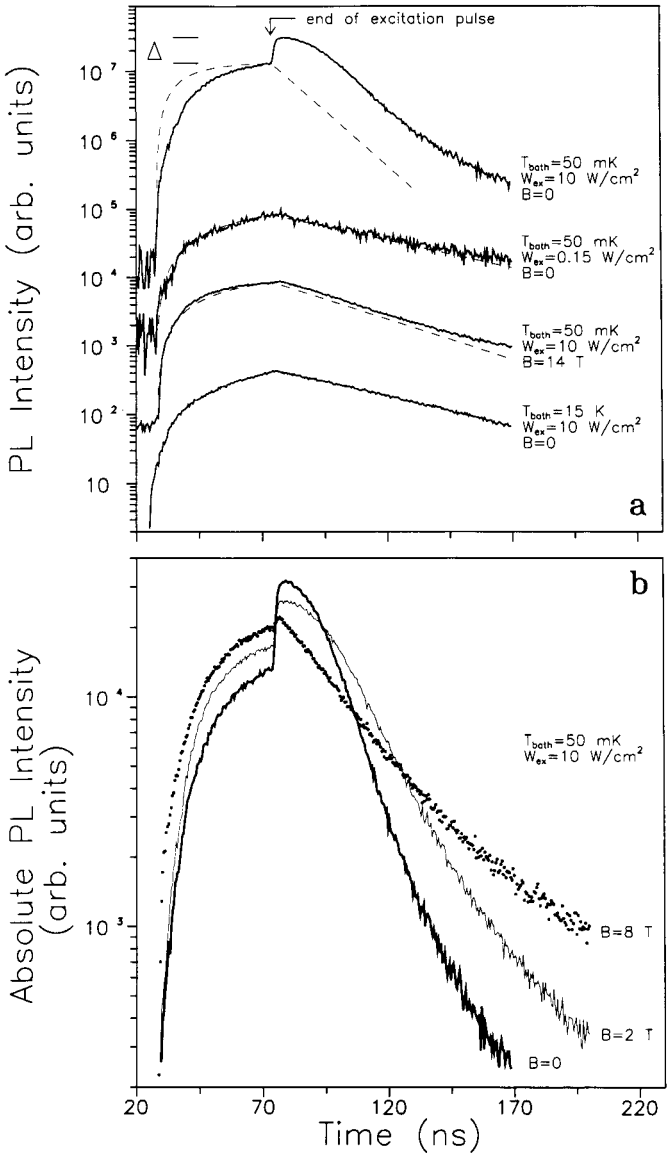

Fig. 2. Indirect exciton PL kinetics measured at the indirect exciton line maximum at $V_g = 1$ V. The excitation pulse has a step-like shape ≈50 ns wide. The dashed lines represent the monoexponential PL rise/decay with time constants corresponding to the fastest PL decay rate. In a) the kinetics are shifted in vertical axis for clarity, while in b) the absolute PL intensities are presented. The sharp enhancement of the indirect exciton PL intensity right after the excitation is switched off and a consequent fast PL intensity decay is observed at high excitation densities, low magnetic fields, and low temperatures

The integrated exciton PL intensity remains almost constant with V_g variation while the decay time varies by several orders of magnitude [4]. Hence, the radiative recombination is dominant in the CQW studied. For delocalized 2D excitons only the states with small center-of-mass momenta $k \leq k_0 \approx E_g/\hbar c$ (where c is the speed of light in the medium) can decay radiatively [5]. The exciton PL kinetics is determined by the kinetics of occupation of the optically active exciton states with $E \leq E_0 = \hbar^2 k_0^2/2m \approx 1$ K. The occupation of these states is increased through the energy relaxation of photoexcited high energy excitons and decreased as a result of exciton recombination. The PL jump denotes a sharp increase of the occupation of the optically active exciton states just after the excitation is switched off [4]. It is induced by the sharp reduction of the effective exciton temperature, T_{eff}, right after the excitation is switched off due to the fast decay of the nonequilibrium phonon occupation and energy relaxation of hot photoexcited excitons, electrons and holes.

With increasing magnetic field the PL intensity during the excitation pulse increases (Fig. 2b) which indicates a reduction of T_{eff}. This can be qualitatively explained by the strong increase of the magnetoexciton mass for the interwell excitons with large interlayer separation $d \approx 12$ nm (by an order of magnitude at the highest magnetic fields studied, as calculated in Ref. [6]) which results in the strong increase of the exciton energy relaxation rate via LA-phonon emission: the exciton–phonon scattering time $\tau_{sc} = (\pi^2\hbar^4\varrho)/(D^2m^3v_s)$, where ϱ is the crystal density, D is the deformation potential and v_s is the longitudinal sound velocity. The reduction of T_{eff} during the excitation pulse can be responsible for the observed disappearance of the sharp PL intensity increase at the excitation switch off at high magnetic fields.

No effect is observed for samples with a large in-plane disorder [3], possibly because disorder results in the mixing of exciton states with different momenta and therefore washing out of the border between optically active and dark exciton states [7]. Due to this a sharp increase of the low energy state occupation would not result in the PL intensity increase.

We note that the sharp increase of the occupation of the low energy exciton states is observed right at the range of parameters where occupation of the exciton states is large: (i) in the indirect regime characterized by a slow exciton recombination, (ii) at high excitation densities, (iii) at low magnetic fields characterized by the small exciton density of states due to the small exciton mass, and (iv) at low temperatures (Figs. 2 and 3). For indirect excitons with a lifetime of 10 ns the estimate (based on absorbed laser power assuming the CQW absorption of 0.01) for the quasi-equilibrium exciton density (at cw excitation or during an excitation pulse at times much longer than 10 ns after the turn on) at $W_{eff} = 10$ W/cm^2 is $\approx 10^{10}$ cm^{-2}. At these densities the quantum statistics significantly affects the exciton kinetics [1, 8]. In particular, for quasi-equilibrium 2D exciton gas the ground state occupation number $n_{E=0} = \exp(T_0/T) - 1$ exceeds unity at $T \lesssim 1$ K ($k_B T_0 = 2\pi\hbar^2 n/gm \approx 0.5$ K is the degeneracy temperature, g is the exciton spin degeneracy) [1]. The final state stimulation of the exciton scattering to the optically active exciton states results in the fast built up of the state occupation and can effectively contribute to the observed sharp increase of the state occupation at the excitation switch off. The transition from the Maxwell-Boltzmann to well developed Bose-Einstein exciton statistics with increasing exciton density results in the increase of the fraction of the optically active excitons [1] which can be responsible for the observed increase of the fastest PL decay rate with excitation density.

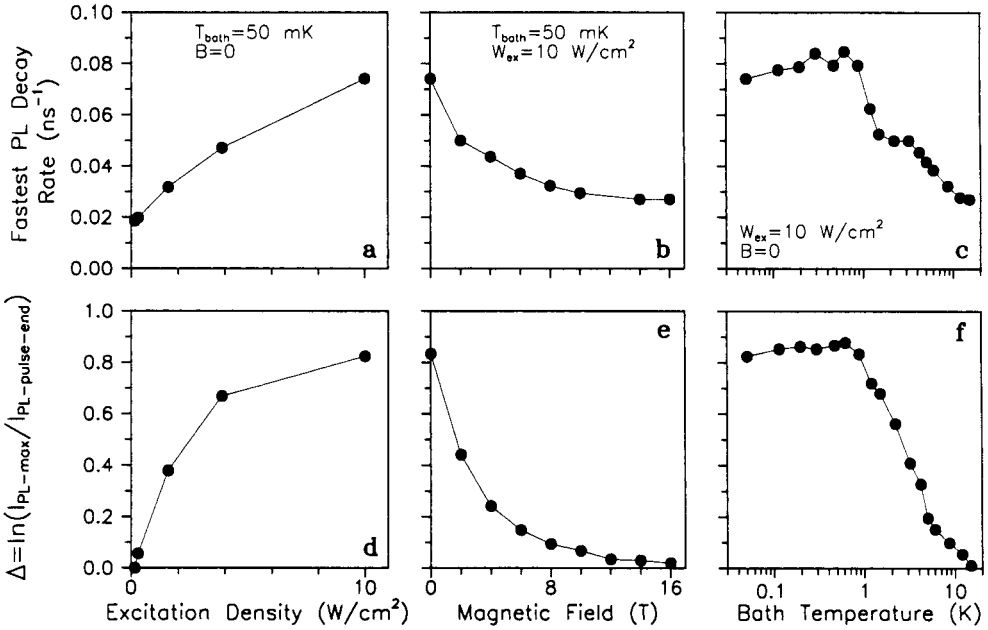

Fig. 3. The fastest indirect exciton PL decay rate part a) to c) and the indirect exciton PL intensity enhancement after excitation switch off; d) to f) $\Delta = \ln(I_{\mathrm{PL-max}}/I_{\mathrm{PL-pulse-end}})$ vs. excitation density, magnetic field and temperature

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