Enhanced Radiative Recombination of Free Excitons in GaAs Quantum Wells

B. Deveaud and F. Clérot
Centre National d'Etudes de Télécommunications, 22300 Lannion, France

N. Roy, K. Satzke, and B. Sermage
Centre National d'Etudes de Télécommunications, 92120 Bagneux, France

D. S. Katzer
Naval Research Laboratory, Washington, D.C. 20375
(Received 15 April 1991; revised manuscript received 15 August 1991)

Radiative properties of free excitons in a single GaAs quantum well are studied under resonant excitation. Enhanced radiative recombination of the excitons, caused by the breakdown of the translational symmetry of the system, is evidenced by the very short lifetime as well as by the strong intensity of the signal. Dephasing mechanisms, by transferring the excitons to nonradiative states, increase the observed lifetime. We deduce a radiative lifetime of 10 ± 4 ps in the absence of dephasing mechanisms.

PACS numbers: 71.35.+z, 73.20.Dx, 78.47.+p, 78.65.Fa

2D confinement of an exciton in a quantum well (QW) leads to a shrinkage of the exciton Bohr radius accompanied by an increase of the oscillator strength and of the binding energy [1,2]. This leads to the observation of strong resonances up to room temperature, allowing a large number of potential applications [3]. The amount of work, theoretical as well as experimental, devoted to the behavior of excitons in quantum wells has therefore been considerable [4]. Among the possible techniques, luminescence, because of its simplicity and sensitivity, and also because of the fact that luminescence transitions in QW's are dominated by excitonic recombination, has been quite widely used [5–8].

In 3D, the time-resolved behavior of excitonic polaritons has been extensively studied (see [9] for a review). Excitonic polaritons are stationary states of an infinite dielectric medium (and, therefore, should not exhibit any temporal evolution). Decay either is observed through phonon scattering [10] (as evidenced for example by Brillouin scattering [11]), or is due to the existence of crystal surfaces where polaritons can transform into external photons (radiative decay) with a rate Γ given by

$$\Gamma = (1-R)V_g/L\,, \tag{1}$$

where R is the reflection coefficient, V_g the polariton group velocity, and L the length of the sample. This picture may explain the "giant oscillator strength" of excitons in thin samples, as observed in GaAs [12].

The above description, however, relies on the translational invariance of the crystal, and implies the conservation of momentum from the exciton to the photon. *For excitons confined in quantum wells, the coupling to photons is profoundly modified by the breakdown of translational symmetry in the growth direction* (hereafter labeled z). This coupling is now very strong for excitons with a wave vector $|\mathbf{k}| < n\omega_0/c$, as they can couple to a whole 1D distribution of photons (here ω_0 is the frequency of the photon at the exciton energy, n the material refractive index, and c the speed of light). Compared to the decay rate of 3D excitons (estimated from the longitudinal-transverse splitting), the decay rate of free excitons in quantum wells has been predicted [13–16] to be enhanced by a factor

$$24\pi(\lambda/a_b)^2\,, \tag{2}$$

where λ is the radiation wavelength and a_b the exciton Bohr radius. This amounts to 8×10^5 in GaAs quantum wells, and radiative decay of near $\mathbf{k}=0$ excitons should be very fast, of the order of 10 ps. On the contrary, excitons with $\mathbf{k}$ above $n\omega_0/c$ should not recombine at all.

This *enhanced radiative decay rate,* which relies on the coherent nature of the exciton, is only possible in the absence of perturbations and, in particular, if the phase coherence of the exciton is preserved long enough. As noted by different authors [15–17], the loss of phase coherence (for example, due to the scattering by acoustical phonons if the temperature is too high) will prevent radiative recombination and thus increase the radiative lifetime from the expected value of about 10 ps to much longer times. An important point has to be raised, as it is generally neglected: The ratio of the luminescence intensity $I_{\text{lum}}(t=0)$ to the excitation power ϕ should be inversely proportional to the radiative lifetime [17,18]. Although luminescence measurements do not allow for an absolute measurement of the intensity, relative measurements bring information of great importance.

Apart from temperature effects, other perturbations may alter the decay of excitons: exciton-exciton or exciton-carrier scattering [19], and also any kind of imperfection in the quantum well and, in particular, interface roughness. Interface roughness will tend to localize the excitons and give rise to bound excitons with a lifetime of the order of 1 ns. As a result, as pointed out by Hanamura [15], the enhanced radiative decay of excitons should be very difficult to observe as it requires one single QW of the highest quality.

In this Letter, we demonstrate for the first time the enhanced radiative decay of free excitons in a single

quantum well. Not only do we monitor the decay of the luminescence intensity, but also the relative value of this intensity and the homogeneous linewidth. Under resonant excitation at 1.7 K, we observe a radiative decay time as short as 24 ps in a high-quality GaAs/AlAs quantum well and show that temperature (above 2 K), excitation density (above 3×10^9 cm^{-2}), as well as excitation detuning (by only 2 meV), by allowing phase-breaking mechanisms, increase the luminescence lifetime and the homogeneous linewidth, and decrease the intensity.

We have studied a single GaAs/AlAs QW grown by molecular-beam epitaxy using growth-interruption techniques [20]. This sample shows a splitting of the excitonic transitions. The well width is 45 Å, and the exciton splitting is 12 meV. The full width at half maximum of the lines is only 2 meV, and no Stokes shift is observed between luminescence and luminescence excitation: Therefore, the quality of the sample is very high. There is currently a discussion about the relation between the luminescence splitting and the interface quality [21,22]: In any case, each of the excitonic lines corresponds to the recombination of excitons in portions of the well with a well-defined average thickness. Three lines are generally present at each spot position. By carefully mapping the sample, it is possible to find zones where one transition is dominant, the two other regions representing less than 10% of the signal. All experiments are carried out on such a zone, where scattering of the exciton to other zones of different thickness (which leads to heating of the excitons [23,24]) may be neglected. At such a position, the line shape of the excitonic transition can be fully analyzed: As we will show in the following, we observe mainly an inhomogeneous Gaussian linewidth at low temperature and low density, and an additional Lorentzian broadening as the temperature or the density is increased.

The sample is excited with picosecond pulses generated by a synchronously pumped dye laser, and the luminescence is detected by a streak camera. The overall resolution of the system is of the order of 20 ps (see the dashed line in Fig. 1). The sample is immersed into superfluid helium for the measurements at 2 K, and is placed on the cold finger of a closed cycle cryostat for the measurements at higher temperatures.

Let us first describe our experimental findings. We show in Fig. 1 the time decay $\tau_{\rm lum}$ at 2.1 K of the excitonic luminescence for an excitation density of 3×10^9 cm^{-2}, as a function of the detuning between the pump and the exciton energies. All curves are plotted in absolute number of counts recorded in the same conditions. For resonant excitation, the decay time is nonexponential, the long-time decay being of the order of 40 ps, and the intensity is quite high [25]. The short-time transient ($t < 20$ ps) is due to residual diffused laser light and can be suppressed by using crossed polarizers. As soon as the laser is detuned above the exciton line, the lifetime increases (up to about 200 ps) and simultaneously the intensity decreases. The same qualitative behavior is observed for resonant excitation when the temperature is raised above 2 K, or when ϕ is increased above 10^{10} cm^{-2}. In this last case, due to the change in excitation power, we do not observe a decrease of $I_{\rm lum}$ but a decrease of the ratio $I_{\rm lum}(t=0)/\phi$. In each case, the lifetime increases up to a limit close to 200 ps.

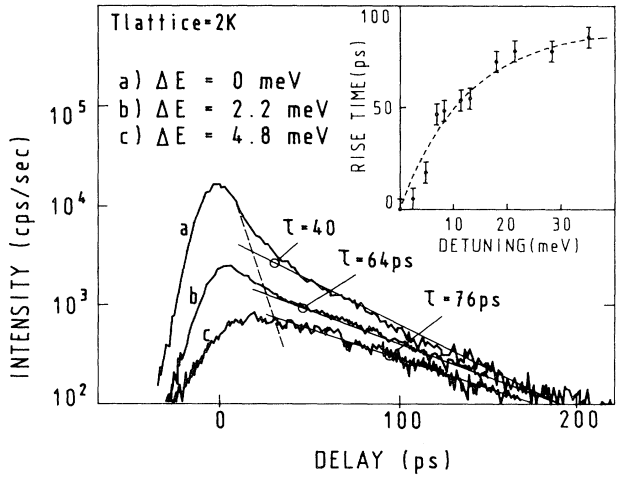

FIG. 1. Exciton luminescence decays for different detunings between the laser and the exciton energy. The same scale is used for the different measurements: A decrease in intensity is observed simultaneously with the increase of lifetime. The laser time behavior is plotted as a dashed curve. Inset: Rise time of the excitonic luminescence for different detunings.

When the detuning is increased above the exciton energy, a long rise time develops which has already been observed by different authors [26,27] and attributed to exciton relaxation and cooling. We observe the same increase of the rise time, but with different features (see the inset of Fig. 1): The absolute value of the rise time is rather short in our case; we consider that this is a consequence of the high quality of the sample, preventing the heating of the exciton subsequent to the trapping into larger portions of the well. The rise time is then only due to thermalization of initially hot excitons, and not to trapping of the exciton into lower-energy regions of the well.

Line shapes of the excitonic transition at 2.1 K, recorded 50 ps after resonant excitation at various densities, are reported in Fig. 2. The increase of the linewidth and the Lorentzian contribution to the line shape are clearly evidenced. At low temperature and low density, the linewidth is mainly Gaussian as a result of residual inhomogeneous broadening (for example, potential fluctuations due to impurities in the barriers or at the interface). A small Lorentzian contribution can be resolved giving a homogeneous width of 0.34 meV. At higher densities, a good fit to the line shape is obtained by keeping the Gaussian contribution constant and increasing the Lorentzian linewidth (see the fits in Fig. 2). A moderate temperature increase, up to 30 K, also leads to a similar Lorentzian broadening.

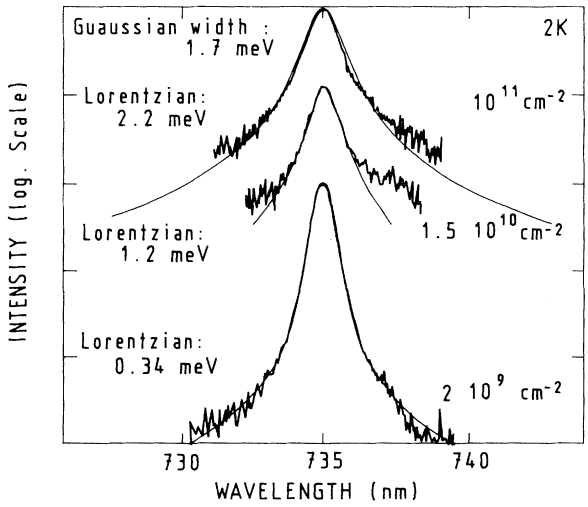

FIG. 2. Excitonic line shapes under resonant excitation at different densities. The fit is a convolution of a constant Gaussian linewidth (1.7 meV) and a density-dependent Lorentzian linewidth.

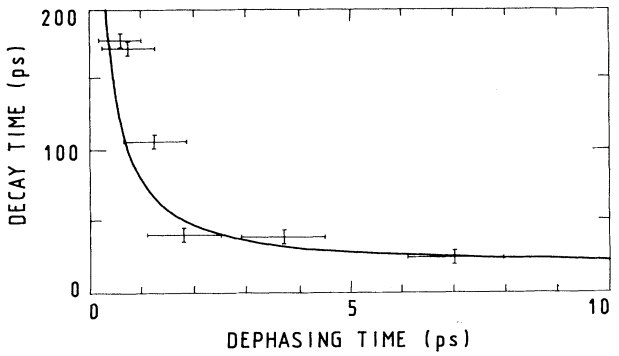

FIG. 3. Measured exciton lifetime, under resonant excitation, as a function of the dephasing time (deduced from fits similar to those of Fig. 2). The fit corresponds to Eq. (3), with $\tau_0=8$ ps and $\tau'=8$ ps, and $\tau_{tun}=1000$ ps.

We now discuss our results. At low temperature, under resonant excitation, upon increase of the excitation density ϕ, the dephasing time decreases because of exciton-exciton collisions [19]. The shorter dephasing time is clearly evidenced by the increased homogeneous linewidth and can be estimated by our fitting procedure. Even if we are at low temperature, the excitons can be scattered to two different nonradiative states: the triplet states and the exciton states with k above $n\omega_0/c$. The respective importance of these two channels is difficult to assert, so at the present stage we can only use a simple calculation where we assume a two-level system, one corresponding to the radiative excitons and one to the nonradiative ones. The excitons can be scattered to the nonradiative states with a dephasing time τ_{deph}, and they come back to the radiative states with a characteristic time τ'. Such a system has a short-time solution decaying as $1/\tau_{lum}=1/\tau_0+1/\tau_{deph}+1/\tau'$, which is not observable with our time resolution, and a long-time solution given by

$$2/\tau_{lum}=(1/\tau_0+1/\tau_{deph}+1/\tau')-[(1/\tau_0+1/\tau_{deph}+1/\tau')-4/\tau_0\tau']^{1/2}, \quad (3)$$

which in our case leads to

$$\tau_{lum}=\frac{1/\tau_0+1/\tau_{deph}+1/\tau'}{(1/\tau_0)(1/\tau')}. \quad (3')$$

The contribution of the nonradiative lifetime also has to be taken into account: The high quality of our samples is obtained through the growth of very thin AlAs barriers (50 Å). Hence our sample structure is analogous to the "resonant tunneling diodes" studied by Tsuchiya, Matsutsue, and Sakaki [28], and tunneling phenomena are included through an "exciton lifetime" τ_{tun} inside the double barrier, leading to a decay time τ given by $1/\tau=1/\tau_{lum}+1/\tau_{tun}$. Our measurements at large densities, detunings, or temperature, exactly on the same spot, give a decay time of the order of 200 ps, in agreement with [27].

The observed decay times are plotted as a function of the observed homogeneous broadening in Fig. 3, together with the results of the best fit using Eq. (3) for $\tau_0=8$ ps, $\tau'=8$ ps. The agreement with the measured values is reasonable. As can be seen from (3), τ_0 and τ' play the same role in the long-time solution so that a set of solutions exists giving a large error bar for τ_0, 10 ± 4 ps. This value is in quite good agreement with the theoretically predicted value of 8.4 ps: For our sample structure, the infinite well is a good approximation and we only correct Hanamura's value of 2.8 ps [15] by a factor of 3 to account for the refractive index of GaAs [16].

Temperature effects are more difficult to model. If we follow [17] or [16], and assume a thermalized exciton distribution, the variation of the luminescence decay time should be linear in T, reflecting the proportion of excitons with wave vector $|\mathbf{k}|<n\omega_0/c$. However, the phonon-scattering time is expected to be a few picoseconds at low temperatures [15] (we estimate 1.5 ps at 9 K and 0.5 ps at 15 K). As a consequence, for resonant excitation, low temperature, and low density, we cannot safely assume a thermalized distribution. In our experiments, the lifetime variation is not linear with temperature but slower (40 ps at 2 K and 65 ps at 9 K) probably because the excitons are initially cold and cannot be considered to be at lattice temperature just after the excitation pulse.

One might argue that our short lifetimes are simply due to localized excitons (on impurities or interface defects, as was assumed by Oberhauser *et al.* [29]). The short lifetime might have two explanations: short radiative lifetime of trap centers (giant oscillator strength of bound excitons [30]) or rapid capture by nonradiative centers. The lifetime increase with power or with temperature would simply be due to the saturation of these lo-

calized states and to the release of free excitons. Indeed, the temperature and density where we observe the changes correspond to reasonable defect densities and localization energy. We discard this explanation for the following reasons.

(i) It is only in the samples of the highest available quality that the very short lifetime of excitons is observed. Reducing the quality of the sample, and thus increasing the density of possible trapped excitons increases the radiative lifetime [24] in agreement with our model and contrary to what a model of localized excitons would predict. Furthermore, the radiative lifetime of localized excitons is expected to be of the order of 1 ns. It could be shorter only for highly localized traps, then giving rise to large energy shifts. This is contrary to our observation of a negligible shift compared to the linewidth of only 2 meV. In the low-density regime, we do not observe any change in line shape or in peak position with time.

(ii) The assumption of rapid trapping on nonradiative centers is contradicted by our observation of a diminution of the ratio $I_{\mathrm{lum}}(0)/\phi$ upon density increase: In such a model, this ratio should stay approximately constant.

In conclusion, we have clearly evidenced the very fast radiative decay of excitons caused by the breakdown in translational symmetry in quantum wells. The deduced radiative lifetime is 10 ± 4 ps in quite good agreement with theory. Our results easily explain the obvious difference between bulk GaAs samples where bound transitions dominate the spectrum even in the highest-quality samples, and GaAs QW's where free-exciton transitions dominate even in medium-quality samples.

The authors express their thanks to I. Bar Joseph, G. Bastard, A. Chomette, B. Lambert, A. Regreny, and C. Weisbuch for useful discussions. D.S.K. is an Office of Naval Technology Post Doctoral Fellow.

[1] G. Bastard, E. E. Mendez, L. L. Chang, and L. Esaki, Phys. Rev. B **26**, 1974 (1982).
[2] D. S. Chemla, Helv. Phys. Acta **56**, 607 (1983).
[3] See, for example, D. S. Chemla, D. A. B. Miller, P. W. Smith, A. C. Gossard, and W. Weigmann, IEEE J. Quantum Electron. **20**, 265 (1984).
[4] For a review, see, for example, C. Weisbuch, in *Semiconductors and Semimetals,* edited by R. Dingle (Academic, New York, 1987), p. 1; S. Schmitt-Rink, D. S. Chemla, and D. A. B. Miller, Adv. Phys. **38**, 89 (1989).
[5] C. Weisbuch, R. C. Miller, R. Dingle, A. C. Gossard, and W. Weigmann, Solid State Commun. **37**, 29 (1981).
[6] E. O. Göbel, H. Jung, J. Kuhl, and K. Ploog, Phys. Rev. Lett. **51**, 1588 (1983).
[7] R. C. Miller and D. A. Kleinmann, J. Lumin. **30**, 512 (1985).
[8] J. Lee, E. S. Koteles, and M. O. Vasell, Phys. Rev. B **33**, 512 (1986).
[9] J. Aaviksoo, J. Lumin. **48&49**, 57 (1991).
[10] Y. Toyozawa, Suppl. Prog. Theor. Phys. **12**, 111 (1959).
[11] C. Weisbuch and R. G. Ulbrich, Phys. Rev. Lett. **39**, 654 (1977).
[12] G. W. 't Hooft, W. A. J. A. van der Poel, L. W. Molenkamp, and C. T. Foxon, Phys. Rev. B **35**, 8281 (1987).
[13] V. M. Agranovitch and O. A. Dubovskii, Pis'ma Zh. Eksp. Teor. Fiz. **3**, 345 (1966) [JETP Lett. **3**, 223 (1966)].
[14] M. Orrit, C. Aslangul, and P. Kottis, Phys. Rev. B **25**, 7263 (1982).
[15] E. Hanamura, Phys. Rev. B **38**, 1228 (1988).
[16] L. C. Andreani, F. Tassone, and F. Bassani, Solid State Commun. **77**, 641 (1990).
[17] J. Feldmann, G. Peter, E. O. Göbel, P. Dawson, K. Moore, C. T. Foxon, and R. J. Elliott, Phys. Rev. Lett. **59**, 2337 (1987).
[18] B. Sermage, F. Alexandre, J. Beerens, and P. Tronc, Superlattices Microstruct. **6**, 373 (1989).
[19] See, for example, A. Honold, L. Schultheis, J. Kuhl, and C. W. Tu, Phys. Rev. B **40**, 6442 (1989).
[20] D. Gammon, B. V. Shanabrook, and D. S. Katzer, Appl. Phys. Lett. **56**, 2710 (1990).
[21] B. Deveaud, J. Y. Emery, A. Chomette, B. Lambert, and M. Baudet, Appl. Phys. Lett. **45**, 1078 (1984).
[22] A. Ourmazd, D. W. Taylor, J. Cunningham, and C. W. Tu, Phys. Rev. Lett. **62**, 2317 (1989); B. Deveaud, B. Guenais, A. Poudoulec, A. Regreny, and C. d'Anterroches, Phys. Rev. Lett. **65**, 2317 (1990); A. Ourmazd and J. Cunningham, Phys. Rev. Lett. **65**, 2318 (1990).
[23] B. Deveaud, J. Shah, T. C. Damen, and C. W. Tu, Appl. Phys. Lett. **51**, 828 (1987).
[24] M. Zachau, J. A. Kash, and W. T. Masselink, "Quantum Optoelectronics, Technical Digest," Salt Lake City, March 1991 (Optical Society of America, Washington, DC, to be published).
[25] Improving the temperature down to 1.7 K, and the excitation conditions, allowed us to observe decays as short as 24 ps.
[26] J. I. Kusano, Y. Segawa, Y. Aoyagi, S. Namba, and H. Okamoto, Phys. Rev. B **40**, 1685 (1989).
[27] T. C. Damen, J. Shah, D. Y. Oberli, D. S. Chemla, J. E. Cunningham, and J. M. Kuo, Phys. Rev. B **42**, 7434 (1990).
[28] M. Tsuchiya, T. Matsutsue, and H. Sakaki, Phys. Rev. Lett. **59**, 2356 (1987).
[29] D. Oberhauser, H. Kalt, W. Schlapp, H. Nickel, and C. Klingshirn, J. Lumin. **48&49**, 717 (1991).
[30] E. J. Rashba and G. E. Gurgenishvili, Fiz. Tverd. Tela (Leningrad) **4**, 1029 (1962) [Sov. Phys. Solid State **4**, 759 (1962)].