Linewidth Dependence of Radiative Exciton Lifetimes in Quantum Wells

J. Feldmann, G. Peter, and E. O. Göbel
Fachbereich Physik, Philipps-Universität, 3550 Marburg, Federal Republic of Germany

P. Dawson, K. Moore, and C. Foxon
Philips Research Laboratory, Redhill, Surrey RH1 5HA, England

and

R. J. Elliott
Department of Theoretical Physics, University of Oxford, Oxford OX1 3NP, England
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The fundamental relationship between radiative lifetime and spectral linewidth of free excitons is demonstrated theoretically and experimentally for quasi 2D excitons in GaAs/AlGaAs quantum wells.

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The linear and nonlinear optical properties of semiconductors are strongly affected by excitonic effects in the spectral region close to the fundamental energy gap. In particular, the near-band-edge absorption spectrum at sufficiently low temperatures reveals the effect of the Coulomb interaction of the electron and hole by showing a hydrogenlike series of discrete transition lines below the actual band-gap energy and an enhancement of the band-to-band transition strength which is in quantitative agreement with theory.[1] In emission, however, the intrinsic free-exciton properties are obscured as a result of the strong contribution of bound excitons and the polariton nature of the free excitons in the case of direct-gap semiconductors and because of the phonon participation in indirect-gap materials.

Excitons in quasi two-dimensional (2D) semiconductors exhibit different properties as compared to three-dimensional (3D) excitons as a result of the confinement of the electrons and holes.[2,3] The most evident effect is the increase of the exciton binding energy.[4,5] Since the electron-phonon coupling is basically unaffected by the 2D confinement the exciton scattering is unchanged and this explains why excitons continue to determine the linear and nonlinear optical properties of GaAs-AlGaAs quantum wells (QW) even at room temperature.[3] Most important for the subject of this paper, however, is the minor importance of bound-exciton recombination in high-quality undoped quantum-well structures and the fact that polariton modes are not excited in usual luminescence experiments.[6] Quasi 2D excitons in quantum wells thus allow us for the first time to study the intrinsic recombination behavior of free excitons.

In this paper we demonstrate theoretically and experimentally the fundamental relationship between the radiative exciton lifetime and the exciton linewidth, which corresponds to the phase coherence time in the case of a homogeneous transition. The dependence of the radiative exciton lifetime on the linewidth reflects the fact that the coherence volume of an exciton is determined by exciton scattering.

We first derive the expression for the exciton lifetime for quasi 2D excitons. The oscillator strength per unit cell of the $1s$ quasi 2D free exciton calculated within the effective-mass approximation for the case of a QW with infinite barriers is given by

$$f_x^{2D} = \Omega f_0 |\phi_{1s}^{2D}(0)|^2 / L_z, \tag{1}$$

where Ω is the volume of the unit cell, f_0 the dipole matrix element connecting Bloch states in the valence and conduction bands, and $\phi_{1s}^{2D}(r_e - r_h)$ the 2D $1s$ hydrogenlike wave function describing the in-plane electron-hole relative motion. $|\phi_{1s}^{2D}(0)|^2$ gives the probability for the electron and hole being in the same unit cell:

$$|\phi_{1s}^{2D}(0)|^2 = 2/\pi a_0^2(L_z) = 2/A_x, \tag{2}$$

where A_x is the area of the 2D exciton with Bohr radius $a_0(L_z)$. For $K=0$ excitons which satisfy the k-conservation rule, all unit cells within the quantum well contribute to the optical transition, resulting in a transition strength F_x^{2D} for the $1s$ 2D exciton:

$$F_x^{2D} = N f_x^{2D} = f_0 \times 2A/A_x, \tag{3}$$

where N is the number of unit cells in the volume of the quantum well, which is given by AL_z, with A the area of the quantum-well layer. Actually, L_z cancels in Eq. (3) and the dependence of F_x^{2D} on thickness L_z is determined only by $a_0(L_z)$.

In reality, however, not only $K=0$ excitons contribute to the optical transition as reflected by the fact that the homogeneous exciton linewidth at finite temperatures is not δ-like but has a certain spectral width $\Delta(T)$, basically determined by interaction of the excitons with acoustic phonons.[7-9] The effective transition strength for the ex-

citon may then be obtained by sharing the $K=0$ oscillator strength, which would correspond to a δ-like absorption or emission line, equally among all states within the finite spectral width $\Delta(T)$. In 2D the density of states is constant and the number of states within $\Delta(T)$ is given by

$$D(E)\Delta(T)=A(M/2\pi h^2)\Delta(T) \quad (M=m_e^*+m_h^*), \tag{4}$$

which yields

$$F_x^{2D}=f_0\times 4\pi h^2/A_x\Delta(T)M. \tag{5}$$

This equation can be rewritten by introduction of the quasi 2D exciton binding energy $E_B^{2D}=4h^2/\mu(a_0^{2D})^2$, where μ is the reduced mass $\mu^{-1}=(m_e^*)^{-1}+(m_h^*)^{-1}$:

$$F_x^{2D}=f_0[E_B^{2D}/\Delta(T)]\mu/M. \tag{6}$$

Only the fraction of excitons $r(T)$ within the spectral width $\Delta(T)$ can contribute to recombination. For Maxwell statistics the fraction $r(T)$ in 2D is given by

$$r(T)=\frac{\int_0^{\Delta(T)}D(E)f(E)\,dE}{\int_0^{\infty}D(E)f(E)\,dE}=1-e^{-\Delta(T)/kT}. \tag{7}$$

The decay of the luminescence due to free-exciton recombination thus is determined by an effective transition strength

$$F_{x,\mathrm{eff}}^{2D}=f_0E_B^{2D}\frac{\mu}{M}\frac{r(T)}{\Delta(T)}, \tag{8}$$

which defines the decay constant according to[10]

$$\tau_x^{2D}=2\pi\epsilon_0m_0c^3/\tilde{n}e^2\omega^2F_{x,\mathrm{eff}}^{2D}, \tag{9}$$

where $\tilde{n}$ is the refractive index and the other symbols have their usual meaning. Inserting Eq. (8) into (9) finally yields

$$\tau_x^{2D}\propto(E_B^{2D})^{-1}(M/\mu)\Delta(T)/r(T). \tag{10}$$

The decay constant τ_x^{2D} thus does not exhibit an explicit L_z dependence but depends on the quantum-well thickness only implicitly via the exciton binding energy. This should not be confused with the free-exciton absorption strength. The absorption strength is obtained by multiplying the free-exciton transition strength with the density of "absorbing centers," which brings in an additional explicit L_z dependence as verified experimentally.[11] The absorption strength thus shows a stronger variation with L_z compared to the free-exciton decay times.

A linear dependence between the exciton lifetime and exciton linewidth is predicted by Eq. (10) in addition to the implicit L_z dependence. In order to obtain a physical understanding of the relationship between exciton lifetime and linewidth it is helpful to compare the result for the free exciton with bound-exciton recombination. The oscillator strength for bound excitons in 3D has been calculated by Rashba and Gurgenishvili[12] and was discussed for 2D bound excitons by Rorison and Herbert.[13] For strong electron-hole correlation, the expression for the oscillator strength of a bound exciton can be intuitively deduced from Eq. (3) by our replacing the number of unit cells in the entire quantum well by the number of unit cells covered by the center-of-gravity motion of the bound exciton, N_{BX}. Introducing again the area (A_{BX}) covered by the center-of-gravity motion, we obtain for the oscillator strength per impurity atom

$$F_{Bx}^{2D}=f_0A_{BX}/A_x. \tag{11}$$

The oscillator strength *per impurity atom* for a bound exciton is thus higher than the oscillator strength per *unit cell* of a free-exciton transition by a factor given by the ratio of the "volume" of the bound 2D exciton ($V_{BX}=A_{BX}L_z$) to the volume of a unit cell.

This result, known as the "giant oscillator strength" of bound excitons, is somewhat misleading. The transition probability is much larger than that associated with tightly bound atomic impurities, and accounts for the fact that bound-exciton transitions are observed in 3D GaAs at impurity concentrations as low as 10^{-10} cm^{-3}. However, the oscillator strength of such bound excitons is weaker than that for coherent $K=0$ excitons by a factor A_{BX}/A. As we have seen, scattering of free excitons reduces the effective strength of free excitons and actually Eq. (5) can also be written as

$$F_x^{2D}=f_0A_c/A_x, \tag{12}$$

where $A_c=4h^2/\Delta(T)M$ is an area which can be identified with the area of coherence of the exciton before scattering. In typical samples A_{BX} and A_c are similar in magnitude so that Eq. (11) and Eq. (12) both yield transition strengths of order unity, as recently confirmed experimentally for 3D excitons in GaAs.[14] The dependence of the radiative exciton lifetime on the spectral linewidth as expressed in Eq. (10) thus is mediated by the coherence area of the quasi 2D exciton, which is determined by exciton scattering.

We next describe and discuss our experimental results which demonstrate the fundamental relation between the spectral linewidth and the exciton lifetime. The samples studied are GaAs-AlGaAs multiple quantum wells (MQW) with GaAs thickness (L_z) between 2.5 and 15.0 nm grown by molecular-beam epitaxy at Philips Research Laboratories. The details of the growth procedure are described elsewhere.[15] The quantum wells are excited by pulses of a synchronously mode-locked dye laser (5-ps FWHM) tuned below the band-gap energy of the AlGaAs. The excited sheet electron-hole concentration was of the order of 5×10^{10} cm^{-2}. The low-temperature ($T<100$ K) photoluminescence in all samples can be attributed to exciton recombination by a comparison of photoluminescence and excitation spectra. The Stokes shift is in the range of 0–5 meV; the linewidth (FWHM) varies between 3 and 10 meV. The

temporal variation of the spectrally integrated exciton luminescence was detected with a Synchroscan streak camera with a time resolution of 20 ps.

The dependence of the decay time[16] τ on well thickness L_z at $T=5$ K for the series of MQW samples investigated is plotted in Fig. 1. We observe the same systematic decrease of τ with decreasing L_z as reported previously,[17] although the absolute numbers are slightly larger. The decrease of τ with decreasing L_z can be attributed on the one hand to the increase of the exciton binding energy E_B^{2D}. This is expected to show an increase of a factor of 4 between true 2D excitons as $L_z \rightarrow 0$ with infinite barriers and a true 3D exciton. However, for a quantitative description, the read band structure, in particular the variation of the valence-band structure with L_z, has to be taken into account.[5,18] The recent results of 't Hooft *et al.* indicate that τ would saturate at about 3.3 ns for the fully 3D case with $L_z \rightarrow \infty$.

On the other hand, the change of the exciton linewidth with L_z contributes to the lifetime. In general, however, it is difficult to investigate this quantitatively on the basis of the L_z dependence alone, because the exciton linewidth in quantum wells contains a sample-dependent inhomogeneous contribution resulting from well-width fluctuations.[19] The homogeneous contribution to the exciton linewidth of a particular sample, however, can be easily varied by the temperature. A variation of the radiative lifetime with temperature consequently is only predicted for free excitons because of the variation of $\Delta(T)/r(T)$ in Eq. (10). Instead, the lifetime is independent of temperature for bound and localized[20] excitons and only a decrease of the emission intensity is expected with increasing temperature because of ionization of the bound or localized excitons. In the limit $\Delta(T) \ll kT$, the exciton lifetime is proportional to kT, while for $\Delta(T) \gg kT$, $\tau \propto \Delta(T)$ is expected, according to Eq. (10). In the temperature regime under consideration the width $\Delta(T)$ will be dominated by impurity and interface scattering and by acoustic-phonon scattering and is expected to vary like $\Delta(T)=\alpha+\beta T$. The results for the homogeneous linewidth of excitons in QW recently reported by Schultheis *et al.*[7] yield $\beta \simeq 10$ μeV/K, which gives $\Delta \simeq 0.5$ meV at $T=50$ K. This means that Δ/kT in our earlier expression is $\simeq 0.1$ and so the $\Delta \ll kT$ regime holds and $\tau \propto kT$.

The experimental results of the temperature dependence of τ up to 50 K for different L_z are summarized in Fig. 2. The decay times increase rapidly with increasing temperature for all samples. The photoluminescence intensity instead remains constant which demonstrates that the exciton decay is dominated by radiative recombination. The increase in τ is almost linear with temperature with about the same slope for different L_z as expected from Eq. (10), except for the thinnest sample with $L_z=2.5$ nm. We therefore conclude that at least for the samples with $L_z \geq 5$ nm, the recombination is determined by free-exciton recombination and the results unambiguously demonstrate the fundamental relationship between exciton linewidth and radiative lifetime. The radiative-exciton recombination rate consequently is not solely determined by the exciton oscillator strength but depends on the "coherence volume" of the exciton, which in turn is determined by exciton scattering. For thinner samples, as for the MQW with $L_z=2.5$ nm in Fig. 2, localization in fact may contribute and the change of the slope in τ vs T may indeed be attributed to a temperature-independent contribution to the decay at low temperatures as expected for bound or localized excitons. This point, however, needs further investigation.

In summary, we have studied theoretically and experimentally the dependence of exciton lifetimes in quantum wells on quantum-well thickness and temperature. We have demonstrated the fundamental relationship between the spectral linewidth and the radiative lifetime of excitons resulting from the fact that the coherent extension of an exciton is determined by the exciton scattering. As an important consequence it has to be realized that the radiative decay time of excitons in different quantum wells with the same thickness L_z is not a unique number but may vary from sample to sample as a result of the

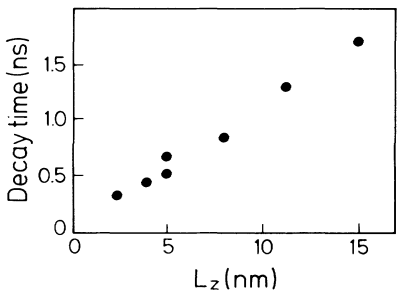

FIG. 1. Exciton decay times vs quantum well thickness L_z for GaAs-AlGaAs multiple-quantum-well samples at 5 K.

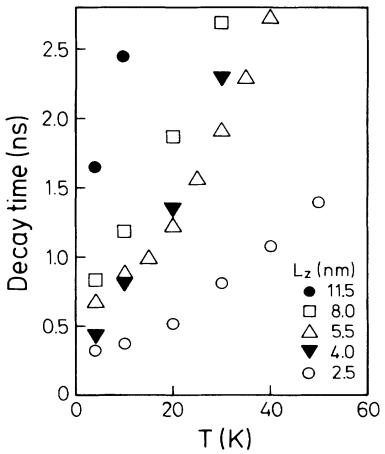

FIG. 2. Temperature dependence of exciton decay times for multiple quantum wells with different thicknesses L_z.

difference in the homogeneous part of the linewidth.

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