Evidence for superradiant decay of excitons in InAs quantum sheets

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Radiative lifetimes are measured for excitons attached to isolated (001) InAs lattice planes interspersed in bulklike GaAs. The broken translational invariance of the system provides an efficient radiative decay channel for excitons. We demonstrate that the radiative lifetime is determined by the spatial extent of areas of continuous In coverage. This dependence directly indicates the superradiant nature of the spontaneous-emission process, since the decay rate is proportional to the area over which the coherent polarization of the exciton extends.

Breaking the translational invariance of bulk semiconductors modifies the interaction of excitons with an incident electromagnetic field. The coupling of excitons to photons in presence of translational symmetry leads to the formation of exciton polaritons, which are quasistationary states, whose energy is resonantly transferred between radiation and polarization fields.[1] Transitions then can occur only via three-step processes, e.g., by the coupling of exciton polaritons to acoustic phonons.[1,2] Lack of wave-vector conservation caused by reduced symmetry introduces a density of final states which allows direct radiative decay of excitons. Since the exciton represents a coherent macroscopic polarization within the whole crystal, its decay rate is predicted to be proportional to the relevant number of unit cells.[3] Excitons in microcrystallites and thin films are thus expected to undergo a superradiant decay in the sense that the decay rate scales with the volume available to the excitons motion.[4] Experimentally, the decay rate of excitons has been related to a phenomenological coherence volume which represents the effect of dephasing collisions with acoustic phonons.[5] An excitonic decay in semiconductors depending on a geometrical coherence volume has been reported, to our knowledge, only for CuCl microcrystallites,[6] where, however, nonradiative recombination at charged surface states may also account for the observed lifetime variation.

We observe a superradiant decay of excitons in III-V semiconductors. The isoelectronic substitution of isolated (001) lattice planes in a GaAs crystal with In breaks the translational invariance of the system, thereby creating an efficient radiative decay channel for excitons attached to the planes. The structures exhibit exceptional crystal quality, which is of vital importance for the phase-sensitive dynamics of excitons. We show that the decay rate is directly controlled by a geometrical constraint, namely, the spatial extent of areas that are continuously covered with In. Experimental control of the size of these areas is achieved which allows us to manipulate the translational symmetry of excitons. Our results reveal the decisive importance of translational symmetry and spatial coherence for the optical response of excitons in low-dimensional systems.

The heterostructures are synthesized by molecular-beam epitaxy on semi-insulating (001) GaAs substrates with various misorientations. Growth conditions are adjusted to ensure that nucleation on terraced surfaces proceeds by step propagation. The final structure con-

TABLE I. Structural sample parameters including the average In coverage δ,[a] the separation of the InAs planes Δ,[a] the angle of misorientation ϕ,[b] the crystallographic direction of the misorientation C,[a] and the mean terrace size L.

Sample No.	δ (monolayer)	Δ (nm)	ϕ	C	L (nm)
1	0.8	11.3	≤ 0.5		$\gg 10$[a]
2	1.2	14.5	≤ 0.5		≈ 10[c]
3	0.8	10.5	1.6	[110]	14[d]
4	0.7	10.5	3.2	[110]	7[d]
5	0.3	27.3	3.2	[100]	7[d]
6	0.3	27.3	6.4	[100]	3.5[d]

[a]Determined by HRDXD.
[b]Determined by HREM.
[c]Average distance of monoatomic steps as taken from HREM images of this kind of structure. The distance is typically 8–12 nm.
[d]Geometrical average as given by the misorientation angle. The actual mean terrace size observed in HREM images is close to this nominal value. The size distribution around the mean value is about $\pm 10\%$.

sists of a GaAs crystal interspersed with ten isolated (001) InAs lattice planes. The actual structural configuration, summarized in Table I, is precisely determined by high-resolution double-crystal x-ray diffraction (HRDXD) and high-resolution transmission electron microscopy (HREM).[7,8] For luminescence, the samples are excited by a synchronously pumped mode-locked dye (Styryl 9) laser. The measurements are taken at 6 K with a maximum excited electron-hole pair density of 5×10^{10} cm^{-2} per pulse. The detection system consists of a 32-cm spectrometer, followed by a two-dimensional (2D) streak camera. The overall time resolution of the system is better than 15 ps.

We first outline results of the HRDXD and HREM investigations. In all samples the high-crystal quality of the GaAs matrix is preserved, unaffected by the inserted InAs lattice planes (7.16% mismatch). In particular, the internal interfaces are coherently strained and thus defect-free. The morphology of the InAs film is controlled by the absolute In coverage. On singular surfaces, an In coverage in excess of one monolayer corresponds to a single InAs monolayer with additional InAs islands on the top. Under special growth conditions,[8] these islands are of single atomic-step height and exhibit a *characteristic* lateral extent of 10 nm. Submonolayer films actually consist of a single InAs monolayer pierced by the GaAs matrix. In the diffraction patterns of submonolayer samples, no detectable broadening of the higher-order satellites is observed, indicating a much higher homogeneity than for films in excess of one monolayer.[7] On vicinal surfaces, the morphology of the InAs film is predominantly controlled by the terrace morphology of the underlying surface. Submonolayer coverages lead to the formation of separated InAs clusters at the terraces, being a consequence of the step-edge nucleation of In adatoms.[9]

In Fig. 1(a) we show the transient spectra of sample 1 after pulsed excitation at 800.0 nm. The two high-energy bands at 818.0 and 830.0 nm are identified as the free-exciton and the free-to-carbon transition in undoped bulk GaAs.[10] The bright emission band at 842.9 nm originates from excitonic states created by the InAs planes.[7] The spectral position of the transition does not shift with time. No Stokes shift is observed with respect to the corresponding transition probed by absorption-related techniques.[7] The decay of the emission takes place on a time scale of 100 ps, which, as will be discussed below, corresponds to the true radiative lifetime of the state.

Both the spectral position and the temporal evolution of the emission are affected as soon as the monolayer coverage is exceeded, as is evident from the transient spectra of sample 2 shown in Fig. 1(b). The InAs-related emission is redshifted to 860.0 nm compared to the corresponding transition of sample 1, reflecting the existence of single atomic steps on top of the continuous InAs monolayer. The atomic-scale morphology of the InAs planes manifests itself also in the spectral diffusion of the transition, which amounts to 5.5 meV, coincident with the observed Stokes shift. The temporal evolution of the emission exhibits a considerably faster rise than for sample 1, namely, 30 ps instead of 90 ps. Furthermore, the

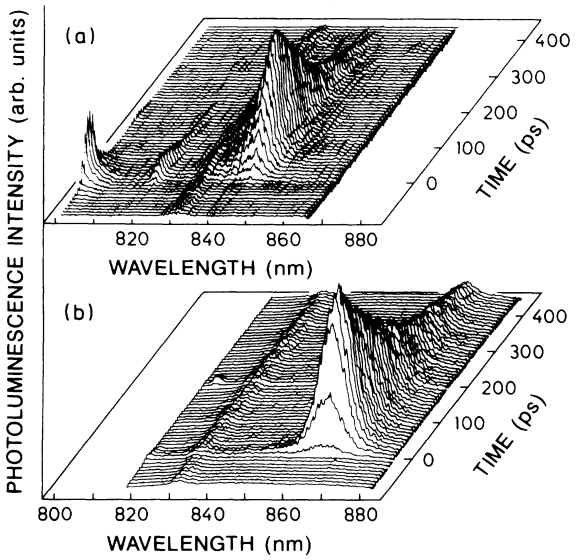

FIG. 1. Transient photoluminescence spectra at 6 K of samples (a) 1 and (b) 2. The spectra are 8 ps apart. The exciting laser pulse is set to 800.0 nm.

decay of the emission takes place on a much longer time scale than that of sample 1, namely, about 300 ps. It is the higher lateral homogeneity of the InAs sheets existing in the submonolayer regime which leads to this pronounced differences in both the temporal and spectral characteristics. In particular, the different rise time is caused by the conservation of the exciton momentum along the layer for perfect interfaces, resulting in a slow (about 400 ps) relaxation to the bottom of the band.[11] In contrast, excitons are rapidly scattered to the radiating state in the presence of interface roughness.[12]

In Fig. 2 we show the time dependence of the spectral-

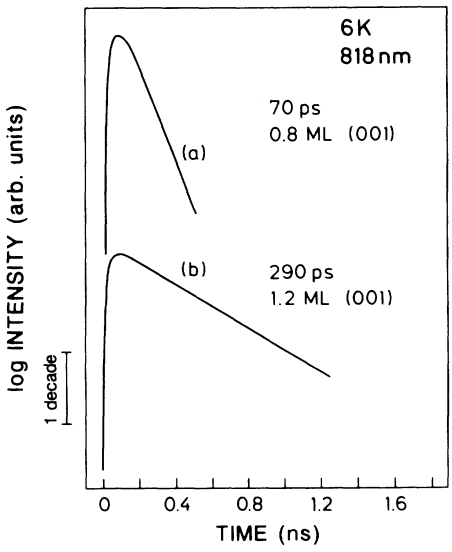

FIG. 2. Time dependence of the spectrally integrated emission at 6 K from sample (a) 1 and (b) 2. The exciting pulse is set to 818 nm, in resonance to the GaAs exciton. Decay times and the In coverage are indicated.

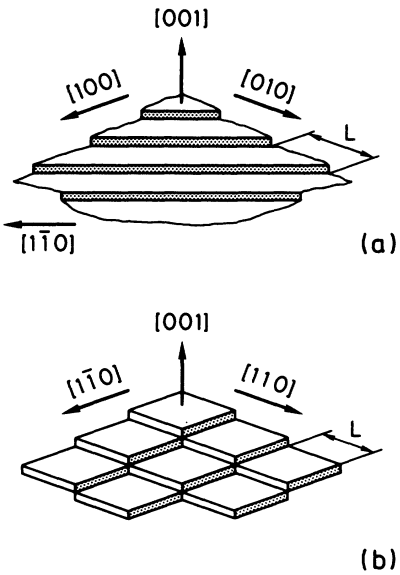

FIG. 3. Schematic representation of terrace configurations for (001) GaAs surfaces misoriented towards (a) [110] and (b) [100]. Steps are running along the misorientation direction C as denoted in Table I. The terrace size L is given by $L=a/(\sqrt{2}\tan\phi)$, where a is the lattice constant and ϕ the misorientation angle as listed in Table I.

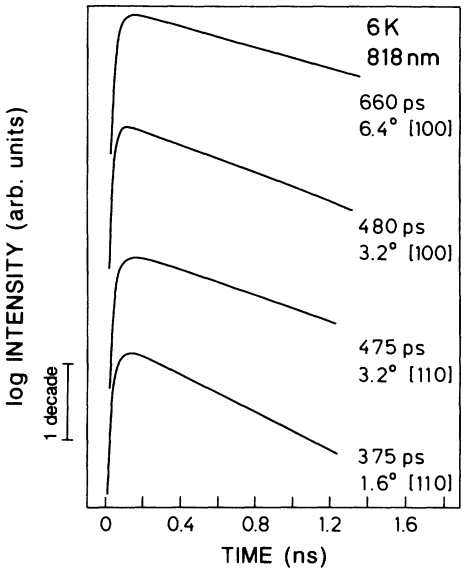

FIG. 4. Time dependence of the spectrally integrated emission from samples 3, 4, 5, and 6. The experimental conditions are the same as for Fig. 2. Decay times and degree and the direction of the misorientation are given.

ly integrated emission of samples 1 and 2. The monoexponential decay allows us to determine the decay times to 70 ps (1) and 290 ps (2). These values can be identified as the actual radiative lifetime of the excitons for the following reasons. The dependence of the luminescence efficiency on excitation density is found to be strictly linear over more than six orders of magnitude. A deviation from linearity only occurs at excitation densities above 5×10^{12} cm^{-2} per pulse, resulting from the progressive filling of the InAs states. Furthermore, the decay time is constant for any excitation density below the band-filling regime. These findings demonstrate that the radiative recombination of the InAs-related excitons efficiently bypasses any concurrent nonradiative channel, resulting in an internal quantum efficiency of unity.[13]

In quantum wells, the spontaneous-emission processes are believed to be dominated by the modified electron and hole overlap.[14] Our observation, however, opposes this intuitive interpretation,[15] which shows that not confinement, but rather the reduced translational symmetry dominates the optical response of 2D excitons.[16] The different radiative lifetimes of the two samples presented above consequently reflect different characteristic areas available for the excitons 2D translational motion. To go further, we consider an exciton having an in-plane radius a_B within a "quantum sheet" of lateral extent L, defined by the area over which the In coverage remains continuous. Denoting the wavelength of the incident radiation as λ, one has to distinguish two fundamentally different regimes: (i) $a_B<L\ll\lambda$ and (ii) $L<a_B\ll\lambda$.

In case (i), the center-of-mass motion of the exciton extends over the whole sheet. The radiative decay rate R is then determined by the characteristic dimension L over which the coherent polarization persists:[17]

$$R\simeq R_0|\langle\chi_e(z)|\chi_h(z)\rangle|^2|\phi_{\rm ex}^{\rm 2D}(0)|^2L^2$$
$$\simeq R_0|\langle\chi_e(z)|\chi_h(z)\rangle|^2\left[\frac{L}{a_B}\right]^2, \quad (1)$$

where R_0 is the decay rate for interband transitions, $\langle\chi_e(z)|\chi_h(z)\rangle$ is the overlap integral of the electron and hole wave functions in the direction perpendicular to the InAs planes, $\phi_{\rm ex}^{\rm 2D}$ is the wave function of the correlated relative motion along these planes, and a_B is the corresponding Bohr radius.

In case (ii), the exciton has no degree of freedom in translation. The radiative transition rate is not enhanced by the factor (L/a_B), hence no superradiant decay occurs but atomiclike transitions are recovered.[18] In the intermediate range between these two limiting regimes the decay rate is predicted to increase continuously with L.[19]

To test the consequences of the above considerations, we fabricated a set of samples (Nos. 3, 4, 5, and 6) with nominally the same In coverage as for sample 1, but on *terraced* substrates (Fig. 3). The spectral position of the excitonic transition is identical to that of sample 1 for all of these samples.[20] Only the *continuity* of the plane potential is restricted artificially by the terrace size, whereas its *strength* is the same as for sample 1. We thus achieve direct experimental access on the excitons 2D motion by tuning the terrace size. In Fig. 4, we show the time dependence of the spectrally integrated luminescence for these samples. The measured lifetimes are drastically longer than the one observed for sample 1, and they increase continuously with decreasing terrace size. Both these phenomena are the sole consequence of the *geome-*

trical constraint imposed on the excitons 2D motion by the interrupted In coverage. In particular, the lifetime observed for sample 6 is close to that of an atomiclike transition, given by $R_0^{-1} \simeq 600$ ps. That is, the planar InAs aggregates realized on the vicinal surfaces represent a true 0D system, in the sense that we actually enter the regime where the translational motion of the exciton is frozen out.[20] In contrast, for sample 1, the In coverage is continuous on a length sale of about three times the Bohr radius [Eq. (1)], and the 2D translational motion of the exciton is preserved within the InAs sheet. All unit meshes within the sheet area contribute to the emission process, and the exciton therefore decays superradiatively via its macroscopic polarization.[4]

In conclusion, we have shown that a planar isoelectronic substitution in a bulk crystal creates an efficient radiative decay channel for excitons by breaking the translational invariance. The decay rate of excitons attached to the planes is determined by the size of areas over which their coherent polarization extends. Direct experimental control of this size has allowed us to manipulate the dimensionality of excitons. In particular, the continuous transition from excitons with a free 2D translational motion to an atomiclike system has been demonstrated. Our experiments has revealed the key role of translational symmetry and spatial coherence for the interaction of excitons with radiation.

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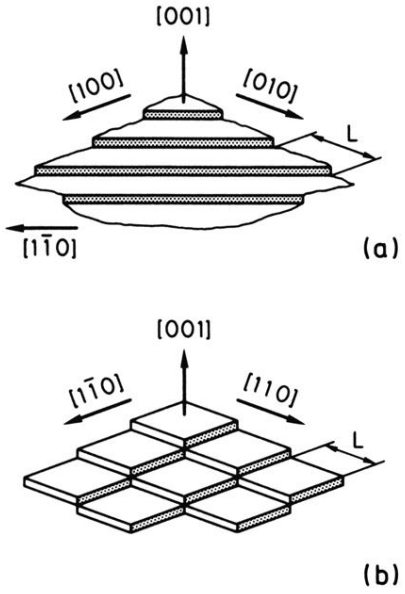

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