Ultrafast Breathinglike Oscillation in the Exciton Density of ZnSe Quantum Wells

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The spatial density profile of a low-density exciton ensemble in ZnSe quantum wells shows a breathinglike oscillation on a 30-ps time scale. This breathing results from the emission of the first acoustic phonon at the end of the quasiballistic transport phase of the excitons which reverses their direction of propagation. Since the scattering destroys the phase of the excitonic wave function, one can deduce simultaneously the coherence length and the coherence time of excitonic transport by evaluation of the oscillation measured from a single experiment. The breathing, which can be modeled by Monte Carlo simulations, is quenched for rising lattice temperature, i.e., increasing phonon absorption, and in samples with significant disorder. These results were obtained by time-resolved nanophotoluminescence with 5 ps and 250 nm temporal and spatial resolution, respectively.

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In the quest of improving the speed of semiconductor devices (e.g., for communications and computing), it is obvious that the continuous reduction of the gate size with every device generation will lead to a natural boundary in the near future. Consequently, one is exploring alternative routes to achieve ultrafast switching or logic operations. One promising way is to utilize the coherence of quantum mechanical states in semiconductors. In particular, one studies the coherent preparation, the coherent superposition, and the coherent manipulation of carriers and optical excitations in semiconductor nanostructures [1].

It is an important ingredient to these new approaches to information processing that coherence can be transported in semiconductor structures. In particular, spin coherence, which can survive under certain conditions up to milliseconds, can propagate over macroscopic distances [2,3]. The coherence of the spatial part of the wave function of carriers or excitons is destroyed much quicker, typically on a few 10 ps time scale or faster [4]. Since this coherence couples strongly to light, it can lead to efficient ultrafast operations. It should be interesting to utilize spatial interference effects for coherent information processing. Such interferences like Young's double-slit experiment have been demonstrated for electrons in doped semiconductors [5]. But also the coherence length of excitonic transport has recently been determined in ZnSe-based quantum wells to be in the range of several 100 nm [6].

The coherence time of excitons in semiconductors has been well studied by linear spectroscopy using the homogeneous linewidth [7] or nonlinear techniques like four-wave mixing [4] and pump-probe experiments [8]. But only recently it was possible to uncover the quasiballistic (i.e., coherent) transport regime of excitons [6,9]. Such experiments require a spatial resolution better than the wavelength of light. A rather efficient and flexible setup to achieve a spatial resolution of 250 nm is based on standard confocal micro-photoluminescence but includes a solid immersion lens (SIL) [10]. The high collection efficiency of the SIL allows one to work at low exciton densities where exciton-exciton interation can be neglected. Since the detection spot (defined by a pinhole in the image plane of the microscope) can be moved independently from the excitation spot, one can follow the lateral spread of the excitonic luminescence over several μm. The coupling of the exciton dynamics in space and in energy as well as the coherence length of excitonic transport have been explored by this technique.

In this Letter we want to document a breathinglike evolution of the exciton density profile in ZnSe quantum wells. The origin of this spatiotemporal oscillation is the emission of the first acoustic phonon by the excitons. So this oscillation is a spectacular marker for the end of the quasiballistic phase of excitonic transport. Further, although we do not measure excitonic coherence directly, this oscillation gives the unprecedented opportunity to determine the coherence time and length of excitonic transport within a single experiment. The breathing is quenched upon increasing lattice temperature or for high interfacial disorder demonstrating the role of phonon absorption and localization, respectively.

We measured the temporal evolution of the photoluminescence spot in two ZnSe/ZnMgSSe multiple quantum-well (MQW) samples with 10 wells of 8 and 5 nm well width, respectively, in a ZnSe/ZnSSe MQW with 120 wells of 7.3 nm well width, and in two ZnSe/MgS MQW samples with 10 wells of 10 or 5 nm well width, respectively. Our experimental setup consists of a picosecond laser for excitation (pulse length $\approx$2 ps; spectral width $\approx$1 meV), a confocal microscope, a spectrometer of 0.5 meV spectral resolution, and a streak camera with 5 ps temporal resolution. In order to improve the spatial resolution and the signal collection efficiency, we adhesively fixed a ZrO_2 solid immersion lens with a refractive index $n = 2.16$ to the sample surface. The spatial resolution of the detection is 250 nm (half width at half maximum) limited by the 20 μm pinhole in the image plane of

the microscope objective. With this method, we can obtain the spatial distribution of the photoluminescence (PL) in the well along the scanning axis at any time after excitation. The samples were kept in liquid helium flow cryostat at a lattice temperature of 7 K unless stated otherwise.

The high collection efficiency of the setup allows us to work on the low-density regime. The excitation fluence used is below 0.01 $\mu J/cm^2$ so that excitonic correlation effects, which have been well studied in the same samples, namely, excitation-induced shift or dephasing [8] as well as biexcitonic effects [11], are negligible. The dephasing time is at its low-density limit which is determined by phonon scattering. We confirmed that experimental data on exciton transport did not show any change when raising the excitation density up to a factor of 10.

In quantum wells based on the polar semiconductor ZnSe, excitons can form efficiently for nonresonant excitation via the emission of LO phonons [12,13]. This is in contrast to the situation in GaAs [14] and results from the strong Fröhlich coupling and much larger exciton binding energy (for a discussion, see [15]). The time scale of the LO-phonon assisted exciton generation is well below our temporal resolution, so we can assume to start with an initial population of excitons with a well defined excess (i.e., kinetic) energy $E_{\mathrm{kin}} = E_{\mathrm{laser}} - E_{\mathrm{PL}} - n \times E_{\mathrm{LO}}$, with $0 \leq E_{\mathrm{kin}} < E_{\mathrm{LO}}$. E_{PL} corresponds to the energy position of the maximum of the photoluminescence and in the present study $n = 2$. Thus, by tuning the laser energy, we can directly vary the initial kinetic energy E_{kin} of the excitons [12,13,16].

From Monte Carlo simulations of a large exciton ensemble excited within a spot of 400 nm diameter with a given kinetic energy and calculating the exciton-acoustic phonon interaction as well as interface roughness scattering, one expects the excitons to move quasiballistically for tens of ps [9]. Since the disorder scattering is predominantly a forward scattering, the excitons will move on average away from their place of generation thus rapidly increasing the size of the exciton density distribution. Even a breathinglike oscillation of the exciton density profile was predicted by such calculations [9] since the emission of an acoustic phonon is predominantly a backscattering process reversing the direction of the exciton motion. On the other hand, in spatially and temporally resolved experiments one notices only a rather slow expansion of the excitonic photoluminescence spot. The reason for this discrepancy is simply the fact that excitons in high momentum states within their parabolic dispersion are dark and do not contribute to the PL signal.

The observation of a spatial breathing requires techniques that detect the whole exciton ensemble instead of monitoring only the bright excitons. It has been demonstrated that THz absorption could be used to detect the whole exciton ensemble [14]. However, the ultralong wavelength makes it hard to achieve high spatial resolution. It was shown that phonon-sideband (PSB) emission reflects the whole exciton population [12,16,17]. Indeed the spot diameter of this emission shows a clear oscillation within the first 30 ps for nonresonant excitation with arbitrary excess energy (see Fig. 1). But this phonon-sideband PL signal is rather weak and too noisy for a detailed and quantitative analysis. Furthermore, only one of our samples provides a strong enough PSB signal above background for deducing a clear oscillation (Fig. 1).

Zero-phonon emission, which is of course visible in all our samples, typically does not show any spot-size oscillations (see Ref. [9]). For initial kinetic energies exceeding 3 meV one finds the typical sublinear increase of the PL-spot area with time [9]. The reason for this behavior lies in the fact that the excitons with high momentum (these contribute to the oscillation) do not contribute to the emission. But under certain excitation conditions even zero-phonon spectroscopy is able to directly monitor an excitonic breathing: the spatiotemporal oscillation of the exciton density profile emerges clearly in the ZnSe/ZnSSe and in the ZnSe/ZnMgSSe samples (an example is given in Fig. 2) when one tunes the laser to a quasiresonant excitation (i.e., resonant with the excitonic PL after the emission of two LO phonons during the exciton generation).

Why can the oscillation now be observed? The answer is related to the movement of the excitons in a fluctuating potential. It is important to note that the observation of the breathinglike oscillation is possible only in samples with a moderate inhomogeneous broadening of the global exciton resonance due to interface disorder. This is the case for the ZnSe/ZnSSe and the two ZnSe/ZnMgSSe quantum-well samples (inhomogeneous linewidth about 3 meV). Quasiresonant excitation within the homogeneous linewidth

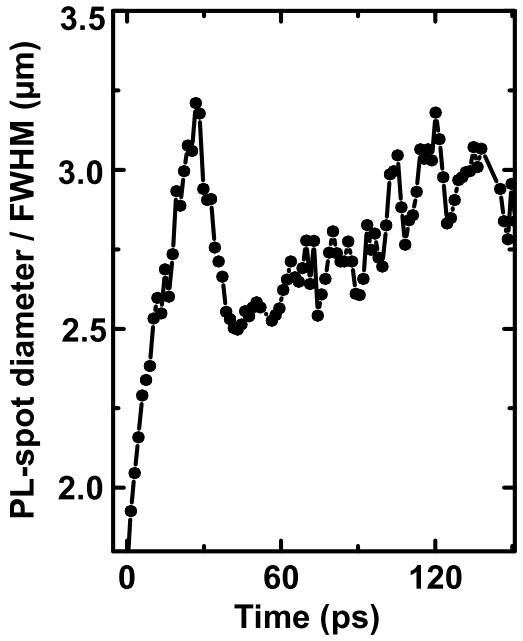

FIG. 1. Temporal evolution of the spatial expansion of the PL spot detected in the phonon-sideband emission form a ZnSe/ZnSSe MQW. The initial kinetic energy of the excitons is 22 meV.

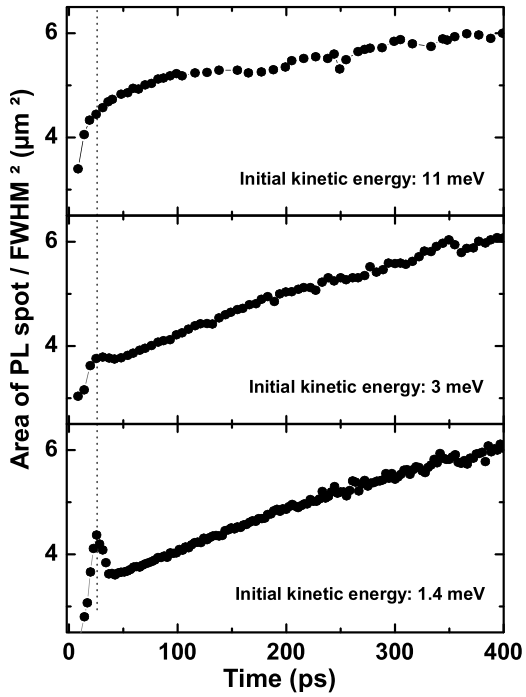

FIG. 2. Temporal evolution of the spatial expansion of the PL spot of the zero-phonon line given by the square of the full width at half maximum in a 8 nm ZnSe/ZnMgSSe MQW sample. The excitons are generated with different initial kinetic energies.

Γ_{hom} of the exciton in a disorder-free sample would leave the exciton with a kinetic energy ($\Gamma_{\mathrm{hom}} = 0.1$–$0.2$ meV in ZnSe-based QWs [8]) much too small for further relaxation via the emission of an acoustic phonon. This relaxation requires an excess energy of at least 0.36 meV as can be calculated from the exciton and phonon dispersions. On the other hand, the oscillation is not observed in samples with significant disorder like ZnSe/MgS QWs (inhomogeneous linewidth typically 10 meV) where trapping into localization sites already dominates on a short time scale. In the ZnSe/ZnMgSSe samples, however, the excitons move quasiballistically several 100 nm within the first few 10 ps and see a moderately varying potential landscape. This easily allows the relaxation of the excitons by emission of an acoustic phonon into a (locally defined) radiative state which explains the observation of the oscillation.

We now want to analyze the results of the temporal evolution of the quasiresonant PL spot. The density profile (which is now essentially identical to the PL-spot profile) and, in particular, the position and amplitude of the spatial oscillation are accurately reproduced by Monte Carlo simulation, as shown in Fig. 3(a). The simulation considers acoustic-phonon absorption, acoustic-phonon emission, and scattering by interface roughness as the three possible processes occurring to an exciton with the initial kinetic energy E_{kin} during its lifetime. Exciton-exciton interactions are neglected because of the low excitation densities. For each of these processes, we calculated the scattering rates according to the theory of Basu and Ray [18] and Takahashi [19]. We reproduce the measured profile by adjusting only one fit parameter, namely, the interface roughness correlation length λ. Here λ is found to be 11 nm, which is about 3 times the exciton Bohr radius, assuming a roughness of 1 monolayer in average. The later assumption is reasonable as can be deduced from high-resolution transmission electron microscopy of ZnSe/ZnMgSSe interfaces. Still, our model for interface roughness is presently not elaborate enough to yield more insight into the role of disorder.

The explanation for the breathinglike oscillation is easily deduced from the angular dependence of the scattering probability for the three involved scattering channels [Fig. 3(b)]. Excitons with $E_{\mathrm{kin}} = 1$ meV are calculated to have a probability of 61% and 68% to propagate in the forward direction after one interface roughness scattering and one acoustic-phonon absorption event, respectively. On the other hand, a majority of excitons (65%) reverse their direction of motion after emitting an acoustic phonon. These findings mean that the spatial oscillation is directly related to the first emission of an acoustic phonon. Since this is the first inelastic scattering event of the exciton at low temperature it leads to the loss of coherence of the individual exciton wave function. The mean excitonic coherence time t_c is then given by the maximum of the oscillation to be $t_c = 29 \pm 2.5$ ps. The coherence length can be obtained from the spatial profile of the PL at the time $t = t_c$. Taking into account the finite detection spot size by performing deconvolution, we found $l_c = 800$ nm. We thus can determine both coherence time and length from one single experiment.

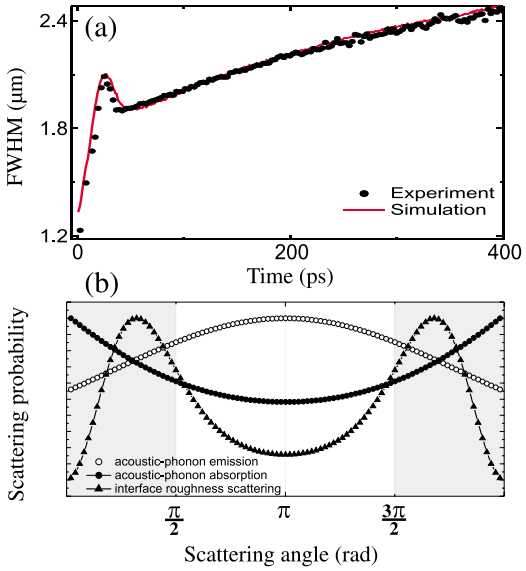

FIG. 3 (color online). (a) Experimental (dots) and simulated (line) temporal evolution of the PL-spot diameter for excitons with 1 meV kinetic energy. (b) Calculated probabilities for various scattering processes of excitons with the kinetic energy in (a). Shaded regions give the areas of scattering angles in the forward direction.

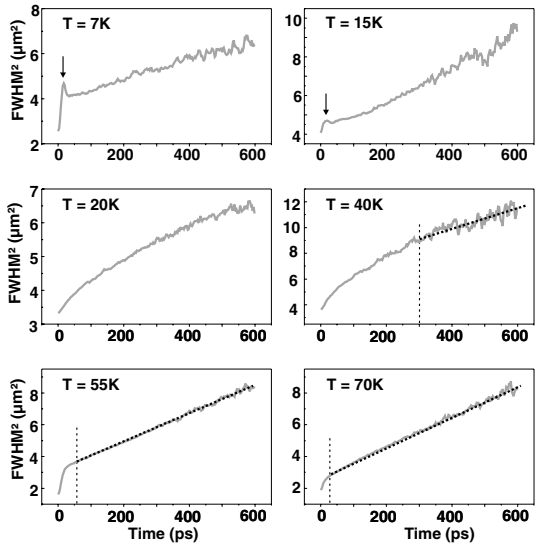

FIG. 4. Temperature dependence of the temporal evolution of the PL-spot size (squared full width at half maximum) at various temperatures in the 8 nm ZnSe/ZnMgSSe MQW. The excess kinetic energy is fixed to 1.5 meV. The arrows mark the oscillation; the dashed lines mark a linear dependence of the spot area on time.

The coherence time is in agreement with four-wave mixing results on the same samples [8]. The coherence length is consistent with earlier cw experiments on the phonon-sideband emission [6]. From the values of t_c and l_c, we can define a quasiballistic transport velocity $v_T = l_c \div t_c = 27\,500\ \mathrm{ms}^{-1}$. The value of v_T is slightly smaller than the group velocity $v_g = 32\,000\ \mathrm{ms}^{-1}$ of excitons with $E_{\mathrm{kin}} = 1$ meV, confirming the dominance of forward scattering due to interface roughness. For the 5 nm ZnSe/ZnMgSSe MQW we find values of $t_c = 14 \pm 2.5$ ps and $l_c = 380$ nm.

Our interpretation of the exciton-acoustic phonon scattering and its influence on exciton coherence is further supported by a thermal quenching of the exciton breathing. When rising temperature the rate for absorption of acoustic phonons strongly increases getting equally probable compared to acoustic-phonon emission at about 40 K. Since the absorption is a forward-scattering channel for the excitons [Fig. 3(b)], it will eliminate the spatiotemporal oscillation. Experimentally, we observe a vanishing of the oscillation between $T = 15$ and 20 K as shown in Fig. 4.

The temporal development of the spot area for times longer than 100 ps is complex. At 7 K it is nearly linear. A linear behavior is found when a Gaussian density distribution is assumed in the diffusion equation. One would expect such a diffusive transport for quasiresonantly excited cold excitons. The behavior of the spot size at 15 K is not understood yet in detail. But we find a clear transition from a sublinear to a linear increase of the PL-spot area for temperatures higher than 40 K. We think that this reflects the transition to a diffusive transport regime for the cold excitons after they have warmed up to the lattice temperature. This transition occurs at earlier time for rising temperature (see the dashed lines in Fig. 4). It is obvious that here more work is needed to clarify the details

In summary, we documented a spatiotemporal breathing of the exciton density after ultrafast generation in ZnSe-based quantum wells. This oscillation allows estimation of both the coherence length and time of the exciton wave function in a single experiment.

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