Reduced Exciton-Exciton Scattering in Quantum Wires

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(Received 14 October 1992)

A nonthermal, quasimonoenergetic exciton distribution is observed in time-resolved photoluminescence measurements up to 150 ps after excitation if a quantum wire is excited resonantly and with low excitation density of 10^2 cm^{-1}. The scattering rate increases only slowly with increasing exciton density. This is the first evidence in optical experiments for strongly reduced exciton-exciton scattering rates in one-dimensional systems.

PACS numbers: 73.20.Dx, 78.47.+p, 78.55.Cr

The investigation of semiconductors with quantum confinement in one, two, or three dimensions is not only of basic physical interest but also of fundamental importance for faster electronic, optoelectronic, or all-optical devices. While confinement in one dimension has been spectroscopically investigated over the last twenty years [1,2] and is in general well understood, the study of confinement in two dimensions is still challenged by the fabrication of homogeneous quantum wires with low nonradiative recombination rates. However, fabrication techniques are improving and one-dimensional systems with good optical properties have been demonstrated [3–7]. Theoretically, novel properties such as increased excitonic binding energy or strong nonlinear effects due to the peaked density of states have been predicted [8,9]. In particular a drastic reduction of particle scattering, for example electron or exciton scattering, has been proposed [10,11].

In a quantum wire the free motion of, e.g., excitons is only possible in one direction which we denote as the x direction. The exciton dispersion curve then reduces to a one-dimensional parabola as shown in Fig. 1(a). Exciton-exciton scattering under strict conservation of energy and quasimomentum K_x is then drastically reduced to one scattering process with $\Delta K = 2K_x$ [see excitons 1 and 2 at energy E_1 in Fig. 1(a)]. This reduction of particle scattering is thrilling in particular for device applications [10]. We will demonstrate here for the first time by optical experiments that indeed excitons created with a specific energy stay for unusually long times at this energy, i.e., show an extremely long-lived nonthermal distribution.

Our quantum wire structures have been fabricated from a multiple quantum well (MQW), consisting of 25 GaAs quantum wells of 10.6 nm width sandwiched between 15.3 nm wide $Al_{0.36}Ga_{0.64}As$ barriers, which were grown by molecular beam epitaxy on top of a 1 μm thick $Al_{0.36}Ga_{0.64}As$ optical confinement layer [3]. We prepared a mask of photoresist stripes oriented along the [110] direction with a 280 nm periodicity by holographic lithography. Rectangular grooves were etched into the MQW layers in a $SiCl_4$ plasma with three reactive-ion etching steps of decreasing depths (for process parameters see Ref. [3]). Scanning electron micrographs show that the geometrical width of the wires amounts to 150 ± 10 nm. The active wire width is 60 nm as proven in Refs. [3] and [12]. The notations are explained in Fig. 1(b). On the samples, a small unpatterned part was left to take reference spectra on two-dimensional (2D) quantum wells. The process technique and characterization of the sample are described in more detail in Refs. [3,14,15].

For our time-resolved experiments, the sample is kept in a helium flow cryostat and excited with pulses from a synchronously pumped mode-locked dye laser. The pulse repetition rate is 80 MHz and the pulse width about 5 ps. The photoluminescence (PL) is spectrally dispersed in a 32 cm spectrometer and temporally resolved in a synchroscan streak camera with a two-dimensional readout

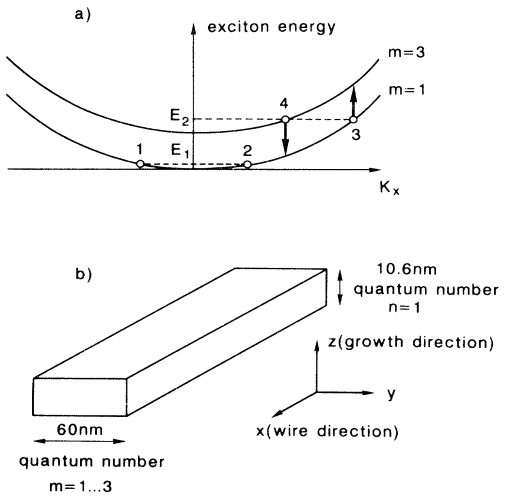

FIG. 1. (a) Schematic diagram of the 1D exciton dispersion in the wire direction. For excitons in the lowest subband (see excitons 1 and 2 at energy E_1) scattering under strict conservation of energy and quasimomentum K_x is no longer possible. For higher exciton energies (see excitons 3 and 4 at energy E_2) intrasubband scattering is still quenched but intersubband scattering is allowed [13]. (b) Notation used for the quantum wire.

system. The temporal and spectral resolution of the system are 10 ps and 0.5 meV, respectively. A polarizer oriented perpendicular to the exciting laser light suppresses the scattered laser light at resonant excitation.

We excite in the low excitation regime, i.e., we only create excitons. For low excitation the quantization in the y direction takes place as a quantization of the center-of-mass (CM) motion of the heavy-hole excitons [14,16]. This becomes evident in the photoluminescence excitation (PLE) spectrum which we reproduce for comparison from Ref. [14] in Fig. 2(a). For a polarization $\mathbf{E}\|\mathbf{x}$, we observe only the $n=1$, $m=1$ and $m=3$ excitons [17]. The $m=1$ and $m=3$ exciton peaks are inhomogeneously broadened. The inhomogeneous broadening is increased in comparison to the QW by a factor of 2, probably due to wire width fluctuations; however, is not necessarily connected with localization.

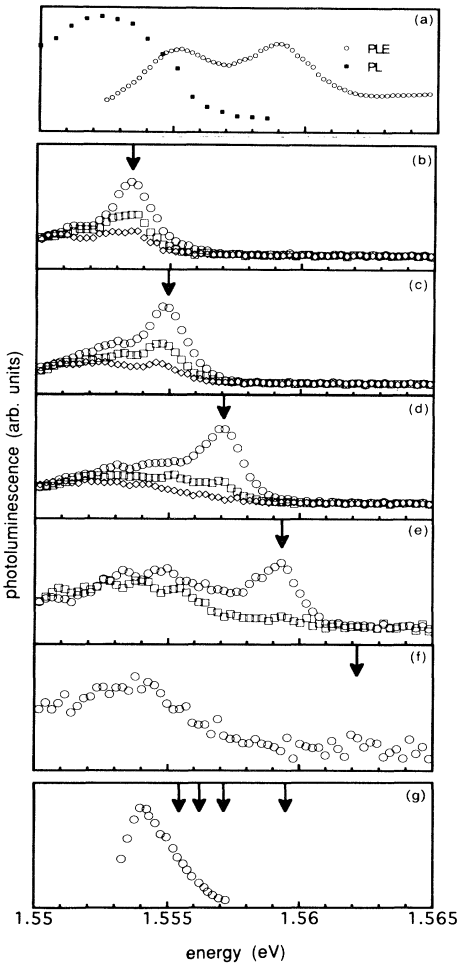

FIG. 2. (a) Time-integrated PLE and PL spectra of the quantum wire from Ref. [14]. Transient PL spectra for resonant excitation energies of (b) 1.5537 eV, (c) 1.5550 eV, (d) 1.5573 eV, (e) 1.5593 eV, (f) 1.5622 eV. The laser excitation energies are indicated by arrows. The spectra for 40 ps (○), 80 ps (□), and 120 ps (◇) time delay after excitation are plotted. (g) Time-integrated PLE spectra of the quantum well reference part from Ref. [14]. The PL peak for the quantum well reference part is identical to the PLE peak and is immediately shifted to lower energies if excited at energies higher than the PLE maximum. In contrast to the quantum wire, relaxation into this luminescence is faster than 10 ps, the time resolution of the streak camera, for different laser excitation energies (as indicated by the arrows).

We now present results for resonant excitation of excitons at a temperature of 10 K for polarization of the electric field $\mathbf{E}$ parallel to $\mathbf{x}$. Qualitatively the same behavior is also observed for $\mathbf{E}\|\mathbf{y}$. Scattering of laser light, however, is reduced in the first case because of sample geometry. Time-resolved transient spectra are shown in Figs. 2(b)–2(f) for delay times of 40 ps (circles), 80 ps (squares), and 120 ps (diamonds) for different excitation energies. The PL spectra exhibit a strong nonthermal component exactly at the excitation energy for times as long as 150 ps which follows the laser photon energy continuously over a range of 6 meV. This nonthermal component indicates slow spectral diffusion of $m=1$ excitons to lower energy islands and slow $m=3$ to $m=1$ transitions within a given spatial regime. Slow thermalization is a special feature of quantum wires, as can be seen by comparison with the fast thermalization at the QW reference sample in Fig. 2(g) (for QW resonant excitation characteristics see Refs. [18,19]). The energy range of this strong nonthermal component of the quantum wire coincides with the center-of-mass quantized heavy-hole excitons (CME) with $m=1$ and 3 as shown by comparison with Fig. 2(a). We define a thermalization time τ as the time in which the ratio of the integrated PL intensity of the nonthermal to the thermal part decreases by e^{-1}. The energy dependence of this time is plotted in Fig. 3 for an excitation density of 10^2 cm^{-1} [20]. Thermalization becomes faster than the time resolution of 10 ps for energies larger than 1.5622 eV. The thermalization time also decreases with increasing excitation intensity; e.g., for an

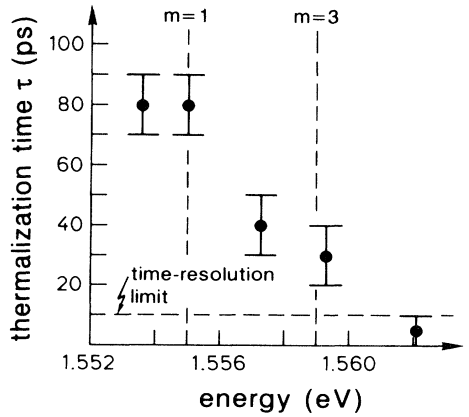

FIG. 3. Energy dependence of the thermalization time τ for an excitation density of 10^2 cm^{-1}.

excitation energy of 1.5571 eV the thermalization time decreases from 85 ps over 20 ps to < 10 ps if the excitation density is increased from 10^2 over 10^3 to 10^4 cm^{-1}.

We now discuss the origin for this long-lived resonant luminescence. First, we exclude an evolution of wire width in vertical (growth) direction as a possible process. PLE spectra taken at different wavelengths between 797.6 and 800.0 nm are identical. An evolution of wire width would clearly lead to different PLE spectra since the wires are certainly completely decoupled in vertical direction. Next, we exclude exciton localization (Ref. [21]) as a possible process. We clearly excite in the regime of the maximum of the PLE spectrum [compare Figs. 2(b)–2(f) with Fig. 2(a)], i.e., we create free, nonlocalized excitons. This is confirmed both by the independence of the PLE spectrum of detection wavelength and the dependence of the CM-quantized heavy-hole-exciton diamagnetic shift in a magnetic field [14,22,23]. Therefore we explain the long-lived nonthermal luminescence at excitation energies corresponding to $n=1$, $m=1$ by the absence of energy-changing exciton-exciton scattering channels under strict conservation of energy and quasimomentum K_x, as discussed earlier. For excitation at energies corresponding to $n=1$, $m=3$ intrasubband scattering is still suppressed for the same reason but intersubband scattering is allowed as depicted in Fig. 1(a) (see excitons 3 and 4 at energy E_2). For even higher excitation energies we get a mixing with $2S$ exciton eigenstates. There, a strong increase of energy-momentum-allowed intersubband and intrasubband scattering by a band-mixing-induced deviation of the parabola energy-momentum dependence causes a strong decrease of τ.

We conclude that exciton-exciton scattering for 1D CM excitons is strongly reduced in comparison to 2D systems. Thermalization time of CM excitons is faster by a factor of 2 if we excite in $m=3$ instead of $m=1$ and is density dependent as expected by comparison to the 2D density-dependent exciton dephasing times [24]. This is a demonstration of the general principle property of scattering reduction in low-dimensional systems. It is important for applications but also offers new fundamental studies, e.g., hole burning in excitonic systems.

We gratefully acknowledge the expert technical assistance of K. Rother and R. Eccleston for a critical reading of the manuscript. We acknowledge support by the Bundesministerium für Forschung und Technologie of the Federal Republic of Germany.

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