RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 47, NUMBER 16 15 APRIL 1993-II

Intersubband relaxation of heavy-hole excitons in GaAs quantum wells

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(Received 25 February 1993)

The temporal dynamics of an exciton in a higher quantum-well subband is observed. The lifetime of the exciton formed by an electron of the lowest subband and a heavy hole of the *second* subband in GaAs quantum wells is determined by time-resolved luminescence as 130±20 ps, in agreement with theoretical estimations of intersubband scattering by acoustic-phonon emission. Consequences for intersubband lasers are substantial.

The various relaxation processes of excitons in quantum wells[1] have been studied recently in several experiments. Energy relaxation within the two-dimensional exciton band,[2] exciton formation,[3] ionization,[4] spin relaxation,[2,5] and recombination[6–9] have been quantitatively measured by time-resolved techniques. In a recent work[10] also the transition rates of excitons below the free carrier continuum from the light-hole to the heavy-hole exciton subband could be successfully measured. Other experiments of intersubband relaxation in quantum wells have been performed exclusively with free electrons, above[11–16] and below the optical phonon energy.[17,14]

In this work we describe the time-dependent measurement of an exciton in a higher quantized subband state. The $(hh_2\text{-}e_1)_{1S}$ exciton, formed by electrons of the lowest ($n=1$) subband and heavy holes of the *second* ($n=2$) subband, is studied by time-resolved luminescence. Our experimental results lead to the conclusion that the exciton lifetime in the second heavy-hole subband is limited by acoustic-phonon emission and considerably longer than the shortest reported values of radiative lifetimes of the ground-state excitons at $k\approx 0$. This finding has promising implications for achieving intersubband inversion by optical excitation of higher subband excitons.

The subband structure of the quantum wells is essential for the experiment. The stucture, grown by molecular-beam epitaxy, consists of 40 periods of 265-Å GaAs wells and 260-Å AlAs barriers. The linewidth of the low-temperature photoluminescence peak is 0.8 meV indicating a high sample quality.[18] The subband structure has been investigated by photoluminescence excitation spectroscopy (PLE), using a cw Ti:sapphire laser and detection at the $(hh_1\text{-}e_1)_{1S}$ exciton peak. The spectra are shown in Fig. 1. The peak closest to the ground state is the light-hole exciton at $k\approx 0$ labeled $(lh_1\text{-}e_1)_{1S}$. The next peak—in Fig. 1 labeled $(hh_2\text{-}e_1)_{1S}$—is the 1*s* exciton formed by an electron of the lowest subband and a heavy hole of the *second* subband. The radiative transition (absorption and emission) of this exciton is a *forbidden* transition in perfectly symmetric wells. The observation is possible due to the built-in electric field,[19,20] which increases the band mixing. The electric field necessary for the observed oscillator strength is calculated[18] as ~3 kV/cm, which is consistent with midgap Fermi-level pinning at the surface. It is important that the energy of the $(hh_2\text{-}e_1)_{1S}$ exciton is well below the energy of free-electron–hole pairs: the energy difference of the $(hh_2\text{-}e_1)_{1S}$ and the $(hh_1\text{-}e_1)_{1S}$ exciton is 4.5 meV compared to the $(hh_1\text{-}e_1)_{1S}$ exciton binding energy of 6.5 meV.[21] In the PLE spectra, furthermore, the $(hh_1\text{-}e_1)_{2S}$ and $(hh_1\text{-}e_1)_{3S}$ excitons[18] and the steplike absorption edge for free-electron–hole pair excitation can be seen.

For the time-resolved luminescence experiments we used a synchroscan streak camera with two-dimensional detection. The excitation was performed by a tunable picosecond dye laser (Styril 8) synchronously pumped by a frequency doubled Nd:YAG (yttrium aluminum garnet) laser. The excitation wavelength was 795 nm for the first series of experiments, the excitation density about 2×10^9 cm^{-2} per layer.

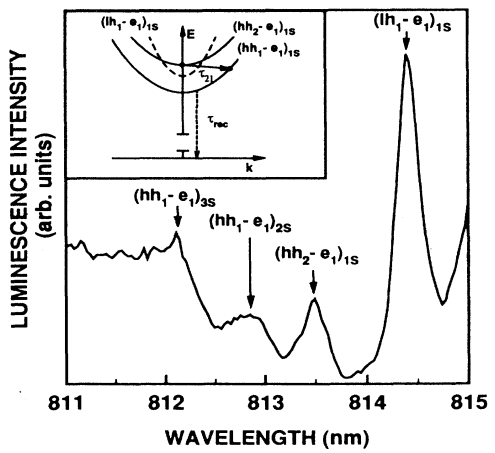

FIG. 1. Excitation spectra obtained with cw Ti:sapphire laser at $T_L=1.8$ K. Detection is at the low-energy side of the $(hh_1\text{-}e_1)_{1S}$ exciton peak. The various transitions are indicated. Inset: relevant part of the excitonic subband structure.

Typical results of the time-resolved luminescence spectra are shown in Fig. 2: The amplitude of the $(hh_2\text{-}e_1)_{1S}$ exciton increases to a maximum and decreases at later times. The temporal evolution of the pure $(hh_2\text{-}e_1)_{1S}$ signal is shown in the inset of Fig. 2, where the data points are the difference of the total luminescence intensity at the wavelength of the $(hh_2\text{-}e_1)_{1S}$ exciton and the background signal. A first quantitative result is obtained by fitting the experimental data with two time constants for the population and depopulation of the $(hh_2\text{-}e_1)_{1S}$ exciton around $k=0$. The best fit is shown in the inset of Fig. 2, using values of 140 ± 30 and 330 ± 30 ps for exponential population and depopulation, respectively. However, as well known for rate equations with exponential decays, the two time constants are interchangeable, so the time constant for the depopulation of the $(hh_2\text{-}e_1)_{1S}$ excitons by intersubband relaxation can be either 140 or 330 ps.

Therefore we modified the experiment for a more direct measurement of the $(hh_2\text{-}e_1)_{1S}$ exciton relaxation: the laser is tuned to *resonant* excitation of the $(hh_2\text{-}e_1)_{1S}$ exciton, and we study the time evolution of the luminescence intensity at the $(hh_2\text{-}e_1)_{1S}$ exciton peak. The results are shown in Fig. 3 for two excitation densities: excitation of the $(hh_2\text{-}e_1)_{1S}$ exciton causes a rise of the $(hh_1\text{-}e_1)_{1S}$ exciton luminescence much slower than the laser pulse width. The maximum of the luminescence is reached at $t\approx350$ ps, the decay is followed until $t>800$ ps. For a lower excitation intensity (curve *b*), the decay of the luminescence is slightly faster.

The quantitative interpretation of this experiment is based on the following model: the laser pulse resonantly excites in the $(hh_2\text{-}e_1)_{1S}$ band around $k\approx0$. Intersubband scattering (with a characteristic time constant τ_{21}) leads to an increasing population of the $(hh_1\text{-}e_1)_{1S}$ exciton band according to

$$n_{(hh_1\text{-}e_1)_{1S}}(t)\propto n_{(hh_2\text{-}e_1)_{1S}}(t=0)\times[1-\exp(-t/\tau_{21})]\ . \quad (1)$$

The average excess energy per exciton in the $(hh_1\text{-}e_1)_{1S}$ band is about 3.9 meV (corresponding to a temperature of 45 K) for acoustic-phonon emission as the dominant scattering process. Thermalization is assumed to be much faster than the subsequent energy relaxation, and leads to a thermal distribution in both exciton subbands with one common temperature varying with time. The average energy per exciton is decreased by energy loss via longitudinal acoustic-phonon emission in both subbands, according to[23,10,22]

$$\langle dE/dt\rangle_{LA}=(D_c-D_v)^2M_{ex}^2 2k_B(T_{ex}-T_L)/\rho L\hbar^3\ , \quad (2)$$

where D_c and D_v are the deformation potentials for electrons and holes,[24] M_{ex} and T_{ex} are the exciton mass and temperature, ρ is the mass density, and L is the quantum-well width. The average energy is increased by the intersubband scattering and also by the recombination from $(hh_1\text{-}e_1)_{1S}$ band at $k\approx0$. Radiative recombination of the $(hh_1\text{-}e_1)_{1S}$ excitons occurs for those excitons with kinetic energy within the homogeneous linewidth.[6] Therefore recombination of the $(hh_1\text{-}e_1)_{1S}$ excitons will act as a heating process since only "cold" excitons are removed from the distribution. From the time dependence of the luminescence at later times ($t>400$ ps), the radiative recombination time at $k\approx0$ can be directly determined from the experiments. We obtain values of

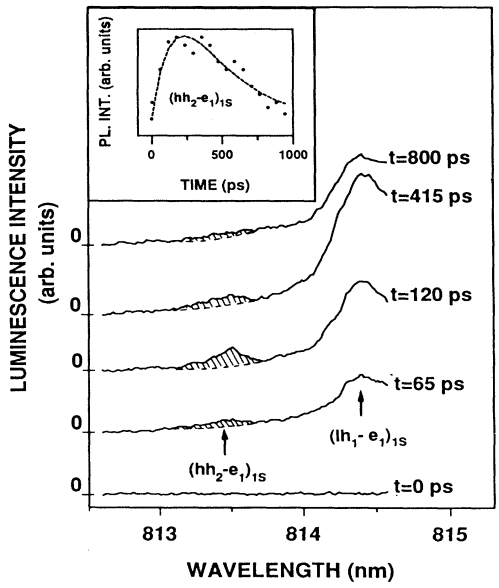

FIG. 2. Time-resolved luminescence data for excitation at 795 nm. $T_L=1.8$ K. Time resolution is 50 ps (full width at half maximum). Inset: $(hh_2\text{-}e_1)_{1S}$ exciton signal as a function of time, determined from the difference of the total signal and the background.

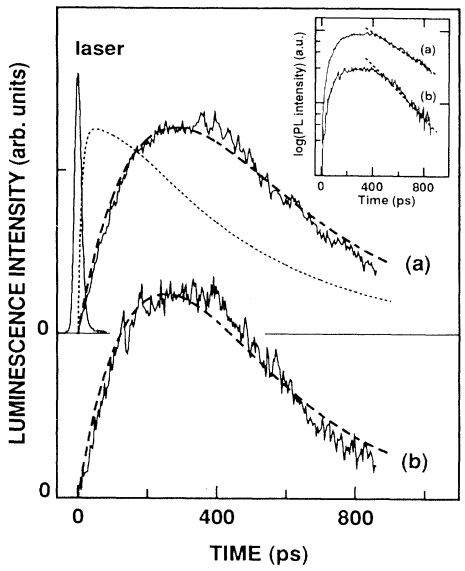

FIG. 3. Luminescence of the $(hh_1\text{-}e_1)_{1S}$ exciton as a function of time after resonant excitation of the $(hh_2\text{-}e_1)_{1S}$ exciton for different excitation densities: (a) $n=1\times10^{10}$ cm^{-2} per layer, (b) $n=4\times10^{9}$ cm^{-2} per layer. Dashed line: fit according to Eqs. (1) and (2) with $\tau_{21}=130$ ps. Inset: Logarithmic scale for determination of τ_{rec}. Dotted line: calculated luminescence for rapid intersubband relaxation (10 ps).

$\tau_{rec}=390$ ps (for $n=1\times10^{10}$ cm^{-2}) and $\tau_{rec}=250$ ps (for $n=4\times10^{9}$ cm^{-2}). The dependence of the recombination time on the excitation density is in agreement with the recently reported direct measurements using resonant excitation of the ground-state excitons.[7,9] The density dependence comes from the smaller coherence volume due to stronger dephasing by exciton-exciton scattering.[6,7] A possible larger homogeneous linewidth Δ at earlier times (higher exciton concentration) and thus a longer radiative lifetime within Δ is compensated by the larger fraction of excitons within Δ [Eqs. (8) and (9) in Ref. 6]. With the known recombination time within the homogeneous linewidth, the time evolution of the $(hh_1\text{-}e_1)_{1S}$ luminescence can be numerically calculated (Fig. 3, dashed lines), *using only one variable parameter τ_{21} for the intersubband relaxation. We obtain the best agreement with our experimental results for a value of $\tau_{21}=130\pm20$ ps for the intersubband relaxation time from the $(hh_2\text{-}e_1)_{1S}$ exciton band at $k\approx0$ into the $(hh_1\text{-}e_1)_{1S}$ band at $k>0$.* The value of τ_{21} is identical for both excitation densities and agrees well with the shorter one of the two time constants (140 and 330 ps) resulting from the first series of experiments. In Fig. 3(a) (dotted line) also the luminescence evolution for very fast intersubband scattering (10 ps) is shown, resulting in a fast rise of the luminescence. This fast rise has been observed experimentally in samples where hot excitons with excess energies within the lowest subband (4.8 meV) comparable to our subband spacing (4.5 meV) have been studied.[8] The delayed luminescence rise observed in our experiments clearly shows the role of intersubband relaxation.

A theoretical estimate of the intersubband relaxation can be made, if we calculate the intersubband scattering rate for excitons at $k=0$ by acoustic deformation potential interaction. An analytical result is obtained for lattice temperature $T_L=0$ (only phonon emission), which is a valid approximation for $T_L=1.8$ K and a subband energy difference of 4.5 meV,

$$1/\tau_{21}=M_{ex}(D_c-D_v)^2\pi/4\rho u_l\hbar^2L^2 \quad (3)$$

(u_l is the longitudinal sound velocity). For our sample parameters Eq. (3) gives a theoretical value of $\tau_{21}=117$ ps. The slightly longer experimental value might be due to the repopulation of the second subband by the high-energy tail of the population in the lowest subband.[14]

Our results should be relevant for the research focused on intersubband inversion.[25] We find two decisive points that are different for the intersubband dynamics of excitons as compared to free carriers: (1) The intersubband scattering time from the second heavy-hole subband $(hh_2\text{-}e_1)_{1S}$ around $k=0$ is considerably longer (130 ps) than the shortest reported values (40 ps in Ref. 9) of the radiative recombination time in the ground state $(hh_1\text{-}e_1)_{1S}$ at $k=0$. Theoretical calculations predict even shorter recombination times.[9] Radiative recombination times below 100 ps are expected also for well width above 150 Å, if the sample quality is sufficiently high. The intersubband scattering dynamics due to acoustic-phonon scattering, however, should not be modified in high-quality samples. Thus the most important condition for achieving inversion can be fulfilled in ultrahigh quality samples with large coherence lengths. (2) Since excitons are optically excited only around $k=0$ (in contrast to free-electron–hole pairs), the selective excitation of a higher subband is possible without exciting the lower subband at $k>0$. For these two reasons subband transitions of excitons below the free carrier continuum represent a very appealing concept for optically pumped coherent sources in the meV range.[26] Radiative transitions between two different exciton subbands have been observed by Olszakier *et al.*[27] with enhanced oscillator strength. In addition, for optical pumping of inverted exciton populations the absence of carrier-carrier scattering[28] should be of importance. Population inversion between light and heavy holes in quantum wells has been recently achieved by tunneling injection.[29]

In conclusion, we have studied the temporal dynamics of an exciton in a higher quantum-well subband, namely, the $(hh_2\text{-}e_1)_{1S}$ exciton formed by a heavy hole in the second and the electron in the lowest subband. This exciton is still well below the free carrier continuum. Time-resolved luminescence of the $(hh_1\text{-}e_1)_{1S}$ exciton after resonant excitation of the $(hh_2\text{-}e_1)_{1S}$ exciton at $k\approx0$ yields a time for the intersubband relaxation into the $(hh_1\text{-}e_1)_{1S}$ exciton band of $\tau_{21}=130\pm20$ ps, in agreement with theoretical calculations of intersubband acoustic-phonon scattering. The lifetime is much longer than the shortest reported values of the radiative recombination time of the ground-state exciton at $k=0$. These results favor concepts for excitonic intersubband lasers in quantum wells.

Experimental work has been performed at RIKEN (Frontier Research Program), Japan. One of us (R.R.) thanks the Conselho Nacional de Desenvolvimento Científico e Tecnológico (CNPq), Brazil. This work has been supported by the Fonds zur Förderung der wissenschaftlichen Forschung (Projects Nos. 7558 and 8704).

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