RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 44, NUMBER 3 15 JULY 1991-I

Exciton dynamics in a GaAs quantum well

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(Received 21 March 1991)

Time-resolved luminescence measurements in a 27-nm GaAs quantum well show that the initial temperature of the photocreated exciton distribution is determined by the excess energy of the excitation photon. Light-hole excitons lying in the band gap transfer to heavy-hole exciton states by density-dependent exciton-exciton scattering. For excitation close to the band edge, excitons cool only via LA-phonon emission. The time-resolved luminescence profile is modeled by evaluating the LA-phonon energy-loss rate.

The rise time of the time-resolved photoluminescence (TRPL) signal in GaAs quantum wells (QW) is determined by (i) the time required for excitons to be generated and (ii) exciton cooling.[1,2] By momentum conservation, only excitons with center-of-mass kinetic energies within one homogeneous linewidth of zero kinetic energy can recombine radiatively.[1] The fraction of excitons satisfying this condition increases as the exciton temperature falls, leading to a slow rise in the photoluminescence (PL) intensity at low lattice temperature due to hot-exciton cooling.[2] TRPL rise times therefore provide information about exciton population densities and temperatures.

In this Rapid Communication, we report TRPL rise-time measurements in GaAs-$Al_{0.3}Ga_{0.7}As$ 27-nm single QW with three-monolayer AlAs interface smoothing layers inserted between the GaAs-$Al_{0.3}Ga_{0.7}As$ interfaces. The growth details and optical characterization of the sample are reviewed in detail elsewhere.[3] The smoothing layers result in an extremely narrow time-integrated PL linewidth (0.2 meV) and negligible inhomogeneous broadening, allowing photoexcitation very close to the PL line without direct resonant creation of heavy-hole excitons. Two important excitation regimes are thereby investigated for the first time. First, we describe results for photoexcitation resonant with the light-hole (lh) exciton photoluminescence excitation (PLE) line which, in this sample, lies 2.4 meV above the heavy-hole (hh) exciton PL line. The lh exciton ground state therefore lies in the band gap.[4] For a lh exciton resonance lying in the electron–heavy-hole band, scattering into free-electron–heavy-hole states is believed to be very fast. Here, lh excitons tend to be stable with respect to the formation of free-electron–hole pairs and the evolution of the lh exciton population can be investigated without significant participation by free carrier states, raising the possibility of slower lh-hh transfer. Then we investigate excitation as a function of excess photon energy near the band edge. In this case, free carriers and excitons can be photocreated at relatively low temperatures and cool only by LA phonon emission. The TRPL rise time can be related to the LA phonon cooling rate without consideration of LO phonon energy-loss processes.

The sample was cooled in a helium cryostat and excited with pulses from a synchronously pumped mode-locked Styryl 8 laser. The PL was spectrally dispersed in a 32-cm spectrometer and temporally dispersed in a two-dimensional (2D) synchroscan streak camera. The temporal resolution of the system was between 15 and 25 ps and the spectral resolution was 0.5 meV.

The inset of Fig. 1 shows TRPL curves at two initial excited carrier densities, n_0, for excitation at the lh exciton line and detection at the hh exciton line at 15 K. For large n_0 [Fig. 1(a)], a fast rise of the hh exciton PL to 80% of the peak value is observed within the time resolution of the streak camera. Scattering between lh and hh exciton bands, therefore, occurs within 20 ps. A weak second component of the rise time is then observed. This is attributed to the rise in excitonic PL due to hot-exciton cooling described earlier, indicating that the scattering between the hh and lh exciton states establishes an exciton population which is initially hotter than the lattice. At lower n_0, the fast rise in the PL disappears [Fig. 1(b)] Changes in the cooling rate only modify the cooling component of the rise in PL amplitude. Therefore the slower rise time must be due to a decrease in the lh-hh exciton scattering efficiency. This n_0 dependence indicates that the lh-hh transfer mechanism is exciton-exciton scattering. Figure 1 shows the variation of total rise time, defined as the time between the peak of the (simultane-

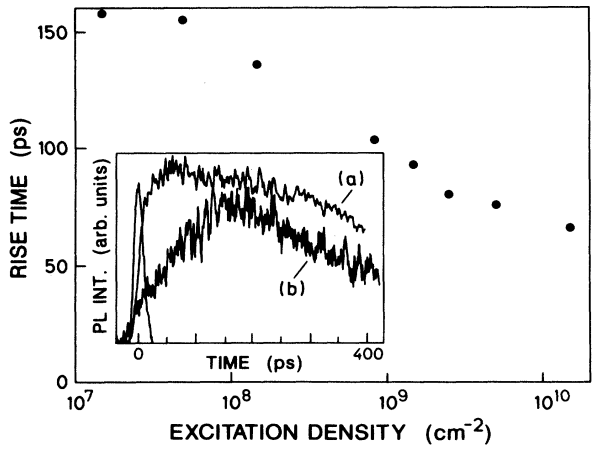

FIG. 1. PL rise time vs excitation density for lh exciton excitation and hh exciton detection at 15 K. The inset shows typical TRPL profiles at (a) $n_0 = 1.5 \times 10^{10}$ cm^{-2}, and (b) $n_0 = 1.5 \times 10^8$ cm^{-2}. Also shown is the laser pulse indicating the time resolution.

ously measured) laser pulse and the peak of the TRPL, with n_0. At the lowest densities investigated, the PL rise-time increase saturates. We expect that exciton-exciton scattering then becomes weaker than (density independent) LA phonon and/or impurity scattering processes. At the highest densities, the rise time is dominated by cooling. The filling of the hh exciton states closely parallels the establishment of a thermal distribution within the two (strongly coupled) exciton bands. For free carriers this thermalization process occurs on the time scale of ≤ 1 ps.[5] The much longer times (at low n_0) measured here are consistent with the lower efficiency of exciton-exciton scattering compared to free-carrier scattering.[6]

These ideas are supported by temperature-dependent measurements (Fig. 2) which show that (at large n_0) the hh exciton states are filled without significant interaction with the lattice. As the sample temperature T_L is increased, the rise time due to cooling reduces and near 28 K [Fig. 2(a)] is close to the time-resolution limit, indicating that hh excitons are formed at an initial temperature which is close to 28 K. This value is in good agreement with the temperature expected after thermalization to a Maxwellian distribution in 2D from an initial δ-like energy distribution at an excess energy of 2.4 meV (the lh-hh line splitting). This is consistent with exciton-exciton scattering since the thermalization occurs with no energy loss from the exciton distribution. At still higher T_L [Figs. 2(b)-2(d)], an initial fast decay is observed, which becomes faster as T_L increases. This fast decay is the exact inverse of the slow rise due to warm-exciton cooling observed at low temperature. The exciton population internally thermalizes to a temperature near 28 K and is then heated to the lattice temperature with a corresponding fall in the radiative recombination rate. Since hh excitons can be formed initially colder than the surrounding lattice, lattice-related processes such as phonon scattering cannot contribute significantly to the filling of the hh exciton states. From the $1/e$ time of the initial fast decay in Figs. 2(b)-2(d), we estimate heating times of 60 ps for 40 K, 40 ps for 50 K, and 30 ps for 65 K. The decrease in times reflects the increase in the LA phonon density as the temperature is raised. To our knowledge, this is the first time that the dynamics of heating of cold excitons by LA phonons has been observed.

Now we describe TRPL measurements (Fig. 3) at 15 K for increasing excess excitation photon energy ΔE above the hh exciton line. Note that $n_0 \approx 1.5\times10^{10}$ cm^{-2}, which is below the threshold where hot-phonon effects become significant.[7] Figure 3(a) shows the resonant lh exciton pumping result (i.e., $\Delta E = 2.4$ meV). Figure 3(b) is obtained at $\Delta E = 6.9$ meV, i.e., only slightly above the electron-hh band edge (PLE measurements show that this band edge lies at 5.7 meV). In Figs. 3(a)-3(d), a progressive increase in the PL rise time is observed. This indicates an increase in the time required for the exciton population to cool to the lattice temperature. An increase in ΔE therefore leads to a higher initial exciton temperature. Note that for Fig. 3(b) a fast initial rise in the PL signal is observed, indicating that the formation of hh excitons occurs within the time resolution of the measurements, consistent with recent exciton-formation time measurements.[8] At larger ΔE this fast rise first reduces [Fig. 3(c)], and then disappears [Figs. 3(d) and 3(e)] due to the larger contribution of exciton cooling to the rise time. Beyond $\Delta E \approx 10$ meV the TRPL profile no longer varies strongly with ΔE [compare Figs. 3(d) and 3(e)], implying

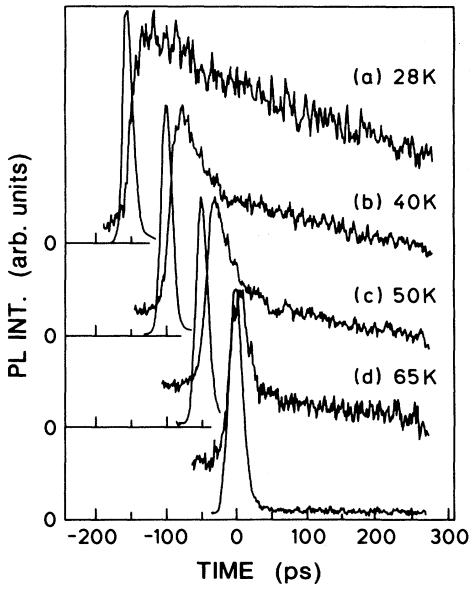

FIG. 2. TRPL profiles for lh exciton excitation and hh exciton detection at different T_L and $n_0 = 1.5\times10^{10}$ cm^{-2}. For clarity each curve is shifted by 50 ps. The laser pulse indicates the time resolution and $t = 0$ in each case.

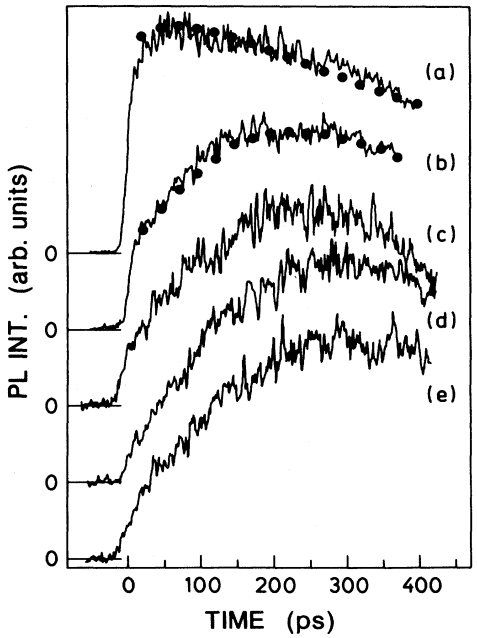

FIG. 3. TRPL profiles at 15 K for detection at the hh exciton line for different excess photon energy ΔE at $n_0 = 1.5\times10^{10}$ cm^{-2}. (a) $\Delta E = 2.4$ meV (lh exciton excitation), (b) $\Delta E = 6.9$ meV, (c) $\Delta E = 8$ meV, (d) $\Delta E = 12.9$ meV, and (e) $\Delta E = 20.8$ meV. (a),(b) Also shown (circles) are the theoretical fit described in the text.

no further increase in the initial exciton temperature. This is characteristic of the onset of very efficient energy loss via LO phonon emission above a certain threshold carrier temperature[9] which causes rapid cooling of the electron-hole and exciton plasma to this threshold within the time resolution of the measurement irrespective of the initial excess energy. No dependence of the TRPL profile on ΔE is therefore apparent beyond this threshold as observed here and previously reported for excitation high in the band.[2] This LO phonon emission threshold has been also observed for free-carrier hot PL in bulk GaAs.[10]

The luminescence intensity $I(t)$ is proportional to the instantaneous exciton recombination rate, given in a QW by[1]

$$I(t) \propto -\frac{dN}{dt} = \frac{N}{K}[1-\exp(-\Gamma/kT_{\rm ex})] . \quad (1)$$

$T_{\rm ex}$ is the exciton temperature, Γ is the homogeneous linewidth, determined from T_2 time measurements to be 0.2 meV,[4] N is the exciton density, and k is Boltzmann's constant. K is a constant determined from the exponential tail of the luminescence beyond 500 ps where $T_{\rm ex}$ is assumed to have reached the lattice temperature, K is then obtained by setting $T_{\rm ex}=T_L$ in (1). Note that an exponential decay can only be obtained according to (1) if the $T_{\rm ex}$ becomes constant. We have used (1) to model the TRPL profile using the variation of $T_{\rm ex}$ calculated from theoretical energy-loss rates. We consider in this paper only the lh exciton and near-band-edge measurements [Figs. 3(a) and 3(b)], so that LO phonon energy loss can be neglected. The principle energy-loss mechanisms are then LA phonon emission by excitons and free carriers. We assume that excitons are formed elastically and instantaneously. At each temperature, the energy loss is evaluated numerically according to the proportion of free carriers and excitons expected at each exciton-free-carrier plasma temperature. The 2D LA phonon deformation potential energy-loss rate in eV/s for a Maxwell-Boltzmann distribution of excitons is[11,12]

$$\left\langle \frac{dE}{dt} \right\rangle_{\rm LA} = -2.14\times10^4 (D_c - D_v)^2 \left[\frac{M_{\rm ex}}{m_0} \right]^2 (T_{\rm ex} - T_L) , \quad (2)$$

where $M_{\rm ex}$ is the total exciton mass, and D_c and D_v are the deformation potentials of the conduction and valence band of GaAs given by 10.7 and -3.4 eV, respectively.[13] Energy loss within the hh and lh exciton bands is calculated by two terms, as in Eq. (2), weighted by the probability of hh or lh exciton occupation at a given temperature. The use of a constant hole mass is always an approximation in carrier cooling studies in GaAs due to the nonparabolicity of the valence bands. Previous studies[7,12] have used values of the order of $0.5m_0$. We find that the best agreement is given using $0.6m_0$, and the usual electron mass ($0.067m_0$). This larger value may be due to neglecting additional energy loss via deformation-potential scattering between the lh and hh exciton bands. For free-electron and hole LA phonon emission, $M_{\rm ex}$ in (2) is replaced by the electron or hole mass, respectively, and the deformation-potential term becomes D_c^2 or D_v^2. Instantaneous carrier-carrier scattering is assumed to equalize the temperatures of the exciton and free-carrier populations after each step in the numerical evaluation of $T_{\rm ex}$.

Note that the temperature of the exciton–free-carrier system can change due to exciton formation even when no energy is lost from the system. For example, if free carriers are created exactly at the band edge, then after electron-hole thermalization, the carrier temperature will be very close to zero. However, if excitons are then formed elastically, the average exciton kinetic energy is equal to the exciton binding energy (E_b) which corresponds to 66 K for $E_b=5.7$ meV. Furthermore, this large average kinetic-energy exciton distribution will heat the remaining cold free carriers via carrier-exciton scattering. For a general ΔE, the final temperature after this thermalization process, but before any energy loss occurs, will lie between $\Delta E/k$ (if only excitons are present) and $(\Delta E - E_b)/2k$ (if only free carriers are present). We calculate the temperature by assuming that excitons with kinetic energies greater than E_b instantaneously become free-carrier pairs. The common temperature of the electron, hole, and exciton plasma is then evaluated by sharing the total kinetic energy (equal to initial free-carrier kinetic energy minus energy lost via phonon emission plus kinetic energy gained by exciton formation) between the total number of particles to obtain the average energy and hence the temperature.

Using (1) and (2), $I(t)$ is calculated and convoluted with a Gaussian of appropriate full width at half maximum to include the time resolution in each measurement. Figures 3(a) and 3(b) show good agreement between theory (circles) and experiment for both $\Delta E=2.6$ and 6.9 meV. We therefore conclude that for ΔE near the band edge, the measured TRPL profiles are quantitatively consistent with the theory of Eq. (1) and cooling via LA phonon emission. The model also confirms theoretically the relationship between PL rise time and exciton cooling. We find that $T_{\rm ex}$ falls to within 15% of T_L after 150 ps for $\Delta E=2.6$ meV and after 300 ps for $\Delta E=6.9$ meV.

In conclusion, for excitation of lh excitons in the band gap, lh-hh exciton transfer occurs via exciton-exciton scattering on a time scale of 150 ps to <20 ps dependent on excitaton density and is much slower than free-carrier interband scattering. Temperature-dependent measurements show that the hh exciton states are occupied at a temperature corresponding to a value of kT close to the lh-hh exciton splitting energy. The exciton population then establishes thermal equilibrium with the lattice by either cooling or heating. These results may aid the interpretation of time-resolved experiments where resonant excitation of lh or hh excitons occurs. Populations which are initially much colder than the lattice can be generated and the effect of subsequent exciton heating on a typical time scale of 30–100 ps should always be considered. Near the band edge, the PL rise time increases with excess photon energy due to a larger initial exciton temperature. Above $\Delta E \approx 10$ meV, no further increase in rise time is observed due to the onset of rapid LO phonon emission. The TRPL profiles in the LA phonon emission regime can be modeled by evaluating the LA phonon energy-loss rate.

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