

Optically induced ultrafast quenching of the semiconductor quantum well luminescence

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We present an experimental configuration that enables the ultrafast, transient quenching of the excitonic photoluminescence in quantum wells. Our scheme is based on two, delayed, short pulses experiment. A first pulse excites carriers in the system, while a second pulse induces an ultrafast redistribution of excitons that results in abrupt dips in the photoluminescence. We present a model that quantitatively accounts for the measured dip depth. The magnitude of the dip, determined by the temperature change of the carriers, can be controlled by varying the power and delay of the second pulse. © 2008 American Institute of Physics. [DOI: 10.1063/1.2857494]

The ultrafast control of the luminescence in semiconductor light emitting devices is a key feature in the development of optoelectronics. The onset and switch-off times of the emission in devices such as vertical-cavity surface-emitting lasers (VCSEL), light-emitting diodes, microdisks, and microcavities are determined both by their external driving mechanism (current injection, light excitation, etc.) and by the relaxation, cooling, and recombination dynamics of the carriers. The speed of the on/off transients limits the operation frequency of the potential applications. The switch-on dynamics are determined by the excitation source and can be on the order of a few picoseconds when pulsed laser excitation is employed.¹ However, the switch-off times are limited by the recombination dynamics of the excited electron-hole (eh) pairs and excitons (X). Typical switch-off times range from several hundreds of picoseconds in bulk based systems,¹ to a few nanoseconds in III-V quantum wells² (QWs) and quantum dot based heterostructures.³ Since this is the limiting factor, it would be desirable to find configurations that would induce faster off transients in these systems.

One way of approaching this issue is by investigating the conditions for the generation of ultrafast changes in the distribution of carriers. These distributions determine most of the emission properties in direct gap semiconductors. In particular, it has been demonstrated the existence of an intimate relation between the X formation time and its subsequent emission dynamics, and the distribution and cooling dynamics of the unbound electrons and holes.^{4–6} Both species (X 's and eh plasma) coexist in a thermodynamical quasiequilibrium^{7,8} and, therefore, it is expected that ultrafast changes in the carrier distributions will affect the excitonic emission dynamics.

In this letter, we show experimentally how thermalized distributions of carriers in QWs can be modified by the pulsed photoexcitation of hot carriers in the bands, resulting in abrupt changes in the exciton temperature (T). The most striking effect of the ultrafast redistribution of carriers is the appearance of sharp dips in the photoluminescence (PL) dy-

namics of heavy-hole (hh) excitons. Although the results presented here concentrate on QWs, the physics and observed phenomena can be directly extrapolated to more complicated systems such as microcavities,⁹ VCSELs, or structures with active media of higher dimensionality, such as bulk semiconductors.

In our experiments, we use two samples based on GaAs/AlAs. The first one is a heterostructure with a single QW (20 nm). The second one contains multiple (50) QWs (7.7 nm). The samples, kept at 9 K, are photoexcited with 1.5 ps long pulses (repetition rate 82 MHz) from a Ti:Al₂O₃ laser with an excess energy of 26 meV above the hh-X. Two consecutive pulses (P_I and P_{II}), whose power and delay can be independently controlled, arrive at the same excitation spot ($\phi \sim 20 \mu\text{m}$). The PL from the X 's and the eh recombination is collected by a streak camera attached to a spectrometer, with an overall time and energy resolution of 15 ps and 0.2 meV, respectively.

Figure 1(a) depicts the time evolution of the hh-X PL when the two pulses of 70 μW each, delayed by 400 ps excite the single QW independently (dash-dotted lines) and when both excite it jointly (solid line). In the latter case, a clear dip appears in the emission of the hh-X at the time of arrival of P_{II} . The abrupt quenching of the PL is limited by our time resolution. The emission from the light-hole (lh) exciton can also be detected 4.7 meV above the hh-X [shown in Fig. 1(a) with a red dotted line]. In contrast to the hh, the lh dynamics do not show the appearance of a dip in the PL.

The origin of the dip can be explained considering the redistribution of carriers that takes place after the absorption of P_{II} . To account quantitatively for the magnitude of the dip depth, we have developed a dynamical quasiequilibrium thermodynamical model of the carriers in the QWs. The absorption of the first nonresonant pulse creates a nonthermal population of eh's at high energies in the bands. In a time scale of ~ 200 fs, the carriers distribute in the bands achieving a well defined T , higher than the lattice temperature.¹⁰ For our carrier densities, far from the degeneration limit, the electron and hole populations can be well described by Maxwell-Boltzmann distribution functions. As time evolves, the carrier distributions cool down due to the interaction with

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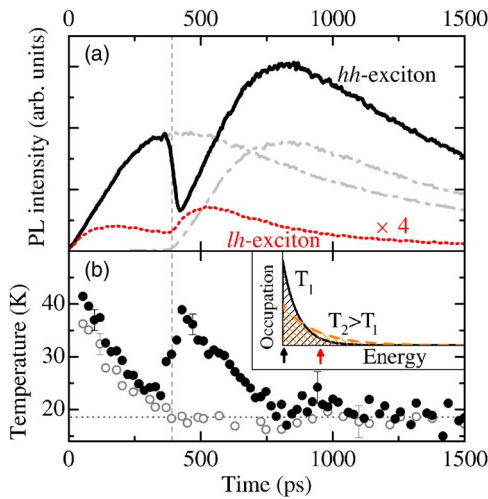


FIG. 1. (Color online) (a) PL dynamics of the single QW hh-X under one pulse excitation (dash-dotted lines) and under two consecutive pulses excitation (solid line; delay between pulses: 400 ps). In red dotted line, the emission of the lh-X (enhanced by a factor of 4) is presented under two pulses excitation. (b) Measured eh temperature for a single (open points) and double (solid points) pulse experiment. Inset: X occupation right before (solid line) and right after (dashed line) the arrival of P_{II} . The black (red) arrow indicates the energy of the $K=0$ heavy(light)-hole X.

phonons, and the populations decrease due to pair recombination and X formation.¹¹ When P_{II} reaches the QW hot carriers are photoinjected at high energy. Due to efficient carrier-carrier scattering, the just created carriers and the pre-existing populations rethermalize in a timescale given by the pulse duration, at a temperature (T_{aft}) higher than that right before the absorption of P_{II} (T_{bef}). Thus, the effect of P_{II} is to warm up the carriers and increase their concentration, in a time scale shorter than a few picoseconds (~ 2 ps).

Concomitantly to the free carrier dynamics, electrons and holes bind to form X's in a time scale of the order of several hundreds of picoseconds for our conditions.^{4,7} The X population obeys also a Maxwell-Boltzmann distribution law. Recent studies⁷ have shown that X's and the coexisting plasma of eh are in thermodynamical quasiequilibrium with the same T . Therefore, an analogous dynamics to that of the free carriers takes place also for the X's: During the arrival of P_{II} , the abrupt warming of the carriers results in an ultrafast warming of the X population. However, in contrast to the plasma, due to the slower formation dynamics of the X's—two orders of magnitude slower than the excitation pulse—the X density is hardly altered within the pulse duration. With these premises and attending to the Maxwell-Boltzmann distributions, the appearance of the dip in the hh-X PL can be understood as follows. The hh-X occupation of the zero momentum states ($K=0$, zero kinetic energy) before/after the arrival of P_{II} is given by

$$f_{MB}^{bef/aft}(0) = \frac{n'}{k_B T_{bef/aft}} \frac{1}{DOS}, \quad (1)$$

where n' is the preexisting density of X's at the arrival time of P_{II} , $T_{bef/aft}$ are the corresponding X (and carrier) temperatures, k_B is the Boltzmann's constant, and DOS is the density of states.

The PL intensity of the hh-X is directly proportional to the occupation of states with $K \sim 0$, as these are the states that can couple to light; therefore, modifications in their occupation at the arrival of P_{II} [see inset of Fig. 1(b)] induce

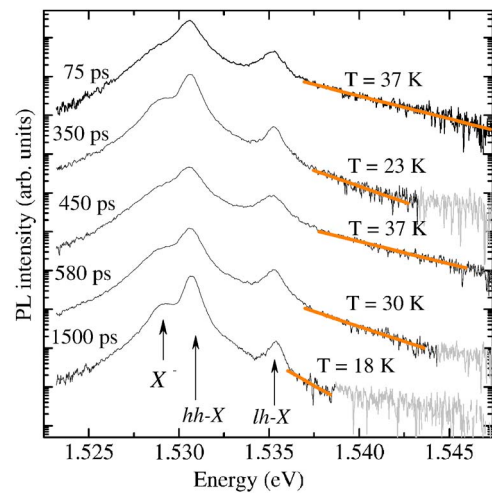


FIG. 2. (Color online) Spectra (rigidly offset) taken at different delays for the conditions of Fig. 1 under two pulses excitation in the single QW. The thick solid lines are Maxwellian fits to the free eh pair recombination. X⁻, hh-X and lh-X denote trions, heavy-hole and light-hole excitons, respectively.

changes in the PL. In particular, the abrupt warming of the carriers ($T_{aft} > T_{bef}$) results in an abrupt drop of the $K=0$ populations [$f_{MB}^{aft}(0) < f_{MB}^{bef}(0)$] and consequently in an ultrafast quenching of the hh-PL (see Fig. 1). The details of the excitonic redistributions also account for the absence of a dip in the lh emission. The lh-X energy is higher than that of the hh-X and the increase in T results in a negligible change of the occupation of the $K=0$ lh states [see inset of Fig. 1(b), red arrow].

In order to gain a deeper insight into the quantitative validity of this model, we have directly measured the carrier temperature in the single QW. Figure 2 shows PL spectra at different times after the arrival of P_{I} for the conditions of Fig. 1. The spectra show the hh-X and lh-X as well as the direct free eh recombination above the lh-X. The low energy shoulder corresponds to weak trion emission (X⁻), whose dynamics do not affect the current analysis.⁷ A Maxwellian fit can be performed at the high energy tail (indicated by solid lines) to extract T . Figure 1(b) shows the temperatures of the eh plasma. In the case of the single pulse, a monotonous cooling of the plasma is observed, with a final equilibrium T (~ 19 K) higher than the lattice temperature (~ 9 K).^{11,12} In the two pulses experiment, an abrupt warming of the plasma and, consequently, of the X's, is observed at the time of arrival of P_{II} . It is worthwhile to mention that the time dependence of the temperature obtained from the ratio of the intensities of the hh and lh excitons follows the trend depicted in Fig. 1(b), confirming the establishment of a thermal equilibrium between free carriers and excitons.

The measured T of the carriers enables us to predict the dip depth following our model. A relative dip depth can be defined as $r \equiv (I_{bef} - I_{aft})/I_{bef}$, where I_{bef} (I_{aft}) is the intensity of the hh-X PL right before (after) the arrival of P_{II} . From this definition and Eq. (1) r is related to T_{bef} and T_{aft} by

$$r \equiv \frac{f_{MB}^{bef}(0) - f_{MB}^{aft}(0)}{f_{MB}^{bef}(0)} = 1 - \frac{T_{bef}}{T_{aft}}. \quad (2)$$

The inset of Fig. 3(a) shows in solid dots the values of r directly measured from the hh-X PL as a function of the delay between the two pulses for the single QW. The high

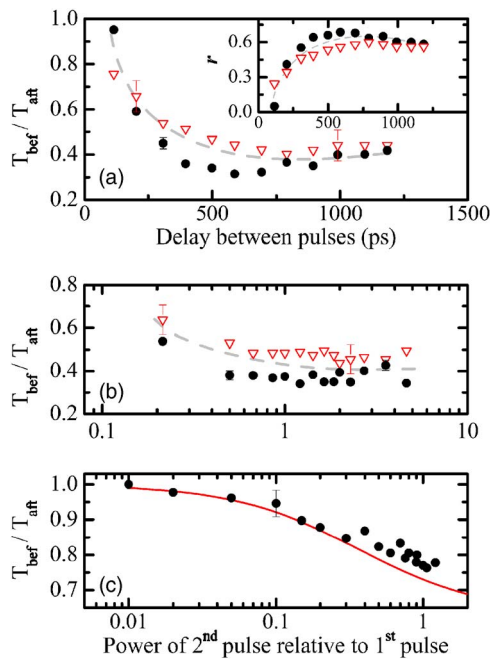


FIG. 3. (Color online) $T_{\text{bef}}/T_{\text{aft}}$ in the *single QW sample* as a function of (a) delay between pulses for a fixed power of both pulses ($70 \mu\text{W}$), and (b) power of P_{II} for a fixed delay (400 ps) and a fixed power of P_{I} ($70 \mu\text{W}$). The open [solid] dots quantify the temperature ratio as directly measured from the PL spectra [from the measured r and Eq. (2)]. The dashed lines are guides to the eye. The inset of (a) shows the corresponding values of r . (c) Same as (b) for the *multiple QWs sample* for a delay of 300 ps. The solid line depicts the result of the model.

values of r demonstrate the capability of P_{II} to quench the PL. The open points depict r obtained from the measured carrier temperature ratio¹³ $T_{\text{bef}}/T_{\text{aft}}$ and Eq. (2). In a symmetrical manner Fig. 3(a) depicts in open points $T_{\text{bef}}/T_{\text{aft}}$ directly measured, while the solid dots compile the temperature ratio as obtained from the measured dip depth in the hh-X PL intensity and Eq. (2). Figure 3(b) shows $T_{\text{bef}}/T_{\text{aft}}$ as a function of the power of P_{II} relative to that of P_{I} . In this case, the signal to noise ratio of the spectra limited the lowest power of P_{II} for which T of the eh plasma could be attained. The good agreement between the temperature ratio obtained from r in the hh-X PL and that measured directly confirms the validity of our model. This is further demonstrated in Fig. 3(c) that depicts (solid points) $T_{\text{bef}}/T_{\text{aft}}$ obtained from the hh-X PL for a different sample (the one containing multiple QWs) as a function of the power of P_{II} (delay=300 ps) in the low power regime. In this case, the broader excitonic linewidth hinders the possibility of extracting T from the spectra. Nonetheless, our model reproduces quantitatively the observed experimental dependence without the need of any adjustable parameters, as shown by the red line, which plots $T_{\text{bef}}/T_{\text{aft}}$ obtained in the following way: the temperature after P_{II} is given by $T_{\text{aft}}=(n_{\text{bef}}T_{\text{bef}}+n_{\text{inj}}T^*)/(n_{\text{bef}}+n_{\text{inj}})$, where n_{bef} is the density of carriers at

the time of arrival of P_{II} , determined from the P_{I} power and the PL decay-time, n_{inj} is the density photoinjected by P_{II} , T^* and T_{bef} , taken as 38 and 24 K, respectively, are the initial carrier T and that measured at a delay of 300 ps in the single QW experiments (see Fig. 1), which were performed under very similar conditions.

In conclusion, we have demonstrated the capability to optically control in a short time scale (<2 ps) the switch-off dynamics of the hh-X emission in QWs. A dip in the emission originates from the redistribution of carriers induced by the sudden warming of the eh plasma when a delayed pulse reaches the sample. We introduce a model that quantitatively reproduces the observed quenching and can be used to obtain the warming of the excitons. This can be exploited to study the carrier relaxation dynamics in more complex systems like microcavities,⁹ and to investigate phase transitions, such as condensation of indirect excitons¹⁴ or polaritons,¹⁵ in which T plays an important role.

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