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By excitation with two, consecutive, picosecond pulses we demonstrate that the distribution and temperature of a photo-created exciton population in semiconductor quantum wells can be dynamically tailored. The changes in the exciton distribution result in abrupt alterations of the photoluminescence dynamics, in particular in the appearance of a sudden decrease in the heavy-hole excitonic emission when the second pulse impinges upon the sample. We present a quasi-equilibrium thermodynamical model that includes the energy distribution of excitons and electron–hole plasma, which quantitatively accounts for the sharp dips observed in the exciton emission dynamics.

1 Introduction The energy distribution of excitons and its temporal evolution determine the photoluminescence (PL) dynamics in semiconductor quantum wells (QWs). Their control is crucial, for example, for the creation and destruction of exciton condensates in semiconductors [1], or for the optimization of the transport properties in two-dimensional heterostructures [2]. Recent studies have shown that the exciton formation and its population dynamics are intimately connected to the momentum distribution of the coexisting free electron-hole plasma created after non-resonant photoexcitation [3, 4]. Excitons and the bath of electrons and holes are in thermal quasi-equilibrium once the populations have thermalized [5]. In this communication we present experimental results on the optical manipulation of a photogenerated exciton population in QWs, by applying a delayed, non-resonant optical pulse. This pulse abruptly warms an already-thermalized free electron–hole plasma in the QW, which, consequently, causes a sudden warming of the exciton populations. The direct effect of the abrupt heating of the exciton distribution is the appearance of a sharp dip in the time-resolved excitonic PL when the delayed pulse reaches the sample.

2 Experimental set-up and samples The sample used for the studies presented in this work is a wide GaAs/AlAs single-QW (20 nm); however, similar results were also observed in GaAs/AlAs narrow multiple QWs (0.7 nm, 50 wells) and in InGaAs/GaAs QWs (10 nm). The sample is kept at 9 K and photoexcited with 1.5 ps pulses (repetition rate 82 MHz) with an excess energy of 26 meV (above the heavy-hole exciton emission). Two consecutive pulses from the same laser impinge upon the sample at the same excitation spot (~20 μm in diameter). The delay between the pulses and their power is controlled independently. For the detection (back-reflection geometry) we use a streak camera coupled to a spectrometer, with an overall time- and energy-resolution of 15 ps and 0.2 meV, respectively.

3 Experimental results and discussion Figure 1 shows streak camera images of the heavy-hole exciton emission after excitation by one single pulse (Fig. 1(a)) and by two consecutive pulses (Fig. 1(b)) delayed by 400 ps. In the latter case a clear dip in the excitonic emission can be observed at the time of arrival of the second laser pulse (marked by a red arrow).

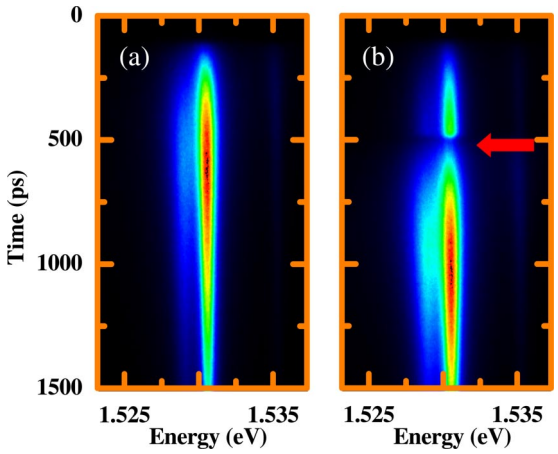

Figure 1 (online colour at: www.pss-b.com) (a) Streak camera images of the heavy-hole exciton emission for (a) a single pulse excitation (70 μW) and, (b) for two consecutive pulses excitation (delay between pulses: 400 ps; power of both pulses: 70 μW). The *z*-scale has been normalized in each panel.

Figure 2 shows in detail the time evolution of the heavy-hole exciton PL in the case of one-pulse experiments (dashed lines) and two-pulses experiment (solid line). The abrupt drop in the luminescence is time-limited by our experimental resolution; nonetheless, its magnitude can be easily quantified. We have carried out a systematic study of the dependence of the dip depth as a function of the power of the second pulse, for a fixed delay and power of the first pulse. The results are depicted in Fig. 3 (solid black dots), where a relative dip depth of 1 would correspond to a vanishing emitted intensity after the arrival of the delayed pulse. A monotonic increase of the dip depth with the power of the second pulse can be observed.

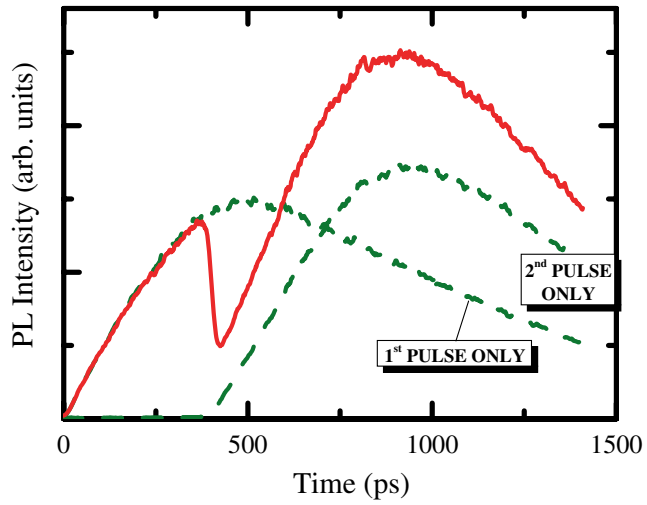

Figure 2 (online colour at: www.pss-b.com) Time evolution traces of the heavy-hole exciton for the case of single-pulse experiments (dashed lines) and the two-pulses experiment (solid line), under the conditions of Fig. 1.

The origin of the dip lies in the ultrafast changes that the exciton distribution undergoes when the second laser pulse is absorbed in the QW. In order to quantitatively explain the magnitude of the dip in the data depicted in Fig. 3, we have developed a quasi-equilibrium thermodynamical model of the excited carriers in the system. The first laser pulse irradiated on the sample creates an excited population of electrons and holes in the conduction and valence bands, respectively. In a time scale of less than 200 fs these populations thermalize and their distributions can be described by a 2D Maxwell–Boltzmann function [6] with a well defined, common temperature [7]. Subsequently, the electron–hole distribution cools down due to interaction with phonons [8], and the populations decrease due to pair recombination and exciton formation. With the arrival of the second pulse new hot carriers are photocreated in the system. Due to efficient carrier–carrier scattering, the injected electron–hole pairs thermalize with the pre-existing carrier populations (achieving a well defined temperature) in a time scale given by the pulse duration. Hence, the second pulse produces an ultrafast increase of the electron and hole populations in the bands and abruptly warms them up – as the newly injected carriers posses an energy greater than the thermal energy of the pre-existing populations. Figure 4(a) shows a schematic diagram of the electron distribution before and after the arrival of the second pulse on the sample.

Simultaneously to the previously described processes, electrons and holes bind to form heavy-hole excitons. The exciton population is in thermal equilibrium with that of the free carriers [5], due to the efficient carrier–exciton interactions, and they can also be described by a maxwellian distribution. Therefore, the abrupt warming of the electron–hole population when the second pulse impinges upon the

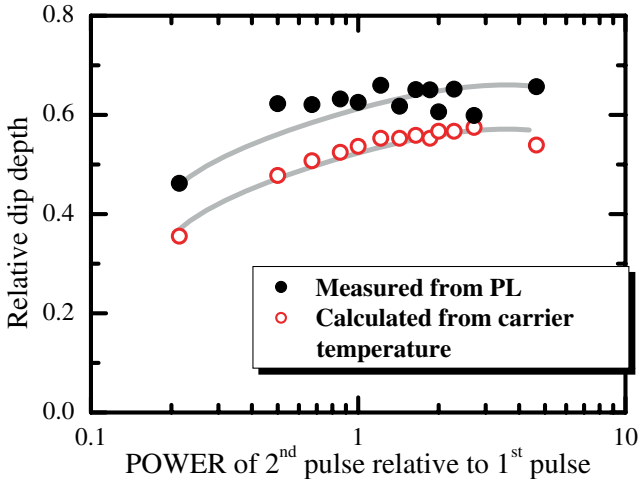

Figure 3 (online colour at: www.pss-b.com) Solid dots: measured relative dip depth as a function of power of the second pulse (first pulse power: 70 μW; delay between pulses: 400 ps). Open dots: calculated dip depth from the occupation of excitons at $K \approx 0$ as extracted from the carrier temperature. The solid lines are two parallel guides to the eye.

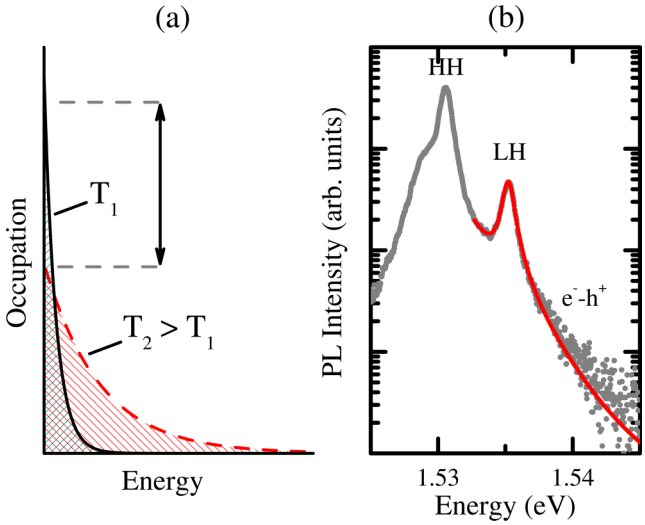

Figure 4 (online colour at: www.pss-b.com) (a) Schematic diagram of the electron distributions before (solid line) and after (dashed line) the arrival of the delayed pulse. The vertical arrow indicates the drop in occupation at the bottom of the band. (b) Emission spectrum 160 ps after the arrival of the first pulse with a power of 70 μW (grey solid dots). The red solid line is the fit, as described in the text.

sample results also in an ultrafast warming of the exciton population. The warming produces an abrupt drop of the exciton occupation in the states with centre of mass momentum $K \approx 0$. These are the optically active states and their sudden depletion produces the observed abrupt drop in the PL.

In order to check the validity of our interpretation, we have directly measured the change in carrier temperature induced by the absorption of the second pulse. Figure 4(b) shows the emission spectra (grey solid dots) at a delay of 160 ps after the excitation pulse. The heavy- and light-hole exciton emission as well as the electron–heavy-hole recombination can be observed. We have performed a fit of the high-energy side of the PL with a model that includes the light-hole exciton emission (lorentzian lineshape) and the free electron–hole pair recombination (following a maxwellian distribution). The result of the fit, for the data shown in Fig. 4(b), is depicted as a red solid line. From analogous fits we can extract the carrier temperature at any given time. In this way, we can measure the warming of the carrier population when the delayed pulse is absorbed, and consequently the warming of excitons and their occupations at $K \approx 0$.

Figure 3 depicts in open dots the results from our model for the drop in the heavy-hole exciton PL when the second pulse reaches the sample. These results follow the same trend as the directly measured dip depth and present a good qualitative agreement (within 20%), confirming the validity of the proposed model.

4 Conclusion We have shown that the distribution of free carriers and excitons in QWs can be dynamically altered with the use of ultrafast optical pulses. These modifications result in dramatic changes in the PL dynamics that can be well reproduced by a quasi-equilibrium thermodynamical model of the carriers in the system.

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