Relaxation of electron-hole pairs by coherent emission of LO-phonons in the quantum kinetic regime measured in CdZnTe quantum wells

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We study the relaxation of excitons in CdZnTe quantum wells by emission of a LO-phonon cascade, ending with a trapping in quantum dots. The state filling of the dots is measured in two-color pump-probe experiments. The observed optical-phonon emission time is found to be smaller (130 fs) than the phonon oscillation period (165 fs), showing that the interaction occurs in a quantum kinetic regime. This is evidenced by measuring the buildup of the phonon replica of an initially photocreated electron-hole pair distribution on a subpicosecond time scale.

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I. INTRODUCTION

The carrier-phonon (CP) interaction is a basic scattering mechanism for highly excited electrons and holes in semiconductors, as is carrier-carrier (CC) interaction. It is fundamental to many semiconductor nanostructure features such as transport properties or relaxation dynamics. LO-phonon emission is among the most efficient relaxation processes for electrons and holes, even if it depends on the semiconductor structure considered. It has been studied for a long time and characteristic parameters such as phonon emission time have been determined from the parameters of the Fröhlich interaction, which couples electrons and LO-phonons. The achievement of lasers that emit femtosecond pulses has allowed temporal resolving[1] of the relaxation of electron-hole pairs, which are excited at high photon energies, with a large kinetic energy, in the semiconductor bands. Nevertheless, singling out the LO-phonon contribution to the relaxation is made difficult by other relaxation processes such as CC scattering or collisions with acoustic phonons, which rapidly spread out the carrier distribution. Here, we present time-resolved experiments in which electron-hole pairs excited in quantum wells (QW) dissipate their energy by emitting a cascade of LO-phonons, which ends by the trapping of the carriers in quantum dots (QDs). As we will see below, this feature allows us to measure the phonon emission time more accurately, as well as to evidence quantum kinetic phenomena better than they have been previously evidenced in bulk or two-dimensional (2D) samples.

There are many theoretical and experimental studies on electron and hole energy relaxations due to CC and CP scatterings on a subpicosecond time scale. Most of these studies treat energy relaxation on a semiclassical level by using the Boltzmann equation. This model assumes a large difference between the time scales of the interaction processes and those of the distribution function dynamics: scattering processes are considered to be infinitely short in time with respect to the variation in the particle distribution. The duration of a collision is roughly given by the oscillation period of the energy quantum that is exchanged and the time evolution of the particle distribution, which is governed by the relaxation rate, is indeed calculated within Fermi's golden rule. When the two time scales become similar, this separation is no longer valid and a quantum kinetic description is necessary. Characteristic features of quantum kinetics are the energy-time uncertainty relation and memory effects.

The study of quantum kinetic effects has attracted considerable interest in the 1990s, and various theoretical as well as experimental papers have been published. (see Ref. 1 and references therein). Generally, such effects are expected to play a significant role in systems where transition rates are of the same order of magnitude or faster than the oscillation frequencies of the involved energy quanta. Several recent theoretical works[2–6] deal with the quantum kinetic processes in quantum-dot systems. These studies treat the influence of carrier-carrier and carrier-phonon interactions on the optical properties of semiconductor quantum-dot systems, as well as the carrier capture processes (carrier transitions from a continuum of states into the QD) and the transitions between QD states. Besides the variety of interesting phenomena it describes and the perspectives to manipulate the interaction dynamics, the quantum kinetic treatment of the carrier-phonon interaction predicts fast carrier capture and relaxation.

In semiconductor nanostructures wherein quantum dots are embedded in quantum wells, various experimental studies[7–10] have shown that, at low excitation intensities, the electron-hole dynamics is dominated by the electron-phonon interaction. Time integrated luminescence spectra exhibit modulations due to carrier-phonon coupling when the quantum dots are quasiresonantly excited, i.e., phonon replica of the photocreated carrier distribution are imprinted in the inhomogeneously broadened emission line of a quantum-dot ensemble. In this paper, we will present ultrafast transmission variation measurements of the photocarrier energy relaxation by optical-phonon emission in CdZnTe QWs. At low excitation intensities, the optical-phonon emission time is measured and characteristic features of a quantum kinetic regime are observed on the transmission variation spectra of QDs embedded in the QWs. We show that there are no well-defined phonon replicas in the first step of the relaxation

process but that they build up on a subpicosecond time scale; that is, we will show that the usually observed phonon replica in luminescence of a quantum-dot ensemble builds up in a quantum kinetic regime.

II. SAMPLE AND EXPERIMENTS

Details about our sample and its linear optical properties can be found in Ref. 8. It contains ten $Cd_xZn_{1-x}Te$ QWs, which are confined along the growth direction by pure ZnTe barriers and whose thickness is equivalent to 6.5 monolayers. These QWs embed QDs, consisting of Cd-rich ($x \simeq 0.6$) CdZnTe islands. The sample optical spectra[8] show the inhomogeneously broadened absorption and Stokes-shifted photoluminescence (PL) lines of the QDs. Electron-hole pairs can be photocreated in the Zn-rich ($x \simeq 0.2$) quantum wells if the excitation is tuned to high photon energies but below the ZnTe barrier band gap. Our previous photoluminescence excitation measurements[8] showed that the relaxation of the electron-hole population in the QW occurs through the emission of a LO-phonon cascade. When the pairs reach an energy above but near the QD ground state, they become trapped in discrete QD states by emission of a LO-phonon whose frequency is characteristic for the QD. No further thermalization occurs after this event. In luminescence, as well as in transmission experiments, this phenomenon gives rise to a spectral modulation of the QD lines. One should point out here that relaxation in the QW and trapping in the QD obey different wave-vector conservation rules.

By performing time-resolved transmission pump and probe experiments, we are able to resolve the phonon cascade in time. We use an amplified titanium-sapphire laser system that injects an optical parametric amplifier (OPA). The OPA output acts as the pump beam. Depending on their chosen duration, the pulses either go through a Fabry–Pérot interferometer or they are only filtered in a two-prism system, ensuring the compensation of the group-velocity dispersion. In both cases, their spectral widths are made narrower than that of the LO-phonon energy. We are thus able to excite spectrally narrow electron-hole distributions and to resolve their replicas. These pump pulses have an asymmetric temporal shape with a duration of 350 fs [full width at half maximum (FWHM)] when filtered with the Fabry–Pérot filter but a Gaussian shape with a duration of 150 fs in the second case. Their spectral shape can be seen in Fig. 3(a) or Figs. 5(a). As probe pulses, we use the remaining part of the white light produced in the OPA. It is a broadband continuum with a duration of 35 fs (FWHM). Great care is taken to avoid any spectral chirp in the spectral region of interest by using pulse compressors with prisms. This is measured by autocorrelation of the probe pulse and cross correlation between pump and probe pulses. The sample is cooled down to 5 K in a liquid helium cryostat. The transmitted light of the probe beam is dispersed in a spectrometer before being detected by a cooled charge coupled device camera. The differential transmission spectra are calculated for each delay between the pump and probe pulses.

As explained above, an initial electron-hole distribution is excited by the spectrally narrow pump pulses in the CdZnTe

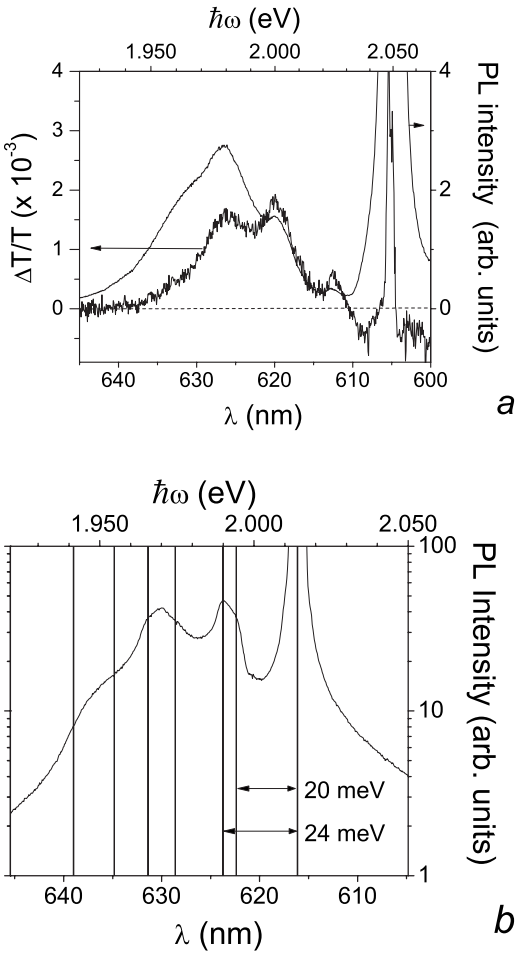

FIG. 1. (a) Transmission variation spectrum for a delay $\tau = 1.3$ ps and time integrated PL spectrum for a pulsed excitation at 2.050 eV. (b) PL spectrum measured for a cw spectrally narrow excitation at 2.014 eV, which is close to the CdZnTe QD emission. The vertical lines are guides for the eye for the two phonon modes at 20 and 24 meV, respectively, of a $Cd_xZn_{1-x}Te$ with $x=0.6$ showing the spectral positions whose distances from the excitation are integer numbers of LO-phonons. The experiment spectral resolution is about 1 meV.

QWs and decays by emitting a cascade of LO-phonons. Replicas of the initial distribution are thus induced at energy intervals that correspond to an integer number of phonons. Trapping of the carriers in the QDs also occurs by emission of LO-phonons. Therefore, only QDs that have levels at the energies of the initial excitation replicas are populated and we observe (Fig. 1) spectral modulation of the inhomogeneous absorption line of the dots. The oscillations are replicas of the initial carrier distribution with a distance between maxima given by the LO-phonon energy. In the following, we will analyze the rise dynamics of these modulations in order to determine the LO-phonon emission time of the cascade and we will study their buildup in the framework of a quantum kinetic description.

When compared to previous studies of the intraband carrier relaxation,[1] our measurements here take advantages of the electronic level scheme of our nanostructure wherein the QWs contain QDs. First, the phonon emission cascade ends

by the trapping of the electrons and holes in QDs. Because these dots show discrete electronic levels, the intradot relaxation occurs on a few picoseconds and the radiative recombination time is a few hundreds of picoseconds, i.e., both are longer than the time scale of the cascade we want to measure. Therefore, after having reached the bottom of the bands, the energy distributions of the electrons and holes do not undergo spectral diffusion, which would spread them out. Second, the differential transmission signal that we measure does not correspond to the saturation of continuum states[1] and does not call for a very high intensity of excitation. Instead, we probe discrete levels that slowly relax and are more readily saturable. Measurements can thus be performed at low intensities and, as will be shown below, avoid a large part of relaxation processes due to carrier-carrier scattering that occur at high densities.[1] Now, if we compare our sample to other structures wherein trapping arises on localized impurities instead of QDs, the dot size dispersion and stoichiometry fluctuations induce a useful inhomogeneous broadening of the energy level distribution: it allows the carriers to find dot levels that are at the right spectral position to end the cascade.

III. PHONON ENERGIES AND CARRIER RELAXATION

In Fig. 1(a), we plot the time-integrated PL spectrum and a time-resolved differential transmission (TR-DT) spectrum for a pulsed excitation tuned at 2.050 eV. The pump pulse intensity is 1.6 $\mu J/cm^2$ (5×10^{12} photons/cm^2). The TR-DT spectrum is measured at a delay of $\tau=1.3$ ps after the excitation by the pump pulse. For such a long delay, the absorption (DT spectrum), like the PL emission, shows a well-defined spectral modulation. This evidences that the emission of LO-phonon cascade gives rise to the buildup of replicas of the initial photoexcited carrier distribution. The modulation period is about 25 meV which, in the time domain Fourier transform, corresponds to a phonon oscillation period of $T_{LO}=165$ fs.

Continuous-wave (cw) excitation measurements [Fig. 1(b)] give us more accurate information about the frequency of the phonons that are involved in the cascade process in the $Cd_xZn_{1-x}Te$ alloy. Indeed, this ternary compound was shown[11] to be a two-mode system wherein two types of phonons, which are CdTe-like and ZnTe-like, coexist over the whole stoichiometric range from $x=0$ to $x=1$. The modulations observed in our PL spectra nicely show these two modes [Fig. 1(b)]. They do not reproduce the excitation linewidth, which is close to 1 meV, but are broader and present bends, which are more pronounced in the first replica and gradually softened in the following ones. Using the value of $x\simeq0.6$ from high-resolution transmission electron microscopy measurements for the QD composition, we find that the values of 20 and 24 meV, respectively, for the two LO-phonon mode energies taken from the data of Ref. 11 are in good agreement with the spectral distances deduced from our measurements. Concerning now the composition of the QW, it is close to $x=0.2$: only the Zn-like phonon mode[11] is observable at 25.5 meV. The main part of the cascade thus emits phonons at this frequency and only the few last steps, when carriers are trapped in the QDs, involve the Cd-like mode at 20 meV.

The fact that we do observe spectral modulation of the signal in a pump and probe transmission experiment is a clear indication that the electrons and hole are jointly trapped in the same quantum dots. Indeed, as we discuss below, it can be shown that the saturation of the transition that is measured in such an experiment cannot be induced by the trapping of a single carrier (or an electron or a hole) localized in the dot.

The separate relaxation of electrons and holes was shown to be dominant[12] in III-V semiconductors, while in II-VI compounds such as CdTe, the relaxation of exciton was shown to be more efficient.[13] Electron and holes undergo a fast binding into an exciton by emitting a phonon after their optical excitation and jointly relax. In both cases, the relaxation process by emission of LO-phonons does selectively populate QDs. For a single carrier relaxation, an electron and a hole state will be occupied at the energy determined by the cascade, i.e., at an integer number of LO-phonons energy from the excitation. Nevertheless, the size and composition fluctuations give rise to separate fluctuations from one dot to the other of the electron level and of the hole level. The selective population of a subclass of QDs by the trapping of one carrier at a given energy will thus not set the energy of the other carrier involved in the optical transition, whose photon energy is thus not determined. A broad absorption change should be observed in the case of a single carrier trapping. In other words, one should expect a weak correlation of the electron and hole level inhomogeneous broadenings. The selective level population by the phonon cascade of only electron or holes states would consequently not give rise to a selective saturation of the optical transitions at given photon energies. When, however, electron-hole pairs are bound into an exciton, they can be jointly trapped in the same dot. Thus, they saturate a transition at a well determined photon energy, which is set by the energy of the trapped exciton. This gives rise to the spectral structures that we observe.

The comparison between the modulation period and phonon frequency[13] cannot be used here to further support the hypothesis of a relaxation of excitons, in opposition to a separate relaxation of electrons and holes. For this last case, when a photocurrent excitation spectrum[14] or an absorption saturation[1] is measured in a quantum well or in a bulk crystal, the LO-phonon is multiplied by a factor involving the electron and hole mass ratio to give the spectrum modulation period. Indeed, when after having emitted a phonon cascade, a single electron blocks an optical transition, the empty hole state that is involved has the same k vector: the ratio between the electron and hole kinetics energy corresponds to their effective mass ratio. In our experiment, the modulation period is equal to the phonon energy. Nevertheless, in quantum dots, the optical transitions do not experience k vector selection rules and this is not another proof of an excitonic recombination.

IV. PHONON EMISSION TIME

Time-resolved pump-probe experiments allow us to estimate the emission time of the LO-phonons from the rise time

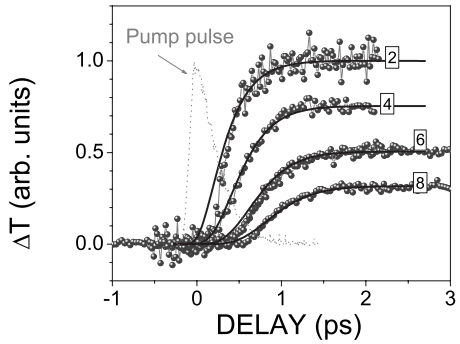

FIG. 2. Transmission variations with their fits for cascades involving different numbers of phonons (shown by the numbers in the square boxes). ΔT is measured for a fixed detection energy E_{det} and for various excitation energies E_{exc} satisfying $E_{\mathrm{exc}}-E_{\mathrm{det}}=iE_{\mathrm{LO}}$, where E_{LO} is the energy of the Zn-like optical-phonon mode. The dotted curve shows the temporal pump pulse shape.

of the transmission variation signal at different excitation photon energies: we compare several cascades (Fig. 2) that end at the same spectral position, i.e., in the same QD subset, but which start at different excitation energies that differ by an integer number of phonons. By tuning the pump pulse from 2.033 to 2.138 eV, we resolve cascades which involve up to eight phonons. The rise time of the different curves we obtained are delayed in time from each other by about 100 fs, which gives us a first approximation for the phonon emission time. In Fig. 2, the zero delay time has been carefully adjusted by cross correlation of pump and test pulses.

Let us first use a Boltzmann-type rate equation to fit our data. For a step in the cascade, the time evolution of the carrier distribution at the energy E can be written as

$$\frac{df(E,t)}{dt}=g(t)\delta(E_{\mathrm{exc}}-E)-\frac{f(E,t)}{\tau_{\mathrm{LO}}}-\frac{f(E,t)}{\tau_{\mathrm{loss}}}+\frac{f(E+\hbar\omega_{\mathrm{LO}},t)}{\tau_{\mathrm{LO}}}, \tag{1}$$

where $\hbar\omega_{\mathrm{LO}}$ and E_{exc} are the LO-phonon and excitation energies, and $f(E,t)$ and $g(t)$ are the occupation function of the exciton and the generation rate, respectively. The two middle terms describe the decay of the distribution $f(E,t)$ by the emission of a phonon, τ_{LO} being the phonon emission time, and by other channels. τ_{loss} is the inverse of the loss rate, which is due to radiative recombination, trapping on interface states, etc., or that, perhaps, should also represent the rate to create an unbound electron-hole pair in the relaxation process. This last parameter can be removed by using the definition of the quantum efficiency of the phonon emission $\eta=\tau_{\mathrm{loss}}/(\tau_{\mathrm{loss}}+\tau_{\mathrm{LO}})$. By using our previous measurement,[8] wherein $\eta=0.8$, we find $\tau_{\mathrm{LO}}=130\pm10$ fs. It is easy to show that neglecting the quantum efficiency would lead to underestimating the phonon emission time by a factor η, namely, $\tau_{\mathrm{estimated}}=\eta\tau_{\mathrm{LO}}\simeq100$ fs.

Our value can be compared to that expected from calculations,[15,16] which consider the Fröhlich interaction between electrons or holes and phonons, the energy and momentum conservations of the particles, and the electronic density of states, which is reduced in a 2D QW. By using the ZnTe parameters for a 30 nm thick QW, we obtain a value that slightly depends on the excess energy but whose mean value is around 130 fs, which is in very good agreement with our experimental measurements.

By considering the value of the quantum efficiency[8] η and the optical-phonon emission time τ_{LO}, the optical-phonon emission process leading to an excitation of the CdTe quantum dot after nonresonant excitation in the quantum well is thus shown to be very efficient and fast. Indeed, by using a very low excitation intensity and for an excitation photon energy at about 200 meV (eight LO-phonons) from the quantum-dot ground state energy, the dots are populated in less than 2 ps. During their relaxation, electron-hole pairs do give rise to a PL emission or to a differential absorption because excitons, which are rapidly bound after their excitation, have nonzero k vector and are not probed before they are trapped in the dots.

Our experiments give valuable indications on both the exciton formation time and the phonon emission time. After the excitation of free pairs, electrons and holes are rapidly bound into excitons by emission of a LO-phonon.[17,18] The excitons relax by emitting LO-phonons before being trapped into quantum dots. Figure 2 shows that these processes occur on a subpicosecond time scale. While Permogorov[14] estimated a phonon emission probability between 10^{-12} and 10^{-11}, corresponding to phonon emission time between 1 and 10 ps, our measurements confirm that both the exciton formation time and the LO-phonon emission time are of the order of 100 fs in II–VI compounds, as measured by Kalt and co-workers[17,18] in ZnSe quantum wells.

V. QUANTUM KINETICS OF THE CARRIER-PHONON INTERACTION

The LO-phonon emission time we determined above shows that the process has to be described in the framework of quantum kinetics[1,19–23] and not by a Boltzmann equation. Indeed, the value $\tau_{\mathrm{LO}}=130$ fs is shorter than the phonon oscillation period, which is $T_{\mathrm{LO}}=2\pi/\omega_{\mathrm{LO}}=165$ fs. This means that the carriers would lose energy and emit an optical-phonon energy before the end of the first phonon oscillation period. As a consequence, we expect that carrier relaxation processes show strong deviations with respect to the semiclassical description and that the energy conservation rule, which is used above in the rate equations, needs to be relaxed. This is nicely evidenced in the TR-DT curves shown in Fig. 3, which were obtained for a spectrally narrow excitation similar to that of Fig. 2. At the earliest times, the signal is not spectrally structured but shows a broad and smooth shape. For increasing times, the modulations build up and we observe an increase in the spectrum modulation contrast, which is defined as

$$C=\frac{\left.\frac{\Delta T}{T}\right|_{\mathrm{max}}-\left.\frac{\Delta T}{T}\right|_{\mathrm{min}}}{\left.\frac{\Delta T}{T}\right|_{\mathrm{max}}}, \tag{2}$$

where $\Delta T/T|_{\mathrm{max}}$ and $\Delta T/T|_{\mathrm{min}}$ indicate the transmission variations at a maximum and the following minimum, respectively. After about 1.5 ps, the carrier distribution gradu-

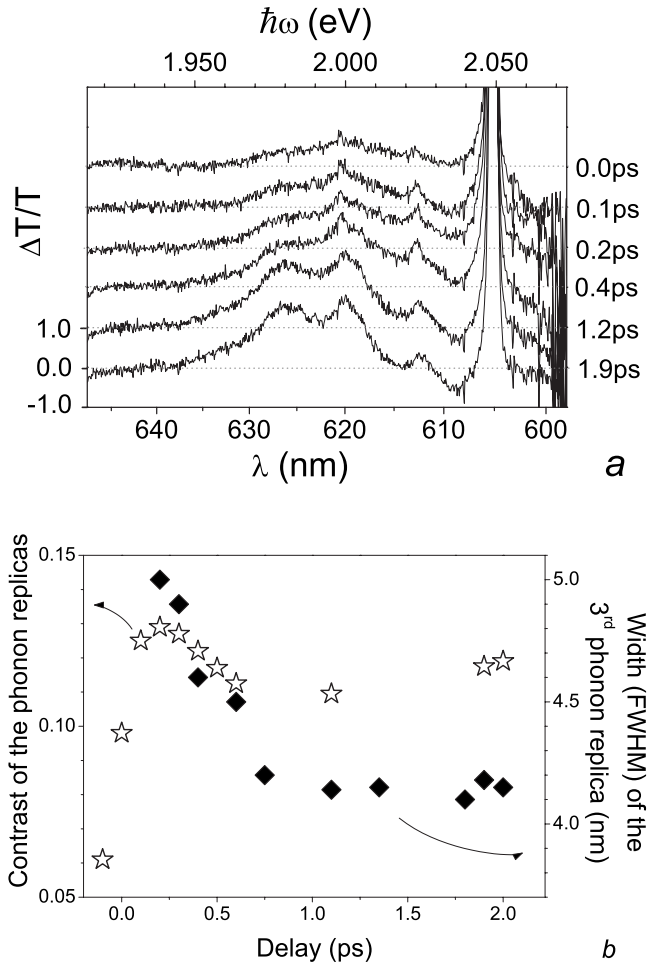

FIG. 3. (a) Transmission variation spectra for different delays between pump and probe pulses measured with long but spectrally narrow pulses. The excitation intensity is 3 $\mu J/cm^2$ (10^{13} photons/cm^2), its spectral position is 605 nm, and the pulse duration is 350 fs. The vertical lines on the upper graph show the two phonon mode energies of the CdZnTe alloy. The Rayleigh scattering of the excitation can be seen at 605 nm. (b) Time evolution of the phonon replica contrast (stars) and time dependent width of the third phonon replica (diamonds).

ally meets the shape that is predicted by the Boltzmann equation and the energy conservation condition.

Considering LO-phonon emission only, the presence of a signal at arbitrary photon energies cannot be explained by a semiclassical description, as it would correspond to a population by carriers of all the electron-hole states, including even those that cannot be reached in a relaxation process that fulfills energy conservation. The signal is, at first, a natural consequence of the energy-time uncertainty, which is important on a short time scale wherein instantaneous scattering processes cannot occur with a well-defined energy exchange and which is seen as a line broadening. Increasing times reduce the latter and restore the energy conservation fulfillment. Second, the carrier distributions gradually build up and narrow. This buildup of the modulation develops over the full duration of the pump pulse, which is longer than both the period and the emission time of the phonons. At the end, the width of the phonon replica reflects the excitation spectral width.

A change in the carrier distribution can modify the response even after the arrival of the probe pulse,[24] which is within the microscopic dephasing time of the probed transitions. It would thus modify the transient transmission spectrum. In our case, this effect should be integrated over the different arrival times of the excitons in the QDs. It would thus give rise to a broadening of the modulation, as observed for very short time delays. Nevertheless, a comparison between the phonon emission time and the phonon period gives a strong argument to suppose that the cascade cannot be described by a simple population transfer between QW levels, as in the Boltzmann equation.

At every step in the cascade, the fast transfer to the lowest energy level by emission of a phonon is compensated by the population transfer from the highest level. However, the phonon replica formation process that we observed is not only the superposition over the long excitation pulse of such successive population transfers. This could not give rise to the formation of the phonon replica without another feature of quantum kinetics: the system preserves the excitation coherence. The phase of the emitted phonons is not destroyed on a short time scale and the corresponding transitions between exciton states do not dephase. The resulting memory of the excitation pulse allows the different spectral components of the electron and hole polarizations to constructively or destructively interfere depending on the detection energy, enabling it to construct the replica.

The main features of our experimental data can be nicely reproduced by using a simple model that is appropriate for a relaxation with wave-vector conservation in a QW. We additionally assume the presence of localized states, trapping the carriers, whose absorption can be saturated. Here, the kinetic equation includes a memory kernel:[25]

$$\frac{df(k,t)}{dt} = \sum_{\mathbf{k}'} g^2(\mathbf{k}-\mathbf{k}')\{f_{k'}(t)[1-f_k(t)]D(\Delta_+) - f_k(t)[1-f_{k'}(t)]D(\Delta_-)\}, \quad (3)$$

where

$$D(\Delta) = \frac{2}{\Delta^2+\gamma^2}[\Delta \sin(\Delta t)e^{-\gamma t} - \gamma\cos(\Delta t)e^{-\gamma t} + \gamma] \quad (4)$$

and where $\Delta_\pm = \varepsilon_k - \varepsilon'_k \pm \omega_{LO}$. $g^2(\mathbf{k}-\mathbf{k}')$ is the transition amplitude from state k to the k'. Here, we use the carrier-phonon coupling parameters of ZnTe. The excitation pulse is modeled by a Gaussian envelope with a width of 9 meV and 150 fs duration, corresponding to the experimental situation of Fig. 5. We take into account the spectral distribution of the QD transition. As shown in Fig. 4, the broad initial distribution, as well as the progressive buildup of the phonon replica, is shown to be similar in both sets of curves. The inverse of the damping parameter was set to 300 fs, which is nearly twice the phonon emission time that we have previously determined.

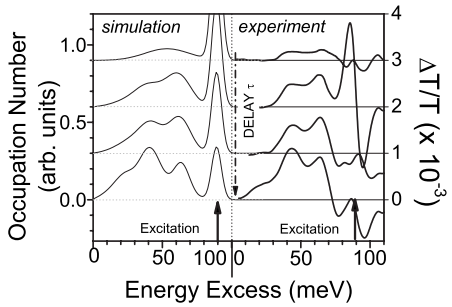

FIG. 4. Comparison between curves obtained by using a quantum kinetic model and experimental results (see text). The experimental data are identical to those shown below in Fig. 5.

Experiments performed with such short pulses allow a better time resolution (with a reduced spectral selectivity of the excitation). Their duration (150 fs) lies between the optical-phonon emission time and its oscillation period. They display (Fig. 5) a behavior similar to that observed in bulk GaAs:[1] the buildup of the different replicas is delayed from one maximum to the other by a time that is roughly given by the phonon emission time.

In Fig. 3, as well as in other experimental results presented here, one sees that a sharp line is superimposed on the broad modulation. Its energy coincides with the ZnTe mode of the ternary compound. Contrary to the broad modulation, it is present at the earliest times of the pump excitation. This sharp resonance reproduces the pump spectral shape (as shown in Figs. 5 and 6). We attribute it to a stimulated Raman emission induced by the pump pulse, which thus does not depend on the delay between the two pulses.

VI. CARRIER-CARRIER INTERACTIONS

We have also performed similar experiments with a higher excitation intensity in order to evaluate the effects of the CC interactions. Some of our results are reported in Fig. 6.

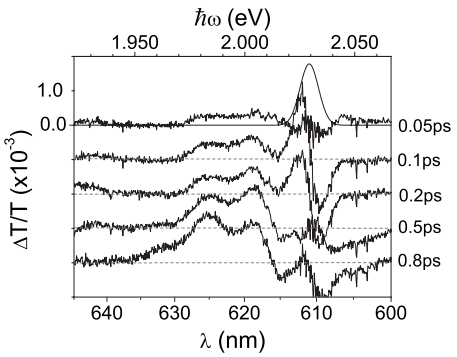

FIG. 5. Transmission variation spectra for different delays between pump and probe pulses measured with short pump pulses (150 fs). The excitation intensity is 1.6 μJ/cm^2 (5×10^{12} photons/cm^2), the spectral position is 610 nm, (2.033 eV). The spectral shape of the excitation is superimposed on the first spectrum.

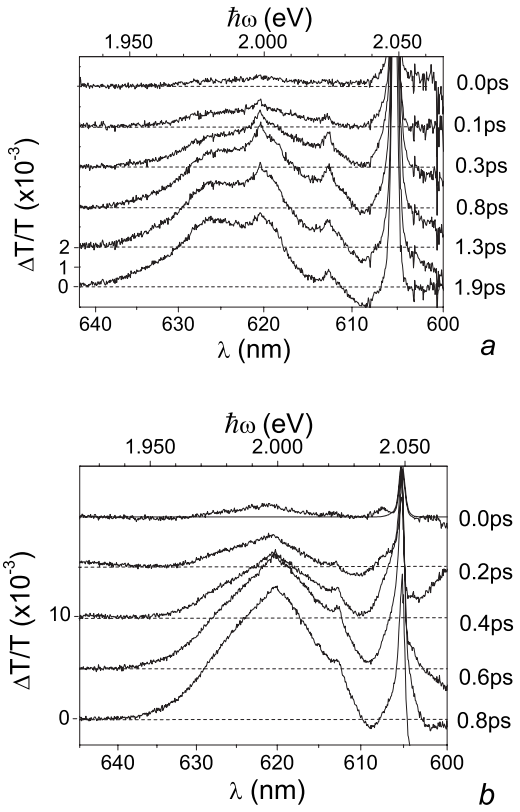

FIG. 6. Transmission variation spectra for increasing excitation densities, which are plotted for different delays between the pump and probe pulses. The excitation intensities are (a) 6 μJ/cm^2 (2×10^{13} photons/cm^2) and (b) 32 μJ/cm^2 (10^{14} photons/cm^2), its spectral position is 605 nm (2.050 eV), and the pulse duration is 350 fs. The Rayleigh scattering of the excitation can be seen on the spectra.

The data previously discussed, which were measured with an excitation density of 3 μJ/cm^2 (Fig. 3), can be first compared to those obtained with twice this density [6 μJ/cm^2 for Fig. 6(a)]. By taking into account the overall sample absorption and the number of QWs, the maximum numbers of photoexcited electron-hole pairs per QW are estimated to be 7×10^{11} and 14×10^{11} cm^{-2}, respectively. The time-resolved transmission spectra show the same quantum kinetic features as previously discussed. As observed above for lower excitation intensities, the phonon replicas, which are not visible on the first differential spectra, do appear later. Nevertheless, no pronounced minima between phonon replica, are now observed and the modulation contrast remains very low.

If we still increase the density of the excitation [32 μJ/cm^2 for Fig. 6(b), which has a maximum exciton density per quantum well of 8×10^{12} cm^{-2}], no phonon related structures are observable at any delay time. Moreover, the DT reaches its maximum value at shorter times. This intensity behavior evidences the influence of CC interaction,[26] which randomizes the initially photocreated electron-hole pair distribution and the subsequent phonon replicas. This behavior is a consequence of the fact that,

contrarily to the CP collisions case, the CC interactions scatter the electrons and the holes to a continuum of final states over which the initial distribution is irreversibly spread out.

Our experiments clearly show the transition from the regime where CP scattering predominates when compared to CC interaction (Fig. 3) to a regime where CC interactions predominate [Fig. 6(b)]. We also performed measurements with increasing excitation photon energies. We observed that the effect of CC scattering becomes more efficient when the number of optical phonons that are involved in the cascade process increases. This shows that the phonon replicas become elusive when we excite at higher photon energies. This could be due to the increase in the density of states in the continuum to which the carriers can be scattered through CC interaction.

VII. CONCLUSION

We have taken advantage of the trapping of excitons in QDs to avoid spectral diffusion at the end of the relaxation cascade by emission of LO-phonons. Time-resolved DT allowed us to directly measure the LO-phonon emission time to be 130 fs. This time, which is short compared to the phonon period, shows that the interaction occurs in a quantum kinetic regime. Our experimental data show so clearly the buildup of the carrier distribution without being influenced by carrier-carrier and carrier-acoustic phonon scatterings.

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