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LO-phonon cascade emission in CdZnTe quantum wells: coherent control and quantum kinetics

P Gilliot 1, C Brimont 1, S Cronenberger 1, O Crégut 1, M Gallart 1, B Hönerlage 1, K Kheng 2 and H Mariette 2

1 Institut de physique et chimie des matériaux de Strasbourg – GONLO, UMR 7504 CNRS - ULP (Strasbourg 1), 23 rue du Lœss, B.P. 43CR, 67034 Strasbourg cedex 2, France.
2 Équipe CEA-CNRS-UJF "Nanophysique et Semiconducteurs" 17 avenue des Martyrs, 38054 Grenoble cedex 9, France.

E-mail: Pierre.Gilliot@ipcms.u-strasbg.fr

Abstract. We study in CdZnTe quantum wells (QW) the quantum kinetics of the LO-phonon cascade which is emitted by photo-excited electron-holes pairs. We perform time-resolved photoluminescence (PL) and pump-and-probe (P&P) experiments where we take advantage of the electron-hole pair trapping in quantum dots (QDs), which imprints a spectral signature of the LO-phonon cascade in the inhomogeneously broadened QD absorption and emission lines. Using first spectrally narrow picosecond exciting pulses, we determine, from the rise time of the P&P or PL signals, the LO-phonon emission time $\tau_{LO} = 130$ fs. This measurement shows that the process has to be described in the framework of quantum kinetics. The value $\tau_{LO}$ is, indeed, shorter than the phonon oscillation period which is $T_{LO} = 2\pi/\omega_{LO} = 165$ fs. It means that the energy conservation rule which is used in rate equations is relaxed. Memory effects are evidenced by the build up of the signal modulation. In a second set of experiments, we use a sequence of two femtosecond phase-locked pulses to achieve the coherent control of the phonon emission. The decaying coherence of the carrier-phonon system which is excited by the first pulse is probed by the second one: their interference results in a spectral channeled spectrum of the electron-hole pairs populations excited in the QW, which is replicated at lower photon energies in the QD P&P spectrum. We thus measure the coherence decay of the electron-hole pairs coupled to LO-phonons. We find that it is close to their lifetime.

1. Introduction

LO phonon emission is one of the most efficient relaxation process of electron-hole pair relaxation in quantum wells. It has been studied for a long time and characteristic parameters like the phonon emission time can be determined from the parameters of Fröhlich interaction between electrons and LO phonons. More recently, it has been shown that the carrier dynamics in quantum wells needs to be described by using quantum kinetics models [1],[2],[3].

Using femtosecond pulses, this relaxation of electron-hole pairs, which are highly excited in the semiconductor bands can be temporally resolved [4]. Nevertheless, in 2D or 3D crystals showing a continuum of delocalized states, such measurements are made difficult by other relaxation processes like carrier-carrier scattering or collision with acoustic phonons, which rapidly spread out the carrier distribution. We present here time-resolved differential transmission (TR-$\Delta T$) experiments where electron-hole pairs excited in quantum wells (QW) emit a cascade of LO-phonons, before being
trapped in quantum dots (QD). In a second set of photoluminescence (PL) experiments, the coherent control of the LO-phonon emission in the very first steps of the cascade is achieved by using a sequence of two phase-locked pulses.

![Figure 1](color online): Transmission variation spectra measured at the QD absorption spectral position for an excitation at 2.050 eV by spectrally narrow pulses of 350 fs duration. Modulations appear progressively with increasing delay between pump and probe pulses, with a period given by the LO-phonon energy.

2. Sample and experiments

We study Zn-rich ZnCdTe QWs which are sandwiched between pure ZnTe barriers. These QWs contain Cd-rich ZnCdTe islands. Inhomogeneously broadened lines observed in emission and transmission spectra are attributed to these QDs. Electron-hole pairs can be excited at high photon energies in the Zn-rich quantum wells. Their relaxation within the QW gives rise to the emission of a LO-phonon cascade [5], which ends by a trapping in discrete QD states. A spectral modulation of the QD absorption and emission lines is then observed, with a separation between the maxima which corresponds to the LO phonon energy.

We present here experiments where this phonon emission process is measured by TR-ΔT pump-and-probe experiments. Previous studies of the intra-band carrier dynamics [4] suffered from the continuous density of the states of the excited electron-hole pairs. In our sample, the electronic level scheme of the nanostructure where the QWs contain QDs shows both a continuum of states where the fast relaxation occurs and discrete electronic levels whose relaxation dynamics is slower than 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 experience a spectral diffusion which would spread it out. Moreover our differential transmission experiments probe discrete levels that relax slowly and are more easily saturable than continuum states, which need a very high intensity of excitation. Low intensity measurements allow to avoid a large part of relaxation processes due to carrier-carrier scattering. The inhomogeneous broadening of the energy level distribution, which is due to the dot parameter dispersion, is also an advantage in our experiments. It does not appear in samples where
trapping arises on localized impurities instead of QDs: within the broadened line, a localized level can be found at any spectral position to end the cascade by the emission of a last LO-phonon.

3. Phonon emission time and quantum kinetics of the electron-phonon interaction

In a first set of experiments, we use spectrally narrow picosecond pump pulses. From the rise time of $\Delta T$ or PL signals, we determine the LO-phonon emission time: we compare several cascades which end at the same spectral position, i.e. in the same QD sub-set, but which start at different excitation energies that differ by an integer number of phonons. We determine thus the phonon emission time and find $\tau_{LO} = 128 \pm 8$ fs.

This measurement shows that the process has to be described in the framework of quantum kinetics [6]. Indeed, the value $\tau_{LO}$ is shorter than the phonon oscillation period which is $T_{LO} = 2\pi/\omega_{LO} = 165$ fs. It means that the energy conservation rule which is used in rate equations is relaxed. This is nicely evidenced in the $\Delta T$ curves shown in Fig. 1. At earlier times, the signal is not spectrally structured but shows a smooth shape. For increasing times, the modulations build up and we observe an increase of the contrast. This can be understood in two ways. First, the short time delays impose an uncertainty which results in a line broadening. Increasing times reduces it and restores the energy conservation fulfillment. Second, the coherence of the emitted phonons is not destroyed on a short time scale and the corresponding transition between electron states does not dephase. The resulting memory of the excitation allows the different spectral components of the electron and hole polarizations to interfere. Their distributions are thus gradually built up and they narrow in time.

![Figure 2.](image_url)

**Figure 2.** (color online): *Left:* Fourier transform of the QD PL spectra as a function of the delay between spectrally broad femtosecond pulses whose duration is 90 fs. *Right:* A modulation of the spectra is evidenced when the delay between the two exciting pulses equals an integer number of phonon periods (165 fs).
4. Coherent control of the phonon emission
Using a sequence of two phase-locked pulses, the coherent control of the phonon emission can be achieved [7]. We take here again advantage of the electron-hole pair trapping in QDs, which imprints a spectral signature of the LO-phonon cascade into the inhomogeneously broadened QD absorption line. The coarse delay between the two exciting pulses is tuned to a fixed proportion of the LO-phonon period. A fine delay allows us to precisely phase-lock the pulses. The decaying coherence of the carrier-phonon system which is excited by the first pulse is probed by the second one: their interference results in a spectral channeled spectrum of the electron-hole pairs populations excited in the QW, which is replicated at lower photon energies in the QD $\Delta T$ spectrum. We thus measure the coherence decay of the LO-phonons. We find that it is close to the lifetime of the electron-hole pairs which are coupled to the LO-phonons.

5. Conclusion
We have measured by time-resolved differential transmission experiments the LO-phonon emission time in CdZnTe and found that it is close to be 130 fs. When compared to the phonon period, this relaxation time is very short, showing that the interaction occurs in a quantum kinetics regime. We have evidenced that by measuring the build-up of the signal spectral modulation which occurs within one picosecond. The coherent control of this phonon emission has been achieved by using phase-locked exciting pulses. We have measured the coherence decay of the electron-hole pairs coupled to LO-phonons and have found that it is close to their lifetime.

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