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LETTER TO THE EDITOR

Pump–probe analysis of polaron decay in InAs/GaAs self-assembled quantum dots

F Bras[1], **S Sauvage**[1], **P Boucaud**[1], **J-M Ortega**[2] **and J-M Gérard**[3]

[1] Institut d'Électronique Fondamentale, UMR CNRS 8622, Bâtiment 220,
Université Paris-Sud, 91405 Orsay, France
[2] CLIO-Lure, Bâtiment 209 D, Université Paris Sud, 91405 Orsay, France
[3] CEA-Grenoble, DRFMC-PSC, 38054 Grenoble Cedex 9, France

E-mail: philippe.boucaud@ief.u-psud.fr

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Abstract
We have investigated by pump–probe spectroscopy the spectral and power dependences of the polaron decay in *n*-doped InAs/GaAs self-assembled quantum dots. The polaron which results from the strong coupling for the electron–phonon interaction in the dots is studied in the spectral range between 58 and 64 meV. We show that besides the initial polaron decay which occurs on a ∼30 ps time scale, a component with a long decay time appears in the pump–probe measurements. The amplitude of this component depends both on the excitation wavelength and on the excitation power density. We show that the amplitude of the long-lived component is close to zero at the resonance wavelength of the polaron absorption. We propose that the origin of this long-lived component is associated with the interaction of the exciting field with the bulk phonons of the substrate.

The study of bulk polarons and two-dimensional polarons, i.e. electrons coupled to polar longitudinal optical phonons, has been an attractive field of research in the last few decades [1, 2]. In semiconductor quantum dots, a strong coupling regime is observed for the electron–phonon interaction between discrete states and quasi-dispersionless longitudinal optical (LO) phonons. This strong coupling induces coherent effects and leads to the formation of electron–phonon quasi-particles called polarons, a coherent superposition of carrier and phonon states. The existence of polarons was first theoretically predicted by Inoshita and Sakaki in 1997 [3]. An experimental signature of the polaron existence in InAs/GaAs self-assembled quantum dots was provided by magnetoabsorption measurements in the far infrared in resonance with electronic intersublevel transitions [4]. More recently, it has been shown that the polaron picture is adequate to interpret the carrier dynamics in quantum dots [5]. In [5], the polaron decay was studied by pump–probe measurements as a function of the excitation wavelength and temperature. It was shown that the polaron decay is governed by the weight of the one-phonon component in the eigenstate and the associated finite lifetime of the phonon component which results from the lattice anharmonicity [6]. This finite lifetime is associated with the incoherent decay triggered by two-phonon processes involving LO and transverse or longitudinal acoustical (TA–LA) phonons or two acoustical phonons [7]. The nature of this instability is of prime importance as it leads to the prediction of a bottleneck for the eigenstate relaxation outside a fixed energy window [8, 9]. A striking feature is that even 20 meV above the LO phonon energy (36 meV in GaAs), the relaxation of the polaron is still governed by the LO phonon component.

The polaron dynamics can be studied by optically probing its electronic part. Far from the LO phonon energy resonance, the energy of the intersublevel transitions is mainly governed by its electronic part and therefore by the quantum dot size and geometry. For a typical lateral size around 25 nm and a lens-shaped geometry, InAs/GaAs self-assembled quantum dots exhibit some intersublevel absorption between the ground state (s state) and the first electronic excited state (p state) in the 20 μm spectral range (∼60 meV). This absorption is polarized along two separate directions because of the elongation of the quantum dots along the [−110] direction [10]. We emphasize that even if the weight of the one-phonon component is weak for transitions around 60 meV, the dynamic of the excitation is governed by the phonon part of the polaron. In [5], we have studied the polaron decay in resonance with the low energy absorption of the quantum dots. In this work, we report on polaron dynamic measurements at higher energy in resonance with the optical transition polarized along the [110] direction. The studied energy range goes from 58 meV to 64 meV. We first show that in this energy range, the polaron relaxation still occurs on a tens of picosecond time scale. We have observed that depending on the wavelength and power density excitation conditions, a component with a long decay time appears in the pump–probe measurements. We show that this long component is not associated with a bottleneck in the relaxation but rather results from an interaction between the exciting field and the two-phonon absorption of the substrate.

The investigated sample was described in [10]. It consists of 30 InAs quantum dot layers grown by molecular beam epitaxy on a GaAs substrate. The dot density is 4×10^{10} cm^{-2}. The quantum dots are n-doped with a Si delta doped layer 2 nm beneath each quantum dot layer. From low temperature mid-infrared absorption measurements, the average carrier density was estimated to be 1.2 carrier per dot [10]. The pump–probe measurements were performed with a free-electron laser tuned around 20 μm wavelength. The free-electron laser delivers macropulses with a duration of 10 μs at a repetition rate of 25 Hz. Each macropulse consists of a train of picosecond micropulses at a repetition rate of 32 MHz. The average power incident on the sample was varied using mid-infrared grid attenuators. The pump and probe beams were focused on a 0.3 mm^2 spot size. In order to improve the signal-to-noise ratio, we have performed a normalization between successive macropulses. The difference between the probe transmitted light with or without the pump is normalized by the transmitted light without the pump of the previous macropulse [5]. This procedure allows the measurement of transmission variations of less than 1%. The measurements were performed at low temperature (5 K) in a helium flow cryostat. The investigated polaron absorption which is in-plane polarized along the [110] direction is resonant around 63 meV with a full width at half maximum of 5 meV. 8 band $\mathbf{k} \cdot \mathbf{p}$ calculations of the electronic structure of the dots indicate that this energy is consistent with a lens-shaped InAs dot with lateral sizes of 25 nm along [110], 28 nm along [−110] and a height of 2.5 nm [11].

Figure 1 shows low-temperature pump–probe measurements as a function of the time delay between the incident pulses for three different average excitation power densities with the free-electron laser tuned at 63.24 meV (19.6 μm). At low excitation power (75 μW), the polaron dynamics is governed by a monoexponential decay with a T_1 lifetime around 28 ps. As the excitation density increases, the monoexponential decay is still observed but an additional component with a long characteristic decay time is observed. At the excitation wavelength of 19.6 μm, the increase of the amplitude of the long decay time component versus pump power density is more pronounced than the increase of the differential transmission at $t = 0$. The decay time of this component could not be measured with the present experimental set-up and remains much larger than 300 ps. Meanwhile, the characteristic decay time of the short component remains constant as the average power density increases. The short decay time component corresponds to the polaron relaxation as previously investigated with the polaron absorption polarized along the [−110] direction [5]. The decay is triggered by the instability of the LO phonon and its finite lifetime because of the crystal anharmonicity [6]. The value of 28 ps is shorter than those measured at longer wavelength. We emphasize that the data reported above 60 meV require a careful analysis. The polaron state can no longer be discussed in terms of zero and one-phonon components alone. The polaron level is significantly nearer to the two-phonon resonance and the weight of the two-phonon component S2 in the polaron state is expected to be non-negligible[4]. When the energy of the level increases towards the two-phonon resonance, the weight of the one-phonon component continues to decrease but the weight of the two-phonon component significantly increases and should ultimately rule the relaxation near the 72 meV (i.e. 2 × 36 meV) resonance. Note that the relative weight of the components is very sensitive to the exact position of the two-phonon resonances given by the phonon energy (36 meV in the GaAs matrix). Moreover, the different relaxation pathways involving optical and acoustical phonons that can occur for the polaron decay considerably influence the relaxation rate. As an example, the LA phonons have a broadening of about 30 meV. The combination of two LA phonons thus defines a relaxation window up to 60 meV and a bottleneck should appear above 60 meV for a relaxation mechanism involving two LA phonons [12]. Note that transitions assisted by acoustical phonons can occur between excited polaron states associated with the cross-polarized transitions from the ground state. This mechanism, not

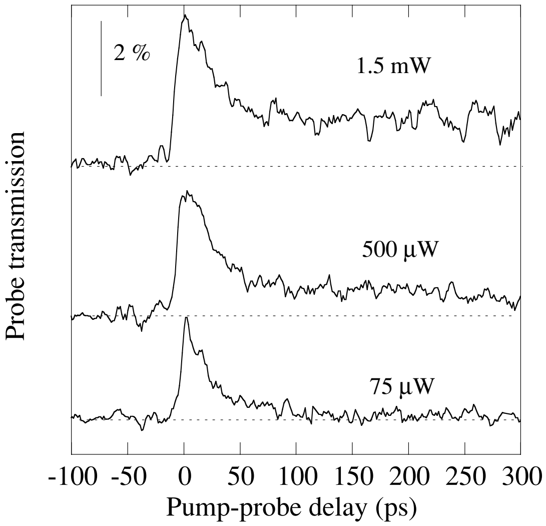

Figure 1. pump–probe measurement along the [110] direction as a function of the time delay between the incident pump and probe pulses for three different average pump excitation power densities. From bottom to top: 75 μW–500 μW–1.5 mW. The free-electron laser is tuned at 63.24 meV (19.6 μm). The temperature is 5 K. A dashed line indicates the baseline at negative time delay. The curves have been offset for clarity.

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[4] The polaron excited state can be written as a linear combination of zero-, one- and two-phonon components: $\alpha|\mathrm{P0}\rangle + \beta|\mathrm{S1}\rangle + \gamma|\mathrm{P1}\rangle + \delta\,|\mathrm{S2}\rangle$.

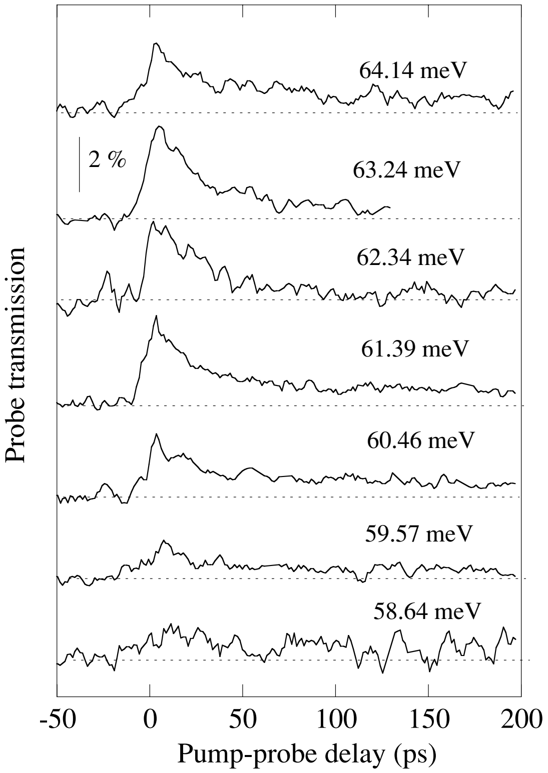

Figure 2. Spectral dependence of the low temperature pump–probe measurements. The average pump power is 120 μW. The energy of the excitation is indicated on the right-hand side of the figure. The curves have been offset for clarity. A dashed line indicates the baseline at negative time delays.

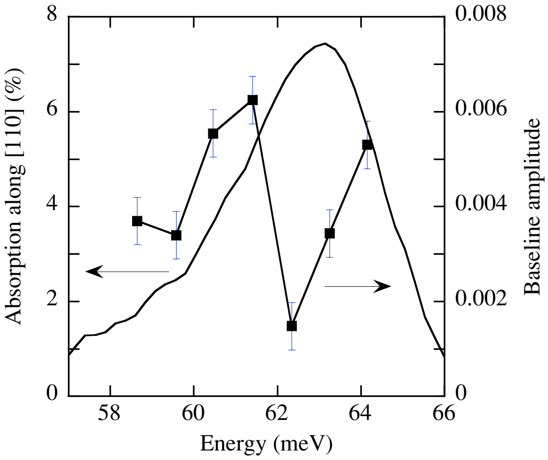

Figure 3. Spectral dependence of the amplitude of the long decay time component (right scale) as compared to the amplitude of the polaron absorption (left scale) measured at low temperature with a Fourier transform infrared spectrometer.

present for the low energy polaron resonance, can provide an efficient relaxation pathway that can contribute to a significant decrease of the relaxation time at high energy [13]. From the data reported in figure 1, we observe that a polaron decay with a finite lifetime is still observed even at energies larger than 60 meV. There arises the question whether the long decay component is a signature of a polaron relaxation bottleneck and thus would correspond to an absence of decay mechanism for the polaron [8].

In order to clarify this issue, we have investigated the polaron dynamics as a function of the excitation wavelength. The average power density is kept constant at a relatively weak value (120 μW). Figure 2 shows the energy dependence of the pump–probe measurements. Two features can be observed on the spectra. First, the amplitude of the variation transmission at $t = 0$ depends on the energy of the excitation and follows the spectral dependence of the polaron absorption. The variation of transmission at $t = 0$ is maximum at 63 meV, an energy which corresponds to the resonance of the polaron absorption. The decay time T_1 of the short component increases from 17 ps to 34 ps as the excitation energy increases from 59.5 meV to 64 meV. As discussed above, further theoretical investigation is required in order to fully simulate this energy relaxation dependence, in particular by taking into account the instability of the two-phonon component of the polaron. Second, the striking feature reported in figure 2 is that the amplitude of the component with a long decay time depends on the excitation wavelength but does not follow the spectral dependence of the polaron absorption. The amplitude of the long decay time component is minimum for an excitation energy ($\sim$62 meV) very close to the resonance of the polaron absorption. At this minimum, the amplitude does not depend on the excitation power density. Figure 3 shows the spectral dependence of the amplitude of the long-lived component as compared to the polaron absorption measured with a Fourier transform infrared spectrometer. There is clearly, as seen in figure 3, an opposite behaviour between the polaron absorption and the amplitude of the long decay time component. For this reason, we rule out that this long component is related to the polaron decay. To understand the origin of this feature, we recall that the present measurements are performed with quantum dots grown on a GaAs substrate. The substrate exhibits a significant two-phonon absorption in this spectral region with a strong resonance at 65.5 meV, smaller resonances at 64 and 55.5 meV and a dip at 60 meV [14]. A plateau is observed around 62 meV in the two-phonon absorption. Note that the absorption coefficient at 65 meV is around 30 cm^{-1} and the substrate thickness in the present experiment is around 300 μm and that the spectral position of the resonances depends on the temperature. We propose that the onset of the long decay time component is associated with a thermal effect in the substrate and a transmission modulation by the pump pulse due to the bulk substrate two-phonon absorption. This interpretation is consistent with the power dependence as reported in figure 1. If the excitation energy matches or is close to the two-phonon absorption resonance of the substrate, the amplitude of the long decay component increases with the excitation density because of the transmission variation induced by the pump. The variation of transmission is reduced for energies falling between the substrate absorption resonances. Note that a quantitative and precise analysis of this thermal effect is difficult to obtain and beyond the scope of this letter.

In conclusion, we have investigated by pump–probe measurements the polaron dynamics in InAs/GaAs self-assembled quantum dots at energies around 60 meV. We have shown that besides the polaron decay which occurs on a $\sim$30 ps time scale, a component with a long decay time can appear on the time-resolved spectra. The onset of this component does not modify the polaron decay time. The amplitude of this long-lived component depends on the excitation energy and on the power density. As the amplitude of the long component does not mimic the polaron absorption, we rule out that this feature is a signature of a polaron relaxation bottleneck. The origin of this long-lived component is attributed to the two-phonon absorption in the substrate.

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