We study Coulomb and carrier dynamics in self-assembled InAs/GaAs quantum dots at room temperature by two-color tunable differential transmission experiments, with resonant excitation in the ground state. Coulomb renormalization of the first excited state in the presence of one electron-hole pair in the ground state manifests as a 6-meV redshift. Several time scales for carrier activation to the first excited state are distinguished, corresponding to activation processes occurring in singly and doubly occupied dots, including carrier thermalization. Electron thermal activation occurs on a 250 ps time scale, confirming the absence of a phonon bottleneck at room temperature.

DOI: 10.1103/PhysRevB.67.235323  PACS number(s): 78.47.+p, 78.67.Hc

Self-assembled semiconductor quantum dots (QD’s) have been attracting much interest in recent years both fundamentally, as an exemplification of quasi-zero-dimensional semiconductor nanostructures, and for applied purposes, as efficient active emitters in the infrared spectrum monolithically grown on GaAs substrates. The carrier dynamics in such systems has been studied extensively using transient optical spectroscopy techniques such as time-resolved photoluminescence (PL) and pump-probe spectroscopy. Early studies in self-assembled QD’s were mostly concentrated on the carrier relaxation dynamics from the host material (barriers) into the confined states of the dots. Over the last decade, consistent efforts have been devoted to the study of the carrier dynamics within the confined states of the dots using resonant injection in the low-lying states. The intradot carrier dynamics is determined by a nontrivial interplay between carrier-carrier and carrier-phonon scattering. Experiments have shown that electron-phonon scattering is very sensitive to the geometry of the dots, which tailors the electron and hole wave functions and thus the electron-phonon interaction strength. In lens- or disk-shaped QD’s with high electronic and hole wave functions and thus the electron-phonon interaction, the electron-phonon interaction is responsible for an effective phonon bottleneck at low temperatures.

In previous differential transmission experiments in long-wavelength InAs QD’s, we showed that thermal escape of carriers resonantly injected in the ground state occurs on a time scale of 100 ps or longer (depending upon the sample structure). Time-resolved PL experiments revealed that electrons are activated to the first excited state on a time scale of 15 ps, as a result of intradot electron-hole scattering following ultrafast hole thermalization. However, excited-state PL emission from thermally activated electrons on a time scale longer than 100 ps was not observed. It is therefore conjectured that at room temperature most carriers (either electrons or holes, or both) disappear from the ground state of the dots, being, for instance, trapped to defect states. Electron thermal activation to the first excited state possibly occurs in dark QD’s where no radiative recombination takes place because the holes escape from the dots on a time scale much shorter than 100 ps. In such a case, a nondegenerate pump-probe technique is required in order to measure the intradot electron thermal activation dynamics. In fact, it is still an open question whether the rate of electron thermalization within the lowest QD states at room temperature is larger or smaller than the radiative recombination rate. The intradot carrier thermalization dynamics has important implications in the design of QD optical devices; therefore, its study motivates further experimental investigations in long-wavelength self-assembled QD’s.

In this paper, we report Coulomb and carrier-activation dynamics in long-wavelength (1.3 μm) self-assembled InAs/GaAs QD’s using a novel tunable two-color pump-probe spectroscopy technique, based on the soliton self-frequency shift in a tapered microstructure fiber. This technique allows us to measure the dynamics of dark QD’s occupied by single electrons or single holes—a clear advantage over PL and PL-excitation techniques. Experiments are done at low excitation fluences where the average electron-hole pair occupation per dot (η) and the fraction of doubly occupied dots (η₂) are smaller than 0.4 and 0.04, respectively. The calibration of the quantities η and η₂ versus excitation intensity is based on previous resonant saturation experiments. The QD’s are pumped in the ground state and probed at various wavelengths across the first excited state for different time delays. The experimental results give a complete picture of the intradot dynamics, including Coulomb renormalization effects and the electron thermal activation dynamics.

The self-assembled InAs/GaAs QD sample we investigated is grown by molecular-beam epitaxy on a GaAs substrate and exhibits ground-state emission at 1300 nm, at room temperature. The estimated dot density in each layer is...
FIG. 1. Spectra of the pulses injected into the microstructure fiber (solid line centered at 1180 nm) and of the fiber output (dotted line). The average power launched into the fiber is $\approx 50$ mW. The output spectrum filtered at 1300 nm is shown as a solid line. The dashed line centered at 1180 nm is the spectral convolution between the input (probe) and the filtered output (pump) spectra. Inset: intensity cross correlation between the pump and probe pulses versus time delay, obtained by two-photon absorption in the GaAs substrate, in a dot-free region of the sample. The trace width (FWHM) is $\approx 200$ fs.

$= 3.5 \times 10^{10}$ cm$^{-2}$. Previous experiments on the same QD's have shown evidence of electronic states with nearly circular symmetry. The first excited state is probed by 140-fs-long pulses from a tunable optical parametric oscillator (OPO). The pump pulses are self-frequency-shifted solitons generated starting from the same pulses, using a tapered microstructure fiber with a core size of 2 $\mu$m and a length of 10 cm for the tapered region. Self-frequency-shifted soliton (pump) pulses centered at 1300 nm are generated starting from various wavelengths of the OPO pulses by varying the input pulse peak power. High tunability ($> 150$ nm) is achieved for the first time in a nondegenerate pump-probe experiment starting from the low-peak-power (kW) pulses of a single oscillator. The pump and probe beams are focused onto the sample at near-normal incidence and are cross-linearly polarized. Nearly-shot-noise-limited sensitivity ($= 10^{-5}$) is achieved in differential transmission ($\Delta T/T$) measurements through the use of balanced detection of the probe beam combined with lock-in amplification of the pump-induced changes. The signal-to-noise ratio of the pump-induced signal is limited by the pump noise in the detection bandwidth.

Figure 1 depicts the spectrum of the OPO (input) pulses and of the pulses at the output of the microstructure fiber for a central input wavelength of 1180 nm. More than 50% of the input pulse energy is downshifted to the wavelength of interest. The pump pulses are obtained by filtering the output beam using an interference filter with a bandwidth [full width at half maximum (FWHM)] of 30 nm. From the intensity cross correlation of the pump and probe pulses we infer a temporal resolution of the experiments of about 200 fs (inset of Fig. 1). Spectral oscillations, due to multimode interference within the microstructure fiber, denote the presence of a satellite pulse delayed by $= 1.3$ ps, which carries only 0.5% of the total energy and therefore does not affect the differential transmission measurements.

In order to determine the transition energies and spectral shapes of the ground state and first excited state, we performed degenerate (one-color) differential transmission experiments in an arrangement where the microstructure fiber is bypassed. The degenerate $\Delta T/T$ signal is proportional to the square of the linear absorption of the dots. Measurements were done as a function of delay for different pulse wavelengths across the two lowest QD resonances. The experimental traces (shown in the inset of Fig. 2) exhibit a three-component exponential decay function that describes the resonant depopulation dynamics. The signal values extrapolated to zero delay are plotted versus wavelength, for an excitation intensity of 0.24 kW/cm$^2$, which corresponds to $\eta \approx 0.1$ and $\eta_2 \approx 3 \times 10^{-3}$ (open circles in Fig. 2). The square root of the spectrum so obtained is proportional to the linear absorption spectrum (solid circles). The absorption spectrum is not deconvolved from the spectrum of the probe pulses and the inhomogeneous line shape of the dots. However, the spectral resolution of the experiments ($= 18$ nm) is smaller than the inhomogeneous linewidth of both the ground state and the first excited state. From the value of the QD linear absorption at the peak of the ground state ($= 4 \times 10^{-4}$ per
layer) we estimate the oscillator strength ($f$) of the QD ground state to be $f = 7.4$, in good agreement with the value reported by Birkedal et al.\textsuperscript{10} in similar QD’s. The minor discrepancy is attributed to slightly different values taken for the linear absorption and the inhomogeneous linewidth of the ground state.

Nondegenerate differential transmission ($\Delta T/T$) curves following resonant excitation in the ground state (at 1300 nm) are shown in Fig. 3 as a function of time delay ($\Delta t$) for a series of probe wavelengths across the first excited state. The excitation intensity was 1.1 kW/cm$^2$ ($\eta_1 \sim 0.4$, $\eta_2 \sim 0.04$); at this excitation intensity the system is in the weak excitation regime, and two-photon absorption in the GaAs barriers is negligible.\textsuperscript{8} The $\Delta T/T$ signal originating from the QD’s occurs only at $\Delta t > 0$, i.e., when the pump pulses anticipate the probe pulses, as confirmed by a control experiment performed on a dot-free region of the sample. The QD response exhibits either induced absorption (negative signal) or induced transmission (positive signal), depending upon the probe wavelength. The early-time ($\Delta t < 60$ ps) dynamics is characterized by an ultrafast component with a characteristic time of $\sim 1$ ps, followed by a slower component that develops on a time scale of tens of ps or longer. The two components are extracted from the data by fitting double-component exponential functions of the form $-A_u \exp(-\Delta t/\tau_u) - A_1 \exp(-\Delta t/\tau_1)$ to the time traces (positive values of the constants $A_u$ and $A_1$ represent signal components that increase with increasing time delay).

We describe the early-time dynamics in terms of two distinct physical phenomena: (i) Electron-hole pairs injected in the ground state by the pump pulses produce an instantaneous renormalization (redshift) of the wavelength of the first excited state through Coulomb interaction, causing a dispersivelike $\Delta T/T$ spectrum. (ii) Carriers are activated from the ground state to the first excited state, generating a (positive) bleaching signal that increases with time delay. According to our previous experimental results,\textsuperscript{7,8} we attribute the ultrafast ($\sim 1$ ps time scale) component to thermalization of the holes; the ultrafast transient is in fact complementary to the ultrafast depopulation component of the resonant dynamics in the ground state (shown in the inset of Fig. 2).

At zero delay, all carriers are still in the ground state and only Coulomb renormalization contributes to the differential signal at wavelengths across the first excited state. Thus, we can extract the Coulomb shift spectrum by extrapolating the spectrum of the $\Delta T/T$ signal to zero delay. The resulting spectrum is shown as solid circles in Fig. 4(a). The dispersivelike shape of the spectrum strongly suggests that Coulomb renormalization occurs instantaneously as a rigid redshift of the first excited state. The magnitude of the redshift is determined by fitting the data to the difference of two Gaussians, one centered at the unperturbed transition wavelength of the first excited state ($\lambda_2 = 1191.5$ nm) and the other centered at the redshifted wavelength $\lambda_2 + \delta\lambda$. The free parameters are the amplitudes and the widths (the same for the two Gaussians) and the redshift $\delta\lambda$. This analysis yields the redshift value $\delta\lambda = (6.7 \pm 1.5)$ nm or, equivalently, $\delta E = (5.8 \pm 1.3)$ meV.\textsuperscript{11}

Since free carrier effects due to two-photon absorption in the GaAs barriers are negligible at the low excitation intensities used in the experiment, the redshift is attributed to QD’s filled with one electron-hole pair in the ground state. This is to the best of our knowledge the first determination of the Coulomb shift in long-wavelength InAs QD’s by resonant optical excitation experiments, at room temperature.
Previous reports on Coulomb renormalization in self-assembled QD’s show either a \( \approx 15 \) meV redshift of the first excited state when two (electrically injected) electrons are present in the ground state\(^{12}\) or a small (\( \approx 1.5 \) meV) average redshift per electron-hole pair in PL experiments with excitation in the dot barriers.\(^{13}\) The experimental value of the Coulomb shift is thus of the correct order of magnitude. We notice a minor systematic deviation of the fit curve in Fig. 4(a) from the data, which is indicative of a slightly more complex Coulomb renormalization spectrum. A detailed analysis using a quantitative estimate of the Coulomb shift would require a microscopic approach and is not attempted here.

In order to determine the spectrum of the activated holes, we deconvolve the contributions of Coulomb renormalization and hole bleaching to the ultrafast transient. We assume that each carrier (either electron or hole) present in the ground state produces a Coulomb shift \( \delta N/2 \) in the dot it occupies: hence a hole leaving the ground state will produce a bleaching spectrum centered at the renormalized wavelength \( \lambda_{N} + \delta N/2 \) and a Coulomb shift \( \delta N/2 \) due to a single hole in the ground state will vanish. The analysis is then performed by fitting the spectrum of the amplitude of the ultrafast component, shown as solid circles in Fig. 4(b), with the sum between a Gaussian (corresponding to the hole bleaching) and the derivative of a Gaussian (corresponding to a decrease in Coulomb shift). Only the respective amplitudes of the two components have to be determined, the other parameters having been derived from the analysis of the Coulomb shift at zero delay. It turns out that Coulomb renormalization and hole bleaching contribute with similar weights to the spectrum of the ultrafast transient, as would be expected. Therefore, the ultrafast component of the QD dynamics is completely understood as the result of the interplay between Coulomb-induced lineshift and intradot thermal excitation of the holes.

The analysis of the system dynamics on the intermediate (tens of ps) time scale is more complex. In the long-wavelength portion of the spectrum, carrier bleaching cannot be unambiguously separated from a Coulomb-induced signal that decays in \( \approx 1 \) ns due to electron-hole recombination in the ground state, tending to zero from negative values. By contrast, at short wavelengths (\( \lambda < 1200 \) nm), a time constant \( < 25 \) ps is clearly identified for the intermediate component, which is thus ascribed to bleaching due to electrons activated via electron-hole scattering after hole thermalization in the doubly occupied dots.\(^{8}\)

We finally address the system dynamics on the time scale of carrier recombination (\( \tau_{\text{rec}} \approx 1 \) ns). In order to determine whether thermally excited electrons reach the first excited state of the dots, we perform a two-color experiment over long time delays (900 ps, limited by the experimental setup) for three probe wavelengths, chosen such that the initial \( \Delta T/T \) signal is large and positive, close to zero, and large and negative, respectively. The differential transmission curves for the three wavelengths are presented in Fig. 5, together with the degenerate pump-probe signal acquired at the peak of the ground state, shown as the thick gray line.\(^{14}\) The signals detected at 1185 nm and 1215 nm decay at long times with a time constant of the order of \( \tau_{\text{rec}} \). By contrast, at 1200 nm, chosen such that the \( \Delta T/T \) signal reaches zero after the ultrafast transient and where the system dynamics is nearly unaffected by Coulomb effects, the signal monotonically increases with delay with a time constant \( \approx 140 \) ps. This signal is attributed to electrons because the holes are already known to thermalize on a \( \approx 1 \) ps time scale. We suggest that this bleaching component originates from QD’s excited with one electron-hole pair in the ground state, where the electron is thermally activated to the first excited state, while the hole is lost by trapping into a defect state or activation into the wetting layer,\(^{15}\) and thus cannot radiatively recombine with the electron in the first excited state. From the intensity of the differential transmission signal, we estimate that approximately 50% of the excited QD’s exhibit this dynamics. The hypothesis of hole activation into the wetting layer is strongly supported by recent temperature-dependent cw PL experiments, where we have measured a thermal activation energy in agreement with the estimate for the hole ground-state to wetting-layer energy spacing in the QD’s \( (\approx 230 \) meV).

We perform a more detailed analysis in order to verify the consistency of the 140 ps component as an electron bleaching signal originating from the first excited state of the dots. The differential transmission time trace at 1200 nm is subtracted from the traces at 1185 nm and 1215 nm, with weights adjusted in such a way that the signals at these two wavelengths decay with the same time constant \( \tau_{\text{rec}} = 1030 \) ps as the \( \Delta T/T \) trace taken degenerately at the peak of the ground state (thick gray line in Fig. 5). Here we are assuming that the dispersive-like Coulomb-induced spectrum is homogeneous: i.e., the signal decay time is wavelength dependent.
independent. The spectrum so inferred (inset of Fig. 5) confirms that the long-living component is carrier bleaching originating from the first excited state of the dots.

To summarize, the electrons have two distinct energy activation mechanisms within the QD states: a fast process with a characteristic time of the order of 20 ps,\(^7\) which relies on electron-hole scattering, and a much slower process due to phonon scattering (direct thermal excitation). Other activation mechanisms such as plasma Coulomb scattering are ruled out due to the weak excitation conditions. At very low fluences (<0.2 kW/cm\(^2\)), \(\eta < 0.1\), \(\eta_2 \approx 2 \times 10^{-3}\), where the influence of intradot electron-hole scattering on the QD dynamics is also negligible, we find an electron thermal activation time of about 260 ps, in good agreement with the results reported by another group on long-wavelength disk-shaped InAs/GaAs self-assembled QD's.\(^4\) These findings are also consistent with the results of our previous studies of the resonant depopulation dynamics of the ground state.\(^5\)

In conclusion, we report a comprehensive study of the carrier dynamics in long-wavelength self-assembled InAs QD's by high-sensitivity two-color pump-probe experiments with subpicosecond temporal resolution, using a novel technique based on soliton self-frequency shift in a tapered microstructure fiber. Resonant excitation in the lowest QD interband optical transition allows the unambiguous determination of the Coulomb-induced shift of the first excited state of QD's occupied by a single electron-hole pair in the ground state. Importantly, we show that intradot electron thermalization takes place on a time scale (\(~250\) ps) which is smaller than that of electron-hole recombination, hence confirming the absence of a phonon bottleneck in self-assembled QD's, at room temperature. Intradot electron thermal excitation occurs in a large class of QD's where the holes cannot radiatively recombine with the electrons, thereby resulting in an electron lifetime longer than the radiative lifetime. These experimental findings are potentially important for future applications of InAs self-assembled QD's.

The authors thankfully acknowledge J. Shah for initial guidance, C. Xu and X. Liu for essential advice and the use of the optical parametric oscillator, J. Perna and L. W. Stulz for technical support, and R.C. Giles for a critical reading of the manuscript.


\(^11\) The amplitude of the Gaussian absorption line extracted from this analysis agrees within a factor of 2 with the linear absorption coefficient inferred at the peak of the first excited state from the degenerate \(\Delta T/T\) measurements. The absorption linewidth is slightly larger (less than 10\%) than the width of the convolution between the pump and probe spectra. This small discrepancy can be explained by taking into account the distribution in size and shape of the dots.


\(^14\) There are minor discrepancies between the long and short time traces in the wavelength region 1200–1215 nm, due to slight variations in the position of the excitation spot on the sample, and thus in QD transition wavelength.