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Coherent acoustic phonons in strain engineered InAs/GaAs quantum dot clusters

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Coherent excitation of the quasilongitudinal and quasitransverse acoustic phonon mode in strain engineered InAs/GaAs quantum dot (QD) clusters grown on (311)B GaAs is monitored by means of time-resolved differential reflection spectroscopy. Carrier capture within the ordered QD clusters initiate coherent acoustic phonon excitation, which induces a transient modulation of the local strain-induced piezoelectric field within the QD clusters. The excited acoustic phonons then modulate the optical properties of the QDs through the quantum-confined Stark effect, causing distinct oscillations of the differential reflection signal. © 2006 American Institute of Physics. [DOI: 10.1063/1.2193460]

In recent years, the formation of well-defined and ordered semiconductor quantum dot (QD) nanostructures by epitaxial growth has attracted considerable attention. Much interest involves the formation and investigation of vertically coupled QD pairs, see, e.g., Refs. 1 and 2, which can serve as the basic ingredients of quantum computation. On the other hand, the formation of nanosized structures in which QDs are laterally coupled provides a challenge in epitaxial crystal growth. High quality QDs in well-defined arrangements, such as QD arrays3–5 and ordered QD groups,6–9 have been realized by self-organized strain engineering. The number of QDs within a single group or QD cluster, formed by using strained-layer superlattice (SL) templates,7,8 is controlled by varying the growth temperature of the SL template and the thickness of the GaAs separation layer between the SL template and the QD layer.8 A consequence of strained nanomaterials is that the strongly enhanced piezoelectric field is 1 μm2.

The ordered QD structure is grown by molecular beam epitaxy on a (311)B GaAs substrate. After deposition of a 250 nm GaAs buffer layer at 580 °C, a (In,Ga)As/GaAs SL template (10 periods) is grown at 520 °C. A 15 nm GaAs separation layer is deposited at 580 °C before the growth of the QDs: between 0.5 and 0.6 nm of InAs at 470 °C. The QDs are capped with 200 nm GaAs. Atomic force microscopy (AFM) images of uncapped QDs reveal the formation of QD clusters with an average density of four QDs per cluster (see Fig. 1). For a detailed description of the sample growth, we refer to Refs. 7 and 8. The inset of Fig. 1 depicts the photoluminescence (PL) spectrum of the QD clusters, at 4.2 and 293 K, with QD ground state peak energies of 1.133 and 1.046 eV, respectively. The peak at 1.327 eV (4.2 K) originates from the SL template.

In order to determine the optical time response of the QDs in the clusters, the sample is investigated by two-color pump-probe TRDR (Ref. 15) at 5 and 293 K. In this configuration, a 76.6 MHz mode-locked Ti:sapphire laser is used as the pump source with a photon energy tuned above the GaAs band gap energy, and is mechanically chopped with a frequency of 4 kHz. Hereby, free carriers are generated within the GaAs barrier layers in which they diffuse toward the SL...
template and the QD clusters. The capture of the carriers into the QDs results in a change of the QD reflection, and is monitored by 2 ps probe pulses generated from an optical parametric oscillator, synchronously pumped by the Ti:sapphire laser. The probe energy is tuned in resonance with the peak energy of the QD ground state transition of the ensemble of QD clusters. The probe pulses propagate perpendicular to the QD plane and parallel to the [311] crystal direction. For more experimental details, we refer to Ref. 15.

Figure 1 depicts the transient differential reflection signal, \((\Delta R/R_0)(t)\), measured at 5 K for an excitation density of 1.7 kW/cm\(^2\). From the rise time of the transient signal, the carrier capture and relaxation time are deduced, which have a value of 19.6±0.2 ps. After the signal has reached its maximum value, the transient signal decays exponentially due to carrier recombination with a characteristic time, i.e., a carrier lifetime, of 724±4 ps. Pronounced oscillations in the TRDR signal are observed, emphasized by the residue obtained by the subtraction of the experimental fit from the original data, as depicted in Fig. 1. In order to determine the frequency of the oscillation, we use fast Fourier transform (FFT) analysis of the oscillating part. From the FFT spectrum we deduce a frequency of 9.09 GHz, which corresponds to an oscillation period \(\tau_{\text{osc}}=110\) ps. We emphasize that only for high excitation density, oscillations in the TRDR signal are observed. In addition, we remark that at a temperature of 77 K, similar oscillations are observed as obtained at 5 K.

Oscillations in differential reflection signals have been reported for semiconductor quantum well structures\(^{16,17,22}\) and heterostructures\(^{18,23,24}\) based on high piezoelectric materials. These oscillations have been identified to originate from coherent acoustic phonons generated by photoexcited carriers within strained epilayers. The carriers screen the PZE field\(^{12,25}\) and subsequently induce a modulation of the refraction index\(^{18,22}\).

Bulk InAs and GaAs have a low piezoelectric constant, however, it has been shown by many groups that self-assembled QDs do induce pronounced piezoelectric fields as a result of the high strain and composition gradients. From excitation density dependent PL measurements, we clearly observe a blueshift of the spectrum with increasing density\(^{13,20}\). Hence, the blueshift reveals the strong PZE field within the QD clusters. Although coherent acoustic phonon excitation in QDs has not been reported before, the strain-engineered QD clusters are good candidates to generate acoustic phonons coherently due to enhanced local strain, related to the high QD density within the clusters as compared to random QD layers. At 5 K the carrier capture time of 19.6 ps is relatively long, such that carrier-induced coherent acoustic phonon generation is largely smeared out. To overcome this problem, we increase the measurement temperature to room temperature where the carrier capture time is significantly reduced.

Figure 2 depicts TRDR signals for various pump excitation densities obtained at 293 K. Pronounced oscillations are observed independent of the excitation density. The residue of the fit for a pump excitation of 28 W/cm\(^2\) clearly reveals the oscillations as depicted in the lower panel of Fig. 2. Using FFT on the oscillating part of the TRDR signals for different excitation densities, the oscillation frequency is determined, as summarized in Fig. 3(a). From the FFT spectra we deduce two frequencies with values of 16.5 GHz \((\tau_{\text{osc}}=112\) ps\) and 28.3 GHz \((\tau_{\text{osc}}=35.3\) ps\) independent of the pump excitation power. For high pump excitation densities a third frequency with a value of 8.95 GHz \((\tau_{\text{osc}}=112\) ps\) is observed, similar to that observed in the high excitation TRDR signal measured at 5 K.

As is reported by Wang \textit{et al.}\(^{23}\), the period of reflection oscillations in TRDR signals due to coherent acoustic phonons in the structure depends on the probe wavelength, and can be expressed by

\[
\tau_{\text{osc}} = \frac{\lambda}{2V_r n(\lambda)}.
\]

Here, \(V_r\) is the velocity of the acoustic mode and \(n(\lambda)\) is the index of refraction at the probe wavelength, \(\lambda\). Using Eq. (1), the sound velocities belonging to the three oscillation frequencies, as deduced from Fig. 2, are calculated. We obtain 5.02, 2.93, and 1.59×10\(^5\) cm/s for frequencies of 28.3, 16.5, and 8.95 GHz, respectively. The first two velocities are in good agreement with the QL and the QT acoustic phonon phase velocity in GaAs propagating along the [311] crystal direction, with velocities of 5.1 and 2.91×10\(^5\) cm/s, resep-
tively, as derived from the Christoffel equation. The third frequency has no direct analogy with acoustic phonons propagating in the [311] crystal direction. Also, this mode is only observed for the high excitation density. The third phonon mode, which is the pure transverse mode, has a phase velocity of $3.2 \times 10^5$ cm/s and is twice the velocity as determined from our measurements. However, the pure transverse mode is polarized along the $[\bar{0}1\bar{1}]$, and is not detectable in the used experimental configuration.

The initial and dominant changes of the QD reflectivity are due to the carrier capture in the QD ground state.15 However, the refractive index can also be changed by a modulated local strain field. That is, the QD reflectivity changes due to a locally higher QD density compared to random QD layers. Hence, the amplitude of the oscillation in the differential reflection signal scales linearly with the periodical change of the refractive index at the probe energy. To locate the origin of the coherent acoustic phonons, the amplitude of the oscillations is measured as a function of excitation density, as shown in Fig. 3(b). The amplitude of the oscillations in the TRDR signals due to the QL and QT acoustic phonons initially increases linearly with increasing pump intensity and saturates for high excitation density. The same effect is observed for the amplitude of the TRDR signal due to the carrier-induced QD bleaching [see Fig. 3(c)]. Dividing the oscillation amplitude by the total TRDR amplitude, the relative oscillation amplitude as a function of the pump excitation density is obtained [see Fig. 3(d)]. The relative amplitude decreases linearly with increasing pump intensity for both acoustic modes, with a mutual ratio of 1:9. The linear dependence of the oscillation amplitude with respect to the QD differential reflection amplitude, especially the agreement of the saturation point, indicates that the coherent phonons are generated within the strained QD clusters and not in the SL template. This result is supported by the PL measurements at 293 K, where the luminescence of the SL template is negligible with respect to the PL of the QD clusters, suggesting that the carriers are mainly captured in the QDs where the electron-hole pairs recombine.

Coherent acoustic phonon generation is confirmed by room temperature TRDR measurements performed on QD clusters with eight QDs on average,8 not shown here. These measurements reveal reflectivity oscillations with corresponding velocities of 4.98 and $2.87 \times 10^5$ cm/s, hence the generation of coherent QL and QT acoustic phonons, respectively. In addition, the oscillation amplitude is decreased with respect to the oscillation amplitude observed for clusters with four QDs on average, which suggests a reduced local PZE field as expected from the reduced local strain field for extended QD groups with random QD layers as the limiting case.

Finally, let us consider the physical mechanism responsible for the oscillations in the TRDR signal. As the pump-generated carriers are captured within the QDs, the electron-hole pairs will be separated by the strain-induced PZE field.13 Subsequently, the electron-hole pairs partly screen the PZE field.16,17,25 The screening of the PZE field locally induces instantaneous strain relief, hereby initiating coherent acoustic phonons. The resultant transient strain pulses,18,22,24 in turn, modulate the local PZE field in the QD clusters, and change the index of the refraction of the QDs through the quantum-confined Stark effect.20,21 The modulation of the optical properties, i.e., the dielectric function of the QDs, is detected by the resonant probe pulses, in resonance with the QD transition energy, which is far below the GaAs band gap and the SL template transition energy excluding a related contribution to the oscillations in the TRDR signal by the GaAs barriers and the SL template.

In summary, we have observed coherent acoustic phonons, the QL and QT acoustic phonon modes, in the TRDR signal of strain-engineered QD clusters grown on (311)B GaAs. The coherent phonon excitation is due to carrier-induced modulation of the locally strain-induced PZE field in the QD clusters, and causes optical oscillations in the strain-engineered QD clusters described by the quantum-confined Stark effect.

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