Polaron-induced lattice distortion of (In,Ga) As/GaAs quantum dots by optically excited carriers

S Tiemeyer\(^1\), M Bombeck\(^1\), H Göhring\(^1\), M Paulus\(^1\), C Sternemann\(^1\), J Nase\(^1\), F J Wirkert\(^1\), J Möller\(^1\), T Büning\(^1\), O H Seeck\(^2\), D Reuter\(^3,4\), A D Wieck\(^4\), M Bayer\(^1\) and M Tolan\(^1\)

1 Fakultät Physik / DELTA, Technische Universität Dortmund, D-44221 Dortmund, Germany
2 Deutsches Elektronen-Synchrotron DESY, D-22607 Hamburg, Germany
3 Optoelektronische Materialien & Bauelemente, Universität Paderborn, D-33098 Paderborn, Germany
4 Angewandte Festkörperphysik, Ruhr-Universität Bochum, D-44780 Bochum, Germany

E-mail: holger.goehring@tu-dortmund.de

Received 6 May 2016, revised 12 August 2016
Accepted for publication 19 August 2016
Published 13 September 2016

Abstract
We report on a high resolution x-ray diffraction study unveiling the effect of carriers optically injected into (In,Ga)As quantum dots on the surrounding GaAs crystal matrix. We find a tetragonal lattice expansion with enhanced elongation along the [001] crystal axis that is superimposed on an isotropic lattice extension. The isotropic contribution arises from excitation induced lattice heating as confirmed by temperature dependent reference studies. The tetragonal expansion on the femtometer scale is tentatively attributed to polaron formation by carriers trapped in the quantum dots.

Keywords: quantum dots, polarons, semiconductor

(Some figures may appear in colour only in the online journal)

Self-assembled (In,Ga)As quantum dots (QDs) are crystalline inclusions on the scale of 10 nm that are embedded in a GaAs matrix. Carriers residing in QDs are three-dimensionally confined, giving rise to discrete energy levels, similar to atoms. The excellent optical quality of such systems has allowed studies on fundamental problems of light–matter-interaction and has paved also the way of QDs into applications as light emitters, ranging from single photon sources to high-power laser diodes \([1\text{–}4]\). An important related problem with multiple facets is the interaction of QD carriers with phonons, which facilitates carrier relaxation into the ground states after non-resonant excitation \([5]\) or mediates also coupling to the optical modes of a resonator \([6]\). On the other hand, this interaction sets limitations to the coherence of confined charge \([7]\) and spin \([8]\) excitations. Despite of their high relevance, carrier–phonon interactions in quantum dots are still not understood in full detail. Theoretically, over the years more and more elaborated models have been developed to account for experimental observations, for an overview see \([5]\). The models range from weak coupling pictures based on (modified) Fermi’s golden rule, involving single- and two-phonon emission events \([9\text{–}12]\), to strong coupling descriptions leading to polaron formation and involving quantum kinetic effects \([13\text{–}15]\). Only the latter model has allowed for a comprehensive description of the entirety of experimental results, concerning carrier relaxations dynamics. Typically, the carrier–phonon interaction has been assessed through intra- or interband optical transitions in QDs \([16\text{–}18]\). For example, nonlinear time-resolved methods like four-wave mixing revealed a drop of coherent exciton polarization on a few ps time scale \([7]\). Through the temperature dependence of this drop, it could be uniquely related to the interaction with acoustic phonons. In the spectral domain, this results in spectral wings on both sides of the zero-phonon exciton spectral line \([7, 19]\). However, the impact of the coupling onto the lattice in form of a possible distortion \([19]\) has not been directly and quantitatively assessed so far in experiment. For optical excitation resonant with the ground state transition the following dynamics were predicted theoretically \([20]\). A quasi-stable polaron, which is a bound state of the injected...
I exhibits a much higher intensity than the difference carrier relaxation towards the ground state by phonon emission, eventually contributes to heating. For non-resonant excitation, carrier relaxation leads to additional heating [21, 22].

(In,Ga)As QDs result from heteroepitaxial, strain driven Stranski–Krastanov growth [23, 24]. The QDs are coupled to the surrounding GaAs matrix, thereby elastically deforming the lattice. Consequently, carrier–phonon interactions inside the QDs translate also into the GaAs crystal lattice. Here we study the GaAs distortion due to optically excited QDs carriers by high-resolution x-ray diffraction (XRD) [25], from which we obtain direct evidence for the polaron-induced lattice expansion. While optically induced changes of bulk systems were considered already by x-ray analysis [26], this has not been achieved so far for nanostructures.

The XRD experiments were performed on an (In,Ga)As/ GaAs QD multilayer structure grown on a (001) oriented GaAs substrate of the size 5 × 5 mm². After deposition of a 500 nm thick GaAs buffer layer, InAs corresponding to a nominal thickness of 1.9 monolayers was deposited at a substrate temperature of 525 °C, resulting in the formation of the wetting layer and the dots. The (001) direction corresponds to the growth axis of the QDs. The QD layer was capped by a 30 nm GaAs spacer. This sequence was identically repeated five times. The last of these layers was capped with 50 nm GaAs. The dot density is about 10¹⁰ cm⁻². The QD size is approximately 8 nm and 30 nm in height and diameter, respectively. The top part of figure 1 shows a sketch of the sample structure and a corresponding dark field cross-sectional transmission electron microscope (X-TEM) image. The X-TEM image displays the stacks of lens-shaped QDs distorting also the surrounding matrix, highlighting thereby the elastic coupling between dots and barriers.

The optical excitation was done by a diode-pumped, continuous wave, frequency-doubled Nd:YAG laser supply- ing radiation at λ₁ = 532 nm wavelength with 1 W maximum power. The laser power on the sample was adjusted by a variable attenuator. The diameter of the laser spot on the sample surface was enlarged to a 4 × 4 mm² in order to obtain homogeneous illumination and therefore homogeneous temperature of the whole sample. Alternatively, a laser diode emitting at λ₂ = 808 nm wavelength was used for excitation with a maximum power of 0.6 W. Both lasers excite electron–hole pairs into the GaAs barriers, however, with distinctly different energies in excess of the band gap. Photoluminescence from the sample as result of the optical excitation was collected by a pair of achromatic lenses and analyzed by a USB spectrometer. The bottom part of figure 1 shows photoluminescence spectra recorded with the two different lasers at the same excitation power while performing structural analysis, resulting in comparable emission intensities. Thus, comparable excitation powers lead to similar carrier densities in the dots, so that any possible impact on the lattice after relaxation into their QD ground states is expected to be comparable. In the applicable power range, the intensity scales linearly with power, suggesting that the number of excited electron–hole pairs per dot remains below unity.

XRD-measurements were carried out at the beamlines BL9 [27] of the synchrotron radiation facilities DELTA (TU Dortmund) and P08 [28] at PETRA III (DESY) using x-ray beam sizes of 0.1 × 1 mm² and 0.05 × 0.2 mm², respectively. The setup developed for simultaneous optical excitation and XRD probing of the QD sample is shown in figure 2 (top). The sample was mounted onto the cold finger of a liquid helium continuous-flow cryostat and kept at a temperature of 100 K.

The left part of figure 2 (bottom) shows a reciprocal space map (RSM) in the vicinity of the GaAs(002) Bragg reflection for the non-excited QD sample, recorded at T = 100 K. The x-ray photon energy was 12.38 keV. At this energy, the penetration depth of the x-rays [29] is in the μm range, so that effects occurring within the InAs QD multilayer structure, located in the top 0.2 μm sheet of the sample, become accessible. The data are scaled to reciprocal lattice units with respect to the GaAs cubic lattice constant of a₀ = 0.565 nm at T = 300 K. Along the L-direction in the RSM, the GaAs(002) Bragg reflection is accompanied by superlattice peaks originating from the layered structure of the QD sample. An analysis of these features by simulations utilizing the kinematical approximation connects the superlattice peaks to the periodic wetting layer-spacing layer system of the sample. However, as shown by the simulations, the QDs do not contribute significantly to the x-ray scattering pattern and can not be clearly resolved by the XRD measurements, in particular because the dot inhomogeneities lead to a strong broadening. On the other hand, changes by carriers trapped in the QDs affect also the surrounding matrix, so that insight into a possible carrier-induced lattice distortion may be taken from a GaAs reflection which is also inhomoge- neously broadened. The middle part of figure 2 gives the difference of two RSMs, one recorded for the excited (I_e) and one for the non-excited (I_n) QD sample within a single scan. To classify the reliability of our measurements, the experiment was repeated but without optical excitation, as shown on the right side of figure 2. The difference scattering pattern I_e – I_n exhibits a much higher intensity than the difference pattern I_ne – I_n, which indicates a peak shift, whereas the remaining intensity variation for I_ne – I_n results from statistical fluctuations of the measurement in general and, therefore, is a measure for the systematical errors of the experiment. A typical peak shift observed for a QD sample in the excited and non-excited state is shown in figure 3 (top left). We would like to note that the width of the Bragg peak

5 A similar power level corresponds to a larger photon flux for the red illumination, while on the other hand the absorption coefficient is smaller than for the green laser. The net effect of these factors is a similar number of electron–hole pairs in the QDs for both excitation wavelengths, as evidenced from the photoluminescence spectra with comparable intensity in figure 1. Therefore, we expect comparable polaronic effects.

6 A laser shutter system was added to the set-up, triggered by the beamline control. Thereby we could record diffraction curves of the sample, both optically excited and non-excited, within a single XRD scan by measuring each data point twice, once with opened and once with closed laser shutter.
hardly changes indicating no significant increase of the inhomogenity of the strain distribution by laser excitation.

The shift of the GaAs\((002)\) Bragg reflection corresponds to an increase of the lattice constant of the GaAs matrix along the heterostructure growth direction. To elucidate the origin of this change, we studied the thermal expansion of the QD sample and, serving as a reference, of a \((001)\) oriented bulk GaAs sample of comparable dimensions. We estimated possible laser excitation induced lattice heating effects by monitoring the energy of GaAs-related emission lines, namely of the band gap as well as defect-related transitions. Within the experimental accuracy of about 0.1 meV we did not resolve a shift of these lines, limiting possible crystal temperature increase to well below 10 K over the whole applied excitation power range. The thermal expansion of both samples was determined by measuring the change of lattice constant \(a\) along the \([001]\) crystal direction from the GaAs\((002)\) and GaAs\((004)\) Bragg reflections in the temperature range between 100 and 125 K in the vicinity of the nominal sample temperature, covering a much larger temperature range than relevant for the studies with optical excitation. The change of the perpendicular lattice constant \(D_{aa}\) normalized by the measured 100 K lattice constant is shown in figure 3 (bottom). Each data point represents the average of four measurements taken at every temperature. We performed a linear regression to the data in order to evaluate differences in the thermal expansion of the two samples (see black line with slope of \((1.26 \pm 1.4) \times 10^{-5} \text{ K}^{-1}\) for QD \([001]\) and red line with a slope of \((1.13 \pm 1.4) \times 10^{-5} \text{ K}^{-1}\) for GaAs \([001]\)) and found, that within the experimental accuracy the thermal lattice expansion is hardly modified by the inclusion of the QDs. The change in slope of the linear fits for GaAs and QD sample is estimated to be \(1.3 \times 10^{-6} \text{ K}^{-1}\) and relates to a lattice constant difference between GaAs and QD sample in the order of \(1 \times 10^{-5}\). Furthermore, the change of the lattice constant due to thermal expansion for \(\Delta T = 10 \text{ K}\) is found to be in the order of \(1 \times 10^{-4}\).

Next we investigated the lattice expansion of the optically excited samples along the \([001]\) and \([100]\) crystal directions as function of laser power density \(P\), where we first focus on the 532 nm illumination. For that purpose high-resolution XRD measurements of the GaAs\((002)\) and GaAs\((200)\) Bragg reflections were analyzed, also at \(T = 100 \text{ K}\). The measurement of the GaAs\((200)\) reflection was performed under a grazing incidence angle of \(\alpha_i = 0.5^\circ\). The relative change of lattice constant, \(\Delta a/a_0\) as a function of \(P\), was obtained from determining the laser-induced shift of the corresponding Bragg reflection. Each \(L\)-value of a Bragg scan was recorded subsequently at closed and opened laser shutter. The \(I_\lambda\)-curves show a displacement to smaller \(L\)-values by \(\Delta L\). Utilizing the method of least squares, \(\Delta L\) was determined by shifting the diffraction curves against each other and minimizing the absolute value of their difference. The relative change \(\Delta a/a_0\) of the GaAs cubic lattice constant then was calculated from \(\Delta L\) according to \(\Delta a/a_0 = \Delta L(P)/L(0)\).
The results of this analysis are shown in figure 3 (right). Note that the excitation power densities are corrected for the reflectivities in the setup. For the GaAs sample, identical linear dependencies of the relative lattice expansion on laser excitation power density are found for the [001] and [100] crystal directions. The solid line in figure 3 (right, top and bottom) shows the corresponding fit to the data with a slope of $(4.53 \pm 1.02) \times 10^{-4}$ cm$^2$ W$^{-1}$. Hence, the laser irradiation of the GaAs reference causes an isotropic lattice expansion, as for thermal heating. In contrast, the QD sample is characterized by a strongly anisotropic lattice expansion, very different from the expected isotropy of a purely thermal effect: while the excitation-induced expansion along the [100] direction (dashed-dotted line in figure 3 bottom right with a slope of $(6.35 \pm 1.54) \times 10^{-4}$ cm$^2$ W$^{-1}$) is similar to that in the GaAs sample maybe slightly steeper as in the purely thermal studies, it is considerably enhanced along the [001] direction (dashed line). This is clearly indicated by a slope of $(11.37 \pm 0.46) \times 10^{-4}$ cm$^2$ W$^{-1}$ which significantly differs from the slopes mentioned above. The crystal therefore undergoes a tetragonal lattice distortion.

The data indicate that the in-plane effect occurs mostly from thermal lattice expansion. Comparing the change of $\Delta a/a_S$ with $P$ ($\sim 0.5 \times 10^{-5}$) and $T$ ($\sim 1 \times 10^{-4}$, $\Delta T = 10$ K) we estimate the laser induced lattice heating to be about 1 K and therefore conclude that the enhanced expansion along the [001] direction is highly unlikely to be assigned to a sole thermal effect. This is further corroborated by power dependent measurements performed with the red laser emitting at 808 nm wavelength, corresponding to a photon energy of $E = 1.53$ eV. The mechanism of lattice heating is quite different then, because the carriers are excited only about one optical phonon energy ($36.7$ meV in GaAs) above the GaAs band gap ($E_{g,GaAs} = 1.50$ eV at $T = 100$ K), while for the green laser with $E = 2.33$ eV photon energy the excess energy of 800 meV corresponds to more than 20 optical phonons.

Figure 3 shows the Raman Scattering Spectroscopy (Raman) measurements performed on the QD and GaAs samples. The top graph shows the Raman spectrum for the QD sample, while the bottom graph shows the Raman spectrum for the GaAs sample. The x-axis represents the wavenumber in cm$^{-1}$, and the y-axis represents the intensity in arbitrary units. The peaks at around 270 cm$^{-1}$ and 300 cm$^{-1}$ correspond to the GaAs and the QD, respectively. The difference between the two samples is clearly visible in the spectrum. The graph also shows the effect of laser excitation on the lattice expansion of the GaAs and QD samples. The solid line represents the fit to the data with a slope of $(4.53 \pm 1.02) \times 10^{-4}$ cm$^2$ W$^{-1}$ for the GaAs sample, while the dashed line represents the fit with a slope of $(6.35 \pm 1.54) \times 10^{-4}$ cm$^2$ W$^{-1}$ for the QD sample. These slopes indicate the lattice expansion caused by laser irradiation.
Once these energies have been released, carriers are trapped in the wetting layer or highly excited QD states. From there, further phonon emission has to occur to bridge the 0.4 eV energy required for relaxation into the dot ground states, where the photoluminescence shown in figure 1 is generated. For red light excitation, crystal heating therefore arises mostly from the QDs, while for green illumination the heating occurs in the whole crystal. Still we find for red light excitation that the lattice expansion along the [001] direction is larger in the QD sample than for the GaAs reference. In combination with the comparable PL intensity for similar excitation powers, we can conclude that certain carrier occupations, independent of the excitation conditions, lead to comparable lattice distortions. Hence, the tetragonal QD lattice distortion is observed for both laser excitation wavelengths which supports that it has an origin different from heating (see footnote 5).

We therefore suggest that the anisotropic contribution to the lattice distortion is mostly induced by the optically injected carriers after relaxation into their ground states. This relaxation occurs on a timescale of a few to a few 10 ps, depending on the excitation power [5]. After about 0.5 ns electrons and holes recombine radiatively, giving rise to the photoluminescence in figure 1. However, the continuous wave excitation maintains on average a steady carrier population in the quantum dot ensemble. The electron–hole pairs couple strongly to the lattice, leading to polaron formation [20].

This suggestion has to be tested regarding compatibility with the experimental findings. Most importantly, there is the
anisotropy of the tetragonal lattice distortion. This anisotropy is in accordance with the expectation from the distribution of the electron and hole wave functions in the QDs. From former studies, it is established that there is a mismatch of the corresponding charge distributions with the electron wave function located below that of the hole along the vertical growth direction of (In,Ga)As self-assembled quantum dots [30]. This implies that electron–hole pairs resemble electric dipoles oriented along the [001] crystal direction. The dipole orientation obviously facilitates and amplifies the polaron formation along this direction. Next, there is the excitation power dependence. The QD population by electron–hole pairs in the ensemble is stochastic so that we monitor an average distortion of the lattice by the polaronic effects. As discussed above, the PL data suggest that similar red and green excitation powers lead to comparable PL intensities, corresponding to comparable numbers of confined carriers. This explains the similar lattice distortion on excitation power for the two different laser wavelengths. With increasing excitation power, more QDs become populated by carriers and contribute to the lattice distortion along the [001] direction leading to an expansion increasing linearly with excitation power, similar to the increase of photoluminescence emission intensity. As function of excitation power it can be described by $\Delta a/\Delta S(P) = (0.5 \pm 0.12) \times 10^{-5} \cdot P$, where the laser excitation power $P$ is measured in W cm$^{-2}$. From this average distortion one may estimate the optically induced deformation at the QD layers. For simplicity, we assume this distortion to be homogeneous in the different layers. An important boundary condition is that we do see a shift of the x-ray reflections but no significant change in peak intensity and width: this limits the local strain differences to about 10$^{-4}$. Together with the average lattice expansion on the order of 2 fm per W cm$^{-2}$ and the exponential x-ray penetration profile into the sample with an absorption depth in the $\mu$m range, we estimate an overall distortion of the lattice constant of 5 fm per W cm$^{-2}$ along the [001] direction at the QD layers. About 60% of this distortion would arise from the polaronic effects. This reasoning is qualitatively further corroborated by considering the expected impact of pure lattice heating on the crystal. The thermal lattice expansion is assumed to occur homogeneously which makes a tetragonal distortion unlikely.

In summary, we found evidence for an optically induced tetragonal distortion of the GaAs crystal lattice into which (In, Ga)As QDs are embedded. The anisotropy of the distortion arises from polaronic effects initiated by QD confined carriers after their optical excitation. We believe that these are proof-of-principle studies and trigger further x-ray studies of ‘condensed matter systems in operation’. So far, condensed matter has been valuable studied by x-rays in passive mode, i.e. without excitation, to understand their structure on an atomistic scale. Many further developments in the analysis of active devices can be foreseen such as higher spatial resolution, temporal resolution where, for example, the polaron formation dynamics in QDs is monitored, wetting layer excitation and nonlinear optical pumping.

Acknowledgments

We acknowledge the machine groups of PETRA III and DELTA for providing synchrotron radiation. We also acknowledge support by DFG through the SFB TRR 160 and by the BMBF through grant no. 05K12PE1. ST and FJW thank the NRW Forschungsschule ‘Forschung mit Synchrotronstrahlung in den Nano- und Biowissenschaften’ for financial support. JM (grant no. 05K10PEC) and TB (FSP-302 with grant no. 05K13PE2) acknowledge financial support from BMBF. ADW, MT (Pr-2013-0001) and TB (AN-2014-0036) thank the MERCU foundation.

References

[1] Bimberg D, Grundmann M and Ledentsov N N 1998 Quantum Dot Heterostructures (New York: Wiley)
[2] Michler P (ed) 2004 Single quantum dots: fundamentals, applications and new concepts Topics in Applied Physics (Heidelberg: Springer)
[3] Wang Z M (ed) 2012 Quantum dot devices Lecture Notes in Nanoscale Science and Technology (Heidelberg: Springer)
[4] Henneberger F and Benson O (ed) 2009 Semiconductor Quantum Bits (Singapore: Pan Stanford Publishing)
[5] Steinhoff A, Kurtze H, Gartner P, Florian M, Reuter D, Wieck A D, Bayer M and Jahnke F 2013 Phys. Rev. B 88 205309
[6] Ates S, Ulrich S M, Ulhaq A, Reitzenstein S, Löffler A, Höflling S, Forchel A and Michler P 2009 Nat. Photon. 3 724
[7] Borri P, Langbein W, Schneider S, Woggon U, Sellin R L, Ouyang D and Bimberg D 2001 Phys. Rev. Lett. 87 157401
[8] Hernandez F G G, Greilich A, Brito F, Wiemann M, Yakovlev D R, Reuter D, Wieck A D and Bayer M 2008 Phys. Rev. B 78 041303(R)
[9] Bockelmann U and Bastard G 1990 Phys. Rev. B 42 8947
[10] Inoshita T and Sakaki H 1992 Phys. Rev. B 46 7260
[11] Vurgaftman I, Lam Y and Singh J 1994 Phys. Rev. B 50 14309
[12] Ferreira R and Bastard G 1999 Appl. Phys. Lett. 74 2818
[13] Inoshita T and Sakaki H 1997 Phys. Rev. B 56 4355
[14] Kral K and Khas Z 1998 Phys. Rev. B 57 2061
[15] Verzelen O, Ferreira R, Bastard G, Inoshita T and Sakaki H 2002 Phys. Status Solidi A 190 213
[16] Hameau S, Guldner Y, Verzelen O, Ferreira R, Bastard G, Zeman J, Lemaitre A and Gérard J M 1999 Phys. Rev. Lett. 83 4152
[17] Kurtze H, Seebeck J, Gartner P, Yakovlev D R, Reuter D, Wieck A D, Bayer M and Jahnke F 2009 Phys. Rev. B 80 235319
[18] Kurtze H and Bayer M 2016 Appl. Phys. Lett. 109 012103
[19] Fan X D, Takagahara T, Cunningham J E and Wang H L 1998 Solid State Commun. 108 857
[20] Yagov A, Axt V M and Kuhn T 2002 Phys. Rev. B 66 165312
[21] Hawker P, Kent A J and Henini M 1999 Appl. Phys. Lett. 75 3832
[22] Bellingham R, Kent A J, Akimov A V and Henini M 2001 Phys. Status Solidi B 224 659
[23] Goldstein L, Glas F, Marzin J Y, Charasse M N and Le Roux G 1985 Appl. Phys. Lett. 47 1099
[24] Tersoff J, Teichert C and Lagally M G 1996 Phys. Rev. Lett. 76 1675
[25] Köhler R, Neumann W, Schmidbauer M, Hanke M, Grigoriev D, Schäfer P, Kirmse H, Häusler I and Schneider R 2008 Structural characterization of quantum dots by x-ray diffraction and TEM Semiconductor Nanostructures (Heidelberg: Springer) pp 97–121

[26] See, for example, Zamponi F, Rothhardt P, Stingl J, Woerner M and Elsaesser T 2012 Proc. Natl Acad. Sci. 109 5207

Korff Schmising C V, Bargheer M, Kiel M, Zhavoronkov N, Woerner M, Elsaesser T, Vrejoiu I, Hesse D and Alexe M 2007 Phys. Rev. Lett. 98 257601

[27] Krywka C, Paulus M, Steremmann C, Volmer M, Remhof A, Nowak G, Nefedov A, Pöter B, Spiegel M and Tolan M 2006 J. Synchrotron Radiat. 13 8

[28] Seeck O H et al 2012 J. Synchrotron Radiat. 19 30

[29] Als-Nielsen J and McMorrow D 2011 Elements of Modern X-ray Physics (New York: Wiley)

[30] Fry P W et al 2000 Phys. Rev. Lett. 84 733