Optical imaging of resonant electrical carrier injection into individual quantum dots

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We image the micro-electroluminescence (EL) spectra of self-assembled InAs quantum dots (QDs) embedded in the intrinsic region of a GaAs p-i-n diode and demonstrate optical detection of resonant carrier injection into a single QD. Resonant tunneling of electrons and holes into the QDs at bias voltages below the flat-band condition leads to sharp EL lines characteristic of individual QDs, accompanied by a spatial fragmentation of the surface EL emission into small and discrete light-emitting areas, each with its own spectral fingerprint and Stark shift. We explain this behavior in terms of Coulomb interaction effects and the selective excitation of a small number of QDs within the ensemble due to preferential resonant tunneling paths for carriers.

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Self-assembled InAs quantum dots (QDs) are an important model system for investigating the fundamental physics of quantum-confined electrons and holes. The selective light emission from a small number of QDs can be achieved, for example, by lithographically defined small-area diodes or apertures, or by the incorporation of QDs into microcavities and nanowires. Such studies have provided information about the electronic properties of the dots and form the basis for novel applications, e.g., optically driven sources of entangled photon pairs for quantum information processing. Of particular interest is the possibility of generating sharp EL emission lines from individual QDs by voltage controlled electrical injection of carriers. Despite many works on resonant tunneling injection of carriers in unipolar QD devices, the simultaneous resonant injection of both electrons and holes required for EL emission from an individual QD has received less attention and is relevant to topical research on electrically driven single QD photon emitters.

Here we use micro-electroluminescence (μEL) spectroscopy and imaging to investigate the optical emission from a single layer of self-assembled InAs quantum dots embedded in the intrinsic region of a p-i-n light-emitting diode. By gradually decreasing the applied bias below the ‘flat band’ threshold voltage at which the bias balances the built-in potential in the i-region, we follow the evolution of the EL spectra and the corresponding spatial form of the EL emission. We show that resonant tunneling of electrons and holes into the QDs at bias voltages below the flat band condition leads to sharp EL lines which are characteristic of individual QDs, accompanied by a spatial fragmentation of the diode emission into small and discrete light-emitting areas, each with its own spectral fingerprint and Stark shift. We explain this behavior in terms of the selective excitation of a small number of QDs within the ensemble due to the presence of preferential resonant tunneling paths for carriers. We also discuss the effect of QD charging and Coulomb interactions on the resonant excitation of the single QD EL emission. This demonstration of bias-controlled excitation of EL from an individual QD within an ensemble of dots could be developed further for use in optoelectronic or quantum information applications.

Our p-i-n diodes were grown by molecular beam epitaxy on a p+ GaAs substrate, which forms the bottom electrical contact of the diode. The layer structure in order of growth on the substrate is as follows: 200 nm and 50 nm p-doped GaAs layer with \( p = 4 \times 10^{18} \text{ cm}^{-3} \) and \( p = 5 \times 10^{17} \text{ cm}^{-3} \), respectively; a 6 nm intrinsic GaAs spacer layer; a 1.8 monolayer (ML) of InAs, which gives rise to a wetting layer (WL) and QDs with a density of about \( 10^{10} \text{ cm}^{-2} \). The QD layer is covered by 16 nm of intrinsic GaAs followed by 50 nm n-doped GaAs (\( n = 2 \times 10^{16} \text{ cm}^{-3} \)) and a 500 nm GaAs top layer with \( n = 4 \times 10^{18} \text{ cm}^{-3} \). The diodes are defined by wet-chemical etching and a ring-shaped gold electrode forms the top electrical contact and provides optical access. Here we focus on large area devices with 200 μm diameter, containing \( \sim 10^8 \) QDs. A schematic band diagram for a bias \( U \) below the flat band condition, i.e. for \( U < 1.5 \text{ V} \), is shown in the inset of Fig. 1a. The μEL spectra were recorded at \( T \approx 15 \text{ K} \) with a spectral resolution of \( \sim 40 \mu\text{eV} \) and a focal spot diameter of about 20 μm. The spatial maps of the μEL were recorded by scanning the focusing mirror along the mesa.

Figure 1(a) shows the EL spectra for a range of applied biases \( U \). As \( U \) is decreased below the flat band condition, the EL spectrum narrows and evolves into single sharp EL lines. To track the evolution of the EL spectrum with \( U \), we show in Fig. 1(b) a color-scale plot of the normalized EL intensity versus \( U \) and the photon energy, \( h\nu \). This reveals clearly the narrowing of the QD emission with decreasing bias and the emergence of two distinct EL features, labeled A and B and indicated in Fig. 1b by dashed lines. A and B both shift to lower en-
The maximum intensity of each spectrum as a function of emission. To demonstrate this effect, we recorded is accompanied by a spatial fragmentation of the EL phonon replica peak, which are reported elsewhere [19].

This method of EL excitation allows the study of such photoluminescence emission of individual InAs QDs [18].

The fragmentation of the EL spectrum into sharp lines consists of a single sharp line only, with a full width at half maximum of $\sim 150 \mu eV$. This linewidth is not determined by the resolution of the spectrometer and is similar to the typical linewidths reported for the low temperature photoluminescence emission of individual InAs QDs [18].

The method of EL excitation allows the study of such sharp emission lines over several orders of magnitudes in intensity and also reveals a characteristic exponential acoustic phonon broadening and a weak but sharp phonon replica peak, which are reported elsewhere [19].

The fragmentation of the EL spectrum into sharp lines is accompanied by a spatial fragmentation of the EL emission. To demonstrate this effect, we recorded $\mu$EL spectra at each position of a square grid covering roughly $1/4$ of the diode surface. Figure 2 shows spatial maps of the maximum intensity of each spectrum as a function of position for a series of bias voltages. For each image, a scale factor relates the maximum of the colorscale to the maximum at $U = 1.415 \text{ V}$.

At $U = 1.415 \text{ V}$ the emission is essentially homogeneous across the scan [20]. At a slightly lower bias, $U = 1.395 \text{ V}$, the diode emission starts to break up into a series of bright spots dominating over a homogeneous background emission. At $U = 1.385 \text{ V}$, the background intensity weakens and several bright spots emerge at distinct positions with a uniform spot size determined by the diameter of the focal spot of the collecting lens. At this voltage the EL spectra fragment into sharp lines. At $U = 1.372 \text{ V}$, the number of bright spots decreases and the relative intensities of the individual spots change compared to the image at $U = 1.385 \text{ V}$. The spectra now consist of individual emission lines with no background EL. At $U = 1.345 \text{ V}$, the maximum intensities are much lower and the number of visible emission spots is reduced to four. At $U = 1.315 \text{ V}$, only one bright spot is visible and the corresponding spectrum, shown in Fig. 1d, consists of a single sharp emission line with an intensity similar to the maximum at $U = 1.372 \text{ V}$. We note that no other EL lines could be observed at this bias, suggesting that this emission center originates from a single QD.

The EL spectra corresponding to the spatial EL maps are presented in Fig. 3 for $U = 1.372 \text{ V}$. Figure 3a shows maps of the normalized EL intensity as a function of position at specific photon energies. The individual scans are distinguished by different colors and the corresponding spectrum, shown in Fig. 1d, is labeled as P1-P7. Each
dependence to the quantum-confined Stark effect in the plotted for several lines L1-L5. We attribute this bias $U_{\text{bias}}$ voltages of $U = 1.372$ V.

In a scan are omitted for clarity. (b) EL spectra recorded for the maximum EL intensity at this energy. Features with intensities below the threshold of 10% of the maximum EL intensity of the emission line L1 of Fig. 1d at the energy $h\nu = 1.2730$ eV. The EL intensity exhibits sharp peaks at bias voltages of $U = 1.32$ V and $U = 1.37$ V.

The resonant tunneling gives rise to an increase in the average charge density in the QD layer, thus screening the local electric field, an effect analogous to the charge build-up effect reported previously for resonant tunneling quantum well diodes [23]. Also note that, although the Stark shifts for various EL lines differ from each other, the rates of shift with bias are very similar. This indicates that the different bias dependences arise from mesoscopic variations of the potential landscape rather than from differences in the electronic properties of the QDs.

Our data demonstrate that at low temperatures the QD EL below the flat band condition is excited by resonant tunneling injection of carriers. The electrons and holes in the doped n- and p-type layers adjacent to the i-region of the diode have an energy spread given by the respective Fermi energies ($< 10$ meV). Since both the electron and hole states have to be aligned with the Fermi seas to resonantly excite EL, one would expect considerably narrower bias resonances than the ones reported here. On the other hand, the Coulomb interaction of a charged QD with the Fermi seas of the contacts and with its nearest neighbor QDs can provide additional tunneling pathways, thus accounting for the relatively large widths ($> 10$ meV) of the resonances in Fig. 4a. We note that this broadening is significantly larger than the width of individual QD EL lines ($\sim 150$ μeV).

Though the number of active QDs is constrained by the conditions of resonant tunneling, this constraint is not sufficient to explain the pronounced spectral and spatial fragmentation of the EL emission revealed in our study. Existing theoretical models [16] do not predict such an effect either. The high density and uniform distribution of QDs ensures that even at the lowest bias several dots could be active. Hence we conclude that some QDs are coupled more strongly to the reservoirs than others.

The emission energy of individual EL lines depends on the applied bias, as can be seen in Fig. 4b, where the energy shift $\Delta E$ relative to the value at $U = 1.41$ V is plotted for several lines L1-L5. We attribute this bias dependence to the quantum-confined Stark effect in the QD [23, 24]. Of particular interest is that the energies of some of the lines remain constant over certain bias ranges. For example, the L1 line does not shift between $\sim 1.36$ V and $1.38$ V, suggesting that the electric field in the intrinsic region remains constant. We propose that resonant tunneling gives rise to an increase in the average charge density in the QD layer, thus screening the local electric field, an effect analogous to the charge build-up effect reported previously for resonant tunneling quantum well diodes [23]. Also note that, although the Stark shifts for various EL lines differ from each other, the rates of shift with bias are very similar. This indicates that the different bias dependences arise from mesoscopic variations of the potential landscape rather than from differences in the electronic properties of the QDs.
which is supported by the different number of observable resonances for different QDs shown in Fig. 4a. Since the emission energies of the investigated QDs are very similar, this variation of the coupling is likely to be due to local variations of the tunnel distance and barrier height. In previous studies, μEL maps of the emission from the ridge-waveguide regions of InGaN quantum well based LEDs have revealed spatial inhomogeneities due to non-uniform carrier injection caused by crystal degradation [26]. In our diodes, the EL spectra are stable with time, but preferential tunneling paths may arise from mesoscopic fluctuations of the n- and p-doped interfaces due to randomly placed dopant atoms in or close to the intrins region, crystals defects or strain-related potential minima associated with the QDs themselves. Such variations would not only explain the spatial and spectral fragmentation of the EL spectra, but could also account for some differences in the bias dependence of the Stark shifts among various QDs.

In summary, we have demonstrated how the homogeneous broad band emission of a large quantum dot ensemble fragments into spatially strongly inhomogeneous sharp emission lines from individual quantum dots. Each EL line exhibits a distinct resonance behavior as a function of the applied bias and a unique Stark shift. These effects can be explained in terms of the selective excitation of a small number of QDs within the ensemble due to the presence of preferential resonant tunneling paths for carriers. Our results provide direct evidence for the resonant and voltage tunable electrical injection of carriers into individual QDs and are relevant for future implementation of such structures into electrically controlled single photon LEDs. In particular, the resonant tunneling excitation of a single dot could reduce quantum decoherence due to interactions with carriers occupying adjacent dots or higher energy continuum states.

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[1] A.D. Yoffe, Advances in Physics 50, 1 (2001).
[2] D. Bimberg, Electron. Lett. 44, 3 (2008).
[3] N.N. Ledentsov, D. Bimberg, and Zh.I. Alferov, J. Lightwave Technol. 26, 1540 (2008).
[4] J.-Y. Marzin, J.-M. Gerard, A. Izrael, D. Barrier, and G. Bastard, Phys. Rev. Lett. 73, 716 (1994).
[5] Z. Yuan, B.E. Kardynal, R.M. Stevenson, A.J. Shields, C.J. Lobo, K. Cooper, N.S. Beattie, D.A. Ritchie, and M. Pepper, Science 295, 102 (2002).
[6] P. Michler, A. Kiraz, C. Becher, W.V. Schoenfeld, P.M. Petroff, L. Zhang, E. Hu, and A. Imamoglu, Science 290, 2282 (2000).
[7] M. Nomura, N. Kumagai, S. Iwamoto, Y. Ota, and Y. Arakawa, Nat. Phys. 6, 279 (2010).
[8] N. Panev, A.I. Persson, N. Sköld, and L. Samuelson, Appl. Phys. Lett. 83, 2238 (2003).
[9] J. Claudon, J. Bleuse, N.S. Malik, M. Bazin, P. Jaffrennou, N. Gregersen, C. Sauvan, P. Lalanne, and J.-M. Gérard, Nat. Photonics 4, 174 (2010).
[10] R.M. Stevenson, R.J. Young, P. Atkinson, K. Cooper, D.A. Ritchie, and A.J. Shields, Nature 439, 179 (2006).
[11] C.L. Salter, R.M. Stevenson, I. Farrer, C.A. Nicoll, D.A. Ritchie, and A.J. Shields, Nature 465, 594 (2010).
[12] M. Narihiro, G. Yusa, Y. Nakamura, T. Noda, and H. Sakaki, Appl. Phys. Lett. 70, 105 (1997).
[13] I. Hapke-Wurst, U. Zeits, H. Frahm, A.G.M. Jansen, R.J. Haug, and K. Fierz, Phys. Rev. B 62, 12621 (2000).
[14] A. Patanè, R.J.A. Hill, L. Eaves, P.C. Main, M. Henini, M.L. Zambrano, A. Levin, N. Mori, C. Hamauchi, Yu.V. Dubrovsiky, E.E. Vdovin, D.G. Austing, S. Tarucha, and G. Hill, Phys. Rev. B 65, 165308 (2002).
[15] D. Reuter, P. Kailuweit, A.D. Wieck, U. Zeits, O. Wibbelhoff, C. Meier, A. Lorke, and J.C. Maan, Phys. Rev. Lett. 94, 026808 (2005).
[16] G. Kiesslich, A. Wacker, E. Schöll, S.A. Vituspevich, A.E. Belyaev, S.V. Danyluyk, A. Förster, N. Klein, and M. Henini, Phys. Rev. B 68, 125331 (2003).
[17] L. Turyanska, A. Baumgartner, A. Chaggar, A. Patanè, L. Eaves, and M. Henini, Appl. Phys. Lett. 89, 092106 (2006).
[18] G. Oertner, D.R. Yakovlev, M. Bayer, S. Rudin, T.L. Reinecke, S. Fafard, Z. Wasilewski, and A. Forchel, Phys. Rev. B 70, 201301(R) (2004).
[19] E. Stock, A. Baumgartner, M. Dachner, T. Warming, A. Schliwa, A. Patanè, L. Eaves, M. Richter, A. Knorr, M. Henini, and D. Bimberg, Conference on Lasers and Electro-Optics, and Quantum Electronics and Laser Science Conference (CLEO/QELS 2009) 1-5, 2418 (2009).
[20] The large scale deviations in the image are mainly due to a misalignment of the focal plane of the lens and the sample surface.
[21] A small number of weak emission lines occur in more than one spectrum due to stray light from overlapping collection areas.
[22] A. Baumgartner, A. Chaggar, A. Patanè, L. Eaves, and M. Henini, Appl. Phys. Lett. 92, 091121 (2008).
[23] P.W. Fry, I.E. Itskevich, D.J. Mowr, M.S. Skolnick, J.J. Finley, J.A. Barker, E.P. O’Reilly, L.R. Wilson, I.A. Larkin, P.A. Maksym, M. Hopkinson, M. Al-Khafaji, J.P.R. David, A.G. Cullis, G. Hill, and J.C. Clark, Phys. Rev. Lett. 84, 733 (2000).
[24] A. Patanè, A. Levin, A. Polimeni, F. Schindler, P.C. Main, L. Eaves, and M. Henini, Appl. Phys. Lett. 77, 2979 (2000).
[25] M.L. Leadbeater, E.S. Alves, F.W. Sheard, L. Eaves, M. Henini, O.H. Hughes, and G.A. Toombs, J. Phys. Condens. Matter 1, 10605 (1989).
[26] M. Rossetti, T.M. Smeeton, W.-S. Tan, M. Kauer, S.E. Hooper, J. Heffernan, H. Xiu, and C.J. Humphreys, Appl. Phys. Lett. 92, 151110 (2008).