Intraband and interband photocurrent spectroscopy and induced dipole moments of InAs/GaAs(001) quantum dots in *n–i–n* photodetector structures

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We have investigated normal-incidence intra- and interband spectra of self-assembled steep InAs/GaAs(001) quantum dots (QDs) with an average height of ~8.0 nm and average base width of ~21 nm placed in $n-i$(QDs)$-n$ photodetector structures. The ground state occupation of the QDs in the $n-i$(QDs)$-n$ configuration is examined and used to assign observed intraband transitions. A photovoltaic effect in intraband photocurrent is observed and shown to arise from induced dipole moments. Stark shift in interband photocurrent spectroscopy reveals the presence and direction of interband transition induced dipoles, making this study the first to determine both intra- and interband dipoles in the same ensemble of QDs.
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Self-assembled semiconductor quantum dots[1] are a potential candidate for infrared (3–14 μm) photodetectors. Quantum dot infrared photodetectors (QDIPs) have intrinsic sensitivity to normally incident infrared light, longer lifetime of excited electrons due to suppressed electron-phonon scattering, and potentially significantly lower dark current.[2] Understanding the QD intraband transitions as well as transport of the photoexcited electrons (holes) is central to realizing the full potential of QDIPs for long-wavelength application. The experimental results on long-wavelength $In_xGa_{1-x}As/GaAs$ QDIPs with $n-n-n$[3–10] and $n-i-n$ configurations[11–14] show promise. In general, there are two types of intraband transitions in QDs, namely, bound-to-bound and bound-to-continuum. It is well known from studies of GaAs/AlGaAs quantum well infrared photodetectors that the highest long-wavelength infrared detectivity is related to bound-to-continuum (or quasicontinuum) intraband transitions.[15] This is so far unclear for the QDIPs. Assignment of intraband transitions has been hindered by the inhomogeneous QD size distribution as well as uncertain QD state occupation. For QDIPs, whether the initial state of intraband transition involves ground state or excited states remains unclear in most cases. Unlike in quantum wells, the state occupation in QDIPs, due to the much smaller real space filling factor of the QDs, could be significantly affected by highly doped contact layers as well as deep-level defects. Laser-induced infrared transmission spectroscopy is a useful method to study the QD intraband transitions.[16] However, it has the disadvantage that electron and hole intraband transitions, which are simultaneously detected, need to be distinguished. Combinations of $C-V$ spectroscopy with infrared transmission spectroscopy[17] or modulation reflection spectroscopy[18] are attractive ways to study QD intraband or interband transitions for different electron populations. Here, we focus on the larger and more uniformly size-distributed InAs/GaAs QDs formed using the innovative punctuated-island-growth procedure[19] which typically gives a very narrow photoluminescence (PL) linewidth of <25 meV for a single QD layer. By combining intraband with interband photocurrent (PC) spectroscopy, and utilizing PL and PL excitation (PLE) spectroscopy, we address the issue of state occupation in the QDIPs, and assign the nature of the intraband transitions observed. In addition, information on QD wave functions has been extracted from bias-dependent photoresponse behavior.

The $n-i-n$ QDIP samples were grown on GaAs(001) ±0.1° substrates via molecular-beam epitaxy. An undoped active InAs QD region is inserted between highly Si-doped top and bottom GaAs contact layers. It comprises a stack of five 3.0 monolayer (ML) steep InAs QDs (grown via the punctuated island growth approach as described in Ref. 19) and 150 ML GaAs barrier spacers. These QDs have an average height of ~8.0 nm and base of ~21 nm.[12,19] Additionally, $n-i-n$ QDIPs containing shallow InAs QDs, formed via continuous 2.0 ML InAs deposition, were also grown and studied. The shallow QDs have the same base width but a lower height of ~3.5 nm as compared to the steeper and larger QDs.[19] The InAs QDs have a density of about $6\times10^{10}\,\mathrm{cm}^{-2}$. Since the InAs QDs were capped at very low temperature (350 °C), the GaAs capping should not dramatically change the QD shape and size. Standard photolithography and wet chemical etching procedures were used for QDIP sample processing. The mesas have a diameter of 250 μm with alloyed AuGe/Ni/Au Ohmic contacts. Long-wavelength and near infrared photoresponse spectroscopy were performed under normal-incidence geometry. All photoresponse spectra were recorded using a current preamplifier and a rapid-scan Fourier transform spectrometer connected to a cryostat with ZnSe windows. The mesa bottom contact was grounded. The photoresponse spectra were calibrated with a pyroelectric detector. The near- and long-wavelength infrared excitation for FTIR PC measurements were $\sim10^{-3}$ and $\sim10^{-4}\,\mathrm{W/cm}^2$, respectively, corresponding to extremely low carrier occupancies of the QDs.

Figure 1 shows PL, PLE, and PC spectra of the QDIP

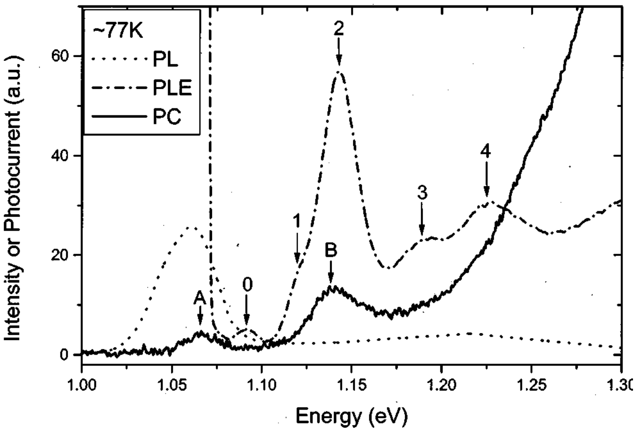

FIG. 1. Near infrared photoluminescence, photoluminescence excitation spectra (at 78 K), and interband photocurrent spectrum (at 77 K and at a bias of +0.58 V) for the steep InAs/GaAs quantum dots. The observed peaks are described in the text.

sample with the steep 3.0 ML QDs. The PL spectrum at 78 K shows a main peak at ~1.060 eV with a full-width at half-maximum (FWHM) of ~36 meV. It is larger than the typical values of <25 meV for single QD layers since the cumulative strain at the GaAs spacer surface affects InAs QD formation in the upper layers, thus leading to the broader QD size distribution. The PLE spectrum at 78 K recorded at a detection energy of 1.060 eV shows several peaks. Comparing with our previously reported PL and PLE data on the steep InAs/GaAs QDs,[19,20] we assign the main PL peak as the ground state interband transitions of InAs QDs. In the PLE spectrum, peak 0 at 1.092 eV is separated by 32 meV from the main PL peak. This is close to the QD LO-phonon energy[21] and the peak is thus identified as a LO-phonon replica. Peak 1 at 1.120 eV, peak 2 at 1.143 eV, peak 3 at 1.188 eV, and peak 4 at 1.227 eV are excited interband transitions of the QDs.[20] The interband PC spectrum at 77 K at a bias of +0.58 V (Fig. 1, solid line) shows two clear peaks and a broad background increasing with energy (within the measured range). Peak A at 1.065 eV can be assigned as the ground interband transition, which corresponds to the main PL peak. Peak B at 1.139 eV can be assigned as the excited interband transitions of the QDs, which corresponds to the PLE peak 2. A slight peak position shift between the PC peak A and the PL main peak (also between peak B and the PLE peak 2) is a manifestation of contribution from the Stark effect[22] and the charging effect on the InAs QD exciton,[23] spatial variation in QD size, and different probing mechanisms between PC and PL and PLE spectroscopies.

Typical normal-incidence long-wavelength infrared PC spectra of the steep QDs at 77 K at different biases are shown in Fig. 2. At a positive bias of +0.58 V, the intraband PC spectrum (top curve) shows a main peak at 174 meV (7.14 μm) with a FWHM of 35 meV (1.37 μm) and a small peak at 115 meV (10.8 μm). At a negative bias of −0.58 V, the intraband PC spectrum (middle curve) shows a main peak at 174 meV with a FWHM of 20 meV (0.79 μm) and a small peak at 110 meV (11.4 μm). Note that even at zero bias (bottom curve) PC is observed at 77 K and shows a peak at 172 meV (7.25 μm) with a FWHM of 29 meV (1.13 μm). This is thus a photovoltaic effect. All the peaks (at 110/115 and ~174 meV) correspond to QD intraband transitions. A slight variation in peak position (<3 meV) and FWHM of the intraband PC spectra between positive and negative bias can be due to differences in gain and QD electron occupation between positive and negative bias. Importantly, from the fact that at 77 K we observe both ground interband transition as well as intraband transition photoresponse at the same bias and temperature, we can conclude that the ground states of the steep InAs QDs in these QDIPs are, on the average, only partly occupied at 77 K (at least at a bias $\leqslant$+0.58 V). The observation of the intraband transitions indicates that the electrons present in undoped QDs are transferred and/or injected from the heavily Si-doped GaAs top and bottom contact layers at 77 K (since undoped epitaxical GaAs layer is *p* type). It is thus concluded that the intraband peaks observed in these structures are due to transitions between electron

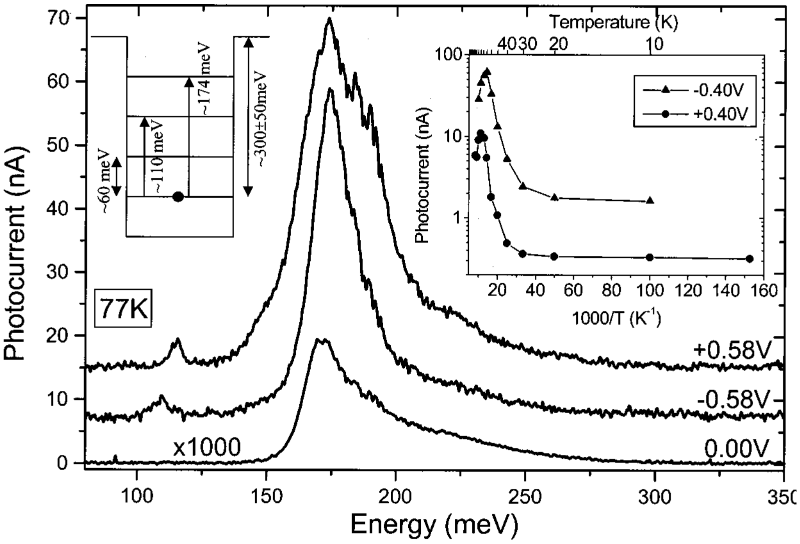

FIG. 2. 77 K intraband photoresponse of the steep InAs/GaAs quantum dots at a bias of +0.58 V (top curve), −0.58 V (middle curve), and 0.00 V (bottom curve). The zero of the upper and middle curves are shifted for clarity. The lower curve (at zero bias) is enlarged 1000 times. Inset (right): intraband peak photocurrent (at bias of ±0.40 V) versus reciprocal temperature. Inset (left): schematic of InAs/GaAs QD electron energy levels.

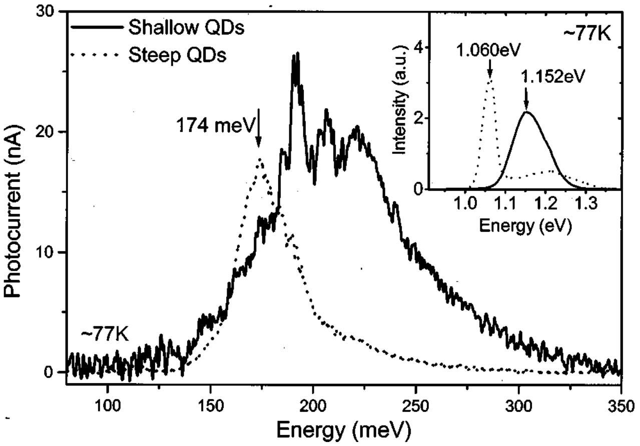

FIG. 3. 77 K intraband photoresponse of steep and shallow InAs/GaAs quantum dots at a bias of +0.41 V. Inset: 77 K photoluminescence spectra of the steep and shallow quantum dots.

ground states and excited states of the steep InAs QDs. In addition, the dominant intraband peak has the same peak position at ~174 meV at 77 K over the entire bias range from −0.80 to +1.00 V.

Our inference about the QD state occupation is further supported by the temperature dependence of the intraband PC at ~174 meV. We find that the long-wavelength photoresponse of the steep QDs does not disappear at zero bias from 7 to 100 K. The inset (right) of Fig. 2 shows the intensity of the main intraband PC peak (at 174 meV) as a function of reciprocal temperature. At positive and negative bias (+0.40 and −0.40 V), the intraband photoresponse shows similar temperature dependence. Below 20 K, the PC is nearly independent of the temperature. This suggests that electrons in the QDs can be contributed via tunneling from a contact (emitter) layer. Between 20 and 77 K, the temperature dependence of the intraband photoresponse corresponds to an activation energy of 25±15 meV. This suggests that the QD ground state energy is located slightly above the local chemical potential, consistent with the statement that the QD ground states are, on the average, partly occupied. At higher temperatures (>80 K), the intraband PC decreases with increasing temperature. This is likely due to a decrease in the photoexcited electron lifetime at higher temperature.

Our systematic temperature and excitation power dependent PL and PLE studies[20] suggest that in the steep QDs the electron ground state binding energy, i.e., the ground state to the GaAs conduction band edge separation, is 300±50 meV. This implies that the intraband photocurrent transitions at 110/115 and 174 meV correspond to bound-to-bound transitions. In order to confirm this bound-to-bound assignment of the transitions in the steep QDs, we compared their behavior to our study of the PL and FTIR intraband PC spectra (Fig. 3) of the shallow 2.0 ML InAs/GaAs QDs (with an average base width of about 21 nm and average height of about 3.5 nm). The PL and intraband transition of the shallow InAs QDs have main peaks centered at 1.152 eV and 206±15 meV, respectively. This indicates that the interband ground transitions and the main intraband transition of the shallow QDs are blueshifted by 92 and 32±15 meV, respectively, relative to the steep QDs. The observed interband blueshift is consistent with the well-established fact that large QDs have larger electron ground state binding energy than small QDs.[24] If the main intraband transitions of the steep and shallow QDs were bound-to-continuum, the main intraband transition of the shallow QDs should be redshifted relative to the steep QDs. This, however, is just the opposite of our above-mentioned observation. Therefore, it can be concluded that the observed intraband transitions of the steep QDs (main peak at 174 meV and weak peak at 110/115 meV) are bound-to-bound transitions (see left inset of Fig. 2). However, it is not clear whether the intraband peaks of the shallow 2.0 ML QD are also due to bound-to-bound transitions.

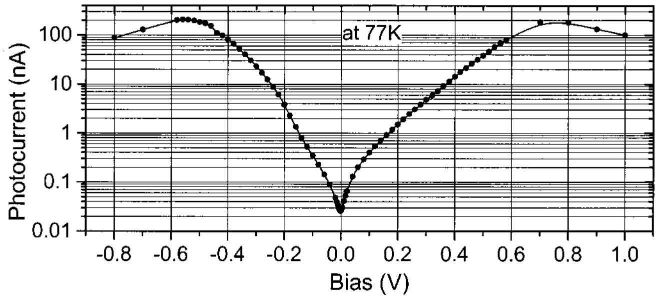

FIG. 4. Bias dependence of the photocurrent of the main intraband peak of steep InAs/GaAs quantum dots at 174 meV at 77 K.

Information on the intra- and interband dipole moments of the steep InAs/GaAs QDs is extracted from the bias dependence of intraband and interband PC peaks. It should be noted that all PC reported here is alternating current generated by modulated infrared light. Figure 4 shows bias dependence of the intraband PC at ~174 meV at 77 K. With increasing bias, the PC first rapidly increases by four orders of magnitude. Beyond a bias of −0.60 and +0.80 V, the PC decreases. This is a negative differential PC behavior, probably due to the influence of electron heating and QD charging upon electron capture process as very recently suggested by theoretical modeling.[25] More importantly, there is no compensation bias observed at which the intraband PC (alternating current) is zero. The absence of a compensation bias is not explicable in terms of an asymmetric built-in potential (diffusion potential) and, furthermore, rules out involvement of bound-to-continuum transitions. The observed intraband bound-to-bound transition related photovoltaic effect can be explained by the appearance of an intraband dipole moment. Namely, the shape and inhomogeneous strain-dependent intrinsically asymmetric potential of the grown pyramid-like QDs gives rise to electron wave function mass center misalignment (along growth direction) between the QD ground states and bound excited states.[26] Upon intraband transition under the modulated FTIR radiation, the wave function misalignment induces dipoles along the growth direction of the QDIP. A displacement between the initial and final centers of gravity will give an intraband dipole moment, $\mathbf{P}=(\epsilon_r-1)\epsilon_0\mathbf{F}$, where ϵ_r, ϵ_0, and $\mathbf{F}$ are the dielectric constant, free space permittivity, and electric field generated by the charge separation, respectively. This intraband dipole can induce a displacement PC. This inference of a displacement PC is also supported by the different dependence of the intraband PC on the FTIR modulation frequency at different bias (not shown). With increasing FTIR scan frequency, the intraband PC at zero bias rapidly increases, while the PC at a high bias (e.g., +0.341 V) remains, by and large, unchanged. We also determined the orientation of the intraband dipole using FTIR interferograms.[27] The intraband transition induced dipoles of the QDs at zero bias under modulated infrared radiation are nearly in phase with the PC at 77 K at high positive bias (e.g., +0.341 V). Therefore, the orientation of the intraband transition induced dipoles in the InAs/GaAs QDs at 77 K is towards the wetting layer from the QD apex. Interestingly, temperature dependent studies of the intraband spectra and interferograms, to be reported elsewhere, indicate that the direction of the 174 meV intraband transition induced dipole reverses as the temperature is decreased below ~40 K, suggesting the significance of ground state electron occupation and escape routes from the involved excited states.

In addition, we measured interband PC spectra of the steep InAs QDs at different bias. The bias dependence of the interband ground transition peak at ~1.027 eV at 200 K reveals a blueshift of 5 meV with varying bias from negative (−0.4 V) to positive (+0.4 V) bias. The interband Stark shift indicates that the center of mass of the QD electron ground state is closer to the wetting layer than that of the hole ground state of the steep InAs QDs. Namely, the orientation of the interband transition induced dipoles is towards the QD apex.

In summary, we have studied normal-incidence intraband and interband PC spectra of InAs/GaAs QDs. The observed PC peaks at ~110 and ~174 meV in the steep InAs QDs grown via the PIG approach, correspond to intraband transitions from the electron *ground* to *bound* excited states. The observed intraband photovoltaic effect can be explained by dipoles induced between the ground and excited electron states in InAs/GaAs QDs. In addition, we have determined, for the first time, the existence and orientation of intra- and interband transition induced dipoles in the same ensemble of InAs/GaAs(001) QDs.

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[1] See, e.g., D. Bimberg, M. Grundmann, and N. N. Ledentsov, *Quantum Dot Heterostructures* (Wiley, Chichester, 1998).
[2] V. Ryzhii, Semicond. Sci. Technol. **11**, 759 (1996); D. M.-T. Kuo, A. Fang, and Y. C. Chang, QWIP2000 Workshop, Dana Point, California, 2000.
[3] S. Maimon, E. Finkman, G. Bahir, S. E. Schacham, J. M. Garcia, and P. M. Petroff, Appl. Phys. Lett. **73**, 2003 (1998).
[4] D. Pan, E. Towe, and S. Kennerly, Appl. Phys. Lett. **75**, 2719 (1999), and references therein.
[5] S. J. Xu, S. J. Chua, T. Mei, X. C. Wang, X. H. Zhang, G. Karunasiri, W. J. Fan, C. H. Wang, J. Jiang, S. Wang, and X. G. Xie, Appl. Phys. Lett. **73**, 3153 (1998).
[6] Q. D. Zhuang, J. M. Li, H. X. Li, Y. P. Zeng, L. Pan, Y. H. Chen, M. Y. Kong, and L. Y. Lin, Appl. Phys. Lett. **73**, 3706 (1998).
[7] S.-W. Lee, K. Hirakawa, and Y. Shimada, Appl. Phys. Lett. **75**, 1428 (1999).
[8] L. Chu, A. Zrenner, G. Böhm, and G. Abstreiter, Appl. Phys. Lett. **75**, 3599 (1999).
[9] J. Phillips, P. Bhattacharya, S. W. Kennerly, D. W. Beekman, and M. Dutta, IEEE J. Quantum Electron. **35**, 936 (1999).
[10] Z. H. Chen, O. Baklenov, E. T. Kim, I. Mukhametzhanov, J. Tie, A. Madhukar, Z. Ye, and J. Campbell, J. Appl. Phys. **89**, 4558 (2001).
[11] Z. H. Chen, O. Baklenov, E. T. Kim, I. Mukhametzhanov, A. Madhukar, Z. Ye, F. Ma, B. Yang, and J. Campbell, Infrared Phys. Technol. **42**, 479 (2001).
[12] Z. Ye, J. C. Campbell, Z. H. Chen, E. T. Kim, and A. Madhukar, Mater. Res. Soc. Symp. Proc. **692**, H9.17.1 (2002).
[13] E. T. Kim, Z. H. Chen, and A. Madhukar, Appl. Phys. Lett. **79**, 3341 (2001).
[14] E. T. Kim, Z. H. Chen, M. Ho, and A. Madhukar, J. Vac. Sci. Technol. B, this proceeding.
[15] B. F. Levine, J. Appl. Phys. **74**, R1 (1993).
[16] S. Sauvage, P. Boucaud, F. H. Julien, J. M. Gérard, and J. Y. Marzin, J. Appl. Phys. **82**, 3396 (1997).
[17] H. Drexler, D. Leonard, W. Hansen, J. P. Kotthaus, and P. M. Petroff, Phys. Rev. Lett. **73**, 2252 (1994).
[18] T. M. Hsu, W. H. Chang, K. F. Tsai, J. I. Chyi, N. T. Yeh, and T. E. Nee, Phys. Rev. B **60**, R2189 (1999).
[19] I. Mukhametzhanov, Z. Wei, R. Heitz, and A. Madhukar, Appl. Phys. Lett. **75**, 85 (1999).
[20] I. Mukhametzhanov, Z. H. Chen, O. Baklenov, E. T. Kim, and A. Madhukar, Phys. Status Solidi B **224**, 697 (2001).
[21] R. Heitz, I. Mukhametzhanov, O. Stier, A. Madhukar, and D. Bimberg, Phys. Rev. Lett. **83**, 4654 (1999).
[22] P. W. Fry *et al.*, Phys. Rev. Lett. **84**, 733 (2000).
[23] R. J. Warburton, C. S. Dürr, K. Karrai, J. P. Kotthaus, G. Medeiros-Ribeiro, and P. M. Petroff, Phys. Rev. Lett. **79**, 5282 (1997).
[24] O. Stier, M. Grundmann, and D. Bimberg, Phys. Rev. B **59**, 5688 (1999); L. W. Wang, J. Kim, and A. Zunger, *ibid.* **59**, 5678 (1999); and references therein.
[25] V. Ryzhii, Appl. Phys. Lett. **78**, 3346 (2001).
[26] Following our observation of the permanent dipole moment and its suggested origin in the underlying asymmetry of the confining potential, theoretical confirmation has been provided by S. J. Sun and Y. C. Chang, Phys. Rev. B **62**, 13 631 (2001).
[27] Z. H. Chen, E. T. Kim, and A. Madhukar, Appl. Phys. Lett. **80**, 2770 (2002).