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Intraband-transition-induced dipoles in self-assembled InAs/GaAs(001) quantum dots

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We present experimental evidence for the existence and directionality of dipoles induced by *intraband* transitions from the electron ground states to *high, bound* excited states in self-assembled InAs/GaAs quantum dots (QDs). Moreover, the orientation of the interband transition induced dipoles is also determined for the same QDs. The findings indicate the potential use of intraband dipoles in asymmetric QDs in proposed quantum gates. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1468896]

Knowledge of electronic energy levels and the nature of the wave functions of self-assembled semiconductor quantum dots (QDs)[1] is central to uncovering the range of physical phenomena and their potential for quantum devices.[2] A variety of theoretical[3–5] and experimental[6,7] studies have established that the QD electron/hole state energies strongly depend on the QD size, shape, and composition, all of which depend upon growth conditions and procedures. However, few experimental results have been reported on the behavior of the carrier wave functions. Recently, information on the electron-hole wave function alignment has been extracted from the Stark shift of interband excitons in AlInAs/AlGaAs QDs embedded in $p-i$(QDs)-n structure.[8–9] In this letter we report on the existence and nature of *intraband* dipoles in pyramidal InAs/GaAs QDs. The observed intraband dipoles are between the QD electron ground states and *high, bound* excited states which are usually not revealed in photoluminescence (PL) and PL excitation spectra.[7]

The InAs/GaAs(001) QDs studied here are embedded in the intrinsic region of n-i(QDs)-n structures, designed as normal-incidence QD infrared photodetectors.[10] A detailed description of the samples and their characteristics was given in Ref. 10. The intrinsic region comprises a stack of five 3.0 monolayer (ML) InAs QDs with 150 ML GaAs barrier spacers. Atomic force microscopy and transmission electron microscopy measurements indicate that uncapped 3.0 ML InAs QDs have an average height of ~ 8.0 nm, average base length of ~ 21 nm, and a density of about 6×10^{10} cm^{-2}. The capped QDs are likely to have an effective height and base not the same as the uncapped islands but given our very low temperature ($<350\,°$C) capping such differences are minimized compared to conventional cap layer growth at $\sim 500\,°$C. In the n-i-n structures used here, the electron ground states of the QDs are only partially occupied[11] by electrons contributed and/or injected from the heavily doped contact layers. All photocurrent (photovoltage) spectra were recorded on 250 μm diameter mesas (with bottom contract grounded) under normal-incidence geometry using a current (voltage) preamplifier and a rapid-scan Fourier-transform infrared (FTIR) spectrometer. The FTIR scan frequency (f_0) corresponds to modulation frequency of a He–Ne laser (0.6328 μm) interference light. The modulation frequency (f) for a wavelength λ (μm) corresponding to an energy E (meV) is equal to $f_0(0.6328/\lambda)\cong f_0(E/1960)$. The photocurrent spectra were calibrated with a pyroelectric detector. The long-wavelength infrared excitation power densities were $\sim 10^{-4}$ W/cm^2 corresponding to extremely low carrier occupancies of the QDs.

Figures 1(a), 1(b), and 1(c) show typical intraband photocurrent spectra of the QDs at 77 K at a bias of 0.000, +0.341, and −0.340 V, respectively, at three scan frequencies each. Two intraband peaks, one at 110/115 meV and the other at 174 meV, are observed, the latter being much stronger. This may be due to a higher gain and the calculated higher available density of the final states[3] of the transition at 174 meV, which are not the first or even second excited bound states but rather very high bound states (most probably fifth or higher states).[3,7] The line shape of the main intraband photocurrent peak (at 174 meV) at different biases is similar. Note the nonvanishing photocurrent at zero bias [Fig. 1(a)] signifying the existence of QD intraband photovoltaic effect. We have earlier[10] suggested that this is due to

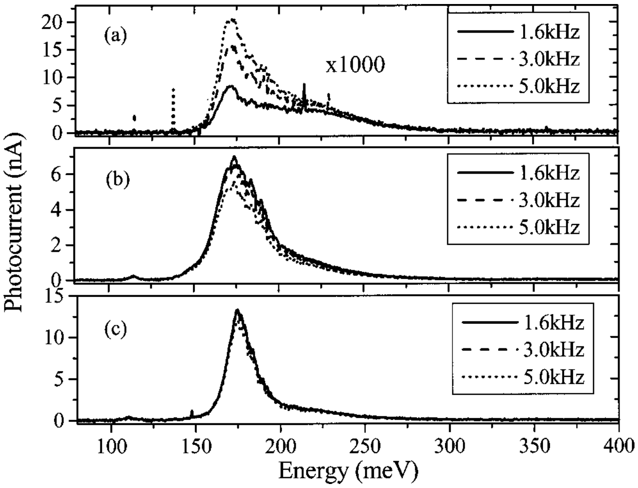

FIG. 1. Scan frequency dependence of photocurrent spectra of the InAs quantum dots at 77 K at a bias of 0.000 V (panel a), +0.341 V (panel b), and −0.340 V (panel c).

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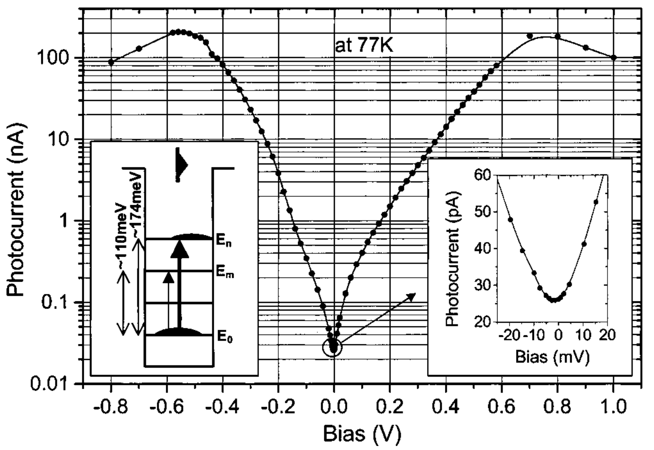

FIG. 2. Bias dependence of peak photocurrent (at scan frequency of 5 kHz) at 77 K. Right inset: enlarged data around zero bias. Left inset: schematic of the InAs quantum dot electron levels. Their wave function mass centers (at zero bias) are indicated.

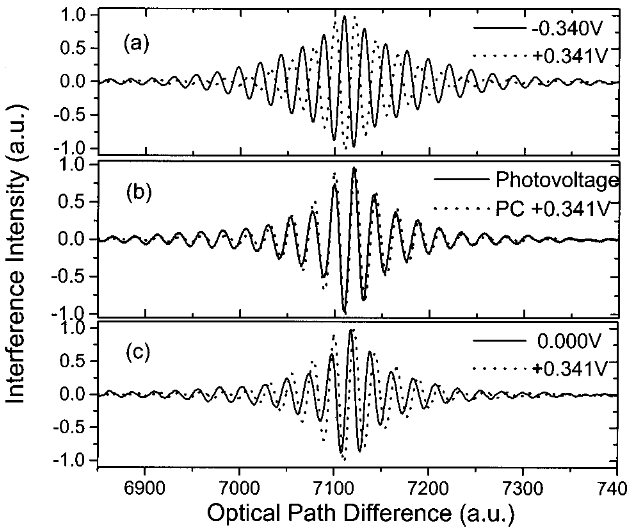

FIG. 3. Photocurrent interferograms at a bias of -0.340 vs $+0.341$ V (panel a) and 0.000 vs $+0.341$ V (panel c). Panel b: Photocurrent (at a bias of $+0.341$ V) and photovoltage interferograms. These interferograms correspond to Fourier transform infrared scan frequency of 5 kHz.

intraband dipoles. In the following we provide detailed experimental evidence for the existence and the nature of these intraband transition induced dipole moments.

To shed light on the nature of the intraband photovoltaic effect, we have examined intraband photocurrent spectra of the QDs as a function of bias from -0.800 to $+1.000$ V (roughly ~ 30 kV/cm) with very small bias increments of 1.2 meV. Figure 2 shows the bias dependence of the main intraband photocurrent peak at 174 meV at the scan frequency of 5 kHz. The inset in Fig. 2 shows a smooth dependence of this photocurrent peak on bias around zero bias. At high bias ($\sim \pm 0.7$ V) a negative differential photocurrent behavior is seen and may be due to the influence of electron heating and QD charging on the electron capture process as very recently suggested by theoretical modeling.[12] The near zero bias measurements indicate that, under modulated infrared radiation, there does not exist a compensation bias at which the photocurrent at ~ 174 meV peak is zero. This cannot be explained by a random bias fluctuation around zero as then the FTIR interferogram at zero bias would be distorted, but this is not found. The absence of compensation bias is also 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 transition related photovoltaic effect can, however, be explained by the relative spatial alignment of the charge center of gravity of the bound ground state and a bound nature of the excited state electron wave functions involved. A displacement between the initial and final centers of gravity will give an intraband dipole moment, $\mathbf{P} = (\varepsilon_r - 1)\varepsilon_0 \mathbf{F}$, where ε_r, ε_0, and $\mathbf{F}$ are the dielectric constant, free space permittivity, and electric field generated by the charge separation, respectively. From the viewpoint of current, the absence of a compensation bias under modulated infrared radiation implies that the photocurrent around the minimum consists of two components with similar amplitudes and a phase difference different from 180°. If the phase difference were exactly 180°, a compensation bias should be observed. The two alternating current components can arise from the photoexcited electrons in the *high* (~ 174 meV), *bound* excited states having (i) a certain life time which induces intraband dipoles or, (ii) being thermally excited and/or tunneling into the GaAs conduction band. The former generates a displacement photocurrent and the latter an extended photocurrent.

Returning to Fig. 1, the presence of a displacement photocurrent, or intraband dipole, is also supported by the observed dependence of the intraband photocurrent at zero bias on the FTIR modulation frequency. With increasing FTIR scan frequency (from 1.6 to 5.0 kHz), the intraband photocurrent at zero bias rapidly increases, while the photocurrent at a high bias of $+0.341$ and -0.340 V remains, by and large, unchanged. The different frequency dependence of photocurrent between zero bias and high bias (e.g., $+0.341$ and -0.341 V) indicates different RC time constants between zero and high bias, which is consistent with the explanation of the existence of a displacement current at zero bias. It is thus apparent that the displacement photocurrent component at the higher biases of $+0.341$ and -0.341 V becomes negligibly small compared to the extended photocurrent component due to greatly enhanced electron tunneling out of the excited states. In addition, the value of the intraband dipole moment could be slightly affected by the employed electric fields as small intraband peak shifts of <3 meV are observed.

We next address the question of the orientation of the intraband transition induced dipole. Figure 3 shows typical 77 K FTIR interferograms of the QDs at different bias. It is expected that the intraband photocurrent at negative/positive high bias, where displacement photocurrent is negligible, should have a phase difference of 180°. Indeed, our FTIR photocurrent interferograms [Fig. 3(a)] at a bias of $+0.341$ and -0.340 V exhibit a phase difference of $180 \pm 20°$. The interferogram signals far away from their peaks (at ~ 7120) have a phase difference deviated from the out-of-phase due to the difference in the linewidths of the photocurrent spectra shown in Fig. 1. Furthermore, we measured FTIR photovoltage (PV) interferogram [shown in Fig. 3(b)] with a voltage preamplifier. The intraband transition induced dipoles of the InAs QDs at zero bias under the modulated infrared radiation are nearly in phase with the photocurrent at high positive

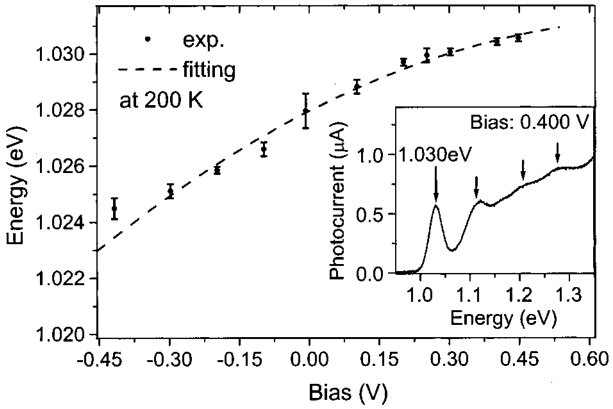

FIG. 4. Bias dependence of interband ground transition energy at 200 K. Inset: Interband photocurrent spectrum at a bias of +0.400 V at 200 K.

bias (e.g., +0.341 V). Therefore, the direction of the intraband transition induced dipoles in the InAs/GaAs QDs at 77 K and zero bias is towards the wetting layer from QD apex [namely, the electron center of mass of the ground states (E_0) of the InAs/GaAs QDs is closer to the wetting layer than that of the *high, bound* excited states (E_n), as schematically shown in the left inset of Fig. 2]. This dipole orientation at 77 K implies that the *high, bound* excited states are not wetting-layer-like but dot-localized since the center of mass of the former states is well below the QD ground state.[3–5]

Figure 3(c) shows interferograms at a bias of 0.000 and +0.341 V, exhibiting a phase difference of 74±20°. The fact that the phase difference is close to 90° indicates that the QDIP at zero bias has dominantly capacitive characteristic. In addition, we observe (not shown here) that the interferograms at all measured positive (negative) biases larger than 0.04 V (0.06 V) are in phase. In the bias range from zero to +0.040 V, the phase deviation gradually decreases from 74° to zero. An additional gradual phase change of ~106° occurs in the negative bias range from zero bias to about −0.06 V. These findings at 77 K imply that at high bias the extended photocurrent dominates, while at zero bias the displacement photocurrent is dominant. 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 rates from the involved excited states.

By studying the Stark shift of the interband ground photocurrent of the same QD ensemble, we also determined that the charge center of mass of the hole ground state is above that of the electron ground state. This is confirmed through our observation of the presence of interband ground transition induced nonzero photocurrent at zero bias and also a Stark shift upon applied bias. The bias dependence of the interband ground transition peak at 1.028 eV (at zero bias) at 200 K reveals a blueshift of 5 meV with varying bias from negative (−0.4 V) to positive (+0.4 V) bias, as shown in Fig. 4. The direction of interband transition induced dipoles found for these InAs/GaAs QDs is opposite to that indicated by the reported calculations based upon the model assumption of abrupt chemical change from binary InAs to binary GaAs across the InAs/GaAs interfaces involved.[3–5] Recent model calculations that allow for In and Ga intermixing through the QD bottom interface indicate that for sufficient intermixing and appropriate composition profile through the QD, the charge center of mass of the hole ground state can be above that of the electron ground state, i.e., the spatial ordering involved in ground state interband transition can get reversed as compared to the no intermixing case.[9,13,14] These calculations, however, do not provide information on the charge distributions associated with the high bound excited electron states in the transition energy neighborhood of ~174 meV involved in the *intraband* dipole observed in our studies.

Our finding on the existence of intraband dipoles in these self-assembled QDs ~77 K supports the feasibility of the proposed optically driven quantum gates based on electron–electron dipoles in asymmetric QDs.[5,15] Unlike the severe technological limitations faced in fabricating vertically stacked asymmetric QDs via lithography as proposed in Ref. 15, the concept is more readily implemented in the experimentally demonstrated vertically self-aligned InAs/GaAs island QDs with different sizes.[16] Such pairs of QDs give rise to different sets of energy levels[16] relevant for different qbits, i.e., conditional transition energies.[15] Additionally, the suppressed electron-phonon scattering in zero-dimensional QDs is known to contribute to long coherence time (~ns)[17] compared to that in quantum wells. This should also help in the potential implementation of the notions proposed in Ref. 15 using vertically self-organized epitaxical quantum dots of the type examined here.

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