Two photon absorption in quantum dot-in-a-well infrared photodetectors

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Two photon absorption processes in $InAs/In_{01.5}Ga_{0.85}As/GaAs$ quantum dot-in-a-well photodetectors are studied using free electron laser excitation. Two photon induced, normal incidence photocurrent, observed in the range of 20–30 μm, arises from sequential near-resonant two-step transitions involving electron ground to first excited states in the dot, to quantum well final states. We find a two photon absorption coefficient of $\beta \sim 1 \times 10^7$ cm/GW at 26.5 μm (47 meV) and 0.8 V applied bias. Second-order autocorrelation measurements exhibit two characteristic time constants of ~3 and ~40 ps. The latter is associated with the intermediate state electron lifetime, whereas the short decay is explained by the involvement of acoustic phonon assisted transitions. [DOI: 10.1063/1.2833691]

Quantum dot infrared photodetectors (QDIPs) have been an active area of research in recent years. It has been demonstrated that these devices are starting to reach the performance levels of the well established quantum well infrared photodetector (QWIP) technology.[1] Motivation for the development of QDIPs comes from their intrinsic advantages such as normal incidence detection and low dark current arising from the three dimensional electronic confinement in QDs.[2]

During the same time, the development of emitters in the midinfrared and far infrared (FIR) such as quantum cascade lasers, that are capable of pulsed and mode-locked operation with pulse widths in the order of approximately picoseconds, have created the necessity of the development of fast and sensitive detectors for monitoring and characterization of short optical pulses in these regions.[3,4] Standard methods where the second-harmonic generated light in a nonlinear crystal is measured by a slow linear detector, can be potentially substituted by nonlinear infared photodetectors.[5]

Nonresonant two photon (2P) processes have been studied in QWIPs,[6] and more recently optimized structures for 2P absorption were investigated, where energetically equidistant subbands result in resonantly enhanced second-order intrinsic nonlinearities.[3] As a result, high sensitivity 2P detectors of midinfrared subpicosecond pulses have been demonstrated.[4,5,7] The development of 2P QDIPs would be highly significant as they are capable of true normal incidence operation, have long intermediate state lifetimes resulting in high 2P absorption coefficients and have an intrinsic energy level configuration which makes them particularly attractive for operation in the terahertz region. Nonlinear studies in nondetector QD structures have demonstrated second-harmonic generation with very high nonlinear susceptibilities.[8] However no studies of nonlinear optical processes have yet been reported for QDIPs.

In this letter, we report on two photon normal incidence detection of FIR picosecond pulses using dot-in-a-well (DWELL) QDIPs. We observe a quadratic increase of the photocurrent with the incident power for bias range of 0.2–0.8 V. For higher applied field (1.2 V), we observe a change of this behavior to linear, as the escape probability increases and the one photon (1P) photocurrent overcomes the 2P process. Finally, we perform second-order autocorrelation measurements of short FIR pulses of ~3 ps at λ ~26.5 μm.

The DWELL sample was grown by molecular beam epitaxy (MBE) upon a semi-insulating GaAs substrate. The device design was the same as for previously reported DWELL QDIPs.[9] The DWELL active region consists five periods of 2.9 ML of InAs quantum dots placed within an 8 nm $In_{0.15}Ga_{0.85}As$ quantum well, Si δ-doped to a concentration of 6×10^{10} cm^{-2}, corresponding to approximately 1 electron/dot.

Figure 1 shows the photocurrent at +1 V measured by a vacuum Bruker IFS-66v/s Fourier-transform infrared spectrometer with a broadband globar light source. In this configuration, 1P transitions dominate and the possible DWELL conduction band transitions are indicated in the inset of Fig. 1. The observed photocurrent arises either due to excitation

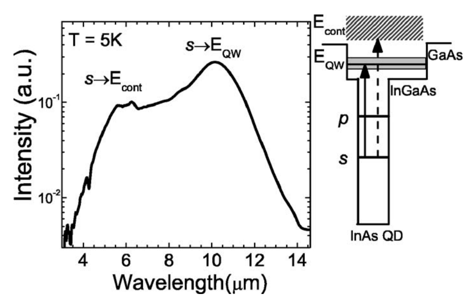

FIG. 1. One photon photocurrent measured by FTIR spectroscopy at 0.8 V and 5 K. The inset shows a conduction band diagram with the relevant transitions in DWELL QDIPs; $s \rightarrow E_{QW}$ (solid arrow), peaked at ~10 μm, $s \rightarrow E_{cont}$ (dashed arrow), peaked at ~6 μm.

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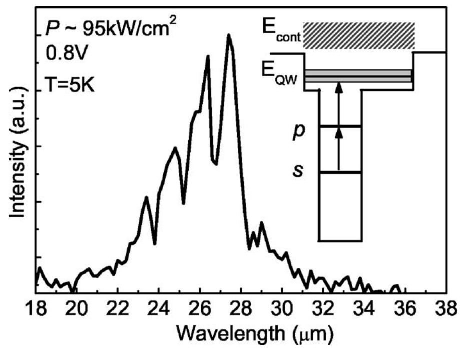

FIG. 2. Two photon (2P) photocurrent spectra at 5 K for DWELL QDIP illuminated with $\sim 95\ \mathrm{kW/cm^2}$ peak power at normal incidence. The dips in the spectrum are due to atmospheric absoprtion in this spectral region. The inset shows the possible 2P process involving intermediate p states.

of electrons from the QD ground state to a quasiconfined state in the QW (Ref. 9) ($s \rightarrow E_{\mathrm{QW}}$ peaked at $\sim 10\ \mu$m) or to the continuum states above the GaAs band edge ($s \rightarrow E_{\mathrm{cont}}$ peaked at $\sim 6\ \mu$m).

To investigate the spectral dependence of 2P transitions, we use a high power FIR source; the free electron laser, FELIX. Figure 2 displays the photocurrent spectra at 5 K measured by tuning the FELIX wavelength from 18 to 35 μm, showing a photocurrent peak at $\lambda \sim 26.5\ \mu$m (47 meV). This energy corresponds closely to the dot ground (s) to first excited (p) state transition energy observed in previous absorption studies[10,11] and 1P photocurrent studies of DWELL QDIPs,[12] thus strongly indicating that we observe a photocurrent associated with s-p transition. As described below, power dependent measurements show that under these conditions a 2P electron transition can be excited involving the s state of the QD, the intermediate p state in the QD and states in the QW (shown schematically in the inset of Fig. 2).

By investigating the dependence of the photocurrent on incident power for different bias conditions, exciting with FELIX output at $\lambda = 26.5\ \mu$m (resonant with the s-p transition), we are able to determine whether a 1P or 2P process is involved. For these measurements, the QDIP was connected to a current preamplifier and the output signal was measured using an oscilloscope.

Figure 3 shows the photocurrent dependence on peak power density P for 0.2, 0.5, 0.8, and 1.2 V, at $T=5$ K. For 0.2 up to 0.8 V, we observe a quadratic dependence of P in the range of $10-400\ \mathrm{kW/cm^2}$, showing that 2P processes dominate. The photocurrent magnitude increases with bias due to the increased escape probability from the final photocurrent state. When the applied bias was increased to 1.2 V, the dependence became more linear (over what range), indicating that tunneling directly from the p state becomes significant and therefore 1P processes give rise to the photocurrent. We also investigated additional samples with a 1P photocurrent peak (i.e., energy of final state) of 160 meV, far from twice the s-p transition energy (~ 50 meV), which showed no quadratic behavior.

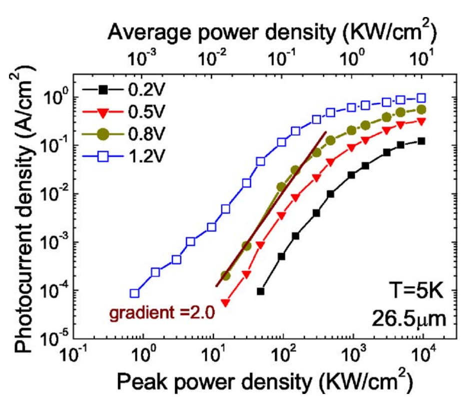

FIG. 3. (Color online) Photocurrent power dependence at 5 K for 0.2 V (black closed squares), 0.5 V (red triangles), 0.8 V (dark yellow circles), and 1.2 V (blue open squares). The dependence is quadratic from ~ 10 up to $\sim 400\ \mathrm{kW/cm^2}$, after which it saturates.

From Fig. 3, it can also be seen that a gradual decrease of the quadratic behavior occurs with increasing peak power density. A further increase in P leads to saturation of the photocurrent at $\sim 4\ \mathrm{MW/cm^2}$, which could be attributed to space charge saturation,[7] absorption saturation,[13] or contact effects.[14]

We can extract the 2P absorption coefficient β via the 2P photocurrent density (Fig. 3), $j_{2\mathrm{P}}$, by[4]

$$j_{2\mathrm{P}} = \frac{e\beta L f_{\theta} g}{h\nu} P_{\mathrm{ave}}^2, \tag{1}$$

where L is the thickness of the active region, which for our five layer structure is $L=40$ nm and P_{ave} is the average power density. The factor f_{θ} is related to the angle of incidence and is equal to unity for normal incidence. Under the assumption that the gain is near unity,[15] from Eq. (1), β is estimated to be $\sim 1 \times 10^7$ cm/GW, close to the values reported for resonant 2P QWIPs (Refs. 4 and 7) which are six orders of magnitude higher than for bulk materials such as GaAs or Si. It is therefore clear that even for unoptimized DWELL QDIP structures such as those studies here that large values of β are possible. Growth of DWELL QDIP samples in which the s-p and p-E_{QW} transitions are resonant would result in approximately order of magnitude larger values of β. DWELL QDIPs offer several advantages for designing such structures as the energy level configuration can be controlled by varying either the well width[16] or number of InAs monolayers deposited during QD growth.[9] In addition, narrow s-p and s-E_{QW} linewidths[11] (~ 5 and 18 meV, respectively) suggest that intermixing techniques[17] can be applied to tune the transition energies using postgrowth rapid thermal annealing. We have also demonstrated that the s-E_{QW} energy can be varied in DWELL structures via the Stark effect,[9] while the s-p energy should be less sensitive to applied bias, providing a further method for tuning the relative energy level separations and hence the 2P absorption coefficient.

The time evolution of the 2P photocurrent signal was analyzed using second-order autocorrelation measurements. The FELIX output is split into two identical pulse trains and the 2P photocurrent measured with the DWELL QDIP sample as a function of delay time between the two pulse trains. At zero delay, we observe an autocorrelation peak with a ratio of $\sim 3:1$ with respect to the signal way from zero delay (the 1P signal), as shown in Fig. 4. This ratio is close to that expected for a 2P process.[3,4,18] We can also extract information about the relaxation times of electrons involved in the the 2P process from our autocorrelation measurements. Following the approach of Nessler et al.,[19] we numerically fit the data and extract information about the carrier lifetime of the intermediate state. We observe a biexponential decay of the autocorrelation trace and use the above approach to fit

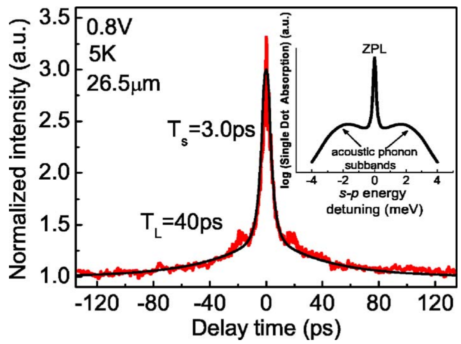

FIG. 4. (Color online) Autocorrelation trace of DWELL QDIP at $\sim$95 kW/cm^2 and 0.8 V at 26.5 μm measured at normal incidence (red line). A biexponential fit (black line), yields a T_s=3 ps and T_L=40 ps. The inset shows a schematic of the single dot intraband absorption line composed of the zero phonon line (ZPL) and acoustic phonon sidebands.

the trace with a short time T_s and a longer time constant T_L and find times of $T_s \sim 3$ ps and a $T_L \sim 40$ ps. T_L is associated with the lifetime of the *p*-like intermediate state, which has been extensively studied[8,10] using pump-probe spectroscopy yielding typical times between $\sim$30 and 60 ps, depending on the energy of the *s*-*p* transition. The origin of T_s is not well understood, but can be explained by the following. The single QD absorption spectrum typically consists of zero phonon line (ZPL) which is significantly broadened by acoustic phonon sidebands (APSs). This was originally observed for QD excitons[20] and more recently also in *s*-*p* absorption spectrum using four-wave mixing spectroscopy.[21] The presence of acoustic phonon sidebands in inhomogeneously broadened QD ensemble, can result in resonant 2P absorption not only involving ZPL but also via APS. The characteristic time in autocorrelation measurements in the latter case will be determined by a very short (approximately picoseconds) decoherence time due to emission of coherent acoustic phonons. Although the absorption of APSs is much smaller compared to ZPL, the number of dots for which the resonant 2P absorption via APSs is possible will be significantly larger than the number of QDs with 2P absorption via ZPL due to the relatively large $\sim$2 meV broadening of the absorption line. However, in order to explain the origin of T_s more explicitly, further detailed studies are required.

In conclusion, we have demonstrated the two photon, normal incidence, detection of ultrashort optical pulses in the far-IR using DWELL QDIPs. We observe a quadratic increase of the photocurrent with incident power tuned into resonance with the ground-first excited state transition in the quantum dots. We also perform second-order autocorrelation measurements and use the results to determine the dynamics of the photodetector. Our results demonstrate the potential of normal incident nonlinear DWELL QDIPs for the characterization of few picosecond far-infrared laser pulses and provide insight into the relaxation dynamics in these devices.

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