Terahertz emission by quantum beating in a modulation doped parabolic quantum well

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(Received 5 February 2008; accepted 24 March 2008; published online 10 April 2008)

We use ultrafast terahertz spectroscopy to observe terahertz-frequency electron oscillations in a modulation-doped InGaAs/AlGaAs parabolic quantum well with subband spacing 0.01 eV. Our study examined how the extrinsic electron density in the well influences terahertz emission efficiency and we found no strong dependence. This indicates that terahertz emission in this structure arises from quantum beating of the photogenerated electrons. Terahertz emission from the cold extrinsic electrons due to ultrafast field screening plays at most a secondary role. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2908868]

Intersubband transitions in semiconductor quantum wells have attracted wide interest during the last two decades as the basis of quantum cascade midinfrared and far-infrared lasers and of quantum well infrared photodetectors. Optically pumped ultrafast terahertz emission was originally used to probe intersubband transitions in quantum wells by Roskos *et al.*[1] and Planken *et al.*[2] They observed terahertz frequency electromagnetic radiation due to coherent electron intersubband oscillations following femtosecond optical excitation of a coupled quantum well. The finite bandwidth of the *fs* optical pulse allows interband photoexcitation into multiple subbands. Terahertz emission arises from quantum beating of excitons initially excited into a coherent superposition of interband subband states. In nonsymmetric structures this leads to time-dependent expectation value for the dipole moment and, thus, radiation. Intersubband quantum beating has also been observed in step quantum wells at midinfrared frequencies.[3]

Coherent charge oscillations have also been observed in modulation-doped quantum wells. Bratschitsch *et al.*[4] reported terahertz intersubband charge oscillations in a modulation-doped parabolic quantum well (PQW). The authors argued that these could not arise from quantum beating primarily because a parabolic well is a symmetric system. In addition, they observed strong coherent emission even for excitation at energies as large as 0.1 eV above the band edge, with no significant increase in the dephasing rate. The authors suggested that intersubband coherence of the photocarriers should decay very rapidly when their kinetic energy is larger than the optical phonon energy.

Instead, they attributed terahertz emission to excitation of the extrinsic electrons by ultrafast field screening. In this model, photocarriers screen a built-in electric field across the quantum well, leading to intersubband plasma oscillations of the extrinsic carriers. Notably, this mechanism does not require phase coherence among the photogenerated carriers. In the limit of weak excitation, the amplitude of the oscillations should be proportional to the magnitude of the screened field and the density of carriers. The authors did note that an electric field would break the symmetry of the structure, and make terahertz emission by quantum beating allowed, but suggested that this would be a small effect. However, we show below that this suggestion is not well supported.

More recent studies have shown that electron-optical phonon scattering is, in fact, not effective at destroying the coherence of intersubband charge oscillations. Huggard *et al.*[5] examined terahertz emission from undoped asymmetric double wells and reported dephasing times of several picoseconds even for pump energies $h\nu - E_G \gg \hbar\omega_{LO}$. Eckardt, *et al.*,[6] observed terahertz collective oscillations of ballistic electrons in wide parabolic *n-i-p-i* potential wells. In these experiments, which probe the semiclassical limit of quantum beating, photoelectrons are excited at the edge of the confining potential, producing terahertz emission as they coherently oscillate across the well. Although the carriers experience multiple optical phonon scattering events during each cycle, only a small fraction of scattering events destroy intersubband coherence. Their results were supported by detailed Monte Carlo simulations.

In this work, we have investigated terahertz intersubband charge oscillations in a modulation-doped PQW and have carefully examined how the extrinsic electron density, electric field, and pump photon energy influence terahertz emission. We found no strong dependence on carrier density and observe strong terahertz emission even when the well is depleted of extrinsic carriers. We conclude that ultrafast field screening is not the dominant excitation mechanism for intersubband charge oscillations in our structure.

Our modulation-doped InGaAs/AlGaAs PQW sample was grown by molecular beam epitaxy. Starting at the surface, the structure contains a 235 nm $Al_{0.3}Ga_{0.7}As$ buffer, the modulation doped 138 nm InGaAs/AlGaAs PQW, a 1000 nm $Al_{0.3}Ga_{0.7}As$ buffer, a 250 nm *n*-InGaAs quantum well designed as a back gate, a smoothing layer and the semi-insulating-GaAs substrate. The composition at the center of the PQW is $In_{0.125}Ga_{0.875}$ As and at the edge is $Al_{0.3}Ga_{0.7}As$. We deposited a 70 Å semitransparent NiCr metal gate on the surface and diffused indium Ohmic contacts to the PQW to form electrical contacts. The Ohmic contacts also contacted the back gate. Capacitance-voltage measurements determined the zero-bias charge density in the well $n = 3 \times 10^{11}$ cm^{-2}. The electron subband spacing is 0.01 eV.

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The optical system used for terahertz emission measurements comprised a time-domain terahertz spectrometer. The optical pump was a mode-locked Ti-sapphire laser producing $\Delta t = 10$ fs laser pulses, with center wavelength $\lambda = 800$ mm, and a spectral bandwidth [full width at half maximum (FWHM)] $\Delta\lambda = 110$ mm. The average pulse power was 10 nJ/pulse and the pulse repetition rate was 75 MHz. Tunable pulses in the energy range $h\nu = 1.45-1.65$ eV were obtained by filtering the pump beam with 10 nm (0.02 eV) FWHM band pass filters. The sample was mounted in a liquid helium flow cryostat (4–300 K) with optical access. The pump pulses were incident on the sample's surface at 60° and terahertz radiation was collected in the pseudotransmission geometry. The photon flux per pulse after the band pass filters was $\sim 1 \times 10^{11}$ cm^{-2} at the sample surface. Terahertz emission from the sample was collected and focused using a pair of parabolic mirrors and detected using free-space electro-optic sampling with a 1 mm ⟨110⟩ ZnTe detector. We measured terahertz emission as a function of gate voltage and pump-photon energy.

We also performed supporting intersubband absorption, interband absorption, and time-resolved photoluminescence measurements on our sample. We measured intersubband absorption in a gated PQW sample from the same wafer using a Nicolet Magna 530 Fourier-transform infrared (FT-IR) incorporating a 4.2 K Si bolometer as a far-infrared detector. In these measurements, the sample was mounted in the bolometer cryostat at $T = 4.2$ K and transmission measurements were performed as a function of gate bias. Terahertz radiation was coupled into the edge of the gated sample and was polarized with the electric field perpendicular to the surface to maximize intersubband absorption. Interband absorption measurements (1.0–1.7 eV) were performed using our FT-IR with near-IR optics and Brewster-angle coupling. Interband absorption was recorded for an as-grown PQW sample mounted in a flow cryostat at $T = 10$ K. Room temperature measurements were performed on a second PQW sample with the substrate removed. Spectrally and temporally resolved photoluminescence (PL) measurements were performed on our PWQ sample at 10 K, using the *fs*-Ti:sapphire laser as an excitation source. We used an Acton-300I spectrograph in spectrally resolved measurements of the PQW PL peak. Bandpass filters were used in time-resolved measurements to isolate the quantum well luminescence. The luminescence intensity was measured with a 1 GHz Si avalanche photodiode module, yielding a time resolution of ~0.5 ns.

Figure 1 shows the terahertz emission from our PQW at $T = 6$ K and gate-channel voltage $V_g = -1$ V, recorded with an 800 nm band pass filter. We observe an initial spike followed by rapid oscillations that decay with a time constant of ~3 ps. Figure 2 shows the Fourier transform of this waveform. The sharp emission peak at 2.82 THz matches the intersubband absorption line observed in far-infrared transmission. The broadband emission at lower frequencies is associated with the initial spike in the waveform. We also observe a broad absorption line at 1.3 THz due to donor impurity transitions in the substrate.

Measurements of terahertz emission as a function of gate voltage found the peak frequency to be voltage independent as expected for a parabolic well. The amplitude of the intersubband oscillations (Fig. 3) nearly linearly increases with bias over the range $V_g = +1$ to -1 V and saturates for $V_g <$

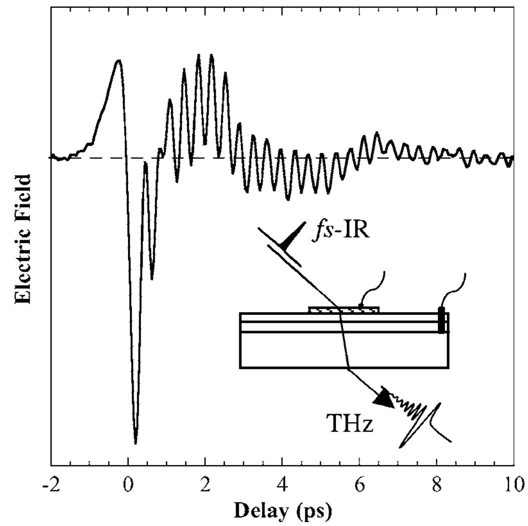

FIG. 1. Terahertz emission from our PQW ($T = 4.2$ K, $V_G = -1.0$ V, $h\nu = 1.55$ eV). Intersubband charge oscillations are observed following impulsive optical excitation at $t = 0$. Inset shows experimental geometry.

-1.5 V. In comparison, $C(V)$ measurements of the extrinsic carrier density show depletion of the well at $V_g = -1.5$ V. The amplitude of the initial spike is independent of gate voltage, suggesting it is not associated with our quantum well. As electrical depletion of modulation doped quantum wells can leave pockets of donor electrons isolated from the contacts, we also probed the carrier density optically using intersubband absorption. The absorption strength decreases with increasing negative gate bias consistent with $C(V)$ measurements, vanishing when the well is depleted. We also determined that photogenerated electrons and holes do not accumulate over successive laser pulses in our sample. Time-resolved photoluminescence measurements determine the $T = 10$ K recombination time to be $\tau_r = 1.0 \pm 0.2$ ns, much shorter than the ~13 ns interval between laser pulses. We conclude that we can excite strong intersubband charge oscillations even when the sample is depleted of extrinsic electrons.

We furthermore investigated how terahertz emission changes as we vary the excitation wavelength (Fig. 4). When the terahertz oscillation amplitude is normalized with respect to the *absorbed* photon flux, as determined by interband ab-

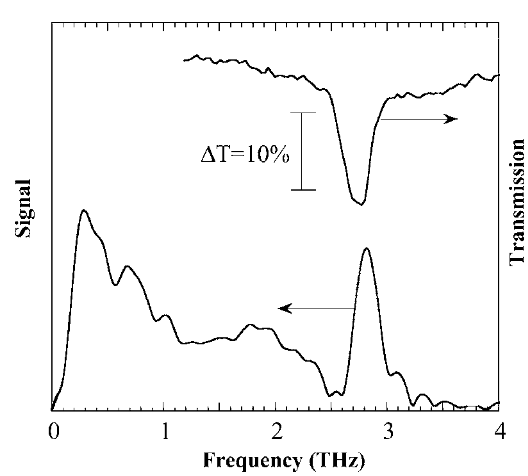

FIG. 2. Fourier transform of waveform from Fig. 1 (lower trace). The sharp emission feature at 2.82 THz is in agreement with the intersubband absorption line observed in far-infrared transmission (upper trace).

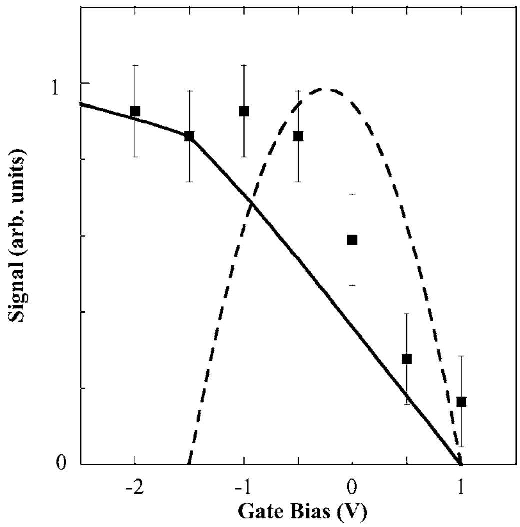

FIG. 3. Terahertz oscillation amplitude vs gate bias (solid points). Solid line shows the quantum well electric field obtained from $C(V)$ measurements, while dashed line shows the product of the electric field and the extrinsic carrier density. Both curves have been normalized to aid comparison to experiment.

sorption measurements, the emission efficiency is seen to decrease to $\sim$20% of its initial value as the pump-photon energy is increased from $h\upsilon - E_G = 0.08-0.24$ eV.

The observed dependence of the terahertz charge oscillations on gate bias indicates that the terahertz emission primarily arises from quantum beating of the photogenerated electrons. In a three-level model of quantum beating, ultrafast excitation can produce a time-dependent dipole moment[7] $P(t) \propto z_{32}z_{21}z_{31}e^{i\omega_{32}t}$, where z_{32}, z_{21}, and z_{31} are dipole matrix elements between the different levels. While $z_{21}z_{31}=0$ in a symmetric structure, the quantum beating processes is strongly allowed in the presence of the $\sim$kV/cm fields typical in quantum wells. Taking (1) to be the highest hole subband of a parabolic confining potential, and taking (2) and (3) to be the lowest electron subbands, we find $z_{31} \propto \langle \varphi_3 | \varphi_1 \rangle = 0$ for $E=0$. For a finite electric field $\langle \varphi_3 | \varphi_1 \rangle = \left(\frac{4}{\pi}\right)^{1/4} u_0 e^{-u_0^2}$, where $u_0 = 2eE/\sqrt{hm\omega_0^2}$, ω_0 is the classical frequency of the oscillator and we have taken $m_h = m_e$ for simplicity. For weak fields the oscillating dipole moment is thus proportional to electric field. A numerical simulation of our quantum well yields $z_{31}/z_{21} \approx 0.5E/(1\ \text{kV/cm})$.

Our data are in good agreement with the prediction of the quantum beating model. The solid line in Fig. 3 shows the electric field determined from capacitance-voltage measurements where we have taken the total field (external field plus internal polarization) to be zero at $V_G = -1$ V. On the other hand, our results are inconsistent with the ultrafast field screening model in which terahertz emission arises from oscillations of extrinsic carriers. This model predicts that the signal should be proportional to the product of the electric field and the density of extrinsic elections, $S_{\text{THz}} \propto nE$ (dashed line in Fig. 3), reaching zero when the well is depleted.

The observed decrease in terahertz emission efficiency with increasing excitation photon energy also qualitatively supports the quantum beating model. Increasing the photon energy produces photoelectrons with high initial kinetic energies. We suggest that these carriers rapidly cool by emitting optical phonons ($\tau \sim 0.1$ ps), but that this reduces, but does not completely destroy the intersubband coherence. In contrast, the ultrafast field screening model has no obvious mechanism that would produce the strong dependence on excitation energy we observe. In particular, heating of extrinsic electrons by photocarriers does not appear to be important: for the measurements shown in Fig. 3, $n_{\text{photo}} < 0.05 n_d$, $h\nu - E_G \leqslant 0.25$ eV, thermalization of the carrier excess energy among the extrinsic and photogenerated carriers would yield electron temperatures $T_e < 100$ K. However, temperature dependent terahertz emission measurements on this sample show no decrease in the amplitude of charge oscillations for lattice temperatures up to $T=100$ K.

In conclusion, we have used ultrafast terahertz spectroscopy to observe intersubband charge oscillations in a modulation-doped InGaAs/AlGaAs PQW. We found that the amplitude of the terahertz intersubband emission is proportional to electric field, but independent of the extrinsic carrier density in the well. We conclude that the charge oscillations primarily arise from the quantum beating of the photogenerated electrons, and that oscillations of the extrinsic electrons due to ultrafast field screening play at most a secondary role.

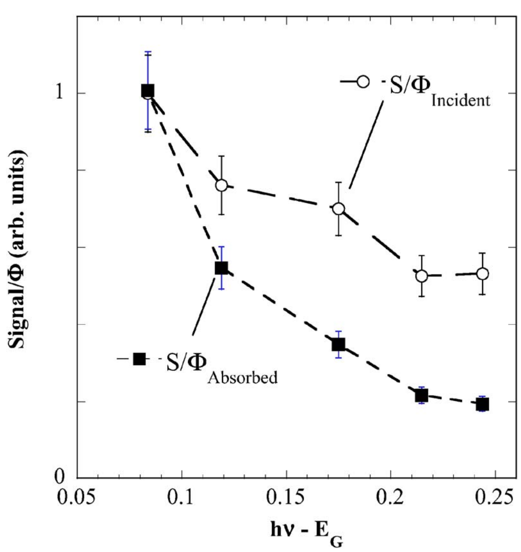

FIG. 4. (Color online) Terahertz oscillation amplitude vs versus pump-photon energy. The open circles show the ratio of the signal to the photon flux incident on the sample. The solid squares show the ratio of signal to photon flux absorbed in the quantum well. Both data sets are normalized at $h\nu - E_G = 0.085$ eV.

The authors wish to acknowledge the technical assistance of C. Papillia and T. Myint in this work. We acknowledge support from the National Science Foundation through the NSF-RUI program (DMR-0606181).

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