Terahertz Emission in Single Quantum Wells after Coherent Optical Excitation of Light Hole and Heavy Hole Excitons

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We report on the first observation of coherent terahertz radiation, tunable from 1.4 to 2.6 THz, emerging from GaAs/$Al_{0.3}Ga_{0.7}As$ *single* quantum wells after the coherent optical excitation of both light hole and heavy hole excitons. We attribute the radiation to charge oscillations following the coherent excitation of the excitons in an electric field. Terahertz radiation is also emitted when no electric field is present in the sample suggesting that valence band mixing leads to a significant far-infrared transition dipole moment between the light hole and heavy hole subbands.

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In the study of carrier dynamics in quantum wells, beating phenomena can be observed when pulsed lasers are used to coherently excite two energy levels in the system. In a two-pulse self-diffraction four wave mixing (FWM) experiment, the beating can be observed as a modulation of the decay of the induced optical macroscopic polarization [1-3]. In a recent experiment, a new technique was introduced with which we can more directly observe beats involving spatial charge oscillations [4]. Systems that have a dipole moment that changes as a function of the beating will emit electromagnetic radiation at the beat frequency. Experimentally, this was demonstrated by observing terahertz radiation from a double coupled quantum well (DCQW) after optical excitation of a superposition of the symmetric and antisymmetric eigenstates of the coupled wells. The superposition of the two states leads to the coherent tunneling of electrons between the two wells and therefore to a time-varying dipole moment. Such an experiment offers the advantage that the observation of terahertz oscillations forms a direct proof of the existence of charge oscillations associated with the tunneling electrons. Charge oscillations are a prerequisite for observing more than one cycle of terahertz radiation in these systems and imply the existence of a far-infrared transition dipole moment on which FWM provides no information. In addition, the proportionality of the signal to the far-ir transition-dipole moment after optical excitation in single quantum wells offers us the possibility to study the mixing between the heavy hole and light hole valence subbands.

In this Letter we present the first observation of *tunable* terahertz radiation in *single* quantum wells from the coherent excitation of *light hole (lh)* and *heavy hole (hh)* excitons, implying the existence of charge oscillations in this system. In Fig. 1(a) we sketch the lh and hh envelope states of the quantum well in an electric field. The charge oscillations are the result of the time evolution of the superposition of the lh and hh exciton states. The measured frequencies agree very well with the lh-hh splitting as measured in photocurrent spectra. We find that the oscillations persist even when there is no dc electric field in the sample (flatband), indicating that lh-hh valence band mixing leads to a significant transition-dipole moment between the lh and hh subbands.

The sample is a metal-*i*-*n* Schottky diode with a multiple quantum well (MQW) in the intrinsic region. The MQW consists of fifteen periods of 175 Å GaAs wells, separated by 150 Å $Al_{0.3}Ga_{0.7}As$ barriers. Between the MQW region and the n^+-doped substrate there is a su-

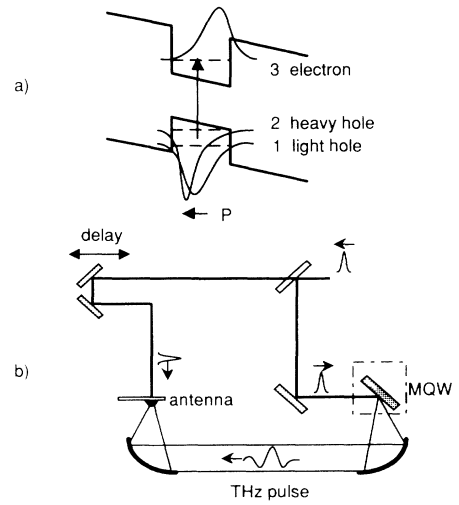

FIG. 1. (a) Wave-function envelopes of the electron, lh, and hh in a quantum well biased with an electric field E. (b) Experimental setup for the THz generation measurements with multiple quantum well (MQW).

perlattice for strain relief that is completely transparent in the wavelength region of interest. The sample is capped with a 200 Å GaAs layer and finally 50 Å of semitransparent chromium is deposited. We can control the electric field in the sample by applying a voltage between the Schottky contact and the doped substrate. The designed lh-hh splitting at flatband is 6 meV or 1.5 THz. The actual splitting as a function of electric field is taken from the measurement of photocurrent spectra at 4 K. We obtain the electric field in the sample by comparing the shift of the hh exciton due to the quantum-confined Stark effect with the calculated shift as a function of electric field. The sample is mounted in a continuous-flow liquid helium cryostat and the temperature is kept at a value around 10 K except when temperature-dependent measurements are done.

Our experimental setup [Fig. 1(b)] uses pulses from a mode-locked, 82 MHz repetition rate, argon-ion–pumped Ti:sapphire laser, tunable around 800 nm. The pulses have a duration of approximately 80 fs and a maximum energy of 15 nJ. The laser beam is split in two. One beam passes a variable attenuator and is focused onto the quantum well sample under a 45° angle of incidence with its polarization parallel to the plane of incidence. The spot size at the sample is roughly 1 mm and the average power of the beam is several tens of mW. The generated THz beam emerges from the sample in the same direction as the partially reflected pump beam and leaves the cryostat through a high-resistivity silicon window. A pair of off-axis parabolic mirrors collimates and then focuses the radiation onto a 50 μm photoconducting dipole antenna [5] with a silicon hyperhemispherical substrate lens. The second laser beam is sent through a variable delay and is used to gate the dipole antenna. By measuring the photocurrent from the antenna versus the delay between the gating pulse and the THz signal, we measure the electric field $E(t)$ of the signal. In this way we obtain both the phase and the amplitude of the signal. The excitation bandwidth is larger than the lh-hh splitting and, when we tune the laser to the excitons, we coherently excite them both.

Detected THz wave forms as a function of wavelength at an electric field strength of 4 kV/cm are shown in Fig. 2. For reference, the inset shows the measured photocurrent spectrum with the clearly resolved hh and lh exciton transitions. Immediately apparent in Fig. 2 is the oscillatory tail of the signal when the laser overlaps both the lh and hh excitons at 1.527 eV, implying the existence of charge oscillations. The oscillations have a frequency of 1.4 THz equal to the measured lh-hh splitting and dephase within a few picoseconds. The oscillatory tail is *only* present when we excite both the lh and hh excitons. When the laser is tuned above the excitons towards higher energies, the oscillations disappear and only a single cycle electrical transient remains. An important contribution to the single cycle transient results from the electron-hole pairs already being in a polarized state upon

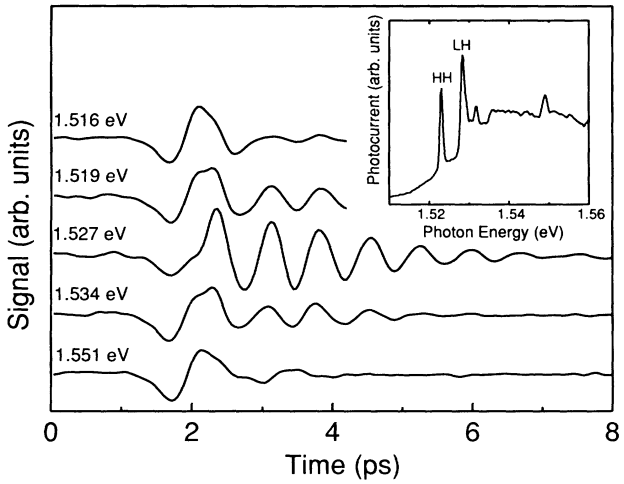

FIG. 2. Measured THz wave forms for several excitation wavelengths. The electric field in the MQW is 4 kV/cm. Inset: The measured photocurrent spectrum.

creation [6,7]. This leads to a time-dependent polarization $P(t)$ that grows with the integral over the pulse energy and hence radiates an electrical transient according to $E \propto \partial^2 P/\partial t^2$. The amplitude of this contribution increases with electric field because the net dipole moment due to the electron and hole separation increases with electric field. When we tune the laser below the band edge of the quantum well sample, as indicated by a very small measured photocurrent, again a small amplitude, single cycle electrical transient remains. We believe that this transient, which shows very little electric field dependence, originates from the substrate although its precise physical origin is still under investigation. At or above the band edge, a fraction of the laser light also reaches the substrate due to the small number of quantum wells. The THz signal coming from the substrate therefore also contributes to the single cycle transient generated above the band edge.

The detected THz wave forms as a function of electric field in the sample are shown in Fig. 3 when the laser is tuned to the lh and hh exciton resonances (1.527 eV for weak fields). The oscillatory part of the signal, which follows the initial single cycle discussed above, has a frequency that increases with electric field. The frequency of the oscillations changes from 1.4 THz at flatband to almost 2.6 THz at the highest electric field. This can be seen more clearly when we calculate the Fourier transforms of just these oscillations as shown in the inset of Fig. 3. To our knowledge this system constitutes the first tunable terahertz emitter. The apparent decrease in amplitude of the oscillatory part of the wave form with increasing applied electric fields is in part an artifact of the decreasing sensitivity of the dipole antenna for higher frequencies [5]. Interestingly, the oscillations are still strongly present at flatband when there is no electric field in the sample. In Fig. 4 we plot the measured terahertz

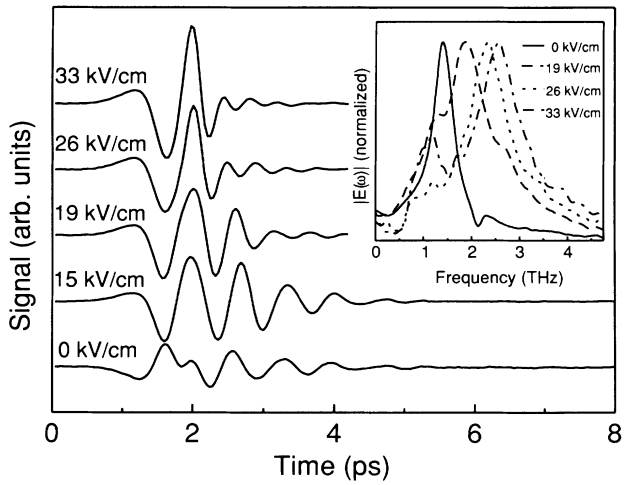

FIG. 3. Measured THz wave forms for several electric fields in the MQW. The laser is tuned to the lh and hh excitons. Inset: The Fourier transforms of only the oscillatory part of the signals.

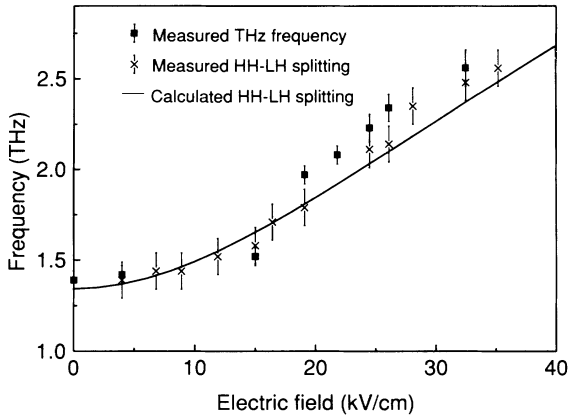

FIG. 4. Measured THz frequencies, lh-hh splittings taken from photocurrent spectra, and calculated lh-hh splittings as a function of electric field in the MQW. The calculation includes excitonic effects.

oscillation frequencies as a function of electric field. In the same figure we also plot the measured lh-hh splittings as observed in photocurrent spectra taken at 4 K. We can clearly see that there is an excellent agreement between the two. The solid line is a calculation of the lh-hh splitting as a function of electric field that includes the Coulomb interaction of the hole with the electron. The calculation does not use any adjustable parameters and reproduces the measurements well.

The origin of the terahertz radiation is that in an electric field, a coherent superposition of the hh and lh excitons leads to charge oscillations in the well at the lh-hh splitting frequency. This can be seen from the following. For simplicity we concentrate on the induced far-ir polarization in the z direction. This polarization can be written in the three-band model [Fig. 1(a)] using a density-matrix formulation as

$$P(t)=\frac{|e|}{V}\sum_k [(z_{11}-z_{33})\rho_{11}+(z_{22}-z_{33})\rho_{22} +2\,\mathrm{Re}(z_{12}\rho_{12})], \quad (1)$$

where the self-dipole moments are defined as $z_{11}=\langle \mathrm{lh}|z|\mathrm{lh}\rangle$, $z_{22}=\langle \mathrm{hh}|z|\mathrm{hh}\rangle$, and $z_{33}=\langle e|z|e\rangle$, and the intersubband transition dipole moment as $z_{12}=\langle \mathrm{lh}|z|\mathrm{hh}\rangle$. ρ_{11} is the generated lh density, ρ_{22} the generated hh density, and ρ_{12} the coherence between the hh and lh states. In Eq. (1), $z_{11}-z_{33}$ and $z_{22}-z_{33}$ are the net displacements between the lh and the electron, and between the hh and the electron, respectively. The last term in Eq. (1) $z_{12}\rho_{12}(t)$ is proportional to $e^{i(\omega_2-\omega_1)t}$, with $\hbar(\omega_2-\omega_1)$ the hh-lh energy splitting, and is responsible for the charge oscillations. The radiated electric field is $E(t)\propto \partial^2 P/\partial t^2$, and contains both the initial transient due to the creation of polarized electron-hole pairs [6] and the oscillatory tail $\propto\cos(\omega_1-\omega_2)t$ caused by the charge oscillations. We can therefore expect to see terahertz radiation as long as there is a nonvanishing transition dipole moment z_{12}.

Because of the broad bandwidth of the exciting laser pulse we may excite not only excitons, but also uncorrelated electron-hole pairs. The following arguments, however, show that the terahertz radiation originates from the beating between the lh and hh *excitons*, not simply the uncorrelated holes. First, the wavelength dependence in Fig. 2 demonstrates that terahertz emission is only present when we tune the laser to both the lh and hh excitons. Second, the dephasing time of the radiation is on the order of a few picoseconds. This rules out uncorrelated holes as the source of radiation since it has been shown [8] that uncorrelated carriers dephase much faster (probably faster than 100 fs) in GaAs quantum wells than excitons, whose typical dephasing times for our carrier densities less than 10^{10} cm^{-2} are on the order of a few picoseconds [8,9]. Third, as shown above, there is an excellent agreement between the measured lh-hh exciton splitting and the measured terahertz frequency as a function of electric field.

The experimental results in Fig. 3 show that oscillations are still strongly present at flatband, when at $k_\parallel=0$ there is no transition-dipole moment between the wave functions of lh's and hh's. The nonzero transition-dipole moment at flatband is caused by the mixing between the lh and hh valence bands for wave vectors $k_\parallel\neq 0$ in the plane of the quantum wells [10]. The mixing leads to a finite value for the transition-dipole moment even when no electric field is applied. Note that our model is based on the dipole approximation and that an integration over $k_\parallel$ has been carried out. Interesting questions concerning the possible contributions at $k_\parallel=0$ due to higher-order moments such as the quadrupoles requires further investigation. Since an integration over $k_\parallel$ has been carried out

in our dipole model (at $k_{\parallel} \neq 0$, transitions are dipole allowed), the dipole moment should be sufficient.

We can look at the generation process as a resonant second-order nonlinear optical frequency conversion process in which light at visible frequencies within the bandwidth of the laser pulse is mixed to generate the difference frequency in the far-infrared region. Generally speaking, such a $\chi^{(2)}$ process can only take place in noncentrosymmetric media. From Fig. 1(a) it can be seen that in our quantum well sample the inversion symmetry is broken by the application of the electric field that allows the frequency conversion to take place. The electric field induces a $\chi^{(2)}$ that has a resonance at the lh-hh splitting frequency. At flatband the field-induced $\chi^{(2)}$ is obviously zero and we are therefore led to conclude that in this case the (resonantly enhanced) $\chi^{(2)}$ responsible for the conversion process is the intrinsic $\chi^{(2)}$ of the host material. It is a well-known fact that bulk GaAs has a $\chi^{(2)}$ although this property is often thought to be of little importance in quantum wells.

It is worth mentioning here that the valence-band mixing at flatband for nonzero in-plane wave vectors is also responsible for the existence of intersubband transitions in *p*-doped semiconductors [11], and semiconductor heterostructures [12] that are designed to be used as midinfrared detectors. The most obvious difference with our experiment is of course that we measure coherent emission of radiation from a nonlinear mixing process, not linear absorption. Furthermore, our transitions take place in the far-infrared region, allowing us to study the sample in a wavelength region otherwise not easily accessible.

In conclusion, we have observed terahertz radiation tunable from 1.4 to 2.6 THz after the coherent optical excitation of light hole and heavy hole excitons in *single* quantum wells. The source of the signal is that coherent optical excitation of both excitons gives rise to charge oscillations and therefore to a time-varying dipole moment that radiates in the far-infrared region. The measured terahertz frequencies are shown to be in excellent agreement with the measured hh-lh splittings. We also observe terahertz radiation at flatband and attribute this to the existence of a lh-hh transition dipole moment caused by valence band mixing.

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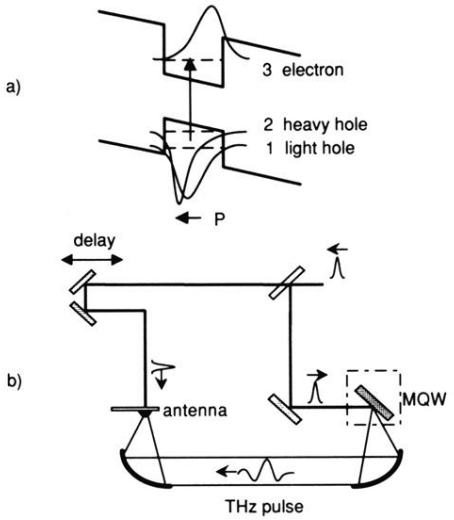

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