Coherent Submillimeter-Wave Emission from Charge Oscillations in a Double-Well Potential

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We directly observe the electromagnetic radiation emitted by electrons coherently oscillating between the two wells of a semiconductor coupled-quantum-well structure. Using time-resolved coherent detection of the submillimeter-wave radiation from these spatial charge oscillations, we trace up to fourteen oscillations at 1.5 THz before phase relaxation destroys the coherence of the oscillating wave packet. In addition to the oscillatory electromagnetic signal, we also observe an instantaneous signal from electric-field-induced optical rectification in the semiconductor structure.

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The possibility to observe charge oscillations in solids has been discussed extensively in the literature and has seen a revival as novel semiconductor heterostructures promise the observation of such oscillations at last. Recently, some of us have seen strong evidence for charge oscillations in a coupled-quantum-well structure from optical pump-probe and degenerate four-wave-mixing (DFWM) experiments [1]. A much more direct probe of such charge oscillations—because it directly measures the time-dependent electric dipole moment and thus discriminates against quantum and phase beats that do not involve any spatial motion of the charge—is the observation of electromagnetic dipole radiation emerging from these charge oscillations [2]. In this Letter, we report the first observation of oscillatory coherent submillimeter-wave emission from a coupled-quantum-well structure and thereby provide a direct proof for spatial charge oscillations in this system. Using time-domain terahertz spectroscopy, we observe up to fourteen cycles of the electromagnetic radiation emitted from the oscillating charge before phase relaxation destroys the coherence of the electronic wave packet. Furthermore, we clearly identify for the first time an instantaneous signal from field-induced optical rectification in the semiconductor structure [3].

Our coupled-quantum-well structure [Fig. 1(a)] consists of a 51-monolayer (145 Å) thick wide well (WW) and a 35-monolayer (100 Å) thick narrow well (NW), separated by a 9-monolayer (25 Å) thick $Al_{0.2}Ga_{0.8}As$ barrier. The pairs are isolated from each other by 200 Å wide $Al_{0.2}Ga_{0.8}As$ buffer layers. Ten of these pairs are sandwiched between nominally undoped 3500 Å thick $Al_{0.2}Ga_{0.8}As$ layers. A reverse-bias field can be applied

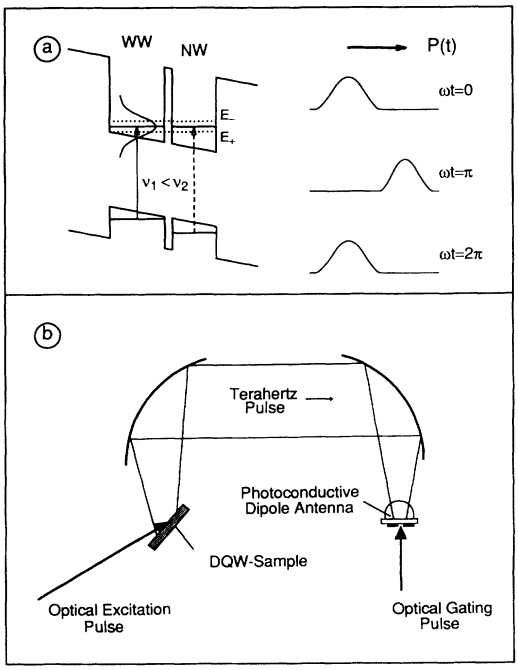

FIG. 1. (a) Schematic band diagram of the asymmetric coupled-quantum-well structure. Interband transitions in the wide well (WW) occur at lower photon energies than in the narrow well (NW), so that a short laser pulse with spectral width $\Delta\nu > (E_- - E_+)/h$ can excite an electronic wave packet that is mostly localized in the WW. A time-varying polarization $P(t)$ accompanies the tunneling of the wave packet back and forth between both wells that follows the excitation in the WW. (b) Schematic of the experimental setup to detect dipole radiation from the oscillating charge in the double quantum well.

between a semitransparent chromium contact on the sample surface and the n-doped substrate, with higher fields corresponding to the wide well being at a higher electron potential than the narrow well. In this asymmetric coupled-quantum-well structure, the first ($n=1$) electron levels in the wide well and the narrow well can be energetically aligned with each other by applying a field, while the corresponding $n=1$ hole levels [both heavy holes (hh) and light holes (lh)] are never aligned for this polarity of the field [Fig. 1(a)] [4]. The electrons then delocalize over both wells while the holes remain localized. The energy levels E_+ and E_- of the new bonding ($|+\rangle$) and antibonding ($|-\rangle$) eigenstates of the system follow anticrossing rules as a function of field, with the minimum splitting $\Delta E = E_- - E_+$ at resonance being proportional to the barrier coupling. A unique feature of the asymmetric double well is that the optical interband transition frequencies ν_1, ν_2 are different in the two wells. This allows us to prepare an electronic wave packet selectively in the wide well using a femtosecond optical pulse at ν_1, as long as its spectral bandwidth is larger than the splitting ΔE but smaller than the difference in the optical transition frequencies $\nu_2 - \nu_1$. This wave packet is a coherent superposition of the two eigenstates and tunnels back and forth between the two wells with a frequency $\Delta E/h$. A time-dependent polarization $P(t)$ is associated with the coherently oscillating wave packet, leading to dipole radiation with a radiated electromagnetic field $E(t) \sim \partial^2 P(t)/\partial t^2$.

The setup of our experiment to observe the dipole radiation from this oscillating charge is schematically shown in Fig. 1(b). In our geometry, the unfocused optical femtosecond beam from a self-mode-locked Ti-sapphire laser with a pulse duration (sech^2) of roughly 160 fs, and a full width at half maximum (FWHM) bandwidth of 7 nm, strikes the sample at an angle of roughly 45°. The sample is held in a continuous-flow cryostat at a temperature of 10 K. The optically excited sheet carrier density is kept below 5×10^9 cm^{-2} per double well to minimize phase relaxation from free carrier and exciton-exciton scattering. The terahertz radiation, emitted collinearly with the reflected optical excitation beam [5], leaves the cryostat through a high-resistivity silicon window, and is collected with a pair of off-axis paraboloids and detected by a 50-μm subpicosecond photoconductive dipole antenna [6]. This photoconductive dipole antenna is gated by a second, time-delayed portion of the femtosecond laser beam.

Figure 2 shows the measured coherent electromagnetic transients as a function of bias field obtained at a laser photon energy of 1.54 eV. Starting at 1.4 kV/cm, a small oscillatory electromagnetic signal with eight oscillation cycles can be detected. At 6.7 kV/cm, the amplitude of the oscillations has increased by a factor of 2 and we can distinguish fourteen cycles of the electromagnetic radiation. At this field, we deduce a dephasing time of 7 ps

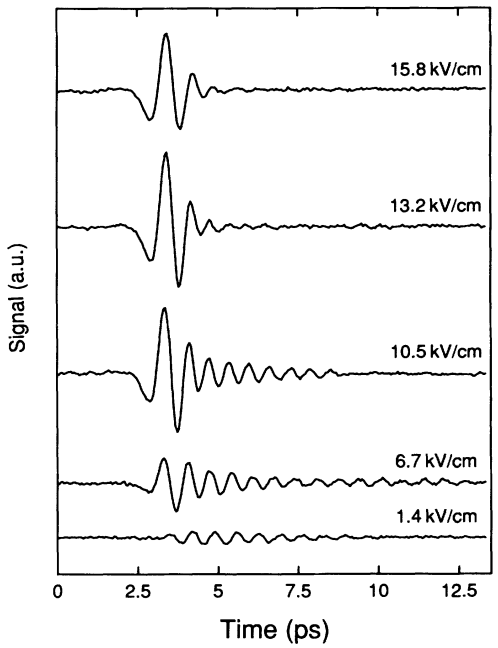

FIG. 2. Measured coherent electromagnetic transients emitted from the coupled quantum well for different bias fields for a photon energy of 1.54 eV.

from the exponential decay of the coherent electromagnetic signal. We observe the strongest oscillations around 10.5 kV/cm. For larger fields, the initial transient consisting of the first radiation cycle still rises, but the oscillations in the trailing part of the electromagnetic signal rapidly disappear and are absent at the largest field. The frequency of the oscillations is roughly constant up to 10.5 kV/cm, but increases by about 20% at 13.2 kV/cm. At 15.8 kV/cm, the amplitude of the oscillations has become so small that we cannot determine their frequency reliably. We note that the signal amplitude of the coherent terahertz transient at 10.5 kV/cm is comparatively large and amounts to roughly half the signal observed from a bulk indium phosphide surface [5] under identical conditions.

It is remarkable that the oscillation frequency changes so little over the reported range of fields, while we expect a parabolic dependence of the oscillation frequency as a function of field away from perfect alignment of the electron levels. The origin of this behavior lies in the excitonic interactions between the holes and electrons in the different wells, which lead to two distinct resonance conditions at different fields: For our sample, the holes remain localized for most of the field range [4], while the electrons can be either in the same well as the holes or in the adjacent well. The different excitonic binding energies of the associated direct and "spatially indirect" optical transitions [7,8] significantly change the fields at which a level resonance occurs for optical transitions in the WW and the NW, respectively [7,8]. Figure 3(a)

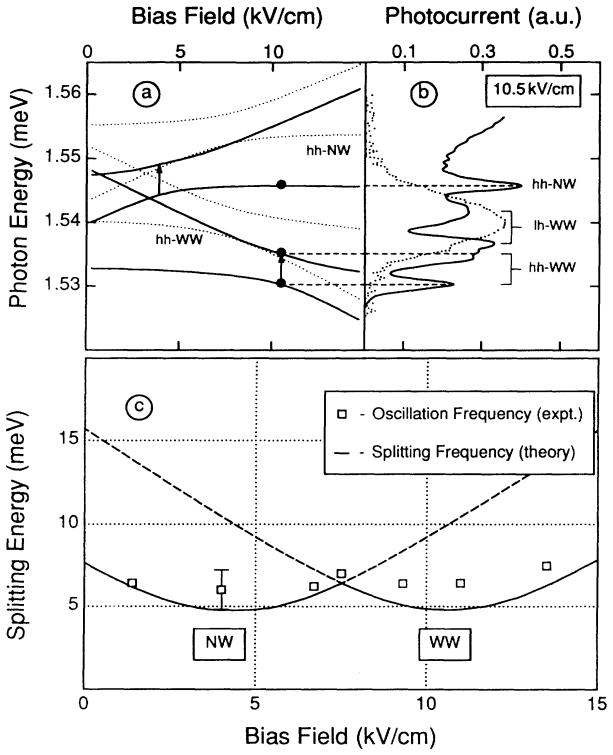

FIG. 3. (a) Energy diagram of the hh transitions in the NW and WW with (solid) and without (dotted) excitonic effects. (b) Photocurrent spectrum at a field of 10.5 kV/cm, showing the resonance splitting of the hh and lh transitions in the WW. The transition in the NW is not in resonance at this field. The dotted curve is the spectrum of the femtosecond optical pulse. (c) Experimentally measured oscillation frequency of the submillimeter-wave transients from all data, together with the theoretical splitting of both NW and WW transitions as a function of field.

shows a calculated level diagram for the heavy-hole transitions with and without the excitonic corrections. While we observe anticrossing of the excitonic transitions in the WW (both hh and lh) at 10.5 kV/cm, we obtain anticrossing of the NW excitons at 4 kV/cm. In Fig. 3(b), we plot the photocurrent spectrum of our sample at 10.5 kV/cm for the same photon energies as in Fig. 3(a), showing the resonance splitting for both hh and lh transitions in the WW, but no splitting for the NW transition. Similarly, at 4 kV/cm (not shown), the NW transitions show resonance splitting, while we only observe a single peak for both the hh and lh transitions in the WW. We do not distinguish other peaks from higher-order hole transitions within the displayed frequency range. The dotted curve is the spectrum of the optical excitation pulse. At this laser frequency, not only the heavy-hole but also the light-hole resonances in the wide well are excited [9]. The level diagram for the lh-WW transitions, which is omitted from Fig. 3(a) for clarity, is almost identical to the hh-WW diagram, but displaced 6.6 meV to higher energies. The lowest hh-NW transition, which is excited by the high-energy wing of the laser pulse, is completely localized in the NW at this bias field and hence does not lead to spatial oscillations of the charge. Figure 3(a) shows that, at 10.5 kV/cm, we mostly excite the delocalized transitions in the WW that set up wave-packet oscillations. However, when lowering the field to 4 kV/cm, we can also excite a coherent superposition of the then resonant transitions in the NW. Thus, for spectrally broad laser pulses, the consequence of the excitonic interactions is an increase in the effective field range over which resonance conditions and hence charge oscillations can occur. This view is supported by additional experiments (not shown) with laser pulses of 4-nm bandwidth, where only the WW transitions can be optically excited. Here, we do not observe any oscillations for bias fields smaller than roughly 6 kV/cm. In Fig. 3(c) we plot the calculated energy splitting for the WW and NW excitonic transitions as a function of field, together with the experimentally observed oscillation frequencies of the submillimeter-wave radiation. Within experimental uncertainties, the agreement of the observed oscillation frequencies with the excitonic model is quite good. We would also like to mention that, although there should be a field-induced dipole moment from the different polarizability of light and heavy holes, estimates show that this dipole moment should be much smaller than the dipole moment from the charge oscillations between coupled wells.

Fourier transformation of the time-domain data (Fig. 4) shows that the electromagnetic transient really consists

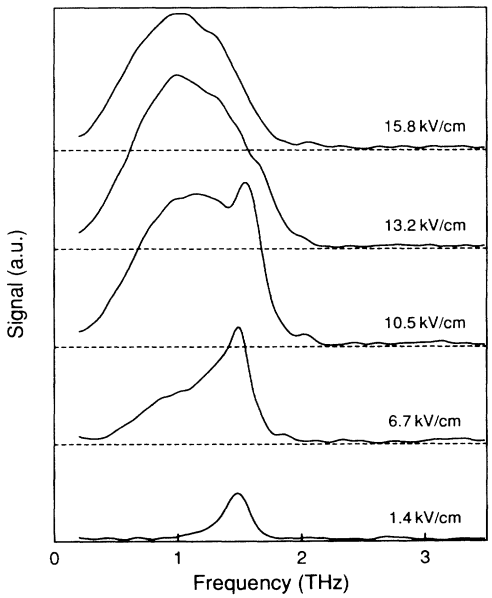

FIG. 4. Spectra of the wave forms in Fig. 2 obtained by Fourier transformation. In addition to the narrow-band signal from the charge oscillations, a broad-band contribution from the initial transient in the time domain can be distinguished.

of two distinct contributions: first, a spectrally narrow line around 1.5 THz corresponding to the oscillatory signal in the time domain, and second, a broad-band signal limited only by the bandwidth of the terahertz receiver dipole. The latter contribution corresponds to a short transient in the time domain similar to the one observed at the highest field in Fig. 2. At the WW resonance field (10.5 kV/cm), we can clearly distinguish between the narrow feature at 1.5 THz and the broad-band contribution. These two contributions to the signal behave completely differently as a function of field. The broad-band, instantaneous signal is only present at large electric fields and vanishes more or less linearly with field. When no field is present in the structure, the broad-band signal is absent from the spectrum and only a Lorentzian line at 1.5 THz corresponding to the charge oscillations remains.

The broad bandwidth and the associated short pulse width of the instantaneous contribution to the signal, together with the disappearance of this contribution when no field is present, suggest that this signal is the first observation of the nonresonant, field-induced contribution to the optical rectification signal that has recently been predicted theoretically [3]. We believe the origin of this signal is that, in an electric field, the electrons and holes of the created excitons are spatially displaced, meaning that the excitons are created in a polarized state. Hence, there is a polarization that follows the integral of the laser pulse energy, causing a radiated electromagnetic wave with a duration close to the optical pulse duration. The experimentally observed duration of this short transient is thus only limited by the finite response time of the photoconductive antenna, leading to a frequency spectrum that rolls off above 1 THz [6]. In a quantum well, this polarization is linear in field at low field but saturates at high field when the electron and hole wave functions accumulate at opposite well interfaces. In the linear regime, the polarization can be described by a field-induced optical rectification process:

$$P_0=\chi^{(3)}(0;0,-\omega,\omega)E_\omega^2E_0\,, \tag{1}$$

where P_0 is the static (submillimeter-wave) polarization, $\chi^{(3)}$ the third-order nonlinear polarizability [3], E_ω the optical field, and E_0 the dc bias field. We note that the excitons being created in a polarized state in a biased quantum well are also responsible for the quantum-confined Stark effect (QCSE) [10]. The dominant term in the QCSE shift comes indeed from the polarization of the exciton in the electric field [7]. The instantaneous electromagnetic pulse observed here and the QCSE are therefore closely related. Of course, the bulk equivalent of this effect must also be present in the terahertz signal emitted from bulk semiconductor surface fields [5], but it is experimentally difficult to distinguish from the transport term resulting from the acceleration of the photoexcited carriers. In our experiment, the quantum-well barriers do not permit free perpendicular transport on these time scales and we can separate the instantaneous polarization from transport contributions.

In conclusion, we have for the first time directly observed the electromagnetic radiation emitted from an oscillating exciton wave packet in a coupled-quantum-well structure after preparing the exciton in a single well with a femtosecond laser. In addition to this oscillating signal, we also identify for the first time an instantaneous contribution to the terahertz signal that is proportional to the electric field in the structure. This signal has its origin in the electron-hole pairs being created in a polarized state in an electrical field.

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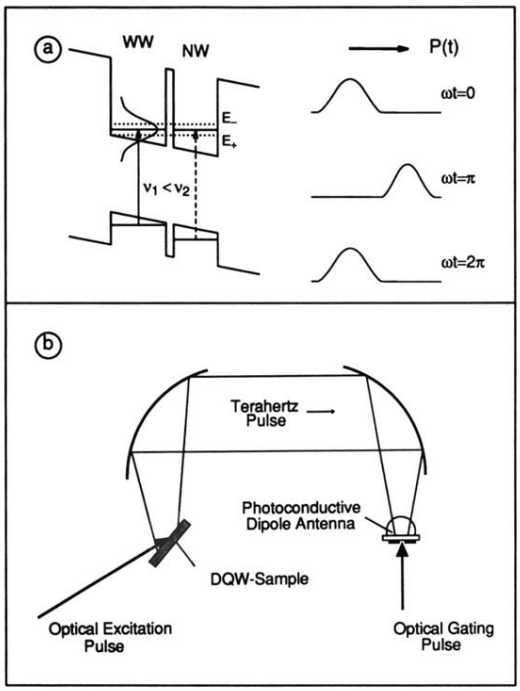

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