THz radiation from coherent population changes in quantum wells

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We identify the contributions to the terahertz (THz) signals from both the creation of polarized *uncorrelated electron-hole pairs* and polarized *excitons* in GaAs/$Al_{0.3}Ga_{0.7}As$ quantum wells. We find that the THz radiation from the creation of polarized excitons is comparable in magnitude to that from the creation of polarized uncorrelated *e-h* pairs. Unlike the THz transient from uncorrelated *e-h* pairs, the signal from polarized excitons is also strongly influenced by the combined effects of detuning and finite pulse duration.

Recently, there has been a lot of interest in the generation of terahertz (THz) bandwidth electromagnetic transients in semiconductors and semiconductor heterostructures.[1-4] When femtosecond optical pulses are used to resonantly excite quantum wells, freely propagating electromagnetic transients are generated with frequency components in the THz domain. Generally, there are two mechanisms that contribute to the emission of THz transients from quantum wells. The first is the optical excitation of excitonic quantum beats accompanied by charge oscillations that radiate THz-frequency wave forms, which typically show several oscillations of the electric field $E(t)$ before the phase coherence is destroyed.[1,2] The second is the optical excitation of excitons in polarized states, in which the center of charge of the electron and the center of charge of the hole are displaced with respect to each other. This gives a dc polarization $P(t)$ that grows as the integrated pulse energy and hence radiates only a single-cycle electrical transient $E(t)$ according to $E \propto \partial^2 P/\partial t^2$.[3,4] The excitonic origin of the THz radiation emitted by the quantum beats is evident from the decay times of a few picoseconds of the oscillatory radiation. This is because in quantum wells (at low temperatures), excitons dephase typically in several picoseconds whereas uncorrelated *e-h* pairs dephase in a few hundred femtoseconds or less.[5-7] For the single-cycle THz transients generated by the excitation of polarized excitons, long dephasing times are not essential as it is the change in the *population* of polarized *e-h* pairs, be they correlated (excitons) or uncorrelated, that counts. Once excitons are created however, their long dephasing time allows us to optically control their coherence and population, as was recently demonstrated in a coupled quantum well.[8]

Here we show how the same technique can be used to identify and distinguish between the *creation of polarized excitons and uncorrelated e-h pairs* as mechanisms for the generation of single-cycle THz transients. Two identical phase-locked optical pulses, having a time separation of a few picoseconds stabilized to within a fraction of a femtosecond for the duration of the experiment, excite a quantum-well sample. We find that the shape and magnitude of the THz transient, generated by the second optical pulse, depends on the phase difference between the exciting optical pulses. For overall pulse separations of a few picoseconds, this effect is very strong when the laser is tuned around the hh exciton but it is absent for excitation of uncorrelated *e-h* pairs in the two-dimensional (2D) continuum. The dependence of the THz signal on the optical phase difference results from the coherent changes in the exciton population when the optical coherence excited in the medium by the first optical pulse interacts with the electric field of the second. These coherent effects are practically absent for uncorrelated *e-h* pairs because they have a very short dephasing time of a few hundred femtoseconds or less. Our results prove that the single-cycle transients observed in Refs. 1, 2, and 4 for excitation around the band edge have significant contributions from *both* the creation of polarized excitons and polarized uncorrelated *e-h* pairs. When the laser excites the lowest hh-exciton state, we also find that a small detuning below the hh-exciton transition strongly modifies the emitted THz transient.

The sample is a metal-*i*-*n* Schottky diode with a multiple quantum well (MQW) in the intrinsic region. The MQW consists of 15 periods of 175 Å GaAs wells, separated by 150 Å $Al_{0.3}Ga_{0.7}As$ barriers. The sample is capped with a 200 Å GaAs layer and finally 50 Å of semitransparent chromium is deposited. We apply an electric field of 10.5 kV/cm to the sample by applying a voltage between the Schottky contact and the doped substrate. The light-hole–heavy-hole splitting at flatband is 6 meV or 1.4 THz. More information on the sample can be found in Ref. 2. The sample is mounted in a continuous-flow liquid-helium cryostat and the temperature is kept at around 10 K.

Our experimental setup is shown in Fig. 1. It uses

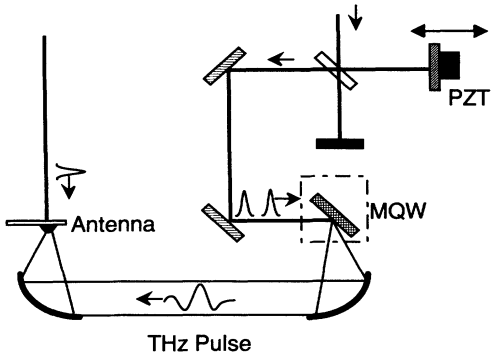

FIG. 1. Schematic of the experimental setup. Two phase-locked pulses from a Michelson interferometer illuminate the MQW sample and generate THz radiation. One of the end mirrors is mounted on a PZT to fine tune the optical delay.

pulses from a mode-locked, 82-MHz repetition rate, argon-ion pumped Ti:sapphire laser, tunable around 800 nm, and with a pulse duration of 80 fs. The laser beam is split in two. One beam enters a Michelson interferometer setup. The arms of the Michelson interferometer can be set to different delays so that the pulses emerging from the interferometer are separated in time. One of the end mirrors of the Michelson interferometer is also mounted onto a piezoelectric transducer (PZT) to actively stabilize the length of the arm to within a fraction of a wavelength. The pulse pair that emerges from the Michelson interferometer is weakly focused onto the quantum-well sample with its polarization in the plane of incidence. The generated THz beam is detected with a 50-μm photoconducting dipole antenna[9] with a silicon hyperhemispherical substrate lens, gated by the second laser beam. For excitation around the hh exciton we avoid the excitation of lh-hh quantum beats[2] by cutting off the high-energy side of the pulse spectrum between the lh- and hh-exciton transitions (see also Fig. 2) with a spectral filter. This process lengthens the pulses to a value of around 300 fs. We have verified that the electric fields of the two pulses emerging from the Michelson interferometer did not overlap in time for this pulse duration, in the experiments described below. For a given overall delay between the two optical pulses, we measure the THz signals for several values of the optical phase difference Φ between the two exciting pulses by fine tuning the delay with the PZT.

Figure 2 illustrates the two different excitation conditions presented here. We excite the quantum well (i) around the hh exciton, and (ii) 30 meV *above* the hh exciton, where we mainly excite excitons and uncorrelated *e-h* pairs, respectively. The effect of spectral filtering on a single laser pulse is evident at the high-energy side of the laser spectra in Fig. 2. For excitation of the hh exciton, the measured THz wave forms generated by the pulse pair are plotted in Fig. 3, for five overall delays between the two optical pulses and two different values of the optical phase difference. The laser is tuned to an energy approximately 5 meV below the hh-exciton transition where the laser pulse still has sufficient overlap with the hh exciton. The upper wave forms correspond to an optical phase difference where the THz amplitude from the second pulse is maximal, the lower wave forms correspond to a phase difference where it is minimal. The single-cycle transient around $t=1$ ps is generated when the first optical pulse strikes the sample. It neither depends on the overall delay between the two pulses nor on their phase difference. The situation is different for the transients generated by the *second,* delayed optical pulse. For example, for a pulse separation of 1.6 ps, the optical phase difference can be adjusted so that the THz transient (upper curve) generated by the second optical pulse

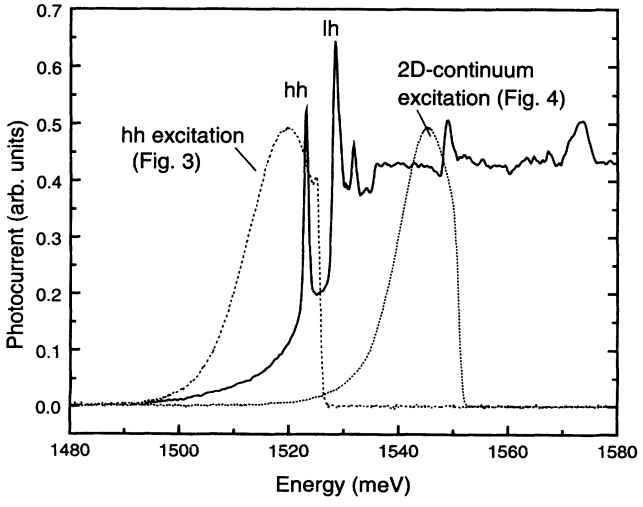

FIG. 2. Photocurrent spectrum of our sample (solid line) and filtered spectrum of a single laser pulse for the two different excitation conditions of Figs. 3 and 4 (dashed lines).

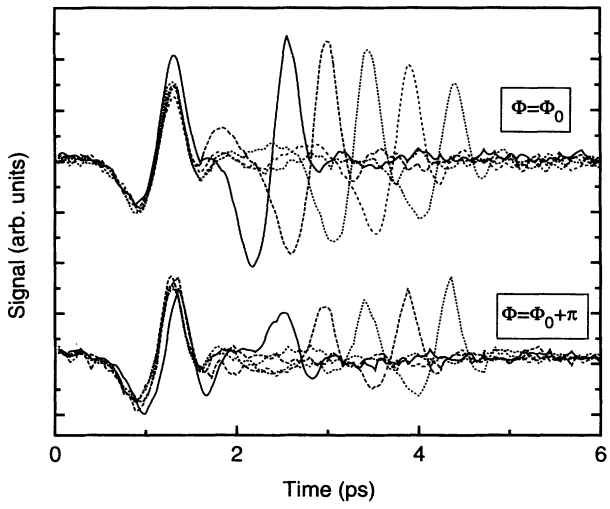

FIG. 3. Measured THz wave forms for excitation of hh excitons, for two phase differences and five pulse separations. The upper wave forms correspond to a phase difference for which the THz amplitude has a maximum. The lower wave forms correspond to a phase difference for which the THz signal has a minimum.

has a much larger amplitude than the transient generated by the first. Alternatively, the phase difference can be adjusted so that the THz amplitude of the second transient is minimal (lower curve) and smaller than the amplitude of the transient generated by the first pulse. When the time separation increases, these coherent effects gradually disappear. For a pulse separation longer than 3.5 ps, a change in the optical phase difference does not significantly influence the THz wave form anymore. In addition, the signal generated by the second pulse begins to resemble the one generated by the first. Note that a change in the optical phase differences of π is obtained by adjusting the time separation between the two pulses by 1.33 fs. This is a negligible change on the time scale depicted in Fig. 3.

For excitation of uncorrelated *e-h* pairs, 30 meV above the hh-exciton transition energy, the detected THz wave forms are plotted in Fig. 4. The upper curve is the THz signal generated when only the first pulse excites the sample. The wave form shows some ringing of the electric field caused by what we believe to be lh-hh absorption. The lower two wave forms are generated when both pulses excite the sample with a time separation of 1.66 ps and two different values of the optical phase difference. Care is taken to keep the temporal profile of the exciting laser pulses the same by cutting off a similar fraction of the laser pulse spectrum with the spectral filter as was done for excitation around the band edge (Fig. 2). The amplitude of the generated THz signals is comparable to the amplitude measured for excitation of hh excitons (Fig. 3). However, in sharp contrast to the results described above, Fig. 4 demonstrates that for a pulse separation of 1.66 ps there is *no* measurable influence of the optical phase difference on the THz signal generated by the second pulse.

To further elucidate the difference between the two excitation conditions, we plot in Fig. 5 the peak amplitude of the THz pulse generated by the second pulse as a function of the phase difference between the two optical pulses. The time separation between the laser pulses is 1.83 ps. The figure clearly shows the strong modulation of the THz amplitude for excitation around the heavy-hole exciton (black squares), and the absence of modulation for excitation 30 meV above the heavy-hole excitons in the 2D continuum (open circles).

The origin of the observed coherent effects in the emission of the THz transients is the interaction of the electric field of the second optical pulse with the coherence excited in the medium by the first. This gives rise to phase-difference-dependent changes in the *population* of polarized excitons and hence leads to the emission of a THz transient. In a two-level system, using a density-matrix formulation, we can write for the time-dependent far-infrared polarization in the z direction: $P(t) \propto |e|(z_{22}-z_{11})\rho_{22}(t)$, where the self-dipole moments are defined as $z_{22}=\langle \mathrm{hh}|z|\mathrm{hh}\rangle$ and $z_{11}=\langle e|z|e\rangle$, with $(z_{22}-z_{11})$ the net displacement between the heavy hole and the electron, and $\rho_{22}(t)$ is the time-dependent heavy-hole exciton population. From $P(t)$, we obtain the electric field of the far-infrared emission according to $E \propto \partial^2 P/\partial t^2$. Assuming δ-pulse excitation, infinite exciton lifetimes, and writing for the envelope function of the optical electric field of the pulse pair $E_L(t) = E_0\delta(t)+E_0 e^{i\Phi}\delta(t-\tau)$, an expansion of ρ_{22} to second order in electric field under the rotating-wave approximation yields[10]

$$\rho_{22}^{(2)}(t)=\frac{|\mu_{21}|^2|E_0|^2}{\hbar^2}\{\theta(t)+\theta(t-\tau)+e^{-i\Phi}e^{-(i\Delta_{21}+1/T_{21})T}\theta(t-\tau)+e^{i\Phi}e^{(i\Delta_{21}-1/T_{21})T}\theta(t-\tau)\}\ , \tag{1}$$

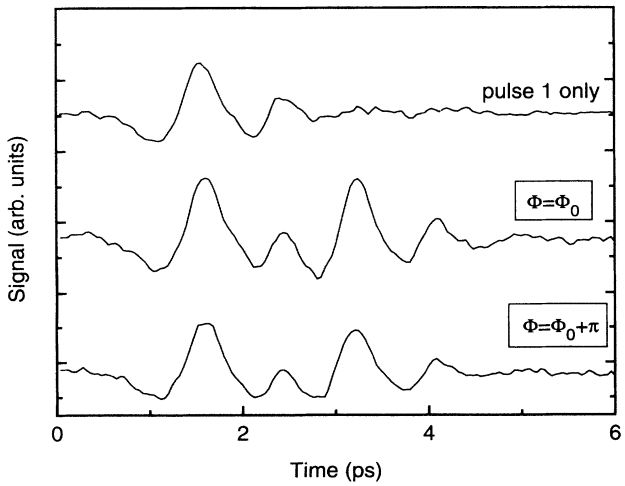

FIG. 4. Measured THz wave forms for excitation 30 meV above the hh exciton. The upper curve is generated by only the first pulse. The lower two are generated by the phase-locked pulse pair for a time separation of 1.66 ps and two phase differences.

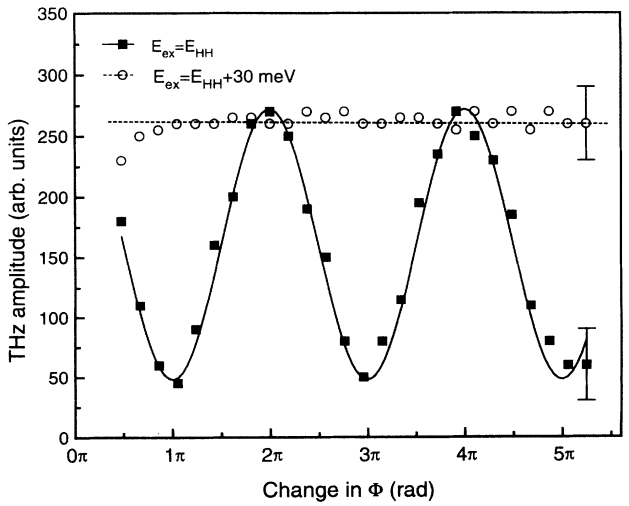

FIG. 5. Variation of the generated THz transient with the phase-difference Φ between the two phase-locked optical pulses, for excitation of hh excitons (squares) and for excitation in the 2D continuum, 30 meV above the hh-exciton transition (circles).

where Φ is the optical phase difference, $\theta(t)$ and $\theta(t-\tau)$ are Heaviside step functions, $\Delta_{21}=\omega_{21}-\Omega_1$ the frequency detunings of the laser, μ_{21} the interband optical transition-dipole moment, and T_{21} the dephasing time. The first two terms within the curly brackets in Eq. (1) describe the successive increases in the exciton population due to the excitation with the two pulses, not taking coherent effects into account. The other two terms give changes in the exciton population that result from the interference between the second pulse and the coherence excited in the medium by the first one. They depend on the optical phase difference Φ and are only significant when the decay time T_{21} of the coherence is comparable to or longer than the time-separation τ between the laser pulses. For $\Phi=0$, $\Delta_{21}=0$, and $T_{21}\rightarrow\infty$, all four terms add up constructively, corresponding to a population of polarized excitons that is larger by a factor of 4 compared to the population excited by only the first pulse. For the THz signal, only the population *change* rather than the *absolute value* of the population is relevant. Accordingly, the THz signal will have an amplitude *three* times larger (and not four times) than that generated by the first pulse. For $\Phi=\pi$, $\Delta_{21}=0$, and $T_{21}\rightarrow\infty$, the last two terms exactly cancel the first two terms corresponding to the effective depopulation of polarized hh excitons by the second pulse and the subsequent emission of a THz transient identical in shape to the one generated by the first pulse, but inverted in sign. Note that the population changes and the emitted THz radiation amplitudes will be smaller if we allow the dephasing time in Eq. (1) to have a finite value of a few picoseconds, consistent with our measured results (Fig. 3). We are now also in a position to understand the absence of any coherent effects for excitation of uncorrelated *e-h* pairs. Dephasing times of uncorrelated *e-h* pairs are several hundred femtoseconds or less, giving a rapid decay of the optical coherence excited in the medium by the first pulse.[5-7] There will, therefore, be no coherent interaction with the electric field of the second pulse and both pulses will excite practically identical numbers of polarized *e-h* pairs.

Equation (1) describes many features observed in our experiments, but for a more complete analysis we have to incorporate effects of detuning *and* a finite pulse duration in our description. For instance, the absence of a clear sign reversal in our measurements shown in Fig. 3 can only be explained by the time-domain picture of the combined effects of detuning, a narrow exciton linewidth (or long dephasing time), *and a finite pulse duration.* When the central frequency of the laser pulse is tuned a few meV below the hh-exciton transition energy, the exciton optical coherence excited by the first pulse oscillates at a different frequency than the electric field of the second pulse. The phase difference between the two optical pulses changes within the duration of the laser pulse and complete deexcitation becomes impossible. A numerical calculation of the exciton population versus time that demonstrates the importance of detuning and a finite pulse duration is shown in Fig. 6 for initial optical phase differences of $\Phi=0$ and π. The calculation is based on a two-level density-matrix formulation. We assume an exciton dephasing time of 2 ps and a pulse separation of 1.66 ps. The calculation clearly shows the more complicated time evolution of the exciton population for a detuning of 5 meV below the hh exciton, such as the absence of complete deexcitation of the exciton population for $\Phi=\pi$.

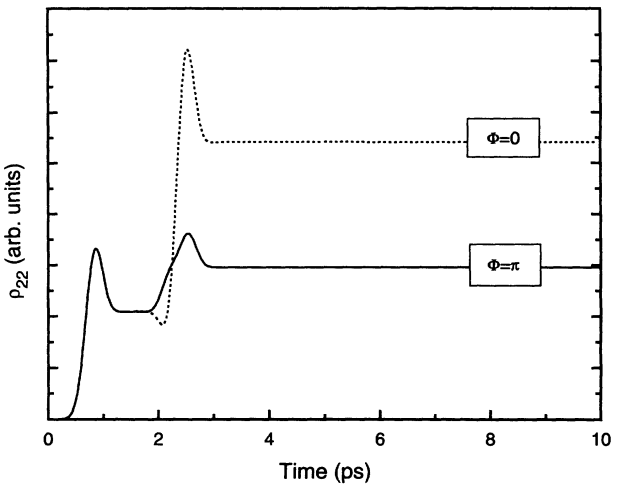

FIG. 6. Calculated time dependence of the hh-exciton population ρ_{22} for excitation with a phase-locked pulse pair, for two values of the optical phase difference Φ. The pulses are tuned to an energy 5 meV below the hh-exciton transition energy and have a time separation of 1.66 ps. The hh-exciton dephasing time is 2 ps.

For large enough detunings, few excitons remain after the laser pulse is gone. *During* the excitation, however, excitons are excited and deexcited again giving a temporary (virtual) population at any time during the pulse proportional to the instantaneous intensity. Although little net population remains when the pulse is gone, the change in the population of polarized excitons during the excitation should lead to the emission of a tetrahertz transient. The concept of virtual photoexcitation in semiconductors was described previously.[11,12] Some experimental evidence that this leads to the emission of a THz transient in bulk GaAs was reported in Ref. 13.

Our measurements show that fast changes in the population of polarized excitons will generate THz radiation. This makes a time-domain description of the excitation necessary. The generation of THz radiation is a resonant, *nonlinear,* second-order optical rectification process. A frequency-domain picture that compares the intensity spectrum of the exciting pulse pair with the absorption spectrum of the quantum-well sample is therefore not sufficient to understand our results.

In conclusion, we have used femtosecond phase-locked optical pulses to identify both the creation of polarized excitons and polarized uncorrelated *e-h* pairs as sources of single-cycle THz transients. For broadband excitation around the band edge this implies that both mechanisms contribute to the transient, but behave rather differently. For example, for exciton excitation, we show that detun-

ing below the narrow exciton line gives rise to the emission of more complicated wave forms.

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