Repetitive excitation of charge oscillations in semiconductor heterostructures

I. Brener, P. C. M. Planken, and M. C. Nuss
AT&T Bell Laboratories, Holmdel, New Jersey 07733

L. Pfieffer
AT&T Bell Laboratories, Murray Hill, New Jersey 07974

D. E. Leaird
Bellcore, Red Bank, New Jersey 07701

A. M. Weiner
Purdue University, School of Electrical Engineering, West Lafayette, Indiana 47907-1285

(Received 28 June 1993; accepted for publication 16 August 1993)

We create forced excitonic charge oscillations in semiconductor heterostructures by excitation with periodic optical pulse sequences. The far-infrared radiation that accompanies the charge oscillations shows temporal interference patterns for pulse sequences with different optical phase profiles, providing evidence for phase induced population transfers between exciton levels in a semiconductor.

Femtosecond spectroscopy has been traditionally employed to measure ultrafast processes in a variety of physical systems. More recent applications of short laser pulses concentrate on controlling vibrational and chemical processes through arbitrarily shaped pulses.1,2 These ideas are mostly proposed and tested on atomic and molecular systems. However, excitonic levels in semiconductor heterostructures can provide a test ground for some of these ideas for a variety of reasons: band gap engineering allows for a relatively easy design of a many level system; dephasing times of these levels can be as long as a few picoseconds at low temperatures. Also, breaking the dipole selection rules is easily achieved and thus more optical fields can be involved in the experiment. Two recent examples of a quasi-three-level system in heterostructures are the bonding and antibonding exciton states in a double coupled quantum well (DCQW)3,4 and the heavy- (hh) and light-hole (lh) exciton states in a single quantum well (QW).5 In the two latter structures, quantum beats have been observed when a coherent superposition of both exciton states was excited with a short, broadband laser pulse. However, due to the presence of an allowed dipole transition between these two exciton states, these quantum beats are followed by emission of far-infrared radiation (FIR). The combination of quantum beats and an oscillating dipole moment has been termed charge oscillations. The presence or absence of the extra electromagnetic field makes a clear distinction between quantum beats with and without charge oscillations.

In this letter we report on the repetitive excitation of excitonic charge oscillations in single and double coupled semiconductor QWs using pulse sequences with variable repetition rates. When the time separation between the pulses in the sequence is less than the dephasing time, we find that coherent buildup of the oscillation amplitude only occurs when the optical phase variations within the pulse sequence are small. In all other situations, we observe temporal interferences in the photoexcited wave functions. These quantum interferences not only influence the temporal behavior of the emitted FIR radiation, but also the exciton populations. Our experiments provide the first evidence for such phase-dependent population transfers between exciton levels in a semiconductor. Furthermore, our results show that the concepts of molecular and coherence control through arbitrarily shaped pulses1 can be further tested in properly designed semiconductor heterostructures simulating an n-level system.

Two GaAs/Al,Ga1-xAs samples were used in this work: (a) a DCQW consisting of ten repetitions of a 10-nm narrow well (NW) and a 14.5-nm-wide well separated by a 2.5-nm Al0.2Ga0.8As barrier, and (b) a multiple QW sample consisting of 15 periods of 17.5-nm GaAs wells separated by 15 nm of Al0.3Ga0.7As barriers. Both samples had semitransparent chromium contacts and a variable electric field could be applied between the Schottky contact and the n-doped substrate. All experiments were carried out at 10 K. A schematic representation of the energy levels in both samples is presented in Figs. 1(a) and (b). The exciton levels in both samples behave as quasi-three-level systems. In sample (a) the two excited upper states |1⟩ and |2⟩ are the bonding and the antibonding exciton levels of the NW; in sample (b) these are the heavy- and light-hole exciton levels. In these samples, dipole transitions are allowed between all three levels (even with no applied electric field). This is in contrast with atomic and molecular systems where, in most cases, optical selection rules makes a dipole transition between the upper two states forbidden. When levels |1⟩ and |2⟩ are excited coherently with a broadband short laser pulse, charge oscillations due to quantum beats between these two levels occur with a beating period of ΔE/h, where ΔE=E1−E2. These charge oscillations produce a time-dependent polarization P(t) which in turn leads to a radiated electromagnetic field given by E(t)∝δP(t)/∂t2.

The optical pulses used in our experiments are generated in a Ti:sapphire laser which produces ~80 fs pulses tunable around 800 nm. Part of the beam passes through a pulse shaper used to generate the periodic pulse trains as depicted in Fig. 1(c). It consists of two gratings and two...
lenses arranged as a unity magnification system. A fused silica phase mask which contains phase patterns fabricated using microfabrication and reactive ion etching is placed in the Fourier plane of the pulse shaper in order to modify the electric field spectrum so as to produce the desired pulse train. The spatial frequency of the phase mask determines the frequency of the pulse train which can be tuned from 1.4 to 2.6 THz by translating different phase patterns with different spatial frequencies into the beam. The pulse train is slightly focused on the sample. The generated THz radiation is collected and collimated with a pair of off-axis parabolic mirrors and focused onto a 50-μm photoconducting dipole antenna with a hyperhemispherical silicon substrate lens. A fraction of the original laser beam is delayed and used to gate the dipole antenna. The electric field $E(t)$ emitted from the sample is measured by recording the photocurrent from the antenna as a function of the delay between the gating pulse and the pulse train.

Figure 2(a) shows the detected THz transient radiated from the DCQW sample after optical excitation with a single pulse and when no internal electric field is present. The excitation wavelength is chosen so that the laser spectrum overlaps the $E_+$ and $E_-$ exciton states of the NW. We observe quantum beats with a period of 1.52 THz which decay with a time constant of $\sim 3$ ps, then the phase mask is adjusted so that the repetition rate of the pulse train matches the period of the quantum beats. The resulting pulse train is shown in the inset of the lower pane. Figure 2(b) shows the measured THz radiation when a 1.52-THz pulse train having the same integrated intensity as in (a) excites the sample. The delay axes are identical for both cases. Two features are immediately striking: (i) the THz radiation emitted in case (b) reaches almost the same maximum value as in the case of single pulse excitation, despite the fact that the individual pulses in the pulse train have less than $1/12$ of the intensity of the single pulse used in (a)—this is a clear indication of forced coherent charge oscillations; (ii) there is almost no THz radiation for negative delay times even though the pulse train extends symmetrically to both positive and negative delay times. The former fact is critically dependent on the lateral position of the phase mask, as evidenced by Fig. 2(c). The signal in Fig. 2(c) was obtained under the same conditions as the trace in Fig. 2(a), but with the phase mask translated horizontally in the Fourier plane by a few μm. This operation changes the optical phases of the individual pulses; however, the repetition rate, fluence, and envelope of the pulse train remain constant. Thus, the far-infrared radiation signature strongly depends on the optical phase of the excitation sequence. As shown in Fig. 2(b), the quantum beats extend to longer delay times as compared to the case of single pulse excitation. This happens only when the pulse sequence repetition rate matches the quantum beat period. The coherent addition of the THz radiation when exciting with the pulse train and the longer duration of the beats are indicative of sustained charge oscillations. All the main features shown in Fig. 2 are also reproduced in the single QW sample when the laser spectrum overlaps the $hh$ and $lh$ exciton transitions, with the only differences of a shorter dephasing time and a quantum beat frequency of 1.4 THz.

In order to understand the results displayed in Fig. 2, we have to look in more detail into the nature of the pulses...
states and caused by excitation with a train with this particular phase sequence; (b) calculated populations of the excited states.

The absence of THz radiation for almost half of the excitation sequence is due to absence of population buildup of the excited states \( |1\rangle \) and \( |2\rangle \) caused by excitation with a train with this particular phase sequence; (b) calculated populations of the excited states.

FIG. 3. (a) Experimental THz transient (dots) for a particular horizontal position for the phase mask and the model calculation as derived from \( \rho_{12}(t) \) (solid line). The absence of THz radiation for almost half of the excitation sequence is due to absence of population buildup of the excited states \( |1\rangle \) and \( |2\rangle \) caused by excitation with a train with this particular phase sequence; (b) calculated populations of the excited states.

The amplitude and phase of the optical pulse train are generated by phase filtering and into the Bloch equations which describe the response of these quasi-three-level systems to the optical excitation. Each pulse in the train has a different optical phase determined by the design of the mask and its relative position in the Fourier plane. When a train of pulses with different optical phases excites an n-level system, quantum interferences of the excited states wave functions are expected if the delay between the individual pulses is less than the dephasing time. The measurement of the amplitude and phase of the far-infrared radiation provides a direct probe of the time evolution of the different wave functions. We solve numerically the linearized optical Bloch equations for a three-level system using the density matrix formalism. For simplicity, we assume an equal dephasing time for all transitions (\( T_{12} = T_{13} = T_{23} \)). The amplitude and phase of the optical pulse train are calculated using Fourier analysis and assuming a Gaussian pulse entering the pulse shaper with a full width at half-maximum (FWHM) of 80 fs. As the lifetime parameters are orders of magnitude longer than the dephasing time, our calculations have only one adjustable parameter, the dephasing time \( T_{12} \). The radiated THz field is given by \( E_{\text{FIR}} \propto \partial^2 \text{Re} (\rho_{12}) / \partial t^2 \), with \( \rho_{12} \) being the off-diagonal density matrix term. Figure 3(a) shows the fit to the experimental data for one particular position of the phase mask. We obtain an excellent agreement with \( T_{12} = 2.7 \) ps. The important features are remarkably well reproduced by the theory such as the canceling of the THz radiation for the first half of the pulse train and the sudden buildup around \( \Delta t \sim 0 \). We would like to point out here that the cancellations in the coherence \( \rho_{12} \) of the wave functions are also accompanied by a cancellation in the exciton populations. This can be seen from Fig. 3(b), where we plot the calculated time evolution of the populations of both excited states, \( \rho_{11} \) and \( \rho_{22} \). Note the cancellation of the populations for the first half of the pulse train which is caused by quantum interferences between the wave functions. This lack of population buildup despite optical excitation by many pulses is the cause of the absence of THz radiation for almost half of the pulse train. In our experiment, we can demonstrate the influence of the optical phases on the emitted THz radiation and thus on the exciton populations by displacing the phase mask horizontally in the Fourier plane, and the result is seen in Fig. 2(c). Our experiments provide the first evidence for phase-dependent population transfers between exciton levels in a semiconductor.

In summary, we have observed forced charge oscillations in single and double QWs. They are obtained after optical excitation by periodic pulse sequences with a repetition rate that matches the quantum beat period in these quasi-three-level systems. The dependence of the emitted far-infrared radiation on the optical phases of the pulses provides the first evidence for phase-induced population transfers between exciton levels in semiconductors. Thus, the concepts of molecular and coherence control through arbitrarily shaped pulses can be further tested in properly designed semiconductor heterostructures.

We thank K. Köhler for providing the DCQW sample, G. E. Doran and B. Tell for technical assistance, and W. H. Knox, D. A. B. Miller, and S. L. Chuang for valuable discussions.

8. In previous experiments using similar periodic pulse trains (Ref. 2) phase-dependent effects were absent since the experiments were performed off resonance and thus the matter-radiation field interaction was proportional to the intensity of the optical field.
9. Obviously these three dephasing times are related to each other and \( T_{13} = T_{23} \). Calculations assuming different parameters show that the physical picture is unchanged.