Coherent nonlinear propagation of ultrafast electric field transients through intersubband resonances

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Amplitude and phase-controlled midinfrared field transients at a wavelength of 12.5 μm induce resonant intersubband excitations in n-type modulation-doped GaAs/AlGaAs quantum wells. The transmitted electric field transients are directly measured by ultrafast electro-optic sampling. Coherent control of intersubband excitations is demonstrated by applying two phase-locked pulses with variable relative phase. A coherent nonlinear response corresponding to partial Rabi flops of up to 60° in the Bloch sphere is observed with excitation pulses of only 1 pJ energy. © *2001 American Institute of Physics.* [DOI: 10.1063/1.1384898]

Low-dimensional semiconductors are important model systems for implementing optical switching[1] and modulation[2] as well as coherent control of material excitations on ultrafast time scales.[3] So far, most research has concentrated on interband and/or excitonic excitations in quasi-two-dimensional quantum wells (QWs) for which different schemes of nonlinear optical switching and coherent control of optical polarizations and photoinduced electric currents[4] have been realized.

Intersubband (IS) transitions between consecutive valence or conduction subbands in QWs display a very high absorption cross section of up to 10^{-13} cm^2 at midinfrared (MIR) wavelengths and—for an energy separation larger than an optical phonon—a carrier lifetime in the excited subband between several hundreds of femtoseconds and a few picoseconds.[5] These properties make IS transitions interesting for optical switching with ultrafast switch-on and switch-off times. Moreover, recent developments like controlled shaping[6,7] and time-resolved detection[8] of electric field transients in the MIR spectral range ($\lambda = 5-20 \ \mu$m) allow to investigate and directly manipulate coherent IS excitations in the time domain.

So far, concepts for optical switching involving IS excitations have been investigated in a number of theoretical studies.[9,10] In quasistationary experiments, the incoherent saturation of IS absorption[5] as well as coherent nonlinear coupling of IS excitations and excitonic transitions[11] and coherent control of photocurrents by simultaneous one- and two-photon excitation of IS transitions[12] have been demonstrated. In contrast, only few experiments addressed the properties of coherent IS polarizations[13,14] and their application for coherent optical switching. In this letter, we demonstrate coherent nonlinear manipulation of IS polarizations in GaAs/AlGaAs QWs by extremely weak ultrashort electric field-transients resonant to the IS transition. Coherent polarizations on the transition between the $n=1$ and $n=2$ conduction subband are generated by shaped MIR transients at a megahertz repetition rate and their coherent emission is amplitude- and phase-resolved by electro-optic sampling. Even for subpicojoule pulse energies, we find a strong nonlinear saturation of the polarization amplitude with increasing excitation field and an excitation-independent polarization decay with a dephasing time of 320 fs.

Our experimental setup has been detailed in Fig. 1(a) of Ref. 7. Near-infrared 13 fs pulses centered at a wavelength of 780 nm are generated in a cavity-dumped mode-locked Ti:sapphire oscillator working at a 2 MHz repetition rate.[15] The pulses are amplitude- and phase-shaped in a programmable pulse shaper[16] and generate tailored MIR transients by phasematched difference-frequency mixing in an 0.5-mm-thick GaSe crystal.[6,7] The time-dependent electric field of such transients is measured directly by ultrafast free-space electro-optic sampling.[8] Near-infrared reference pulses of 17 fs duration are derived from the Ti:sapphire laser and transmitted through a 10-μm-thick ZnTe crystal in which the MIR transients induce a polarization rotation of the reference pulses proportional to the momentary MIR electric field. This signal measured in a balanced photodiode setup as a function of time delay between reference and MIR pulses is directly proportional to the MIR electric field. In the present study, shaped MIR pulses at $\lambda = 12.5 \ \mu$m excite a n-type modulation-doped sample consisting of 51 GaAs QWs of 10 nm width separated by 20-nm-thick Al$_{0.35}$Ga$_{0.65}$As barriers (electron density $n_s = 5 \times 10^{10}$ cm^{-2}). The sample was processed into a prism [Fig. 1(c)] to achieve a strong coupling between the p-polarized pulses and the IS transition dipoles. The $n=1$ to $n=2$ IS absorption spectrum [Fig. 1(a)] displays a very small linewidth of $\Delta E_{IS} = 3.7$ meV (full width at half maximum). Experiments are performed at $T = 15$ K.

First, we discuss the coherent response of the sample in the linear regime. In Fig. 2, we present data for applying two phase-locked MIR pulses with 400 fs separation and relative phases of (a),(b) $\Delta\Phi = 0$ and (c),(d) $\Delta\Phi = \pi$. In the upper and middle panels of Figs. 2(a) and 2(c), electric field transients sent through an undoped reference sample of identical prism shape and pulses propagated through the QW sample are

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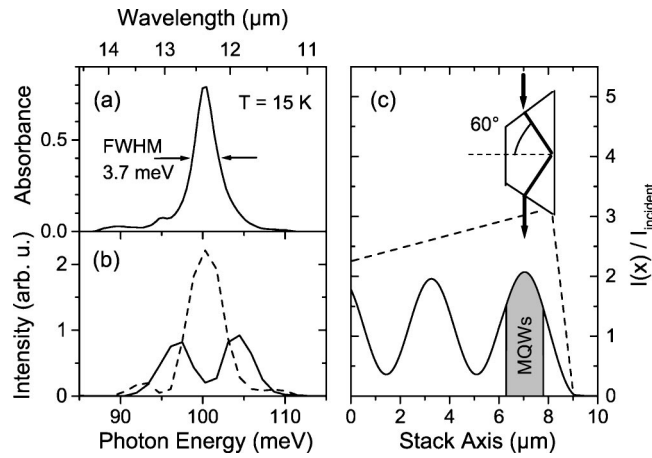
FIG. 1. (a) Intersubband absorption line of n-type modulation-doped GaAs/AlGaAs quantum wells measured with a Fourier-transform spectrometer. (b) Spectra of two phase-locked MIR pulses with 400 fs separation and relative phases of $\Delta\Phi=0$ (dashed line) and $\Delta\Phi=\pi$ (solid line). (c) Calculated intensity pattern of the standing wave arising close to the right interface of the prism shaped sample due to total internal reflection. In this geometry the electric field and consequently the actual Rabi frequency experienced by the electrons are almost identical for all individual quantum wells.
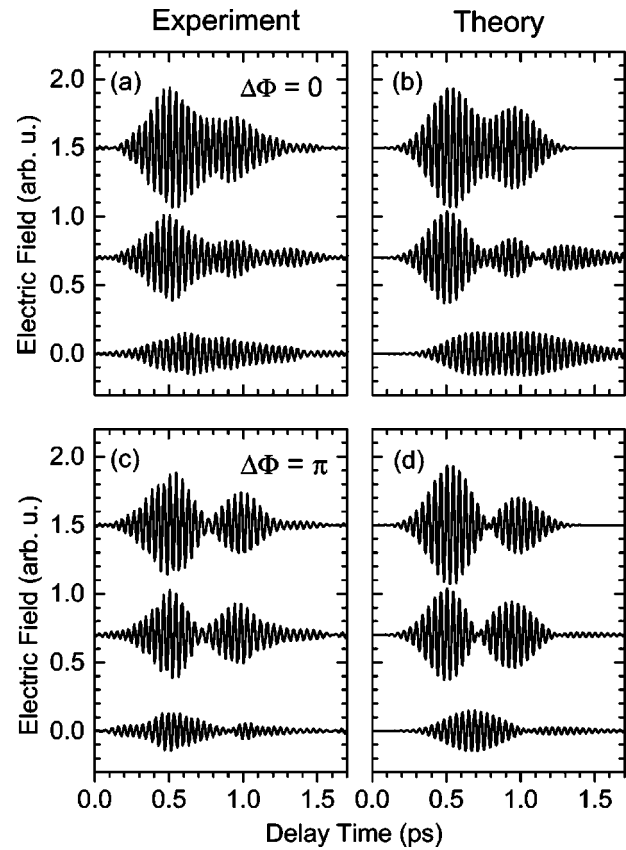

FIG. 2. Coherent control of IS excitation using two phase-locked MIR pulses with 400 fs separation and relative phases of (a),(b) $\Delta\Phi=0$ and (c),(d) $\Delta\Phi=\pi$. Each panel shows the incident pulses (upper traces), the pulses sent through the quantum well sample (middle traces), and the respective difference (lower traces) being the reemitted FID of the coherent IS polarization. Left panels: Electric field transients obtained with ultrafast electro-optic sampling. Right panels: Model calculations based on Maxwell–Bloch equations.

presented, respectively. In the lower panels, we plot the difference transients which represent the re-emitted free induction decay (FID) of the excited IS transition. For phase-locked pulses with a relative phase $\Delta\Phi=0$ [Fig. 2(a)] the coherent polarization created by the second pulse adds to the coherent excitation left from the first pulse resulting in a pronounced prolongation of the FID. Correspondingly, the spectrum of the applied field transient [dashed line in Fig. 1(b)] has its maximum at the IS resonance [cf. Fig. 1(a)]. For $\Delta\Phi=\pi$ [Fig. 2(c)], however, destructive interference between the FIDs induced by the two MIR pulses results in a strong reduction of the amplitude of the coherent IS excitation after the second pulse. In order to compensate for the partial decay of the coherent excitation in response to the first pulse the amplitude of the second excitation pulse has been adjusted to a smaller value. In our experiment [Fig. 2(c)] we have not achieved a complete cancellation of the two destructively interfering FIDs. The corresponding spectrum of the excitation pulses [solid line: Fig. 1(b)] shows a pronounced dip at the IS resonance.

In Figs. 2(b) and 2(d), the results of model calculations based on Maxwell–Bloch equations are shown. Such calculations, which will be discussed in more detail below, are in excellent agreement with the data of Figs. 2(a) and 2(c). Our results clearly demonstrate the feasability of coherent control of IS excitations with subpicosecond dephasing times by weak midinfrared transients. Related techniques have been applied in the near-infrared for a coherent control of excitonic polarizations which is facilitated by the much longer dephasing times of up to 10 ps.[3]

Next, we present results for excitation with single MIR transients. Experimental data for small electric field amplitude (linear response) and for stronger excitation are shown in Figs. 3(a) and 3(b), respectively (note the *different* ordinate scales). With increasing amplitude of the exciting electric field, one finds a pronounced saturation of the emitted field strength. Amplitude data as a function of the incoming field are summarized in Fig. 3(c) (symbols). Each circle has

been measured in an independent experiment. We want to stress the fact that the observed nonlinearity corresponds to an excitation of $\approx$30% of ($n=1$) electrons to the ($n=2$) subband and is caused by *extremely weak* excitation pulses with energies of ≤ 1 pJ.

In contrast to the saturation of the FID amplitude, the decay time of the coherent reemission of approximately 300 fs shows only minor changes with increasing excitation field. This behavior is in very good agreement with recent four-wave-mixing experiments performed with the same sample.[17] Such measurements give a decay time of the macroscopic IS polarization of $T_2=320$ fs which remains unchanged for excitation densities between 0.5% and 30% of the total electron concentration. An analysis of the time-resolved four-wave-mixing signal demonstrates a predominant homogeneous broadening of the IS transition [Fig. 1(a)] with a linewidth of 3.7 meV fully accounted for by the measured T_2 value. Consequently, the coherent reemission studied here exhibits a decay determined by T_2. In MIR pump-probe experiments a population relaxation time of $T_1=550$ fs was determined.[17]

The experimental results were analyzed by model calculations using the Maxwell–Bloch equations for independent homogeneously broadened two-level systems. This single particle approach is appropriate as both subband nonparabo-
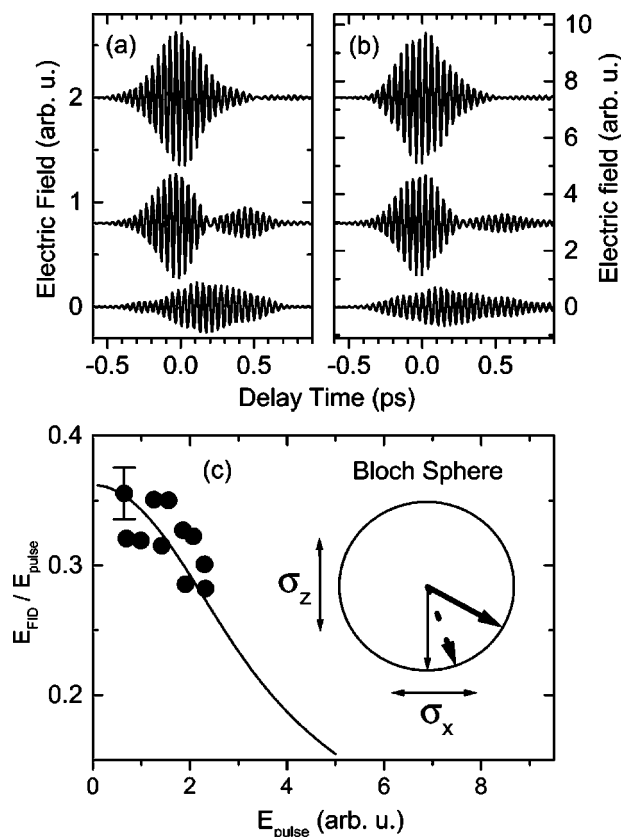
FIG. 3. (a),(b) Measured electric field transients and respective FID of the IS excitation in response to a single pulse for (a) low and (b) high excitation fields. (c) Symbols: Nonlinear ratio between the field amplitudes of the reemitted E_{FID} and excitation pulses E_{pulse} as a function of E_{pulse}. Solid line: Model calculation using Maxwell–Bloch equations. Inset: Schematic of Bloch sphere.

licities and many-body effects caused by the Coulomb interaction among the electrons play a minor role for the low electron concentration of our sample ($n_s = 5 \times 10^{10}$ cm^{-2}, Fermi energy 2 meV at 15 K).[17]

Our theoretical description is similar to that describing self-induced transparency experiments.[18] In the prism geometry applied in our measurements [Fig. 1(c)], the electric field and consequently the actual Rabi frequency experienced by the electrons is nearly identical for all individual quantum wells, as is evident from the calculated intensity pattern in Fig. 1(c). This fact allows for a simplified theoretical description with a position independent electric field for all QWs and—a posteriori—justifies the subtraction of the transmitted from the incident field in order to directly obtain the FID of the coherent IS polarization. We find good agreement between experiment and theory [solid line in Fig. 3(c)] using the following parameters for the highest excitation

density in our model calculations: electric field amplitude: 10^6 V/m, dipole moment of IS transition: 5×10^{-28} C m, duration of Gaussian electric field: 300 fs. A direct comparison of our nonlinear experiments with the model allows to determine the degree of excitation quite accurately. In a Bloch sphere picture [inset of Fig. 3(c)], the strongest observed saturation of the FID corresponds to a partial Rabi flop of up to 1/6 (60°) of the full cycle (360°).

In conclusion, we demonstrated coherent control of linear intersubband polarizations with subpicosecond dephasing times by weak ultrafast electric field transients in the midinfrared. A nonlinear response, i.e., saturation of the polarization amplitude by up to 20%, can be induced with pulses of 1 pJ energy. This nonlinear response follows exactly the prediction of an ideal homogeneously broadened two-level system. This fact and the field-independent dephasing times may facilitate realization of ultrafast switching devices based on coherent IS polarizations.

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