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Coherent nonlinear dynamics of intersubband excitations in a two-dimensional electron gas

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Abstract

Femtosecond midinfrared four-wave mixing is used to study the ultrafast dynamics of coherent intersubband polarizations in n-type modulation-doped GaAs/AlGaAs quantum wells. Time-integrated four-wave mixing yields a dephasing time of $T_2 = 320$ fs, independent of excitation density. Time-resolved four-wave mixing unambiguously shows that the intersubband transition is purely homogeneously broadened by intraband Coulomb scattering within the low-density ($n = 5 \times 10^{10}$ cm^{-2}) electron plasma. The nonlinear propagation of tailored midinfrared pulses is studied by electro-optic sampling, which allows direct time-resolved measurements of the electric field transients. Excitation pulses of only 1 pJ energy are needed to observe partial Rabi flops of up to 60° in the Bloch sphere. © 2002 Elsevier Science B.V. All rights reserved.

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A quantitative understanding of the intersubband (IS) absorption spectra of quantum wells (QWs) requires knowledge both about the dynamics of coherent IS polarizations, which determine the homogeneous width of an IS transition, and the incoherent relaxation processes of carriers. The knowledge of dephasing processes of coherent IS polarizations is still very limited [1], as most studies so far have concentrated on carrier dynamics. Since state-of-the-art GaAs/AlGaAs QWs have a very high structural quality, disorder-induced inhomogeneous broadening of IS resonances is reduced considerably. This allows to directly measure irreversible dephasing, caused, e.g., by carrier–carrier scattering. A powerful technique to characterize such dephasing processes is femtosecond midinfrared degenerate four-wave mixing [2].

In addition to analyzing ultrafast dynamics, the generation and optical control of coherent IS excitations provides interesting perspectives for fast optical switching. Such experiments have become possible by the recent advent of controlled

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shaping [3] and time-resolved detection [4] of electric field transients in the midinfrared spectral range ($\lambda = 5$–$20\ \mu m$).

Here, we present data on the dynamics and optical manipulation of coherent nonlinear IS polarizations in QWs with negligible disorder-induced inhomogeneous broadening. The sample investigated consists of 51 GaAs QWs of 10-nm width (modulation doped to an electron density of $n_S = 5 \times 10^{10}\ cm^{-2}$), separated by 20-nm thick $Al_{0.35}Ga_{0.65}As$ barriers. A strong coupling between the p-polarized pulses and the IS transition dipole is achieved by a prism geometry. The $n = 1$ to $n = 2$ IS absorption line in this sample has a very small line width (FWHM) of only $\Delta E_{IS} = 3.7$ meV (see inset of Fig. 1). All experiments are performed at a temperature of 15 K.

Our experiments were performed with midinfrared pulses of 130-fs duration, which were generated by nonlinear frequency conversion of amplified pulses from a Ti:sapphire laser (repetition rate 1 kHz) [5]. They were tuned to the $n = 1$ to $n = 2$ transition at $E = 100$ meV ($\lambda = 12.4\ \mu m$). The excited-state lifetime was determined by midinfrared pump-probe measurements [Fig. 1(a)]. The nonlinear change of IS absorption induced by the pump pulse was monitored by weak delayed probe pulses. In Fig. 1(a), the spectrally integrated absorption change $\Delta A = -\int d\omega \ln[T(\omega)/T_0(\omega)]$ of sample A is plotted as a function of the delay time between pump and probe (T, T_0: sample transmission with and without excitation). We find a population decay of $T_1 = 550$ fs. For the four-wave mixing experiments [6] two midinfrared pulses were incident on the sample under wave vectors $\vec{k}_1$ and $\vec{k}_2$ to generate a transient grating. Third-order signals diffracted from this grating into the direction of $2\vec{k}_2 - \vec{k}_1$ are recorded either by a time-integrating detector or time-resolved by upconversion with a third synchronized 100-fs pulse at $\lambda = 1.5\ \mu m$. Fig. 1(b) shows the time-integrated FWM signals, plotted on a logarithmic scale, as a function of the time delay Δt_{12} between the two pulses generating the transient grating. Data are shown for both high (30% of the electrons initially in the $n = 1$ subband are excited into the $n = 2$ subband) and low (5%) excitation intensity. For all intensities, the signals rise within the time resolution of the experiment, exhibit a delayed peak at ≈ 100 fs, and subsequently decay with a time constant of $\tau = 160 \pm 15$ fs, reflecting the decay of the macroscopic IS polarization $P^{(3)}$. This decay time is significantly faster than the population decay T_1 [compare Figs. 1(a) and (b)].

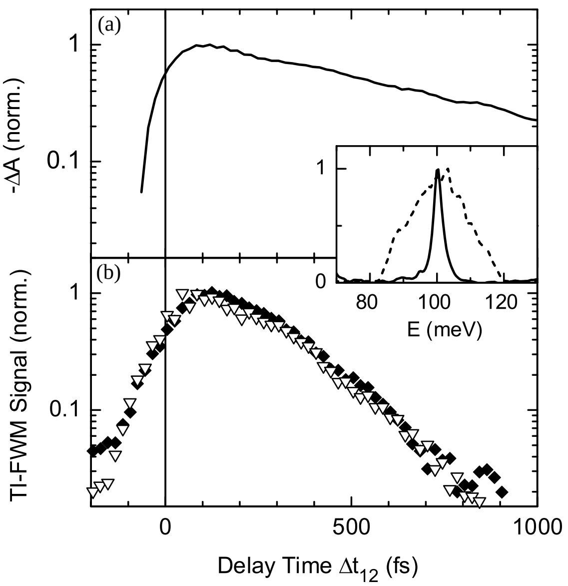

Fig. 1. (a) Pump-probe signal at the intersubband transition, yielding the excited-state lifetime. The spectrally integrated absorption change $\Delta A = -\int d\omega \ln[T(\omega)/T_0(\omega)]$ (symbols) is plotted versus pump-probe time delay. (b) Time-integrated (TI) FWM intensity in the diffracted direction $2\vec{k}_2 - \vec{k}_1$ for pulse intensities $I_0 = 0.7\ MW/cm^{-2}$ (solid symbols) and $6I_0$ (open symbols). Signals are shown on a logarithmic scale as a function of the time delay $\Delta t_{12} = t_2 - t_1$ between the incident pulses. Inset: Spectra of the intersubband absorption (solid line) and of the laser pulse (dashed line).

Homogeneous and inhomogeneous broadening of the IS transition can be distinguished by time-resolving the diffracted signal. In Fig. 2, the time-resolved FWM signals are plotted as a function of the time delay Δt_{23} between the second pulse generating the transient grating and the gating pulse used for upconversion. For all Δt_{12}, the peak of the time-resolved signal stays at the same time Δt_{23}. Inhomogeneous broadening would lead to a photon-echo-like behavior (the peak occurs at $\Delta t_{23} = \Delta t_{12}$), whereas homogeneous broadening results in a free induction decay (FID) of the

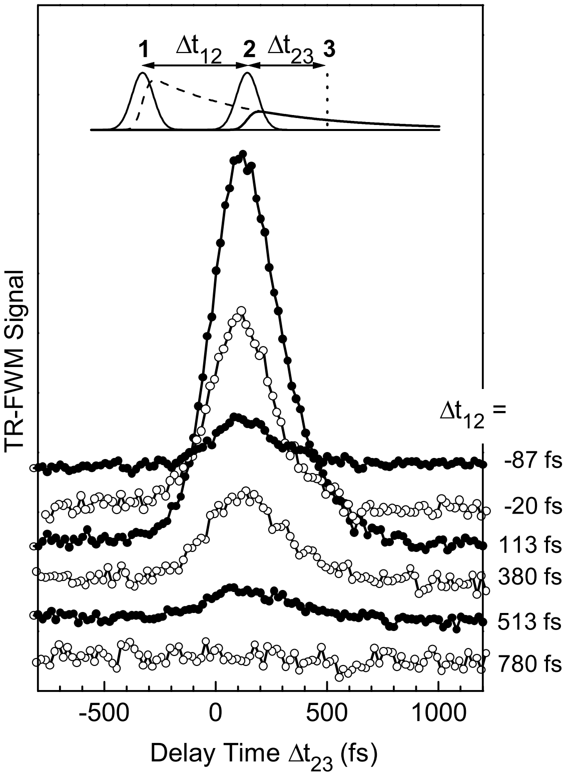

Fig. 2. Time-resolved FWM transients detected by sum-frequency mixing of the midinfrared FWM transients with near-infrared pulses ($\lambda = 1.5$ μm) in GaSe. Signals are shown as a function of the time delay Δt_{23} between the near-infrared pulse 3 and the midinfrared pulse 2, at various fixed delays Δt_{12}. Inset: pulse sequence.

macroscopic polarization (the peak occurs at constant Δt_{23}). Thus, our result gives direct evidence for a predominant homogeneous broadening of the IS absorption profile.

For our case of predominant homogeneous broadening, one derives an IS dephasing time of $T_2 = 2\tau = 320$ fs from the decay of the time-integrated signals [Fig. 1(a)]. This corresponds to a homogeneous linewidth of 4 meV, which is equal to the linewidth of the steady-state IS absorption spectrum. As discussed earlier [2], the irreversible dephasing with a rate of $T_2^{-1} \approx 3$ ps^{-1} is mainly due to carrier–carrier scattering. In the limiting case investigated here, i.e., for low electron concentration, purely homogeneous broadening of the IS transition and a dephasing time which is independent on excitation density, the IS excitation can be modeled by a homogeneously broadened two-level system.

In a second experiment, coherent nonlinear control of IS excitations in GaAs/AlGaAs QWs by extremely weak electric field-transients of sub-pJ energy [7] is demonstrated. A high-repetition-rate (2 MHz) laser system is used for this experiment. By amplitude- and phase-shaping of near-infrared (780 nm) 17-fs pulses in a programmable pulse shaper we generate tailored mid-infrared transients by phase-matched difference-frequency mixing in a 0.5-mm-thick GaSe crystal

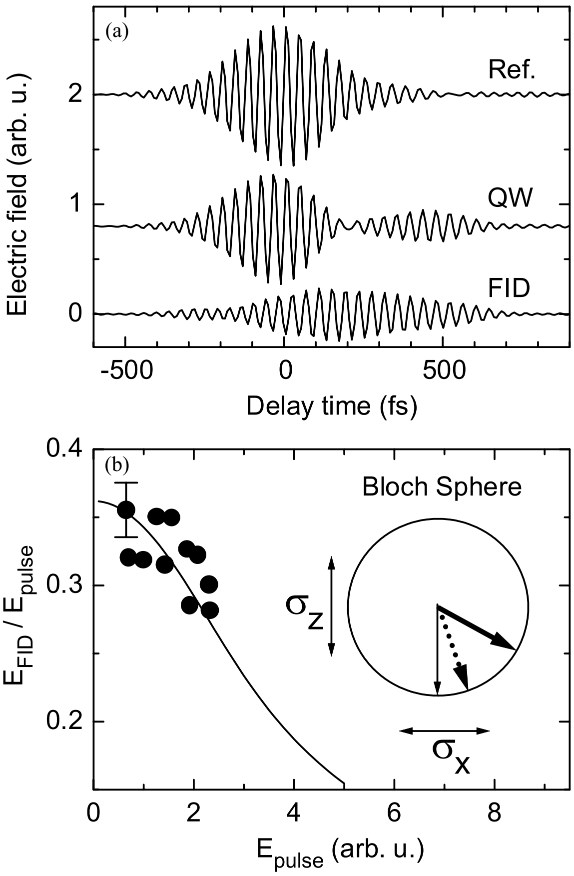

Fig. 3. (a) Measured electric field transients for low excitation through a reference sample (Ref.), through the QW sample, and the free-induction decay (FID) of the IS excitation in response to a single pulse. (b) Dots: ratio between the field amplitudes of the re-emitted E_{FID} and the excitation pulses E_{pulse} as a function of E_{pulse}. Solid line: model calculation using Maxwell–Bloch equations. Inset: schematic of Bloch sphere.

[3]. The time-dependent electric field of these transients is measured directly by ultrafast electro-optic sampling [4]. With this method we have investigated the nonlinear propagation of such electric field transients through the IS resonance. Data for small electric field amplitude (linear response) are shown in Fig. 3(a). This figure shows the electric field transients of pulses transmitted through an undoped reference sample of identical shape in the upper curve and of pulses transmitted through the QW sample in the central curve. In the lower curve, we plot the difference of these electric fields, which represent the re-emitted FID of the excited IS polarization. With increasing amplitude of the exciting electric field one finds a pronounced saturation of the emitted field strength [Fig. 3(b)]. The observed nonlinearity corresponds to an excitation of 30% of the $(n = 1)$ electrons to the $(n = 2)$ subband. We note that this is already caused by extremely weak excitation pulses with energies of less than 1 pJ. Our experimental data are confirmed by model calculations using the Maxwell–Bloch equations [solid line in Fig. 3(b)]. The observed saturation of the FID is explained by a partial Rabi flop in the Bloch sphere of up to 60°. The nonlinear sample response agrees well with the prediction for an ideal homogeneously broadened two-level system.

In summary, we have studied the ultrafast coherent dynamics of IS polarizations in high-quality GaAs/AlGaAs quantum wells with an electron density of $5 \times 10^{10}\ \mathrm{cm}^{-2}$. Time-resolved FWM experiments demonstrate unambiguously that the narrow IS absorption lines of such GaAs/AlGaAs quantum wells are predominantly homogeneously broadened. The dephasing time $T_2 = 320$ fs is governed by electron–electron scattering and is independent of the excitation density. We have demonstrated coherent control of linear IS polarizations by weak ultrafast electric field transients in the midinfrared. A nonlinear response, i.e., a saturation of the polarization amplitude by up to 20%, can be induced with pulses of only 1 pJ energy. This nonlinear response follows the prediction of Maxwell–Bloch equations, assuming 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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