Coherent Control of Absorption and Polarization Decay in a GaAs Quantum Well: Time and Spectral Domain Studies

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Two phase-locked pulses are used to coherently excite excitonic polarizations. It is shown that the second pulse can either be strongly amplified by taking up energy gained from the destruction of the exciton polarization or can be decreased drastically by giving up all its energy to excitons. Both the temporal and the spectral signatures of the transmitted pulse shapes agree well with model calculations.

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The phase of optical light pulses is important in interaction with matter when the dephasing times of the optical transitions are longer than pulse duration and temporal interpulse separation. In this regime, the relative phase between successive incident pulses strongly influences the dynamic evolution of the quantum system. Femtosecond laser sources make it possible to perform coherent control experiments such as coherent control of photocurrent [1–4] and carriers [5–12] in semiconductors. Recently, coherent control of excitons in semiconductors has been demonstrated through THz radiation [5,6], pump-probe reflectivity [7,8], and four-wave-mixing [9,11] experiments.

When the exciton polarization created by a first pulse is coherently controlled by a second pulse, many interesting phenomena are expected to happen to the second pulse. When the second pulse constructively adds to the polarization, we expect its absorption to be greatly enhanced. On the other hand, when it destructively interacts with the polarization, the energy stored in the exciton polarization will be removed from the system and added to the second pulse. To the best of our knowledge, these phenomena have never been observed directly, because *most experiments focused on the coherent control of the exciton system and not on the control of the pulses themselves*. This is partly due to the fact that control experiments, such as THz radiation [5,6], nonlinear reflectivity [7,8], and four-wave mixing [9,11], are nonlinear in nature and do not probe the pulse shapes.

In this Letter, by probing the *linear* propagation of pulses through a GaAs/AlGaAs multiple quantum well sample, we probe the intriguing interaction between the coherent exciton polarization and the controlling pulses. In particular, using up-conversion technique with both short (spectrally broad) and long (spectrally narrow) upconverting pulses, we obtain both temporal and spectral information with high temporal and spectral resolution, respectively. The main thrust of our report is the direct demonstration that the intensity of a phase-controlled second pulse can either be greatly enhanced by gaining energy from the exciton system (thus destroying the exciton polarization) or completely destroyed by giving all of its energy to the system (thus greatly increasing the exciton polarization), respectively. Our results prove that the energy exchange between the excited exciton system and the second pulse crucially depends on the relative phase between the two. We show that *absolute energy gain of the second pulse*, i.e., amplification of the second pulse induced by the propagation through the coherently excited system can be achieved, when both the relative phase and the intensity ratio between two incident pulses are chosen appropriately.

Our experimental setup is shown in the inset of Fig. 1. We use a mode-locked Ti:sapphire laser as the source of transform-limited pulses with a pulse duration of 150 fs. We produce two phase-locked collinear pulses of temporal separation τ with the same linear polarization [13] and a third beam which is used as an upconverting pulse whose bandwidth and pulse duration can be controlled by a prism pair and a slit. The two phase-locked collinear pulses that have passed through the sample are upconverted with short or long pulses. The sample investigated is a GaAs/AlGaAs multiple quantum well which consists of thirty 15 nm GaAs wells separated by 5 nm $Al_{0.36}Ga_{0.64}As$ barriers. Our sample is predominantly homogeneously broadened, as shown by the fact that the polarization decay decreases exponentially in time with roughly the same time constant with that in the four-wave-mixing experiment, fully consistent with the exciton linewidth measured in the photoluminescence excitation experiment. A significant inhomogeneous broadening would lead to a rapid decay of the transmitted intensity on a time scale given by the inverse of the inhomogeneous broadening, which would make it much harder to observe the coherent control. All experiments were performed at 10 K and the laser was tuned to the heavy-hole (HH) exciton energy and the excitation density was kept below 10^8 cm^{-2} throughout all experiments to ensure that we are in the linear regime.

In Fig. 1, transmitted pulse shapes probed by time-resolved up-conversion are shown for $\Delta\phi = \pi$ and 0 for

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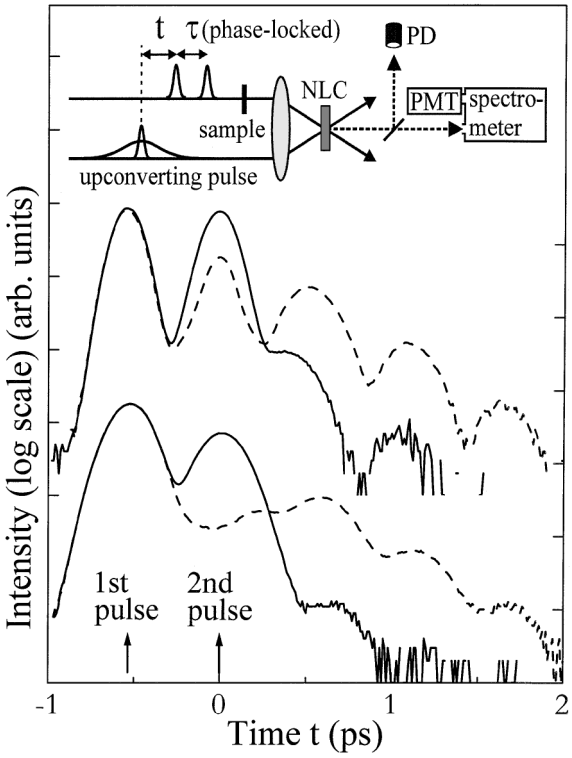

FIG. 1. Inset: Schematic of the experimental setup. NLC stands for nonlinear crystal and PD stands for photodiode. For a time delay $\tau = 550$ fs equal to the HH-LH beating period, transmitted pulse shapes probed by time-resolved up-conversion are plotted for $\Delta\phi = \pi$ (solid) and 0 (dashed) when the intensity ratio between two incident pulses (I_2/I_1) is 0.59 (upper) and 0.12 (lower).

intensity ratios I_2/I_1 between incident second and first pulse of 0.59 and 0.12. The time delay τ of 550 fs is chosen equal to the HH-LH(light-hole) beating period such that the phases of the HH and LH exciton polarizations are identical when the second pulse arrives. In Fig. 1, for an incident $I_2/I_1 = 0.59$, it is shown that for $\Delta\phi = \pi$, I_2/I_1 increases after transmission through the sample to 0.91. Taking into account the one pulse absorption of 34%, our result demonstrates that the second pulse does not lose energy due to absorption to the system but gains slight energy from the system. This is possible only by decreasing the coherent exciton polarization that was prepared by the first pulse. After the transmission and distortion of the second pulse, the upconverted intensity shows a polarization decay with modulation due to the HH-LH quantum beats [14,15], which is a measure of the remaining exciton polarization. Because of the vanishingly small solid angle probed by our experiment and subpicosecond time scale, this polarization decay is by far the dominant contribution in the directions parallel and antiparallel to the excitation, completely overshadowing any contribution from luminescence. For $\Delta\phi = 0$, due to enhanced absorption the second pulse loses much of its energy to the system. In this case, I_2/I_1 decreases to 0.24 after transmission and thus the following decay signal is enhanced due to the increased exciton polarization. The change of coherent exciton polarization caused by the second pulse and the intensity of the second pulse itself therefore depend strongly on the phase of the second pulse.

A dramatic situation is seen in Fig. 1, where the intensity ratio is chosen as $I_2/I_1 = 0.12$ before incidence. In this case, after transmission through the sample, the second pulse is nearly gone for $\Delta\phi = 0$ and virtually no coherent exciton polarization remains for $\Delta\phi = \pi$. For $\Delta\phi = 0$, there is a deep minimum at the position of the second pulse (corresponding to $I_2/I_1 = 0.02$), and the second pulse acquires a large tail. For $\Delta\phi = \pi$, considerable enhancement of the second pulse ($I_2/I_1 = 0.42$) and no free polarization decay tail are seen after transmission. Therefore, the intensity of the second pulse itself and the coherent exciton polarization after the transmission of the second pulse can either be completely destroyed or greatly enhanced if the intensity ratio between the two pulses is appropriately controlled.

To demonstrate that all above experimental findings can be understood with linear optics, we perform model calculations, which describe the HH and LH exciton system as a simple three-level system. The splitting between the excitons (7.5 meV) as well as the ratio of their oscillator strength (1:1.85) was estimated from the photoluminescence excitation spectrum. Dephasing of the HH and LH exciton polarizations is modeled using $T_2 = 1$ ps for both transitions. The propagation through the sample, i.e., the reradiation of the exciton polarization, is included via taking the transmitted field to be the incident field plus a factor α (chosen here to match the experiment) times the total optical polarization $P(t)$ of the HH and LH excitons: $E_{\text{out}}(t) = E_{\text{in}}(t) + i\alpha P(t)$ [16,17]. With this model, we calculate directly the time-resolved intensity of the transmitted field $|E_{\text{out}}(t)|^2$, neglecting the temporal width of the upconverting pulse. The results displayed in Fig. 2 for various ratios I_2/I_1 have been obtained using as in the experiments a time delay τ of 550 fs and Gaussian pulses with a pulse duration of 150 fs tuned to the HH exciton resonance. In agreement with experiments, the calculations show that for $\Delta\phi = \pi$ the transmitted intensity is strong at the temporal position of the second pulse followed by a rather weak polarization decay with some HH-LH beats. For $\Delta\phi = 0$ the situation is reversed, i.e., the intensity is rather small and distorted at the position of the second pulse but the polarization decay is enhanced. The maximum modulation for the coherent control of the polarization decay seems to occur for $I_2/I_1 \approx 0.25$, which is close to the optimal value $|\exp[-\tau/T_2]|^2 = 0.33$. At the temporal position of the second pulse interesting phenomena are observed, which depend on I_2/I_1. The calculated structures are due to interference between the reradiating polarizations induced by the first and second pulse with the second pulse itself. Because of this interference, in agreement with experiment, there is a deep

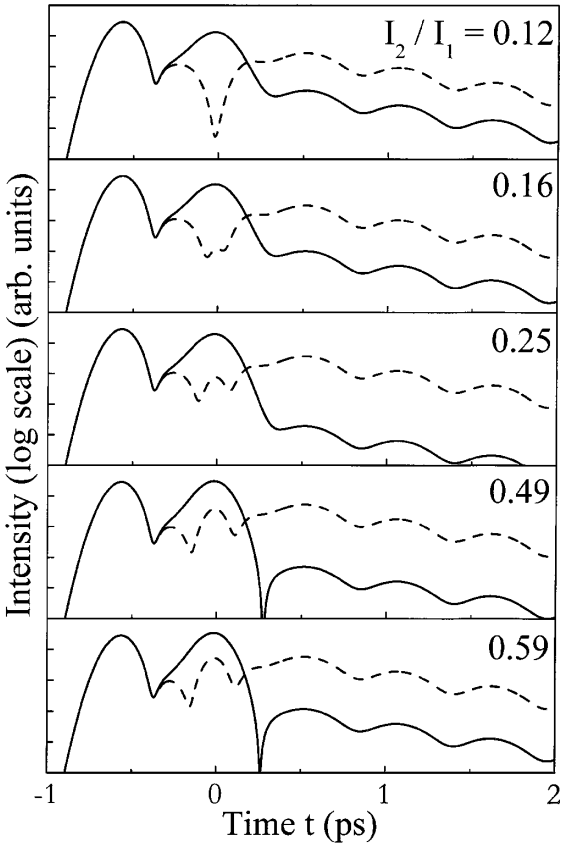

FIG. 2. Calculated time-resolved intensity of the transmitted field for a time delay $\tau = 550$ fs for $\Delta\phi = \pi$ (solid) and 0 (dashed) and various intensity ratios I_2/I_1. From top to bottom: $I_2/I_1 = 0.12$, 0.16, 0.25, 0.49, and 0.59.

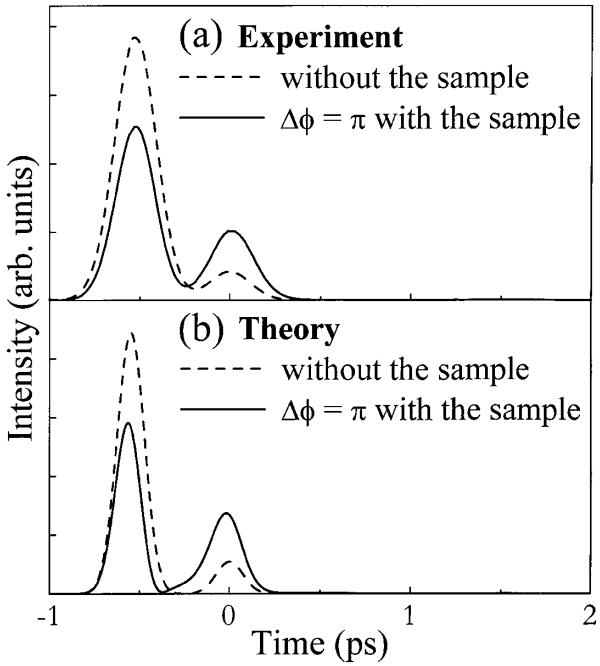

FIG. 3. Temporal shapes of pulses not transmitted or transmitted through the sample with relative phase π when a time delay is equal to the HH-LH beating period and intensity ratio is 0.12, obtained (a) experimentally and (b) theoretically. It is shown that the second pulse is amplified when transmitted through the sample.

minimum at the second pulse for $I_2/I_1 = 0.12$, whereas with increasing intensity ratio the transmitted second pulse itself shows up.

It has been shown in Fig. 1(b) that the second pulse gains significant energy from the system in the case of $\Delta\phi = \pi$. Thus, we note the possibility of *absolute* amplification of the second pulse due to its energy exchange with the excited exciton system. Figure 3(a) shows the result with relative phase π of Fig. 1(b) again, together with the time-resolved transmission obtained without the sample. After the transmission, the intensity of the first pulse decreases by 34%, due to absorption. However, the second pulse is amplified by a factor of 2.5, which is attributed to energy gain from the exciton polarization excited by the first pulse. Thus, the second pulse is not attenuated by absorption but amplified by simultaneous destruction of the coherent exciton polarization. The experimental results are in good qualitative agreement with the model calculations displayed in Fig. 3(b).

Taking advantage of the relatively narrow spectral width of a long upconverting pulse, we furthermore demonstrate both energy gain of the second pulse from the system and enhanced absorption of the system in the spectral domain. Figure 4(a) shows the spectra of the transmitted field with relative phase π or 0, respectively, for a time delay τ of 1.5 ps. The transmission was probed by a long upconverting pulse with a relatively narrow spectral width ($\Delta\lambda \sim 1.5$ nm) and temporal pulse width of about 0.6 ps using zero time delay between the long upconverting pulse and the second pulse; i.e., the upconverting pulse arrives at the same time as the second pulse. The spectrum shows enhanced intensity at the HH exciton energy in the case of $\Delta\phi = \pi$, compared with the spectrum obtained without the sample. *This is by far the most direct evidence for the coherent, linear gain of energy experienced by the second pulse through the instantaneous destruction of the coherent exciton polarization.* For $\Delta\phi = 0$, in contrast, the spectrum shows a dip nearly down to the bottom at the HH exciton energy, which indicates the enhancement of absorption. These experimental findings are again well reproduced by numerical results. As in the experiment, the calculated spectra shown in Fig. 4(b) show at the HH exciton energy an enhanced intensity for $\Delta\phi = \pi$ and a strongly reduced intensity for $\Delta\phi = 0$.

In Fig. 4(c) the intensity at the HH exciton energy is plotted versus the relative phase under the same experimental condition as in Fig. 4(a), for the two cases that the pulses either are transmitted through the sample or not. Whereas there exists very small modulation due to the Ramsey fringe in case the pulses are not transmitted through the sample, much stronger modulation appears when the pulses are transmitted through the sample.

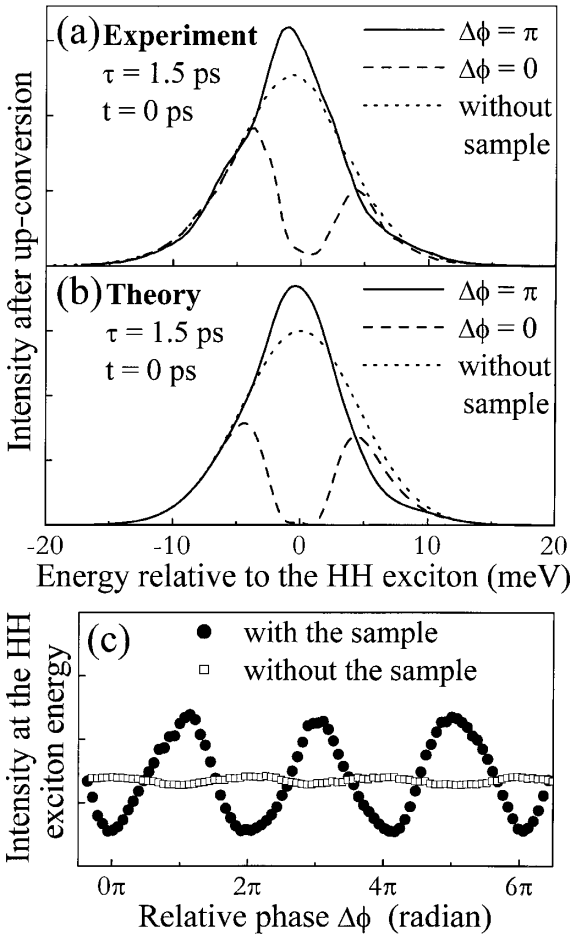

FIG. 4. For a time delay τ of 1.5 ps, shown are the spectra of pulses not transmitted or transmitted through the sample with relative phase π or 0, probed by a long upconverting pulse when a time delay t of upconverting pulse relative to second pulse is 0 ps. In each case of $\Delta\phi = \pi$ or 0, a bump or dip is shown at the HH exciton energy respectively, the agreement between (a) the experimental and (b) theoretical results being shown. (c) The intensity at the HH exciton energy versus relative phase under the same experimental condition as in (a).

Hence, we prove that the considerably large bump and dip in Fig. 4(a) are the result of the interference between the coherent exciton polarization and laser field, apparently.

In conclusion, we have demonstrated directly the interaction between the coherent exciton polarization and the controlling pulses by probing the linear propagation of pulses through a quantum well sample in both the time and spectral domain. We prove that the energy exchange between the excited exciton system and the second pulse crucially depends on the relative phase between the two. The second pulse can be amplified by the propagation through the coherently excited system, when both the relative phase and the intensity ratio between two incident pulses are chosen appropriately. The temporal and spectral domain studies of this coherent control performed by monitoring directly the linear transmitted field are in agreement with model calculations using linear optics.

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