Femtosecond Studies of Coherent Transients in Semiconductors

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The coherent interaction of femtosecond laser pulses and thin CdSe and GaAs samples is investigated experimentally and theoretically. Oscillatory structures in the differential probe transmission around the exciton resonance and around the pump frequency are observed when the probe pulse precedes the pump. Comparison with theory attributes the oscillations to the coherent coupling between the light field and the electron-hole transitions in the semiconductor. For nonresonant excitation, the oscillatory structures around the exciton are identified as the early stages of the optical Stark effect.

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Coherent light-matter interactions have been extensively studied in atomic spectroscopy.[1] These interactions are readily observable in atomic systems as a result of the relatively long dephasing times. In semiconductors, on the other hand, the coherent coupling of the exciting light to the electron-hole transitions disappears very rapidly because of the various interaction processes of the electronic excitations. Until now only a few coherent processes in semiconductors have been observed experimentally. The anisotropic state filling in Ge,[2] and phase coherence and orientational relaxation of excitons in GaAs,[3] as well as the study of coherent coupling effects in pump-probe spectroscopy in dyes,[4,5] have been investigated using picosecond pulses. In the femtosecond time domain, carrier orientational relaxation[6] and band-to-band spectral hole burning,[7–10] as well as the exciton optical Stark effect,[11,12] have been reported.

In this paper, we report the first observation of oscillatory structures in the probe transmission spectra around the exciton when the semiconductor is excited spectrally below the exciton resonance and the probe pulse precedes the pump pulse. For the case of resonant interband excitation, similar oscillations are also observed around the central frequency of the pump pulse. We refer loosely to this class of effects as "coherent transients," including all the nonstationary features observed in semiconductors on time scales short enough that thermalization to quasi-equilibrium due to carrier-carrier or carrier-phonon scattering has not yet occurred.

Experiments were performed with a variety of different semiconductors to show that the observed coherent effects are quite general and do not depend on the specific semiconductor. A semiclassical theory is developed which qualitatively explains all of the experimentally observed features. The experiments were conducted in a pump-probe geometry with $\cong$80-fs pulses. The laser system consists of a colliding-pulse mode-locked dye laser amplified in a dye amplifier using a 10-kHz copper-vapor laser or a 10-Hz neodymium-doped yttrium aluminum garnet laser. A portion of the amplified pulses was focused on an ethylene glycol jet to generate a white-light continuum probe pulse while the remainder was used as pump pulse. For below-band-gap excitation, with the 10-Hz system, the portion of the continuum was selected with interference filters and was amplified to be used as the pump pulse. Induced changes in the probe transmission were measured over a 125-meV portion of the spectrum. The pump and probe pulses are orthogonally polarized and make an angle of $\cong 15°$ with each other. The pump intensity is approximately 100 times larger than the probe intensity. The normalized transmission difference between excited and unexcited samples $\Delta T/T_0 = (T - T_0)/T_0$ was measured as a function of time delay between the pump and probe pulses.

Figure 1 shows differential transmission spectra for a thin CdSe platelet of $\cong 0.5$-μm thickness at 10 K as a function of probe wavelength for three time delays, where always the probe peak precedes the peak of the pump pulse. The spectra of the pump pulse and of the linear sample absorption are shown in the inset. The pump is centered in the interband absorption region of the sample. Oscillations in the differential transmission spectra are observed around the exciton line (close to 6700 Å) and also around the pump frequency (close to 6250 Å) with no structure in between. The magnitude of the oscillations is larger in the exciton region compared with those in the pump region. The oscillatory features around the pump are within the spectral pump width.

Figure 2 displays the differential transmission spectra for a $\cong 0.5$-μm bulk GaAs platelet at 15 K for three

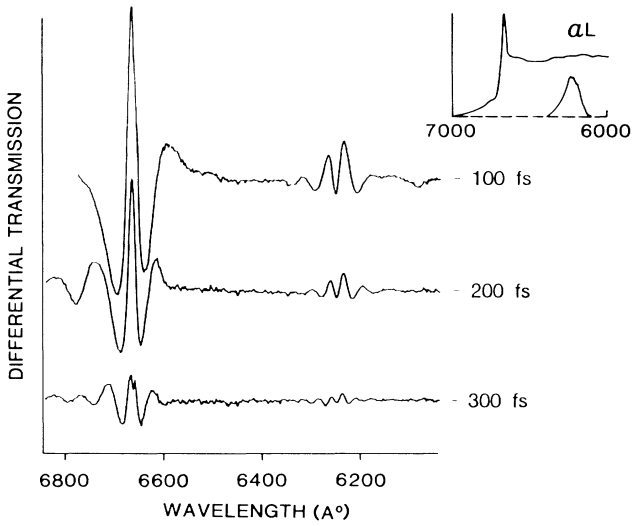

FIG. 1. Differential transmission spectrum for a pump-probe delay of −300, −200, and −100 fs measured for a thin CdSe platelet at 10 K. Inset: Absorbance αL for the sample, and the pump spectrum.

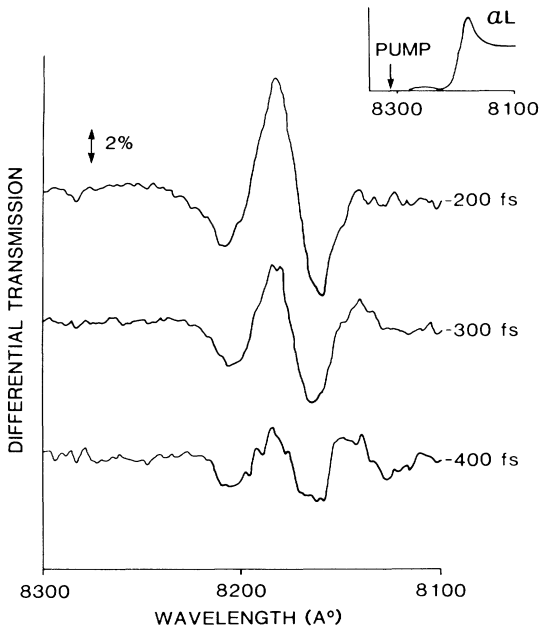

FIG. 2. Measured differential spectra for pumping below the exciton resonance in bulk GaAs at three different time delays of −400, −300, and −200 fs, between pump and probe pulses. The pump pulse has ≅200-fs duration and is centered around 832 nm. Inset: Sample absorption αL.

different time delays. The pump frequency is tuned at 832 nm, below the exciton resonance, such that, in contrast to the situation in Fig. 1, one-photon excitation of carriers is not possible in this case. Oscillations in the differential transmission spectra are observed around the exciton resonance at early times. The structures are asymmetric with the largest positive spike being located on the high-energy side and the largest negative spike on the low-energy side of the exciton, respectively. At later times the oscillations evolve into a dispersive-looking feature.

In order to understand the origin of the observed oscillations at very early times we have developed a semiclassical theory. We assume a weak, short probe pulse interacting with the time-dependent medium polarization which is driven by the strong pump field, $E_L(t)$. The time difference between pump and probe peak is denoted by t_p. The different electron-hole–pair states are modeled as mutually uncoupled transitions, each of which is described by a density matrix. The dissipative scattering processes among the elementary excitations are included through phenomenological decay rates in the equation of motion for the density matrix. We simulate the net effect of intraband scattering as a population decay for the individual k states (diagonal damping Γ) and we include the phase relaxation as a dipole damping rate (nondiagonal damping γ). All other Coulomb effects are neglected. The theory has been evaluated for the two experimentally analyzed cases of (i) the *resonant* pumping where the pump pulse resonantly excites the band-to-band transitions, probing the transmission around the frequency of the pump, and (ii) the *nonresonant* excitation of the exciton in which the pump pulse is detuned from the exciton but the probe measures the changes at the exciton. For the case (i) we treat each k state separately and integrate the result over all k values. The pump-induced changes of the probe transmission in the vicinity of the pump frequency Ω_L are found to be[13]

$$\Delta T(\omega) = T_0 \mathrm{Re}\left[\int_0^\infty dt\, e^{i(\omega-\Omega_L)t} e^{-2\gamma t} \int_0^\infty dt'\, e^{-\Gamma t'} E_L(t_p - t') E_L^*(t_p - t' - t)\right] + T_0 \mathrm{Re}\left[\int_0^\infty dt\, e^{i(\omega-\Omega_L)t} e^{-\Gamma t} \int_0^t dt'\, e^{-(2\gamma-\Gamma)t'} E_L(t + t_p - t') E_L^*(t_p - t')\right], \quad (1)$$

where the prefactor T_0 is a combination of constants. Here, we have assumed that the pump-field amplitude E_L is small in comparison to a π pulse. To derive Eq. (1), we make use of the fact that the pumping occurs high into the band which allows us to neglect bound states. In the k integration, we assumed a flat density of states which simplifies the expression considerably and leads to Eq. (1).

Examples of the computed results for this case are presented in Fig. 3(a) for different time delays between probe and pump. The spectra show clear oscillatory structures when the probe precedes the pump, similar to the experimental results. A detailed inspection of the two additive terms in Eq. (1) shows that the first term, which only involves times before t_p, describes the saturation of the absorption due to the population inversion created by the leading edge of the pump pulse. The second term describes the coherent coupling. It involves not only times before t_p but also depends on the pump field values at times longer than t_p, and it is always oscillatory if the integrand has a peak in the range of integration. Such a peak occurs for negative t_p since $E_L(\tau=0)$ is the maximum of the pump field; however, no peak is possible in the integrand of the first term in Eq. (1). The frequency of the oscillations is determined by the time delay t_p between pump and probe. Increasing the temporal overlap between the pulses causes a reduction of the number and of the amplitude of the oscillations with respect to the central peak, as is the case in the experimental results. The analysis of the relative importance of the two damping processes shows that the amplitude of the oscillatory structures depends on the value of the dipole damping rate γ, but the shape is rather insensitive to the value of γ. Furthermore, the oscillations depend very strongly on the population decay rate Γ (diagonal damping); increasing Γ decreases the amplitude of the oscillations with respect to the central peak until only a broadened peak, the spectral hole, is left.

For the situation where the transmission spectra are measured in the vicinity of the exciton frequency ω_x, case (ii), we model the exciton as a single homogeneously broadened transition and neglect the contributions from the band states. Examples of the results are shown in Fig. 3(b). The oscillations now occur around the exciton frequency qualitatively reproducing the experimentally observed features for below-band-gap excitation. The structures are not symmetric as in the case of the interband transitions because the pump laser is detuned from the exciton resonance. When the delay time approaches zero, the oscillations gradually disappear and the transmission changes assume an almost dispersive shape. The dispersive structure is caused by a frequency shift of the exciton resonance, the so-called optical Stark shift. Hence, the oscillatory structures should be viewed as the early stages of the optical Stark effect in short-pulse pump-probe spectroscopy. To obtain a better understanding of the features shown in Fig. 3(b), we simplify our analytic result for the differential transmission assuming that the pump detuning below the exciton greatly exceeds both the exciton linewidth γ_x and the spectral width of the pump. For this case, the differential transmission is given as

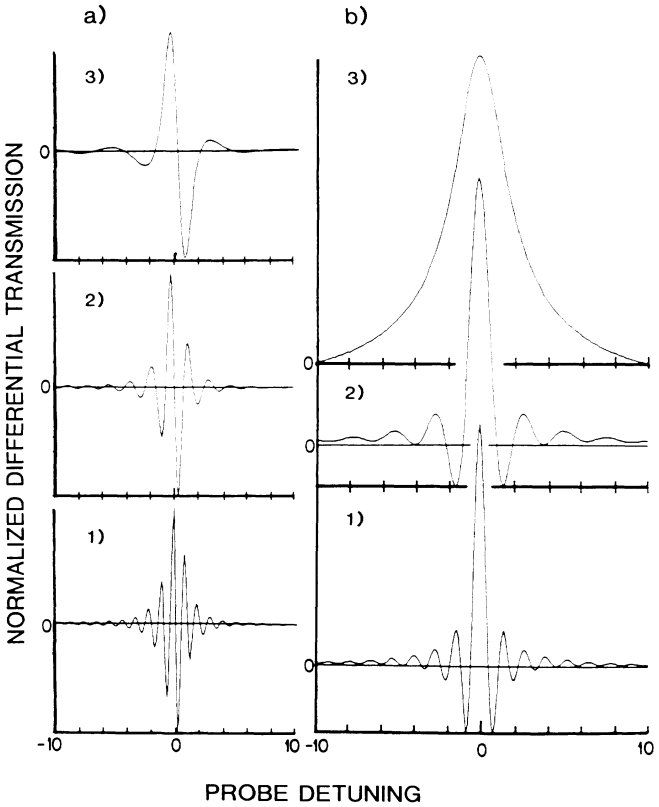

FIG. 3. (a) Calculated normalized differential transmission spectra in the spectral region of the exciton resonance for various time delays, (1) $t_p=-500$ fs, (2) $t_p=-300$ fs, (3) $t_p=-100$ fs, assuming a 120-fs pump pulse. The probe detuning is given with respect to the exciton resonance ω_x, i.e., $(\omega-\omega_x)/\sigma$. (b) Calculated normalized differential transmission spectra in the spectral vicinity of the pump pulse for time delays (1) $t_p=-400$ fs, (2) $t_p=-200$ fs, (3) $t_p=0$. A 120-fs pump pulse has been assumed and the probe detuning is defined with respect to the central frequency Ω_L of the pump pulse.

$$\Delta T(\omega)\cong -T_0\frac{1}{\omega_x-\Omega_L}\,\mathrm{Im}\left[\frac{1}{\gamma_x-i\Delta}\int_0^\infty dt\,|E_L(t+t_p)|^2 e^{i\Delta t-\gamma_x t}\right], \tag{2}$$

where $\Delta=\omega-\omega_x$. Equation (2) can be regarded as generalization of the Stark-shift result obtained in the adiabatic limit.[14] It shows that the amplitude of the oscillatory structures occur around the exciton resonance, and they fall off inversely proportional to the detuning of the pump from the exciton, whereas the shape of the oscillations is detuning independent. The integrand in Eq. (2) has a peak only if t_p is negative, explaining the disappearance of the oscillations for positive time delays.

In conclusion, we have observed oscillatory structures in the differential transmission spectra of semiconductors. These oscillations occur around the exciton and around the central frequency of the pump in the case of resonant interband excitation. The dephasing time of the exciton is clearly longer than the duration of the femtosecond pulses employed in the study. The oscillations around the pump can be viewed as arising from the interference between the probe and that part of the pump which is scattered into the probe direction. The scattering is caused by the excitation grating generated by the probe and the leading part of the pump pulse. As shown in our analysis, this mechanism is not critically dependent on the dephasing time, and can hence be observed also in systems with rapid dephasing.

The oscillations reported here are not artifacts, as they appear not only around the pump but also in the exciton region where the central frequencies of pump and probe are different. It has to be mentioned that oscillatory structures around the pump pulse have been observed also in organic dye solutions.[15] It is possible that the structures are also of the type discussed here. In addition, we have seen similar features also in other semiconductors, specifically in $CdSe_xS_{1-x}$ doped glasses and in GaAs-AlGaAs multiple quantum wells.

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