# Self-Induced Transmission on a Free Exciton Resonance in a Semiconductor

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We observe coherent long-distance propagation of an optical pulse in resonance with the free exciton at high light intensities in an optically thick semiconductor. The experiments show pulse reshaping, pulse breakup, and a high degree of transmission. Microscopic calculations using the semiconductor Maxwell-Bloch equations yield good agreement with the experimental data. [S0031-9007(98)07576-0]

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Propagation experiments with short light pulses are crucial for the simultaneous study of temporal and spatial coherence in matter. In semiconductors, as opposed to atomic systems which can be modeled by noninteracting two-level systems [1], spatial dispersion [2] and excitation-induced nonlinearities of the excitonic resonance [3–5] will result in remarkable modifications of the pulse propagation features. At low light intensities, temporal polariton beating of the transmitted pulse has been observed and shows excellent agreement with linear dispersion theory [6]. In nonlinear experiments, however, many-body effects (such as exciton-exciton collisions), unknown in atomic systems, will destroy the polariton beating after the pulse by interaction-induced dephasing [7–9]. Let us recall the situation in idealized two-level systems: Rabi oscillations (coherent electron density oscillations) lead to lossless soliton propagation of $2\pi$ pulses—so-called self-induced transparency (SIT)—and pulse breakup for input pulse areas larger than $3\pi$ [10]. Following this area theorem [11], SIT was pioneered in atomic vapor about 25 years ago [12]. In semiconductors, SIT has been demonstrated on bound excitons in CdS [13], which resemble ideal noninteracting two-level systems due to the restricted wave function overlap between different excitons. So far, SIT had not been found on free excitons and the chances for its discovery are low because theoretical investigations [14,15] came to the conclusion that interaction-induced dephasing between excitons with overlapping wave functions will reduce the polarization coherence necessary for the establishment of complete pulse transmission. On the other hand, the appearance of Rabi oscillations in semiconductors [14,16–18], as well as propagation over long distances [19], has been predicted at light intensities larger than $\pi$. Thus at least partially coherent propagation should be feasible. In this Letter, we will show experimentally that coherent long-distance propagation and multiple-pulse breakup on a free exciton resonance in a semiconductor at high light intensities is possible. Because many observed features resemble SIT in atomic systems, we will call this newly observed phenomenon *self-induced transmission* in semiconductors.

We used 50–80 fs pulses around 680 nm with pulse energies of about 100 nJ from an optical parametric amplifier [20] pumped by a regenerative Ti:sapphire amplifier (COHERENT REGA) at 200 kHz. Careful alignment using FROG [21] made sure that the pulses were chirp-free, which is essential for the experiment. The pulses were focused onto CdSe samples in a cryostat ($T = 8$ K), and the transmitted beam was imaged onto a pinhole, cutting out the central part of the beam in order to investigate regions of constant intensity. The inset in Fig. 1 shows the experimental setup: After propagation through the sample, the pulses are time resolved by cross correlation with 50 fs pulses in a 1 mm thick beta-barium-borate crystal. The transmitted spectra were recorded simultaneously. Strained hot-wall epitaxial samples [22,23] with $\alpha L = 1.7$ and 6.8 were especially tailored for this experiment ($\alpha$ is the absorption coefficient and $L$ is the sample length). The large strain due to the thermal expansion and lattice mismatch with the $BaF_2$ substrate caused an $A$-$B$ exciton splitting of up to 72 meV. Figures 1a and 1b show the linear absorption spectra. Both samples show substantial inhomogeneous broadening due to strain relaxation.

In order to model the results of the pulse propagation experiment, the semiconductor Maxwell-Bloch equations [14,24] have been solved. These equations contain the wave equation for the field coupled to the material

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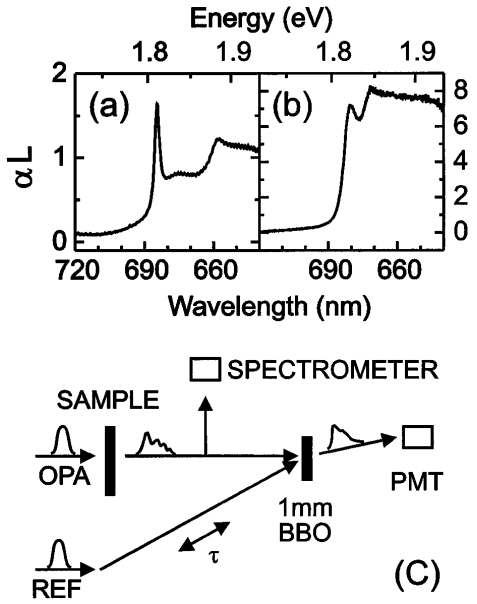


FIG. 1. (a),(b) Linear absorption spectra of the thin and thick CdSe epitaxial samples at $T = 8$ K. (c) Experimental cross-correlation setup. The reference pulse for the cross correlation has a duration of 50 fs.

equations for the polarization. Approximations have to be made because the nonlinear polarization of the semiconductor many-body system is too difficult to be calculated exactly [25]. In the present case, we applied the slowly varying envelope approximation of the field [24], whereas the material equations include mean-field and correlation effects (diagonal and nondiagonal dephasing as well as nonlinear polarization scattering) in the second-order Born approximation for polarization and electron distribution [5]. The resulting material equations are a standard tool in semiconductor optics and have been discussed in Refs. [5,15,26–28]. We use $\Delta_{LT} = 1$ meV, $m_e = 0.125m_0$, and $m_h = 0.431m_0$ as input parameters for the CdSe material.

Figure 2 shows the experimental (a) and theoretical (b) cross-correlation traces and the transmitted spectra of a 180 fs pulse propagating on the $A$-exciton resonance of CdSe with $\alpha L = 1.7$. The lowest trace in Fig. 2a shows the cross correlation of the pulse as it travels through the $BaF_2$ substrate (which is identical with the cross correlation of the incident pulse). Linear propagation through the CdSe shows propagation beats in agreement with the low-intensity excitation presented in Ref. [6]. Increasing the intensity to 12 MW/cm$^2$, which corresponds to a pulse area of $\pi$, the propagation beats vanish, and a trailing shoulder appears about 500 fs after the maximum. The transmitted pulse cross correlation resembles the input pulse at 26 MW/cm$^2$ ($1.5\pi$). At 46 MW/cm$^2$ ($2\pi$), a trailing shoulder appears 250 fs after the maximum, becoming more pronounced at 180 MW/cm$^2$ ($4\pi$). Also, the leading shoulder 100 fs before the maximum becomes visible. The quantitative agreement between the predictions and the measurements is remarkable. The maximum transmitted intensity (corrected for reflection) is more than 90%. Looking at the spectra, one finds that at low intensities, the excitonic absorption leads to the dip in the center of the pulse spectrum. This dip vanishes for increasing intensities, and at 26 MW/cm$^2$, the transmitted spectrum equals the input spectrum. For higher intensities, no changes such as broadening or shifts are observed. The agreement with the theoretical predictions is again very good. The theoretical analysis shows that at low intensities the propagation beats result from the exciton-polariton interference similar to Ref. [6], whereas the shoulders at larger intensities are caused by carrier-density Rabi flopping.

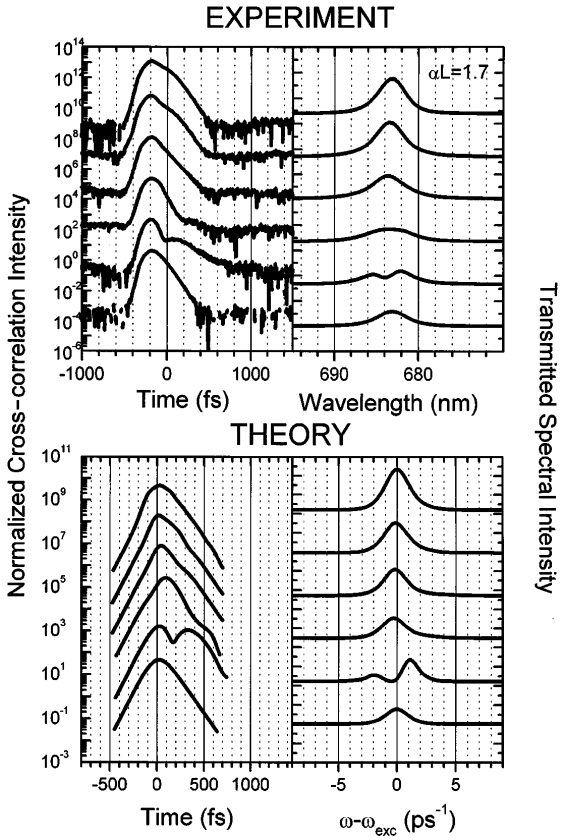


FIG. 2. Propagation of 180 fs pulses through the thin CdSe sample with $\alpha L = 1.7$ for increasing intensities. $\lambda = 683$ nm. The cross-correlation traces are shown at the left and the transmitted spectra are plotted at the right. Experiment, input intensities (top to bottom): 180, 46, 26, 12, and 0.92 MW/cm$^2$, propagation through substrate. Theory, pulse areas (top to bottom): $4\pi, 2\pi, 1.5\pi, 1\pi, 0.27\pi$ pulse area, substrate.

The coherent pulse breakup should manifest itself more clearly in thicker samples due to the accumulation of propagation effects. We therefore use the $\alpha L = 6.8$ sample. However, for 180 fs pulses, multiple pulse breakup as predicted in the theory could not be reproduced [29]. The reason is probably the large spectral width of the laser pulse, which interacts with many excitonic transitions due to the inhomogeneous broadening of the

$\alpha L = 6.8$ sample, each having a different oscillator strength and a different coupling to the light. Similar effects are known from atomic systems, where in this so-called "sharp-line case" [30] the transient propagation behavior can be very irregular. In atomic systems, it was shown recently [31] that the area theorem needs to be rederived for short optical pulses and inhomogeneous broadening.

In order to reduce the influences of these peculiarities, we used a narrow band spectral filter producing 900 fs pulses and thereby selecting a narrow distribution of resonances within the broad inhomogeneous distribution of the $\alpha L = 6.8$ sample. This approach closely matches the original experimental suggestions in atomic systems [12], where a spectrally narrow laser pulse propagated through a Doppler-broadened metallic vapor.

Figure 3a shows the experimental cross-correlation traces and transmitted spectra for increasing intensity. At an intensity of 5 MW/cm$^2$, which corresponds to a $1.5\pi$ pulse, the original transmitted pulse form is already steepened, shortened, and shows a slight shoulder structure about 500 fs after the maximum. Increasing the intensity to 31 MW/cm$^2$ (2.5 times the initial pulse area), a clear breakup into several pulses is observed. About 400 fs after the main peak, a second shoulder can be observed, and about 1000 fs after the maximum, a small third peak is visible. At the highest intensity of 99 MW/cm$^2$ (4.5 times the initial pulse area), a clear breakup into four peaks is visible, which appear about 350, 600, and 1000 fs after the maximum. *Coherent destruction* manifests itself in the extremely steep transient from the maximum to the first minimum. The transmitted spectra show a slight redshift with increasing intensity and a slight asymmetric broadening towards the blue. This broadening, however, is certainly not due to self-phase modulation, because the nonlinear phase shift at our intensities is less than one mrad, and XFROG [32] traces have shown that self-phase modulation occurs in the investigated samples only at much higher intensities. Tuning the laser only 10 meV above the exciton resonance leads to a drastic reduction of transmission but still to a shortening of the transmitted pulse (not shown here), probably as a consequence of the enhanced dephasing rate in the continuum [5,14,15].

Figure 3b shows the results of the theory model for the 900 fs pulse propagation through the thick CdSe sample. At 5 MW/cm$^2$, a breakup into two pulses is visible, with the trailing pulse about 750 fs after the maximum. Additionally, a small prepulse occurs about 500 fs before the maximum. At 31 MW/cm$^2$, a breakup into three main pulses can be seen, with the trailing pulses at 400 and 750 fs after the maximum. Also, a small prepulse is visible 300 fs before the main peak. At 70 MW/cm$^2$, a clear breakup of the pulse into four parts is visible, with the trailing peaks at 300, 700, and 1500 fs. Also, a clear steepening of the leading edge is visible as intensity increases. The transmitted spectra show a slight redshift

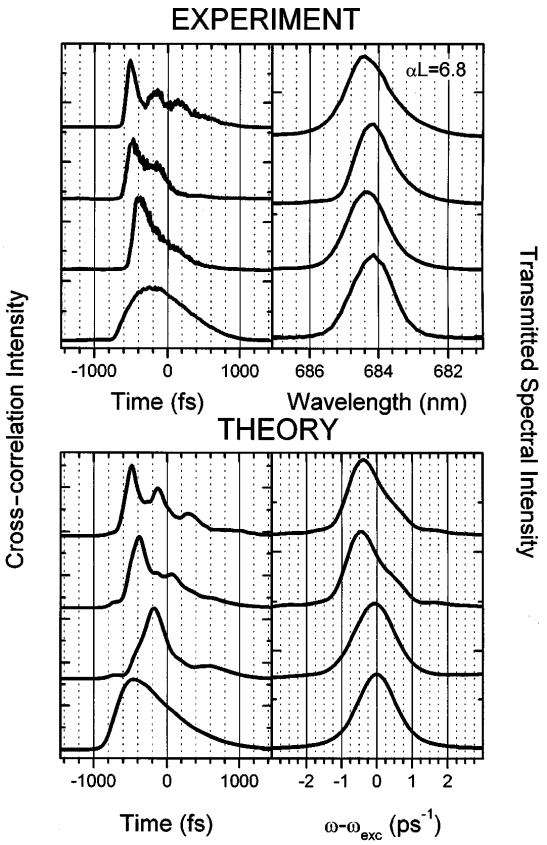


FIG. 3. Propagation of 900 fs pulses through the thick CdSe sample with $\alpha L = 6.8$ for increasing intensities (experiment). $\lambda = 684.5$ nm. The cross-correlation traces are shown at the left and the transmitted spectra are plotted at the right. Experiment, input intensities (top to bottom): 99, 31, and 5 MW/cm$^2$, propagation through substrate. Theory, pulse areas (top to bottom): $6.6\pi, 3.7\pi, 1.5\pi$, substrate.

and an asymmetric broadening at the blue side, as was observed in the experiment. The maximum transmitted intensity is slightly higher than 50%.

The pulse breakup and the spectral distortions can be traced back to carrier-density Rabi oscillations, as theoretical calculations show. The agreement between theory and experiment for the thick sample is qualitatively good, but not as quantitative as in the case of the thin sample. Certainly the different amounts of inhomogeneous broadening in the samples, not taken into account in the calculations, play a role here. The accumulation of nonenvelope effects for long propagation distances as well as approximations in the many-body theory may also be responsible.

The basic experimental features, such as pulse breakup due to Rabi flopping, can already be found in the mean-field approximation [19], but we expect that the additional inclusion of inhomogeneous broadening would yield a reduction of the theoretically predicted pulse shape modulations, thus reducing the agreement between theory and experiment. Therefore, we stress that further theoretical analyis, especially for high-quality samples and high-resolution experiments, should include a better treatment

of correlation effects (excitonic occupation, memory effects) [3,33,34], spatial dispersion [35], the second-order wave equation for strong exciton resonances (nonenvelope effects) [36], and, depending on the sample quality, inhomogeneous broadening. Most of these phenomena, like memory effects and bound states, may increase the coherence and stabilize the pulse shape modulations against the inhomogeneous broadening. Further experimental investigations as well as microscopic calculations will have to follow to clarify the detailed interplay of all effects discussed above. Nevertheless, our calculations show a surprisingly good agreement with all qualitative features of the present experiment.

In conclusion, we have observed strong self-induced transmission and multiple-pulse breakup on a free exciton resonance in optically thick samples. Even at the required high intensities and on a strongly absorbing resonance, coherent long-distance propagation with similar features as SIT in atomic systems is possible. The presented calculations yield a qualitative picture, and our results should be viewed as a first step in understanding coherent high-intensity pulse propagation phenomena in semiconductors.

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