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Femtosecond dynamics of the optical Stark effect in semiconductor-doped glass

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We report the observation of the optical Stark shift in CdS_xSe_{1-x} semiconductor-doped glass excited under nonresonant below gap condition and probed with femtosecond optical pulses. An ultrafast and pure light-induced shift of the band edge is observed. The measured dependence of the band edge shift with the pump pulse peak intensity is linear for pump intensities up to 0.3 GW/cm^2. Above this value, departure from linearity is observed. For a pump intensity of 3 GW/cm^2, the band shifts by 11 meV. The response of the shift tracks the profile of the pumping pulse. © *1996 American Institute of Physics.* [S0003-6951(96)03008-9]

The optical Stark effect (OSE) has been a subject of study for some time, mostly in atomic gases and in solutions of organic dye molecules,[1] and it is mainly characterized by a light induced shift in the energy levels of these systems. In semiconductor materials, however, since the dephasing times are much shorter than in atomic systems, observation of this type of effect was possible only recently due to the development of ultrashort and intense laser pulse generation. In bulk semiconductors, the OSE has been recently observed in CdS[2] and GaSe[3]. For structures with two-dimensional quantum confinement, the optical Stark effect has been thoroughly studied.[4–7]

In this letter, we present femtosecond differential transmission spectroscopy data that show the occurrence of the OSE in a semiconductor-doped glass. Monitoring the absorption edge, from 580 to 680 nm during the excitation by a short pulse below the band gap, we identify a clear rigid shift of the edge away from the pump wavelength towards higher energies, a clear signature of the OSE. The shift of the band edge tracks almost exactly the time profile of the 60 fs pump pulse, and the dependence of the shift with the peak pump intensity agrees well with the optical Stark effect model.

Semiconductor-doped glasses (SDG) have been a subject of increasing interest during the past few years due to the fact that they were shown to exhibit large optical nonlinearities when excited close to the fundamental absorption edge, and were also shown to have fast response times.[8–11] In the case of CdS_xSe_{1-x}-doped glasses, optical absorption and dispersion nonlinearities have both been demonstrated, and in most of the results they are of χ^3 type. In a previous work, using CdS_xSe_{1-x}-doped glasses as active medium, we have described a polarization rotation switch[12] with a response time shorter than 50 fs. The results that we obtained were consistent with an OSE-type nonlinearity.[13]

The sample studied was the $CdS_{0.4}Se_{0.6}$ SDG CS-2.62 from Corning Glass (average μ-crystallite diameter ~10 nm),[14] with a thickness of 500 μm. Measurements were performed using the pump-probe technique. The sample was pumped by 60 fs duration pulses of 7 nm FWHM width, centered at 626 nm and with an energy of 350 nJ, and the

response of the dynamical processes in the system were probed by 12 fs duration probe pulses. The detuning between the central wavelength of the pump and the band gap is 110 meV. The pump and probe pulses were obtained from a colliding-pulse mode-locked laser whose pulses, after being amplified in a copper-vapor laser-pumped dye amplifier, were split into two beams. Pulses from one of the beams are used to excite the sample. The other beam is sent through a short piece of optical fiber where it undergoes spectral broadening and is compressed with a grating pair becoming the 12 fs probe pulse. The pump and probe polarizations are orthogonal and the probe energy is 1/150 of that in the pump. The transmittance of the probe through the excited sample is monitored by an optical multichannel analyzer. Differential transmittance spectra are recorded as a function of the time delay between the pump and probe beams. The differential transmission spectrum is given by $DTS(\omega) = [T(\omega) - T_0(\omega)][T_0(\omega)]^{-1} = \exp(-\Delta\alpha L) - 1$, where L is the sample length, $T(\omega)$ and $T_0(\omega)$ are the probe transmission in the presence and absence of the pump, respectively. $\Delta\alpha$ is the difference between the absorption coefficient when the pump is present and when it is not.

Figure 1 shows the experimental absorption change, $-\Delta\alpha$ for the SDG CS-2.62 at 300 K for different relative time delays between the pump and probe pulses. Negative time delays indicate situations in which the probe peak precedes the peak of the pump pulse. In Fig. 1 we also plot the linear absorption curve (dashed line) and the pump and probe spectra from which we can see that the sample is being excited far below its band gap. For large negative delays the spectra are unchanged from the linear absorption but when the two pulses start to overlap, there is a dynamical increase in the sample transmission. Subtracting the measured absorption change for each delay from the linear absorption spectrum, one can observe that the resulting absorption edge is quickly shifted to the blue as the pump travels through the medium, reaching a maximum at zero delay and then returning almost to the nonperturbed position. For positive delays, changes in the spectra are still observable until 500 fs, which was the largest delay investigated. If absorption edge shifts rigidly, i.e., in the limit of a "pure shift," the measured absorption change $\Delta\alpha$ line shape should be exactly the same as

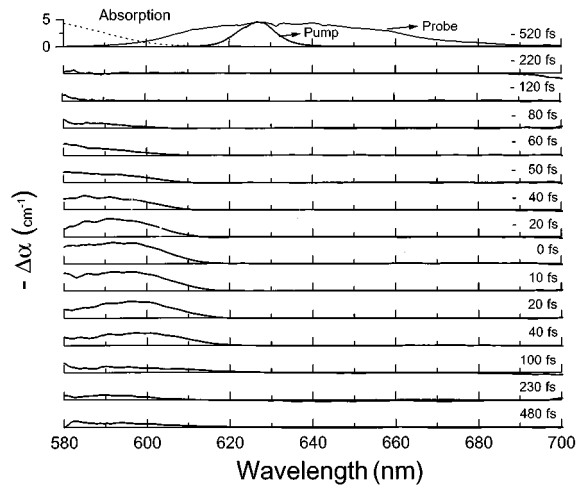[a)]Electronic mail: tsuda@spin.att.com

FIG. 1. Differential absorption spectra ($-\Delta\alpha$ vs wavelength) measured for a sample of SDG CS-2.62. The fast recovery bleaching at small wavelengths indicates the occurrence of the optical Stark effect. The pump and probe spectrum are overlayed in the upper trace, together with the shape of the absorption profile of the sample (dotted line).

that from the derivative of the linear absorption $d\alpha/dE$.[15] In Fig. 2, $d\alpha/dE$ obtained from $\alpha(E)$, measured independently, is compared to the measured $\Delta\alpha(E)$ at zero delay. It can be seen that the shape of the curves are very similar, which allows us to say that to a good approximation, at zero delay, the absorption edge is purely shifted to the blue. This rigid shift, that reinforces the difference between the characteristics of the OSE in atomic systems and semiconductors, has already been reported[2,15] for exciton and continuum states. For the exciton states in quantum wells, this effect is explained by the enhancement of the oscillator strength due to the larger shift experienced by the band edge compared to that of the exciton, which increases its "binding energy." It was also observed that at high intensity, the resonances lose height and broaden. As discussed in Ref. 2, the "pure" Stark shift of the continuum states that we observe should also be expected.

Figure 3 shows the behavior of the absorption edge at different time delays. The energy shift was computed as
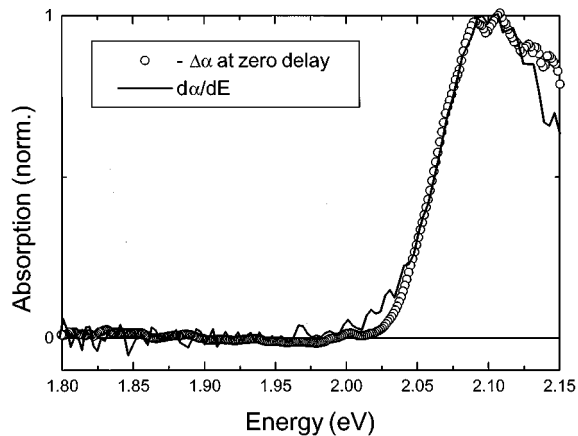

FIG. 2. Measured normalized absorption change, $-\Delta\alpha$, at zero delay time (open circles) and $d\alpha/dE$ (solid line) vs energy for SDG CS-2.62. The good agreement between the two curves indicates that the band edge is being purely shifted.
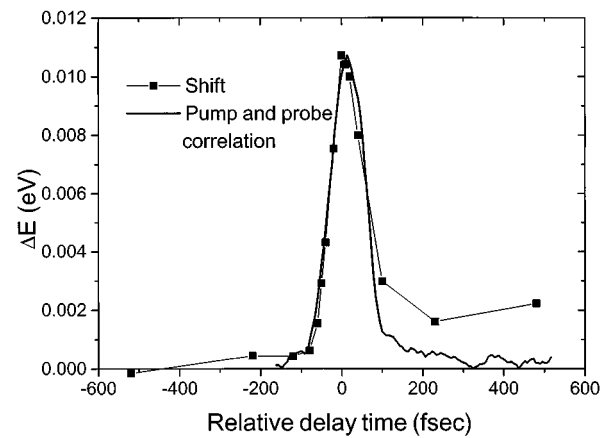

FIG. 3. Measured dependence of the absorption edge shift as a function of the pump and probe delay time.

$\Delta E = \Delta\alpha(d\alpha/dE)^{-1}$, where $\Delta\alpha$ was obtained from data shown in Fig. 1 and $d\alpha/dE$ is the derivative of the linear absorption curve as a function of the energy taken at 601 nm. The absorption edge is rapidly blue shifted and returns to an absorption level that is just smaller than the initial one. As mentioned before, for negative delays no changes in the spectrum are observed, which means that the remaining shift of the absorption spectrum observed at 500 fs time delay eventually recovers fully. Apart from the remaining shift at large positive delays, the profile of the shift as a function of the time delay between pump and probe pulses follows closely that of the pump pulse. This is the behavior expected for an optical Stark shift, where only virtual excitations are created, implying a fast response time. The residual bleaching of the absorption observed at 500 fs time delay, in opposition to what is expected from the OSE, may be attributed to the injection of real carriers into the conduction band due to the occurrence of two-photon absorption (TPA)[13,15] or residual overlap between the pump spectrum and the absorption tail. The pump pulse duration is 60 fs and for this reason the OSE does not contribute to the signal at the 500 fs delay situation. The slow recovery component in the time response of the system must be related to band filling effects. The magnitude of this contribution to the observed signal is comparatively small. The importance of the role of real excitations in this kind of experiment was discussed elsewhere.[2,15] Different from the coherently driven contribution, which adiabatically follows the pump pulse, the absorbed carriers decay on a longer timescale after the pump pulse is gone and prevent the full recovery of the induced absorption change. In this situation, dynamical screening of the electron-hole interaction should be considered, since it can contribute to the bleaching of the energy shift.

We have also investigated the shift of the spectrum as a function of pump intensity. From Fig. 4, valuable information about the behavior of the induced Stark shift with pump intensity can be obtained. At zero delay, the pump intensity was changed using a variable neutral density filter in the pump beam path. Pump pulse intensities inside the crystallites ranged typically from 0.1 to 3.5 GW/cm^2.

In atomic systems, the OSE is treated under the "dressed atom" formalism from which a blue shift of the transition
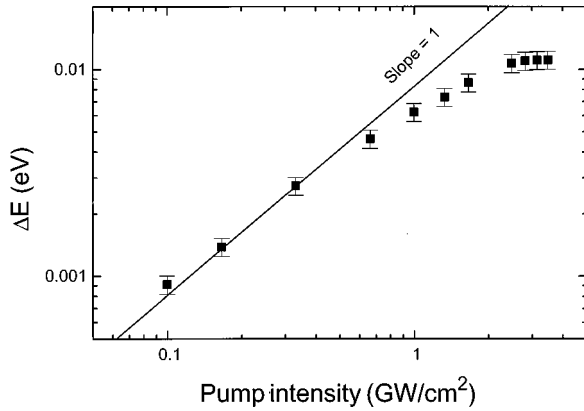
FIG. 4. Absorption edge shift as a function of the peak pump intensity. Squares represent experimental data points. The solid line is obtained from Eq. (1) in the large detuning approximation.

lines is predicted. This picture was used by Mysyrowicz et al.[16] as a first approach to describe an observed excitonic line shift in a GaAs quantum well. In the case of a semiconductor system, it is necessary to take into account the different nature of the interaction due to the formation of bound (exciton) states plus the renormalizations caused by the real or virtual carriers injected which screen the coulombic interaction.[6,7,17] The shift of the band gap due to the presence of a pump field E_p is given, in the low field limit, by[7]

$$\Delta E_g = \frac{2(\mu_{cv} E_p)^2}{\hbar \Delta \Omega} \left(\rho_\infty + \frac{E_0}{\hbar \Delta \Omega} \nu_\infty \right), \qquad (1)$$

where $\hbar \Delta \Omega = (E_g - \hbar \omega_p)$ is the energy detuning, E_g is the energy gap, $\hbar \omega_p$ is the pump photon energy, μ_{cv} is the interband dipolar matrix element, and E_0 is the exciton binding energy. The parameters ρ_∞ and ν_∞ are related, respectively, to the anharmonic exciton-photon interaction and to exciton–exciton interaction. In the case of three-dimensional carriers (no confinement), their values are $\rho_\infty = 8$ and $\nu_\infty = 24$.[7]

As shown above in Fig. 4, the observed shift of the absorption edge in our experiment follows the predicted linear dependence with the pump intensity for intensities up to 0.3 GW/cm². In this low pump intensity limit, we can calculate an effective dipole moment μ_{cv} for the SDG, using Eq. (1). The application of Eq. (1) to the case discussed here requires an additional averaging over the random orientation of the crystallites in the glass matrix, considering that in our experimental conditions the pump field is orthogonal to the probe field. This averaging multiplies the right hand side of Eq. (1) by a factor of 1/15. The obtained value for μ_{cv} is 5.6 $\times 10^{-29}$ C m. This compares well with the estimated values of μ_{cv}(CdS) = 4.4 × 10^{-29} C m and μ_{cv}(CdSe) = 6.1 × 10^{-29} C m obtained from conventional first-order perturbation calculation for direct transitions between parabolic bands.[18,19] In this approximation, μ_{cv} can be directly obtained from the band gap energy, E_g, and the electron effective mass in the conduction band.

At pump intensities above 0.3 GW/cm², deviation from the linear dependence of the shift on the pump intensity is observed. This deviation is predicted in the work of Ell et al.,[7] and is related to many-body effects. As the pump intensity increases, the virtual carriers tend to spend more time in the conduction bandlike states, making the screening of the Coulomb interaction more effective. This causes a renormalization of the interactions, leading to a decrease in the rate of change of the shift with respect to the pump intensity.

In conclusion, we have shown the occurrence of the optical Stark shift in a SDG on a femtosecond time scale. This is the first time, to our knowledge, that the Stark shift is spectrally time resolved in this type of material. The measured dependence of the band edge shift with the pump pulse peak intensity and its response time is in qualitative agreement with results obtained on the study of the OSE in bulk and confined semiconductor structures. Moreover, our results confirm that the optical Stark effect is the dominant contribution to the nonresonant third-order refractive nonlinearity of this type of SDG.

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