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Doppler-Shifted Self-Reflection from a Semiconductor

A. Schülzgen (a), N. Peyghambarian (a), and S. Hughes (b)

(a) Optical Sciences Center, University of Arizona, Tucson, AZ 85721, USA
e-mail: axel@u.arizona.edu; phone: (520) 621 2382; Fax: (520) 621 4442

(b) Department of Physics, University of Tokyo, 7-3-1 Hongo, Bunkyo-Ku, Tokyo 113, Japan

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We report the experimental observation of a self-reflected wave inside a dense saturable absorber. A femtosecond pulse saturates the absorption and causes a density front to penetrate into the semiconductor. The dielectric constant change across the boundary between areas of low and high densities results in internal reflection. Due to the front propagation the self-reflected light is shifted by the Doppler effect. The Doppler shift makes it possible to distinguish between surface reflection and self-reflection and is used to experimentally verify the dynamic nonlinear skin effect. The measurements are in agreement with our theory which is within the framework of the reduced semiconductor Maxwell-Bloch equations.

By interacting with a nonlinear medium, a laser beam can substantially modify the conditions for its own propagation. Well known examples are self-focusing [1] and self-induced transparency [2]. The generation of self-reflected waves inside a very dense saturable absorber has been studied theoretically [3 to 5] over the last decade. In these works, a cw laser beam saturates the absorption in a surface layer, which allows the light to penetrate into the medium. Subsequently, a stationary boundary between regions of low and high absorption can be established. If the spatial transition is sufficiently sharp, the change in the refractive index across the transition region gives rise to a self-reflected wave. However, no experimental verification has been reported so far due to the need for extremely high absorption and nonlinearity of the material.

Very recently, Forysiak et al. [6] combined the concepts of self-reflection and laser induced excitation front propagation [7, 8] to predict the dynamic nonlinear optical skin effect. They investigated the dynamic problem of a short laser pulse incident on a dense saturable absorber described by a two-level system. The calculations show that for sufficiently high irradiances, pulse durations shorter than the carrier lifetime, and a linear penetration depth smaller than the wavelength, a moving carrier density front can be excited. The motion of the boundary between regions of high and low carrier densities causes a shift of the self-reflected light to longer wavelengths due to the Doppler effect. The measurement of this Doppler shifted light is direct evidence for the dynamic nonlinear skin effect.

We report the experimental observation of a Doppler-shifted self-reflected wave close to the exciton resonance of ZnSe. In our analysis, we show that an intense femtosecond laser pulse excites a moving density front in the semiconductor. Simultaneously, absorption saturation allows the pulse to penetrate further while the density front gives rise to a red-shifted reflection due to the moving mirror effect.

Two series of experiments were performed on a ZnSe layer: single beam reflection, and degenerate pump–probe reflection measurements for various time delays. The 3.8 μm thick layer of ZnSe was grown by molecular beam epitaxy on a GaAs substrate. The sample was held in a cryostat at a temperature of 77 K. A frequency doubled, passively mode-locked Ti-sapphire laser with 150 fs pulses was used to perform both kinds of experiments at various photoexcitation densities and under normal incidence. Pump and probe beams were linearly crosspolarized to minimize coherent effects and to allow selective blocking of the reflected pump beam. The central frequency of the laser pulses was always resonant with the ground state exciton transition at 2.792 eV.

We chose ZnSe because of its large exciton binding energy of almost 20 meV and its pronounced ground state exciton absorption well separated from band-to-band transitions. This resonance exhibits high nonlinearities under optical excitation [9 to 11]. Strong saturation of the excitonic absorption due to efficient exciton screening has been observed [9]. Liquid nitrogen temperature provides three desirable conditions: an isolated resonance of high absorption, a carrier lifetime much longer than the incident pulse (as shown below), and a dephasing time yielding comparable spectral widths of exciton resonance and incident pulse. Thus, we investigate a system similar to the two-level model system of Forysiak et al. [6] which meets all criteria for the predicted dynamic nonlinear skin effect. The linear optical skin depth $1/\alpha_0$ at the exciton resonance is about 40 nm, which is much smaller than the wavelength inside the medium $\lambda_m \approx 160$ nm.

Fig. 1 shows the spectra of incident and reflected pulses for various incident energy fluxes in a single beam experiment in which dynamic effects are present. For normalization purposes the reflected pulse spectra have been divided by the corresponding energy fluxes. The incident pulse has been normalized according to the background refractive index. A pronounced ground state exciton feature can be seen in the low density reflec-

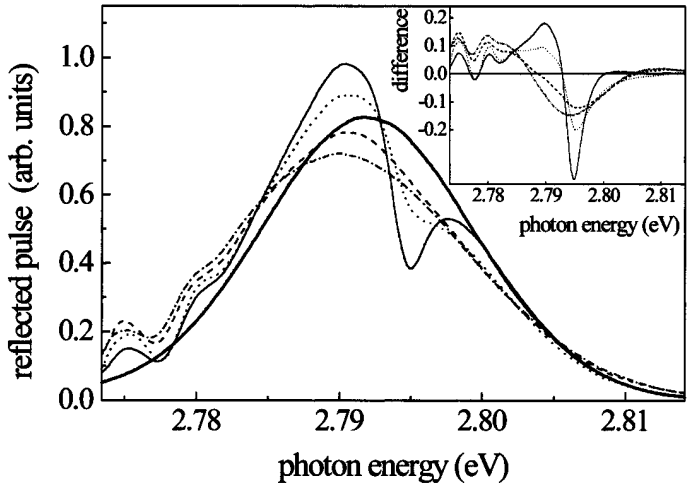

Fig. 1. Normalized pulse spectra reflected at a ZnSe epitaxial layer for incident pulse energy fluxes of 0.1 μJ/cm^2 (solid line), 2 μJ/cm^2 (dotted line), 3 μJ/cm^2 (dashed line), and 5 μJ/cm^2 (dash-dotted line). The thick solid line shows the spectrum of the incident pulse. Inset: Spectrally resolved difference between reflected and incident pulses

tion (see the thin solid line in Fig. 1) accompanied by Fabry-Perot interferences below this transition energy.

With increasing pulse irradiances the excitonic structure becomes less resolvable until the distinct feature disappears. However, even for the highest irradiances with the exciton resonance almost completely bleached, the reflected pulse spectrum does not resemble that of the incident pulse. To emphasize this point, the difference spectra between reflected and normalized incident pulses (thick solid line in Fig. 1) are plotted in the inset of Fig. 1. While at moderate pulse irradiances (up to 2 $\mu J/cm^2$) the bleaching of the exciton structure dominates, a distinct change of the spectral shape of the difference spectrum is observed at the highest irradiances. A negative signal centered at the exciton resonance is found, and an increasing amount of light is reflected at longer wavelengths, extending to more than 15 meV below the exciton resonance. For the highest pulse irradiance the linear reflectivity is almost doubled at photon energies three times the full width at half maximum (FWHM) below the exciton resonance.

In order to distinguish between dynamic and steady-state effects, we have also performed degenerate time-resolved pump–probe reflection measurements. In such an experiment dynamic effects are present only during the pump–probe overlap time. The reflected probe pulse spectra for two different pump–probe delays at a pump pulse energy flux of 5 $\mu J/cm^2$ are shown in Fig. 2. For comparison, the spectrum in the absence of a pump pulse is also depicted by the thin solid line. In the case of optimized temporal pump–probe overlap in which dynamic effects are present, we can almost reproduce the single beam results and observe again additional reflected light well below the exciton resonance. For delays larger than the pulse duration in which dynamic effects are absent, the spectral shape of the reflected pulse resembles that of the incident pulse. Looking again at the spectrally resolved difference spectra (inset of Fig. 2) our interpretation

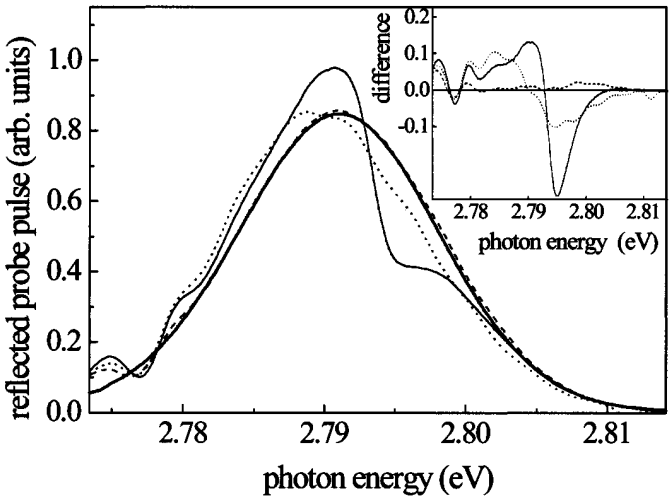

Fig. 2. Spectrum of the reflected probe pulse without pump (solid line), for a pump–probe delay of 0 fs (dotted line), and 200 fs (dashed line). The pump pulse energy flux is 5 $\mu J/cm^2$. The thick solid line shows the spectrum of the incident probe pulse which is also used to calculate the spectral differences between reflected and incident pulses shown in the inset

of the single beam experiments is strongly supported. Only for zero pump–probe delay a structure which corresponds to Fig. 1 is obtained while the flat line for 200 fs delay indicates the absence of dynamic effects.

The data presented in Fig. 2 indicate that we operate in a regime of high densities where the excitonic part of the dielectric constant is saturated to a large degree and the main part of the laser pulse can indeed penetrate into the sample. Using a reduction of 50% of the excitonic contribution to the linear reflectivity, a saturation energy flux of 1 to 2 $\mu J/cm^2$ is estimated. The reflected pulse spectrum remains nearly unchanged between 200 fs and 5 ps pump–probe delay indicating that the carrier lifetime is larger than 5 ps. Thus, effects of a finite carrier lifetime can be neglected in the modeling of our experiments. Additionally, any detection of luminescence can be excluded.

To model the dynamic interaction between the propagating light pulse and the semiconductor medium in detail, we have solved numerically the reduced semiconductor Bloch equations (SBEs), see e.g. [12] coupled to the vector Maxwell's equations. The complexity of the problem made an approach using the full SBEs not feasible at this time. However, for certain limiting cases the full SBEs can be cast into a reduced form, which has been successfully employed recently for a description of Rabi oscillations [13] and polariton solitons [14] in semiconductors. The reduced SBEs may be written in the following form:

$$\frac{\partial P}{\partial t} = i[\Delta + 2\beta N]\,\frac{P}{2} + i2N\Omega - \frac{P}{\tau_{\mathrm{deph}}}\,, \qquad \frac{\partial N}{\partial t} = \Omega\,\frac{\mathrm{Im}\,(P)}{2}\,, \tag{1}$$

which contain the macroscopic polarization P, and the inversion density N, of the semiconductor. Ω is the Rabi frequency, Δ is the detuning from the 1s exciton resonance, and β is a phenomenological parameter which was estimated to be 0.15 by fitting to the full SBEs for the parameters of ZnSe [15]. In the regime of resonant excitation, the reduced SBEs can be applied if the Rabi energy is much smaller than the exciton binding energy and as long as the excited electron–hole density is not too high. By limiting our numerical simulation to Rabi energies up to 8 meV, we meet both criteria simultaneously. We have verified numerically, that for a Rabi energy of 8 meV, the maximum electron–hole plasma density is still less than 10% of the Mott density for ZnSe.

Applying Maxwell's equations, we tacitly ignore some difficulties associated with the half space problem of exciton polaritons, for a textbook discussion, see [16]. Since the finite exciton mass allows polaritons of common energy to propagate with different wave vectors, Maxwell's boundary conditions are not sufficient to describe the wave propagation through the surface. An additional boundary condition (ABC) has to be introduced resulting in a modification of the bulk dielectric function in a thin surface layer. Since the modification is of minor importance if the transition broadening exceeds the splitting of the polariton branches (ZnSe: $\Delta_{\mathrm{LT}} \approx 1$ meV [15]) we neglect this polariton effect.

We study the propagation of linearly polarized one-dimensional (plane wave) Gaussian-shaped pulses with the respective input Rabi energies of 0.2, 2, and 8 meV, which account for increasing pulse irradiances. In agreement with the experimental and material parameters we used a pulse duration of 150 fs, a central frequency resonant to the lowest exciton transition, and started with the condition $\alpha_0\lambda_{\mathrm{m}} = 4$. Fig. 3a shows the calculated reflected pulse spectra. In good agreement with the experiments a vanishing resonance structure and additional reflection well below the exciton resonance are observed for higher irradiances. The difference spectra, again shown in the inset, emphasize

that the applied model is appropriate to describe the nonlinear optical skin effect in a semiconductor. Slight differences between the experimental and the theoretical linear reflection should be attributed to uncertainties of the material parameters, e.g., inhomogeneous sample thickness and transition broadening, and the neglect of the ABC problem.

To show the propagating density front several snap-shots of the carrier densities distribution are shown in Fig. 3b. Although, the density front is not as sharp as in the two-level calculation it still results in an internal reflection clearly visible in the electric field distribution (not shown). The observed front propagation and the calculated reflection verify impressively the validity of the moving mirror picture. Using $(\Delta\lambda/\lambda) = (2v/c)$ for the Doppler shift $\Delta\lambda$ a front velocity v of 10^6 m/s can be estimated. To emphasize the propagation effect we reduced the layer thickness to 300 nm and found that the Doppler-shifted reflection disappeared.

Analyzing the theoretical results, another important difference between a semiconductor and a two-level system becomes apparent. In the latter case, an increasing irradiance always results in more pronounced self-reflection and Doppler shift. In a semiconductor, both effects disappear for too high incident fields, because band gap renormalization

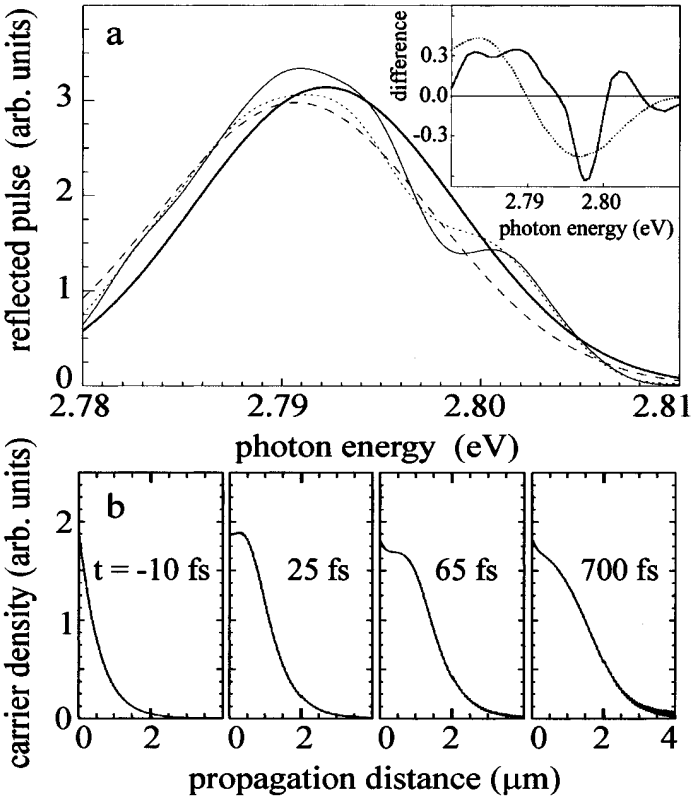

Fig. 3. Model calculation: a) Reflected pulse spectra for increasing Rabi energies Ω: 0.2 meV (solid line), 2 meV (dotted line), and 8 meV (dashed line). The incident pulse spectrum is also given (thick solid line). Inset: Spectrally resolved difference between reflected and incident pulses for $\Omega =$ 0.2 and 8 meV. b) Spatial distribution of the carrier density at different times

finally results in an infinite amount of electron–hole transitions. Furthermore, we verify the importance of a large excitonic binding energy by repeating our calculations using a binding energy of 10 meV with all other parameters remaining the same as above. We find that the Doppler shift of the reflection spectra becomes very small indeed. We anticipate that for semiconductors with smaller excitonic binding energies, e.g., GaAs, front propagation and self-reflection will be extremely difficult to achieve because of the stronger influence of the continuum states and the smaller linear absorption. We conclude that the occurrence of the dynamic nonlinear skin effect in semiconductors is limited to an intermediate range of laser pulse irradiances and to materials characterized by strong electron–hole Coulomb interaction such as ZnSe, and the group III nitrides for example.

In summary, we demonstrated experimentally and analyzed theoretically the excitation of a moving carrier density front in a ZnSe layer by 150 fs optical pulses. We observed red-shifted reflected light, which could be explained in terms of a Doppler-shifted self-reflected wave originating from the propagating density front.

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