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Optical nonlinearity and fast switching due to birefringence and mode coupling in CdS crystals

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Interference and mode coupling between ordinary and extraordinary waves lead in non-isotropic media to a rotation of the plane of polarization of the transmitted light. Placing the specimen between two polarizers, pronounced interference patterns are observed near the isotropic point. Pump and probe experiments with donor doped and undoped CdS crystals demonstrate for the st time a fast blue shift of the interference spectra by the action of intense laser light pulses. This gives rise to a nonlinear optical switching in the ns region. The induced change of the transmission due to saturation effects of the donors leads to a change of the birefringence $\Delta n = n_{\parallel} - n_{\perp}$. Under ps excitation, very fast rotations of the plane of polarization have been observed. Coherent phenomena like two photon absorption or the optical induced Stark effect are apparently responsible for relaxation processes with decay times in the sub-ps region.

1. Introduction

Interference of light waves plays an essential role in linear and nonlinear optical phenomena and related photonic devices. The superposition of reflected light beams is used, e.g., by interferometers, lasers, or in thin films to create dispersive optical bistability. Optical non-isotropic media exhibiting birefringence have also been studied often and used for applications. Nevertheless, only very few investigations on nonlinear optical phenomena are known. Apparently, this is due to the fact that birefringence effects usually are not very sensitive to the photon energy $\hbar\omega$. However, near the isotropic point of some uniaxial crystals like CdS, very narrow spectral structures occur if one places specimen of thicknesses in the mm range between two polarizers [1]. Due to a strong change of birefringence $\Delta n = n_{\parallel} - n_{\perp}$ with photon energy $\hbar\omega$, a series of maxima and minima with meV energy separation is observed. Normally, this effect is only seen for linearly polarized light with the $\boldsymbol{E}$ vector having a finite angle α against the c-axis of the crystal. Near $\Delta n = 0$, a coupling of waves with $\boldsymbol{E} \parallel \boldsymbol{c}$ and $\boldsymbol{E} \perp \boldsymbol{c}$ can also lead to a pronounced interference pattern [2,3]. A change of the refractive indices, their difference Δn, or of the coupling constant ρ under intense light excitation should induce very pronounced and fast nonlinear optical effects. For the first time, this is demonstrated here by experiments and careful theoretical analysis.

2. Experiments

For the two-colour pump and probe experiments with ns time resolution two pulsed dye lasers pumped by an excimer laser are used. The probe signal is detected by a fast photodiode ($\rho_{\text{rise}} = 100$ ps). Delays between pump and probe pulse from -5 to $+10$ ns can be realized. For the one-colour experiments in the ps time domain an active mode locked Nd:YAG laser with a frequency tripling BBO crystal synchronously pumping a dye laser with 3 ps autocorrelation width and 3.8 MHz repetition rate is applied. The transmitted probe signal is detected by a slow photodiode in lock-in technique, thus the signal measures directly the change of the transmission by the action of the pump. The delay time can continuously be varied from -500 to 4500 ps.

3. Experimental results

Fig. 1 demonstrates the interference pattern for an In doped CdS crystal near the isotropic point using incident and detected light, both polarized under $\alpha = 45°$. A blue shift of the structure with increasing pump power is shown in the inset. Maxima and minima change their position. In the region of the isotropic point a relatively high absorption due to the In doping occurs. Here, a pronounced bleaching due to saturation effects is observed at higher intensities, an effect already used to achieve optical bistability in these materials [4].

The nonlinear shift of the interference structure in the doped crystals occurs also at room temperature but has not been observed in undoped crystals.

Time dependent spectra for light transmitted through the same CdS : In sample under the same conditions are drawn in fig. 2. In fig. 2A the time shape of three pulses is given: the dotted line (a) represents the unchanged transmitted probe pulse measured without the analyser. Under the action of an intense pump pulse (line b), delayed by 2 ns to the probe pulse, a bleaching of the absorption coefficient is observed. The third curve (line c) shows the additional effect of the rotation of the plane of polarization ($E_{\text{analyzer}} \parallel E_{\text{polarizer}}$). In fig. 2B the ratio between the pumped probe pulse with and without the analyser is plotted as a function of time. Approximately 1 ns after the beginning of the bleaching, a steep decrease of the transmittance is observed followed by a slower increase. Two results are obvious: First, the pump pulse leads to a shift of the interference pattern of more than just half a period. The maximum changes first into a minimum and shift finally into the next maximum. Second, at the end of the pulse, the transmission signal does not return to the initial status, i.e. the relaxation time of the shift must be longer than 10 ns. This is in accordance with the results of measurements of the induced transmittance of In doped crystals [5].

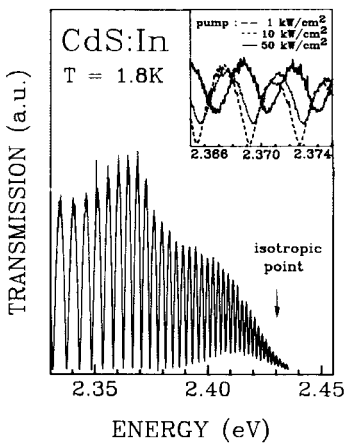

Fig. 1. Transmission spectrum of an In-doped CdS crystal placed between two parallel polarizers with an angle of $\alpha = 45°$ to the c-axis. Inset: shift of the interference pattern for three different pump intensities at $\hbar\omega = 2.435$ eV.

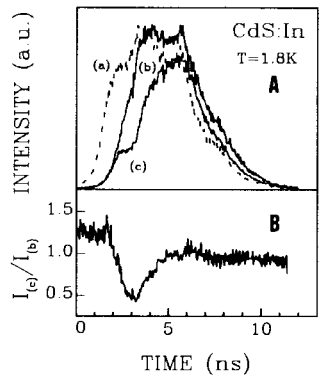

Fig. 2. (A) Time dependence of the transmission of an In-doped CdS crystal at $\hbar\omega = 2.428$ eV. (a) Probe pulse only, polarizer $\alpha = 45°$, no analyser (magnified by 50). (b) Probe pulse under the influence of pump pulse of 50 kW/cm^2; intensity at $\hbar\omega = 2.435$ eV; polarizer $\alpha = 45°$, no analyser. (c) Same as (b) but analyser $\beta = 45°$. (B) Intensity ratio $I(c)/I(b)$.

Under ps excitation, an entirely different behaviour is found. An undoped CdS crystal without the long relaxation times produced by the In donors was used. Fig. 3 shows the differential transmission spectra for probe light $E \parallel c$ with (a) and without (b) the analyser ($E \perp c$) between crystal and detector. Only for a total time overlap of both of the pulses is an effect observed. The measured change of transmittance without the analyser (curve b) is almost independent of the photon energy and can be interpreted as a two-photon absorption (TPA) or a two-step transition

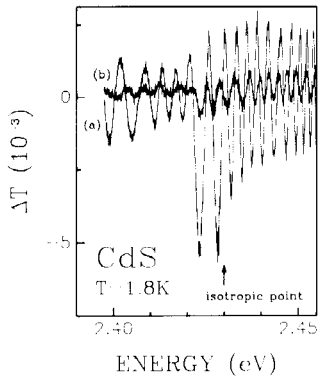

Fig. 3. One colour ps pump and probe spectra of an unpolarized CdS crystal at $\hbar\omega = 2.430$ eV for incident light $\boldsymbol{E} \| \boldsymbol{c}$: (a) with analyser $\boldsymbol{E} \perp \boldsymbol{c}$; (b) without analyser.

(TSA). The picture changes drastically applying the analyser (curve (a) of fig. 3). Now, at the chosen photon energy, an increase of the transmittance is observed. The differential transmission spectrum (DTS) as a function of the photon energy is given in fig. 3. Curve (a) shows the DTS without delay between pump and probe pulse. Here changes during the autocorrelation width are important. Curve (b) with $\delta t = -20$ ps shows the "long-term" effect with relaxation times greater than 250 ns. A strong oscillatory behaviour around the energy of the isotropic point is found.

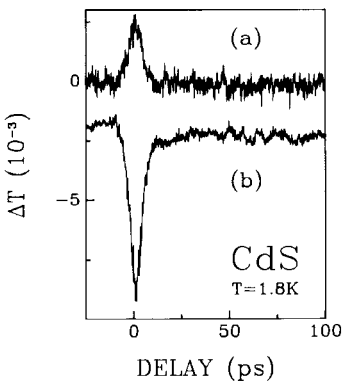

Fig. 4. Differential transmission spectra of an undoped CdS crystal for incident light $\boldsymbol{E} \| \boldsymbol{c}$ and detected light $\boldsymbol{E} \perp \boldsymbol{c}$. Pump intensity: 50 MW cm^{-2}. (a) Without delay between pump and probe. (b) Delay time -20 ps.

4. Discussion

A deeper understanding of the experimental results is achieved using the well-known theory [2,3] of birefringence and mode coupling of non-isotropic media. The dependence of the theoretical parameters on the intensity of the pump pulse should also explain the physical origin of the nonlinear phenomena.

We discuss a general "ansatz" for the optical behaviour of a linearly polarized light beam passing an uniaxial crystal in the x-direction. The amplitude of a wave with components $E_z(d)$ and $E_y(d)$ at $x = d$ for a direction forming an angle β against the z-direction is apparently

$$E_\beta(d) = E_z(d) \cos\beta + E_y(d) \sin\beta. \tag{1}$$

The electric field vector components obey the differential equations

$$\frac{\partial^2 E_z}{\partial x^2} + k_z^2 E_z = \rho E_y, \quad \frac{\partial^2 E_y}{\partial x^2} + k_y^2 E_y = \rho^* E_z, \tag{2}$$

with propagation vectors $\boldsymbol{k}_z(\omega)$ and $\boldsymbol{k}_y(\omega)$. The coupling constant ρ and its conjugate complex value ρ^* are correlated to the non-diagonal matrix elements of the dielectric tensor.

The birefringence is defined as

$$\Delta n(\omega) = n_\perp(\omega) - n_\parallel(\omega) = \frac{c}{\omega}\left[k_z(\omega) - k_y(\omega)\right] = \frac{c\,\Delta k(\omega)}{\omega}. \tag{3}$$

The solution of eq. (2) is:

$$\begin{aligned} E_z(x) &= C_1\, e^{i\gamma_1 x} + C_2\, e^{i\gamma_2 x}, \\ E_y(x) &= C_3\, e^{i\gamma_1 x} + C_4\, e^{i\gamma_2 x}, \end{aligned} \tag{4}$$

with

$$\gamma_{1,2}^2 = k^2 + \left(\tfrac{1}{2}\Delta k\right)^2 \pm \sqrt{k^2\,\Delta k^2 + \rho^2}, \tag{5}$$

$$k_x = k + \tfrac{1}{2}\Delta k, \quad k_y = k - \tfrac{1}{2}\Delta k. \tag{6}$$

The constants C_i are deduced from the initial conditions for a light beam with amplitude E_0

being linearly polarized with an angle α to the z-axis:

$$E_z(0) = E_0 \cos \alpha, \quad E_y(0) = E_0 \sin \alpha. \tag{7}$$

The constants C_i and γ_j in eq. (4) depend on the initial angle α of the plane of polarization at $x = 0$, the final angle β at $x = d$ (d = crystal thickness), the medium value k of the propagation vector, on the birefringence Δk and also on the coupling constant ρ.

First, we discuss the results of the pump and probe experiments in the ns region shown in figs. 1 and 2. In this case α and β are 45° and the coupling of waves can normally be neglected. Then, the solution of eq. (1) is simplified and depends only on Δk and d [2]. A change of the interference pattern with increasing laser pulse intensity, shown in fig. 1, should therefore be connected only with a change of the birefringence Δk. Indeed, the observation that crystals with high absorption in the region of the isotropic point are sensitive to this kind of nonlinearity give this direction: the strong saturation of the absorption is certainly connected with a change of the real part of the refractive index and acts apparently with different strength to k_z and k_y. As the saturation effect is the stronger the nearer the absorption occurs to the band edge, Δk changes the more effective the higher the photon energy is. This can be approximated by shifting the whole $\Delta k(\omega)$ curve by a certain value.

In fig. 5 the interference pattern calculated from our "ansatz" is plotted for values of $\Delta k(\omega)$ taken from literature [6]. By shifting $\Delta k(\omega)$ by about $\Delta\hbar\omega = 1.5$ meV, corresponding to a change of Δk by 15 cm^{-2}, the situation of the inset of fig. 1 (shift by 1/2 period) is simulated. In the short wavelength region this leads to a quite larger change of Δk of 30 cm^{-2} and to a movement of the pattern by more than a full period (right inset of fig. 5). Calculations in the frame of the Kramers–Kronig relations are planned to correlate the change of Δk with the observed saturation of the absorption.

Our results explain also nicely the time dependent experiments of fig. 2. At the beginning of the laser pulse, a shift of the interference structure and a switching from high to low transparency of the arrangement starts. At times greater than 5 ns, Δk rises further and the plane of polarization is tuned about 360° (right of inset of fig. 5). For times longer than 20 ns, where the pump pulse is almost decreased to zero, no backswitching occurs. Indeed, the decay time of the saturation effect is in the range of 250 ns, a time too long to be observed with our experimental set-up.

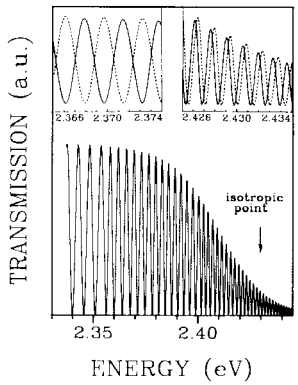

Fig. 5. Calculated transmission spectrum corresponding to fig. 1: $\Delta\hbar\omega$ from literature [6], $\alpha = \beta = 45°$, $\rho \approx 0$, $d = 0.18$ cm. Insets: solid line, original spectra; dotted line, spectrum shifted by $\Delta\hbar\omega = 1.5$ meV.

A more complicated situation holds for the described experiments in the ps region. Here the coupling constant ρ is of importance, and in addition a small deviation of the angles α and β plays an important role for the experimental results. The general form of the solution of eqs. (1) to (7) has to be used to compare experiment with theory. The fast effect turning the plane of polarization and thus the DTS signal, shown in figs. 3 and 4, could have been generated by a fast change of Δk, k, and ρ, as their values determine the interference pattern entirely. A careful analysis shows that neither a change of k alone nor of the coupling constant ρ could explain our results.

As in the case of the ns experiments in the ps time domain, the remaining explanation is to assume a change of Δk with excitation intensity. This could be affected either by TPA inducing a

change of the refractive index, or by the action of the high electric field of the electromagnetic wave on electronic and excitonic states near the band gap. This is in accordance with well-known phenomena like the Franz–Keldysch effect for band–band transitions or the confined Stark effect for free excitons. Assuming an induced shift of the interference pattern of $\Delta\hbar\omega = 0.5$ meV, a satisfying agreement between theory and experiment could be achieved.

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