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NON-LINEAR OPTICAL PROCESSES IN BIREFRINGENT AND DICHROITIC WIDE BAND GAP SEMICONDUCTORS

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Abstract

The influence of both birefringence and dichroism on the shape of optical linear and non-linear interference transmission spectra is demonstrated for undoped hexagonal CdS-crystals. Using two color pump and probe spectroscopy , it is shown that the non-linear differential transmission spectra at the isotropic point are governed by two main effects: Free and bound exciton absorption is responsible for a component decaying with a time constant of about 500 ps, while the very fast relaxing part can be explained by two photon excitation processes.

1. Introduction

Optical non-linear phenomena in semiconductors have been intensively studied, both to understand their basic origin and to find new materials for optoelectronic applications. In general, the optical non-linear response is small and methods to measure the dielectric tensor ε^* and especially its dynamical behaviour very sensitively have to be applied.

In several wide band semiconductors interference spectroscopy allows a very accurate measurement of the complex refractive index $n^* = n + ik$. Due to excitonic effects some hexagonal II-VI-compounds like CdS show near the band edge the optical phenomena birefringence $\Delta n = n_{\parallel} - n_{\perp}$ and dichroism $\Delta k = k_{\perp} - k_{\parallel}$. Of great interest for optical studies is that Δn changes its sign at a certain wavelength ω_0 called the isotropic point. Here,. wave mixing by applying an appropriate strain or magnetic field to the crystal can occur[1-3]. Because of the disappearing of Δn the dichroism Δk governs especially intense[4] the optical properties at ω_0.

In the following we show at first how sensitive the linear interference spectroscopy allows to evaluate the optical constants of pure CdS. Performing pump and probe experiments we demonstrate the influence of the non-linear changes of optical parameters on the interference spectra. In pure crystals the dynamical effects are fast and range from some ps to several ns. From excitation spectra the non-linearity can be demonstrated to origin either by free and bound exciton absorption or by two-photon excitation.

2. Linear Inferference Spectroscopy

In fig. 1 we show linear interference transmission spectra of a CdS crystal in the unstrained (curve A) as well as in the strained (curve B) case for a configuration with crossed polarizers under 45 degrees against the c-axis of the specimen (interference pattern spectra). The strain has been applied to the probe by fixing it at room temperature on a metal plate and then cooling it down to liquid helium temperature. The different expansion coefficients of crystal and metal substrate create an intense strain which is big enough to induce wave mixing in the pass-band configuration (crossed polarizers parallel and perpendicular respectively) (Curve C).From the unstrained curve A, the optical parameters Δn and Δk can be determined very accurately. The spectrum changes drastically if a stress is applied (curve B): Now at the energy of the isotropic point a single deep minimum appears flanked by several higher lying minima. In addition, the spacing of the interference structure has changed as well as the width of the pattern at higher photon energies.Finally, in curve C in the pass-band filter configuration the wave mixing effect is demonstrated. Now a dominant doublet exists, centered by a deep minimum at ω_0 while theory shows that with negligible dichroism only one broad maxima should occur.

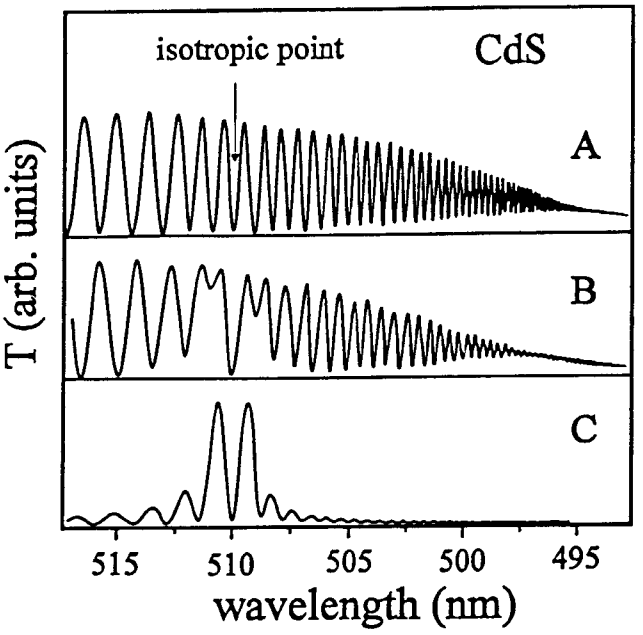

Figure 1. Interference spectra of CdS at 2 K. A) Crossed Polarizers under 45 degrees, unstrained. B) Crossed Polarizer under 45 degrees, strained. C) Pass-Band spectrum.

We have shown that this behaviour can be understood only, if we assume: The appearence. of a wave coupling ρ ,which turns the plane of polarisation at ω_0 by about 90 degrees, and a considerable change of Δn and Δk. A detailed calculation of all this is given elsewhere[3].

3. Non Linear Interference Spectroscopy.

Having understood in principle the linear behaviour of interference transmission spectra in pure CdS crystals we can now use our knowledge to determine the parameters which are most sensitive for non-linear effects. In a former paper[5] we have shown that highly In doped CdS specimen change their spectra dramatically by using high intensity laser light in the interference pattern configuration. The life time was determined to be about 100 ns. For nominally undoped specimen, one-color pump and probe experiments resulted in relatively small effects, with relaxation times not exceeding several ps. The origin of this interesting kind of non-linearity could not yet clearly be determined.

Here, we report on experimental results with a two-color laser system, where the pump and the probe laser with a pulse length of 3 ps can be tuned independently from each other. To be able to measure small non-linear transmission changes in the order of magnitude of 10^{-1} to 10^{-5} a modulation technique is applied.

In fig. 2 the excitation spectra for the probe laser positioned at the photon energy of the isotropic point are shown for two different configurations. The striking effect is the existence of maxima resp. minima at prominent energies of free and bound excitons. Clearly, the creation of excitonic complexes changes the structure of the interference spectra. It is interesting to note that in the configuration A (crossed polarizers), where there is a minium of transmission (see fig. 1), we have a negative signal, i. e. a decrease of the transmission, while in the configuration B where there is a maximum of transmission an opposite behaviour can be seen. It becomes evident that this behaviour must be correlated to the coupling of waves at ω_0.

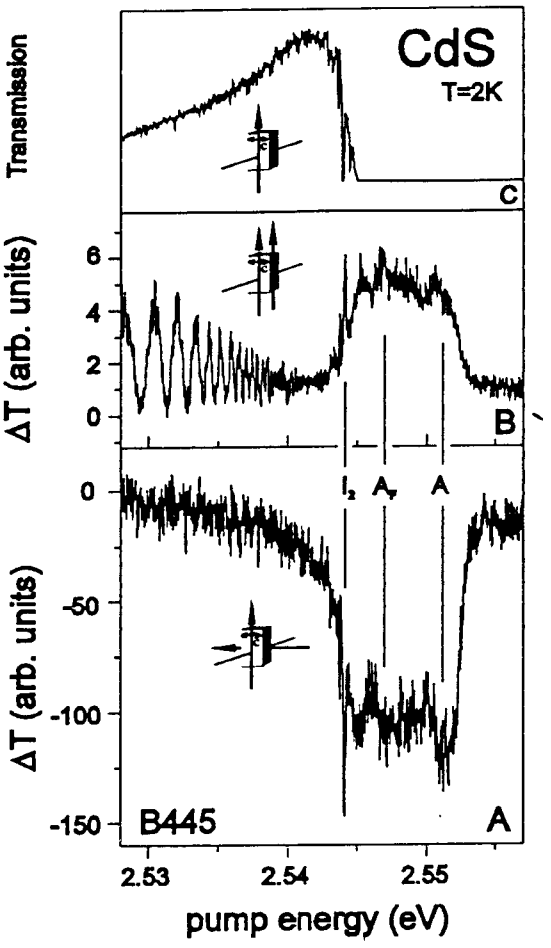

Figure 2.. A) Linear transmission spectrum. B) and C) Excitation-spectra of the DTS-signal for two different configurations at ω_0.

An important tool to understand the mechanism of the non-linear change of the interference pattern is the time-delayed two-color-pump-and-probe spectroscopy (fig. 3). Here, we show the development of a differential transmission spectrum (DTS) in the pass-band configuration from zero delay between the probe and pump laser pulses to a time of about 250 ns, the pulse period of the laser. In the first several ps we observe a fast decay followed - as could be shown by more detailed experiments - by a larger relaxation time of about 500 ps and a rest which exists up to μ-seconds. It is important to note, that the relaxation time of the induced absorption at ω_0 excited in the exciton region could be measured to be about 500 ps.

4. Discussion

The pump and probe spectra show mainly two different phenomena: The observed decrease of the transmission in the region of ω_0, resulting from excitation by a pump-frequency in the free and bound excitation, relaxes with the decay time of these excitonic complexes of about 500 ps. The DTS signal shows the same time dependence but is negative for minima and positive for maxima in the pass-band interference structures. This is understandable if the decrease of transmission is connected with an increase of the dichroism. A decreasing

transmission for both polarisations will clearly increase Δk and thus change the interference spectra especially intense at ω_0. As in the band-pass configuration (fig. 2, curve A) the increase of k and Δk act in the same direction, a strong negative effect is observed.In the case of parallel polarizers (fig. 2, curve B) both phenomena add in opposite directions the positive regime of excitation spectra is much smaller than the positive one in curve B.

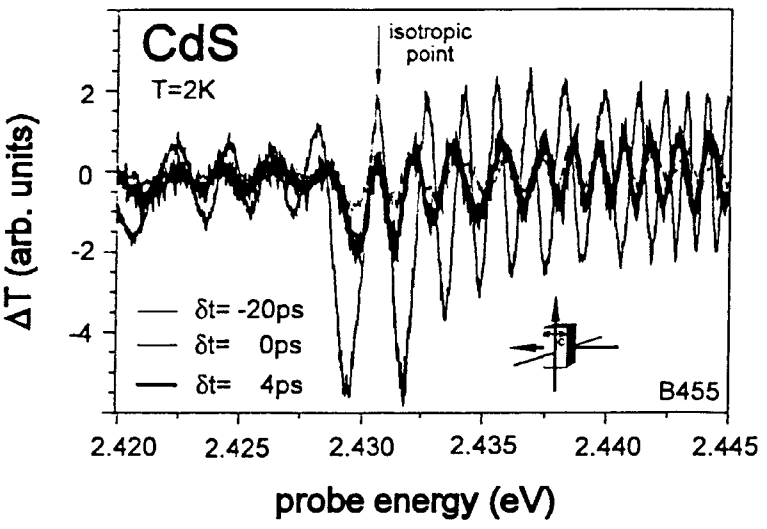

Figure 3. Time delayed DTS-spectra in the band-pass configuration near ω_0.

One possible explanation for the ps relaxation times would be a two-photon exitation process over virtual states in the region of the exciton complexes resulting - according to the Kramers-Kronig relations - in a change of the real part of n*. This gives rise to a short living shift of the maxima and minima connected with a DTS signal as shown in fig. 3. Indeed, it can be demonstrated by theoretical analysis that a shift of the whole spectral position of Δn by a few cm^{-1} can explain not only the oscillations of the DTS signal but also their spectral shift with delay times.

5. Conclusion

In conclusion, it could be shown that the linear and non-linear behaviour of the interference spectra in CdS can be understand only, if both the birefringence and the dichroism and their change at high pump intensities are taken into consideration.

The enlargement of the sensitivity of the non linear effects by applying the methods of interference spectroscopy connected with very fast relaxation times shows possibilities to apply the above discussed phenomena for the development of switching devices in the ps region.One important advantage is, that pumping and probing can be achieved at different photon energies.

6. References

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