Simultaneous Two-Photon Absorption and Second Harmonic Generation in CdSe

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Abstract. A hexagonal CdSe crystal is excited with picosecond Nd:glass laser pulses. Two-photon absorption and resonant non-phasematched second harmonic generation occur simultaneously. Using different crystal orientations all components of the second harmonic susceptibility tensor and some components of the two-photon absorption susceptibility tensor are determined.

1. Introduction

Two-photon absorption (TPA) and second harmonic generation (SHG) occur simultaneously when picosecond light pulses of a Nd:glass laser (wavelength λ_L = 1.054 µm, duration $\Delta t_L \approx$ 6 ps) pass through a hexagonal CdSe crystal (wurtzite structure 6mm point symmetry).

Previous TPA studies are summarized in [1,2]. The simultaneous occurrence of TPA and SHG is studied scarcely [3,4]. Excited state absorption of the two-photon generated carriers was observed for nanosecond excitation pulses [4,5].

Intensity dependent transmission measurements of the pump laser and second harmonic conversion efficiency measurements are carried out to separate TPA and SHG. A detailed theoretical treatment is given elsewhere [6].

2. Experimental

Single picosecond light pulses separated from a passively mode-locked Nd:phosphate glass laser were applied in the experiments. The size of the CdSe crystal (from Cleveland Crystals Inc., selenium treated for high resistivity) was 12 × 12 × 10 mm with the c-axis parallel to a 12 mm edge. Energy transmission measurements of the pump laser (T_E) and SHG energy conversion efficiency measurements (η_E) were performed. Four different interaction schemes of the pump pulses with the crystal were studied (see Table 1) to determine all non-zero SHG susceptibility components d_{ij} [7] and some TPA coefficients $\alpha^{(2)}_{ijkl}$.

3. Results

The T_E and η_E results of the oo→e interaction (wavevector $\mathbf{k}_L \perp$ c-axis and electric field strength $\mathbf{E}_L \perp$c-axis) are presented in Fig.1. The pump pulse reduction by second harmonic generation

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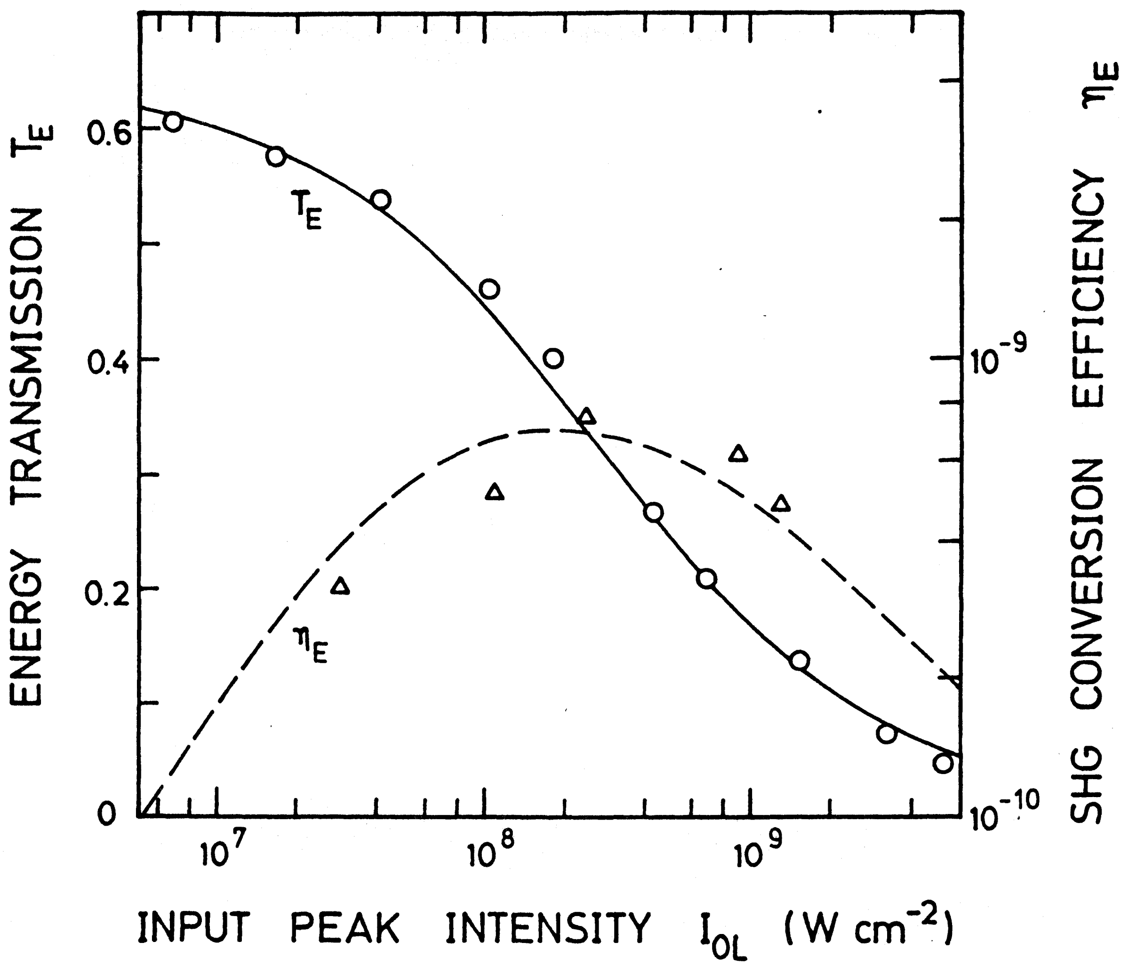

Fig.1: o——o: Energy transmission T_E. Δ---Δ: SHG energy conversion efficiency η_E. oo→e interaction. Sample length ℓ=1 cm. Fit parameters see Table 1.

is estimated to be $T_{E,SHG} \approx 1-\eta_E \ell \alpha_2$ where ℓ is the sample length and α_2 the linear absorption coefficient at the second harmonic frequency (ordinary ray o: $\alpha_{2o} \approx 1.2\times10^5$ cm^{-1}, extraordinary ray e: $\alpha_{2e} \approx 1.5\times10^5$ cm^{-1} [8,9]). η_E is found to be two orders of magnitude too small to influence the energy transmission T_E.

The experimental points in Fig.1 are fitted by theoretical curves [6]. The T_E curves include no excited state absorption and the good fit to the experimental data indicates that an absorption of the two-photon generated carriers is negligible at λ_L = 1054 nm.

In Table 1 the investigated crystal orientations relative to the laser pulses, the SHG interaction processes, and the fitted SHG susceptibility coefficients d_{ij} and TPA coefficients $\alpha^{(2)}_{ijkl}$ are listed.

4. Conclusions

In non-centrosymmetric crystals TPA and resonant non-phase-

Table 1: Interaction schemes and corresponding SHG and TPA coefficients. $T_{E,x}$ ($T_{E,z}$) is transmission of laser light polarized $\perp$($\|$) to c-axis.

Orientation $(\mathbf{k}_L, \mathbf{c})$ (degrees)	$(\mathbf{E}_L, \mathbf{c})$ (degrees)	Interaction	SHG d_{ij} (10^{-11}m V^{-1})	TPA $\alpha^{(2)}_{ijkl}$ (10^{-10}m W^{-1})
0	90			$\alpha^{(2)}_{xxxx}$:1.8$\pm$0.3
90	90	oo→e	d_{31}:2.5$\pm$0.3	$\alpha^{(2)}_{xxxx}$:1.8$\pm$0.3
90	0	ee→e	d_{33}:3.9$\pm$0.3	$\alpha^{(2)}_{zzzz}$:1.8$\pm$0.3
90	45	oe→o	d_{15}:2.3$\pm$0.3	
			$T_{E,x}$: $\alpha^{(2)}_{xxxx}$ and	$\alpha^{(2)}_{xzzx}+\alpha^{(2)}_{xzxz}$:0.3$\pm$0.3
			$T_{E,z}$: $\alpha^{(2)}_{zzzz}$ and	$\alpha_{zxxz}+\alpha_{zxzx}$:0.3$\pm$0.3

matched second harmonic generation occur simultaneously. Energy transmission and second harmonic conversion efficiency measurements are necessary to separate the contributions of the SHG and the TPA to the nonlinear transmission.

The measured energy transmissions of CdSe versus input pump pulse peak intensity may be used as calibration curves for the peak intensity detection of picosecond light pulse by energy transmission measurements [10].

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