Measurements of Self- and Cross-Phase Modulation Coefficients of the Crystals using a modified Z-scan technique

Nonlinear Optics and Spectroscopy Laboratory

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1. Introduction

The third-order nonlinearity is the lowest-order optical nonlinearity that is present in all materials [1,2]. It is responsible for many processes such as third-harmonic generation (THG), nonlinear absorption (NLA), nonlinear refraction (NLR), self-phase modulation (SPM), cross-phase modulation (XPM) and others . In the interaction of strong fields with matter the third-order susceptibility of media can be detrimental to quality of interacting pulses by introducing spatio-temporal phase distortion. Cubic nonlinearity can strongly influence the efficiency of the processes governed by the second-order nonlinearity such as THG [3], high-intensity second harmonic generation (SHG) and optical parametric amplification (OPA) [4]. For determination of the third-order susceptibility of crystals different nonlinear techniques were used. The properties depending from the crystal orientation were omitted in the most of these studies or used incorrectly [5].

2. Symmetry properties of third-order interactions in crystals

It should be noted that the effective nonlinear refraction coefficients of crystals usually depends very strongly on the wave propagation direction and its polarization. They are proportional to the certain combination of the elements of the cubic susceptibility tensor [1,2,6]. The number of independent components of this tensor depends upon the symmetry of crystal and the type of nonlinear interaction. The use of the spatial symmetric properties of the crystals shows that there are 21 nonzero coefficients of which only 11 are independent for crystal class . The number of independent coefficients decreases to 7 for nonlinear optical Kerr effect. In the dispersionless approach this number is equal to 5 for THG and to 4 for optical Kerr effect.

$$\tilde{P}_i(\mathbf{r},t) = \varepsilon_0 \int_{-\infty}^{t}\int_{-\infty}^{t}\int_{-\infty}^{t} \chi^{(3)}_{ijkl}(\mathbf{r},t',t'',t''')\tilde{E}_j(\mathbf{r},t')\tilde{E}_k(\mathbf{r},t'')\tilde{E}_l(\mathbf{r},t''')\,dt'dt''dt'''$$

Spatial symmetry

$$\chi^{(3)}_{ijkl}(\mathbf{r},t',t'',t''') \equiv \chi^{(3)}_{ijkl} \Rightarrow$$

$$422,\quad 4mm,\quad \bar{4}2m,\quad 4/mmm \Rightarrow$$

$$\begin{bmatrix} \chi^{(3)}_{xxxx} = \chi^{(3)}_{yyyy} & \chi^{(3)}_{zzzz} & 0 \\ \chi^{(3)}_{yyzz} = \chi^{(3)}_{xxzz} & \chi^{(3)}_{yzzy} = \chi^{(3)}_{xzzx} & \chi^{(3)}_{xxyy} = \chi^{(3)}_{yyxx} \\ \chi^{(3)}_{zzyy} = \chi^{(3)}_{zzxx} & \chi^{(3)}_{yzyz} = \chi^{(3)}_{xzxz} & \chi^{(3)}_{xyxy} = \chi^{(3)}_{yxyx} \\ \chi^{(3)}_{zyyz} = \chi^{(3)}_{zxxz} & \chi^{(3)}_{zyzy} = \chi^{(3)}_{zxzx} & \chi^{(3)}_{zyyz} = \chi^{(3)}_{yxxy} \end{bmatrix}$$

OKE

$$(ij \leftrightarrow \mu = 1,\dots,6; kl \leftrightarrow \nu = 1,\dots,6$$
$$\Leftrightarrow 11 \leftrightarrow 1,\ 22 \leftrightarrow 2,\ 33 \leftrightarrow 3,$$
$$23 \leftrightarrow 4,\ 13 \leftrightarrow 5,\ 12 \leftrightarrow 6\ ) \Rightarrow$$
$$\Rightarrow \chi^{(3)}_{ijkl} \equiv \chi_{\mu\nu} \Rightarrow$$

$$\begin{bmatrix} \chi_{11} & \chi_{12} & \chi_{13} & 0 & 0 & 0 \\ \chi_{12} & \chi_{11} & \chi_{13} & 0 & 0 & 0 \\ \chi_{31} & \chi_{31} & \chi_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & \chi_{44} & 0 & 0 \\ 0 & 0 & 0 & 0 & \chi_{44} & 0 \\ 0 & 0 & 0 & 0 & 0 & \chi_{66} \end{bmatrix}$$

THG

$$\begin{bmatrix} jkl & xxx & yyy & zzz & yzz & yyz & xzz & xxz & xyy & xxy & xyz \\ m & 1 & 2 & 3 & 4 & 5 & 6 & 7 & 8 & 9 & 0 \end{bmatrix}$$

$$\chi^{(3)}_{ijkl} \equiv \begin{bmatrix} C_{11} & C_{12} & C_{13} & C_{14} & C_{15} & C_{16} & C_{17} & C_{18} & C_{19} & C_{10} \\ C_{21} & C_{22} & C_{23} & C_{24} & C_{25} & C_{26} & C_{27} & C_{28} & C_{29} & C_{20} \\ C_{31} & C_{32} & C_{33} & C_{34} & C_{35} & C_{36} & C_{37} & C_{38} & C_{39} & C_{30} \end{bmatrix}$$

Dispersionless OKE

$$\chi^{(3)}_{xxxx} = \chi_{11} = C_{11} \qquad \chi^{(3)}_{xxyy} = \chi_{12} = C_{18}$$
$$\chi^{(3)}_{yyzz} = \chi_{23} = C_{16} \qquad \chi^{(3)}_{zzzz} = \chi_{33} = C_{33}$$

Wave propagation and OKE in the DKDP crystal

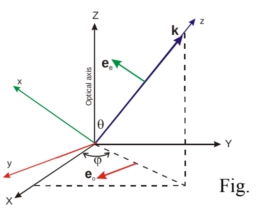

Fig. 1

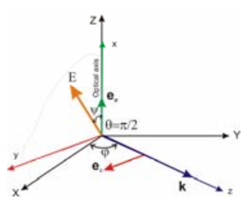

Fig. 2

$$\chi^{(3)}_{oooo} = \chi_{11} - \frac{1}{2}(\chi_{11} - 3\chi_{12})\sin^2 2\varphi$$

$$\chi^{(3)}_{oeoe} = \chi^{(3)}_{eoeo} = \frac{1}{2}(\chi_{11} - 3\chi_{12})\sin^2 2\varphi\cos^2\Theta + \chi_{12}\cos^2\Theta + \chi_{23}\sin^2\Theta$$

$$\chi^{(3)}_{eeee} = \chi^{(3)}_{oooo}\cos^4\Theta + \frac{3}{2}\chi_{23}\sin^2 2\Theta + \chi_{33}\sin^4\Theta$$

3. Z - scan method

One of the simplest methods for the measurement of the effective nonlinear coefficients of refraction is the Z-scan method [2,7-9] .

$$T(x,t) = 1 + \frac{4\Delta\Phi_0(t)x}{(1+x^2)(9+x^2)};\quad x = \frac{z}{z_R};$$

$$\Delta\Phi_0(t) = k\gamma I_0(t)L_{eff};$$

$$\langle\Delta\Phi_0\rangle = \frac{k\gamma I_0 L_{eff}}{\eta};$$

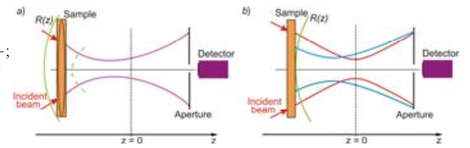

Fig. 3

4. Measurements of the third-order susceptibility tensor elements of the DKDP crystal

In this paper, the effective nonlinear coefficients for SPM and XPM in different crystals are found taken into account crystal dispersion properties and in the dispersionless approach also.

Experimental measurements of these coefficients for some nonlinear and laser crystals were undertaken using the experimental setup shown in Fig. 4. In the experiments the diode pumped minilasers with pulse duration about 1 ns and pulse energy ~ 2 mJ were used. For the measurement of the effective SPM and XPM coefficients additional polarization analyzer A before the diaphragm D is placed. This arrangement allows to find more easily the independent elements of .

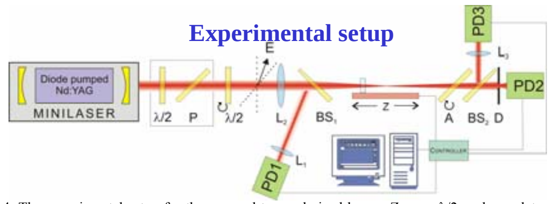

Fig. 4. The experimental setup for the one and two polarized beams Z-scan: λ/2 – phase plates for wave **E** polarization rotation, polarizer P and analyzer A, focusing lenses $L_{1,2,3}$, beam splitters $BS_{1,2}$, diaphragm D and photodiodes $PD_{1,2,3}$.

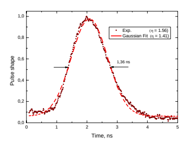

Fig. 5

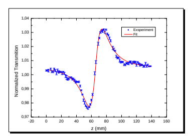

Fig. 6

Table 1: Nonlinear refractive indexes for different polarizations and different beam propagation directions in DKDP crystals

Crystal	Cut Direction	Polarization	n_2 [10^{-16} cm^2 W^{-1}]
DKDP	Θ = 59, φ = 0	o	3.45
DKDP	Θ = 59, φ = 0	e	2.25
DKDP	Θ = 90, φ = 45	o	3.04
DKDP	Θ = 90, φ = 45	e	2.91

Table 2: Measured elements of the DKDP tensor $\chi^{(3)}$

$\chi_{11} = 1.95\ 10^{-14}$	$\chi_{33} = 1.65\ 10^{-14}$	$\chi_{12} = 0.50\ 10^{-14}$

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