THE NONLINEAR OPTICAL PROPERTIES OF $\text{Sn}_2\text{P}_2\text{S}_6$

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Abstract We have studied the optical second harmonic generation (SHG) from a $\text{Sn}_2\text{P}_2\text{S}_6$ crystal for fundamental wavelengths between 1100 to 1200 nm. The SHG signal was compared with that from a KTP crystal and it was found that $d_{21}(\text{Sn}_2\text{P}_2\text{S}_6) = 3.6 * d_{33}(\text{KTP})$.

INTRODUCTION

$\text{Sn}_2\text{P}_2\text{S}_6$ is a monoclinic nonlinear optical crystal and at room temperature it has a transmission range from 550 to 8000 nm. These two qualities make $\text{Sn}_2\text{P}_2\text{S}_6$ a very interesting material for nonlinear optical processes in the infrared.

To determine the nonlinear optical coefficients of $\text{Sn}_2\text{P}_2\text{S}_6$, the optical second harmonic generation (SHG) has been studied as a function of the incident wavelength, and compared with the SHG signal from KTP.

EXPERIMENTAL SET-UP

For the wavelength dependent SHG experiments an optical parametric oscillator (OPO) with a KTP crystal was used. This OPO was angle tuned and pumped with the second harmonic of a Nd:YAG laser, producing 7 ns pulses in the wavelength region from 950 to 1210 nm.

The two generated beams of the OPO were separated with a polarizer and the lowest frequency of these two was used for the experiment. This beam was focused by a 10 cm lens into the sample and the SHG signal was detected with a photomultiplier in combination with a monochromator.

MEASUREMENTS AND RESULTS

The $\text{Sn}_2\text{P}_2\text{S}_6$ crystal, grown by the Bridgmann method at the Uzhgorod State University, was cut parallel to the xy-plane and had a thickness of 2.25 mm. The flux grown KTP reference crystal, was cut parallel to the xz-plane and had a thickness of 3.45 mm.

For the measurements on KTP the fundamental beam was polarized along the z-direction. This gives an SHG signal, which is polarized along the z-direction, due to

FIGURE 1: SHG signal from $\text{Sn}_2\text{P}_2\text{S}_6$ divided by the SHG signal from KTP as a function of the fundamental wavelength. The dashed line is a guide to the eye.

the nonlinear optical coefficient d_{33} . To measure the Maker-fringes the crystal was rotated in the xy-plane. In this way there are no other d-coefficients involved in the SHG process.

For the measurements on $\text{Sn}_2\text{P}_2\text{S}_6$ the fundamental beam was polarized in the xz-plane and the detected SHG signal was polarized along the y-direction. The rotation for the Maker fringes was made in the xz-plane. In this case the SHG signal will depend on d_{21} , d_{23} and d_{25} , because $\text{Sn}_2\text{P}_2\text{S}_6$ is a monoclinic crystal¹. Due to the fact that $\text{Sn}_2\text{P}_2\text{S}_6$ has a high refractive index (table 1) the influence from d_{23} and d_{25} at a small angle of incidence can be neglected.

In figure 1 the ratio between the SHG signal from $\text{Sn}_2\text{P}_2\text{S}_6$ and KTP is plotted as a function of the fundamental wavelength, showing a strong increase of this ratio for higher wavelengths. This increase can be ascribed to the fact that for $\text{Sn}_2\text{P}_2\text{S}_6$ the SHG wavelength lies near its absorption edge of 550 nm.

To calculate the nonlinear optical coefficients of $\text{Sn}_2\text{P}_2\text{S}_6$ we have assumed that at 600 nm the absorption might be neglected. In that case the conversion efficiency η at a Maker-fringe maximum is given by¹:

$$\eta = \frac{P_{2\omega}}{P_{\omega}} = \frac{2 d_{eff}^2}{\epsilon_0 c n_{\omega}^2 n_{2\omega}} \frac{P_{\omega}}{A} \frac{1}{(n_{2\omega} - n_{\omega})^2} \quad (1)$$

where P_ω is the power of the fundamental input beam, $P_{2\omega}$ is the power of the generated SHG beam, λ is the fundamental wavelength, n_ω is the refractive index at ω and A is the spotsize. d_{eff} is the effective nonlinear coefficient and depends on the propagation direction and polarization of the fundamental beam.

In the case of KTP d_{eff} is equal to d_{33} and for $\text{Sn}_2\text{P}_2\text{S}_6$ it is equal to d_{21} .

Calculating $d_{21}(\text{Sn}_2\text{P}_2\text{S}_6)$ with the help of eq. (1), correcting for the Fresnel losses and using the refractive indices as given in table 1, we find for a fundamental wavelength of 1200 nm

$$d_{21}(\text{Sn}_2\text{P}_2\text{S}_6) = 3.6 d_{33}(\text{KTP}) \quad (2)$$

Vanherzeele et al.² measured $d_{33}(\text{KTP}) = 18.5 \text{ pm/V}$ at a fundamental wavelength of 880 nm. Making use of Miller's rule gives $d_{33}(\text{KTP}) = 16.2 \text{ pm/V}$ at 1200 nm and thus it follows that $d_{21}(\text{Sn}_2\text{P}_2\text{S}_6) = 58 \text{ pm/V}$ at this wavelength.

TABLE I. Refractive indices for KTP ³ and $\text{Sn}_2\text{P}_2\text{S}_6$ at 600 and 1200 nm

	KTP		$\text{Sn}_2\text{P}_2\text{S}_6$	
	600nm	1200nm	600nm	1200nm
n_1	1.767	1.734	3.161	2.846
n_2	1.776	1.742	3.070	2.814
n_3	1.872	1.825	2.980	2.739

CONCLUSIONS

The nonlinear coefficient d_{21} of $\text{Sn}_2\text{P}_2\text{S}_6$ is very high compared to KTP, even if we assume that at 600 nm there is no absorption.

So we can conclude that $\text{Sn}_2\text{P}_2\text{S}_6$ is a very promising material for nonlinear optical processes in the infrared, because of its high nonlinear coefficient and its transparency up to 8000 nm.

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