$MgSO_3.6H_2O$ - a new crystal for nonlinear optics

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ABSTRACT

The first investigation on the nonlinear optical properties of Magnesium sulfite hexahydrate monocrystals are reported. The crystal has been grown, for the first time according to our knowledge, by our new original method, that allows them to reach up to 40 - 50 mm linear size. Both Type I and Type II phase matched effective second harmonic generations (SHG) are observed when the crystal is pumped with Nd:YAG laser radiation. First data for $MgSO_3.6H_2O$ indici of refraction dispersion and for its second order susceptibility tensor components are presented.

Keywords: nonlinear optics, second harmonic generation, nonlinear susceptibility, new nonlinear materials, $MgSO_3.6H_2O$.

1.INTRODUCTION

The search for new nonlinear optical media is a very important activity in connection to the fast growing application of nonlinear optical devices both in science and technology. Such devices as harmonic generators, optical parametric oscillators, phase conjugators, correlators are iireplaceable parts in many experimental scientific and technological set-ups.

The requirements to the new nonlinear media include large size, high value of nonlinear susceptibility, high damage threshold, resistance to atmospheric factors, transparency in a wide spectral range and possibility of angular or temperature phase matching.

Another important role of the investigations of new nonlinear media is the verification of existing theoretical models in order to find their possibility to predict the nonlinear properties of the different crystal structures. It is important to point out that there are no reports about other nonlinear optical crystals from the point group C_3 , to which $MgSO_3.6H_2O$ belongs. The results of our investigations of the $MgSO_3.6H_2O$ physical properties so far are presented here. Data for its tranparency range, indici of refraction dispersion and nonlinear susceptibility tensor are reported for the first time. We have observed effective phase matchable second harmonic generation with both Type I and Type II interactions in a wide spectral range.

2.LINEAR OPTICAL PROPERTTIES

$MgSO_3.6H_2O$ large linear size monocrystals, reaching 40-50 mm and more, are grown by our new original method [1]. The only previous publications about that crystal, [2], [3], [4], are dealing with monocrystals, obtained by traditional methods, which produce them not larger than 1-2 mm.

Transmission curves of three different crystal samples, from 6 mm to 14 mm thick, are presented at Fig. 1. The transparency range is from 200 nm to 1500 nm. Losses in the samples vary fron 5 % to 20 % because of different optical quality. Further improvements in our growth method techniques will lead to reduction of those losses.

Table 1		
λ, (nm)	n_o	n_e
435.8	1,5281	1,4822
532	1,5186	1,4752
546	1,5177	1,4744
632.8	1,5124	1,4696
1064	1,5005	1,4587

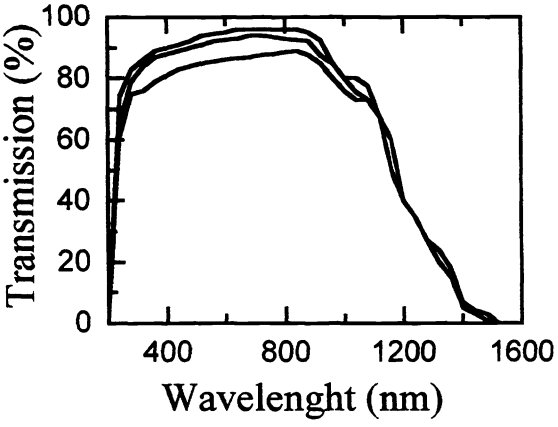


Fig.1 Transmission of the $MgSO_3.6H_2O$ elements

The present equipment in our posession does not allow good accuracy measuremets of refraction index. However our measurements show large enough birefringence that allows phase matchable second harmonic generation in a wide spectral range. The values of $n(\lambda)$ are presented at Fig. 2 and Table 1. Data marked by * and ** are obtained from experimentally measured phase matching angles for Type I and Type II second harmonic generation at 1064 nm fundamental wavelength.

3.SECOND HARMONIC GENERATION IN $MgSO_3.6H_2O$

The experimental setup used for nonlinear optical properties investigation of $MgSO_3.6H_2O$ samples consists Nd:YAG laser that generates nano-second pulses of approximately 20 ns duration with 10 mJ energy at 1064 nm fundamental wavelength. Energy is measured by Energy meter Rj 7200.

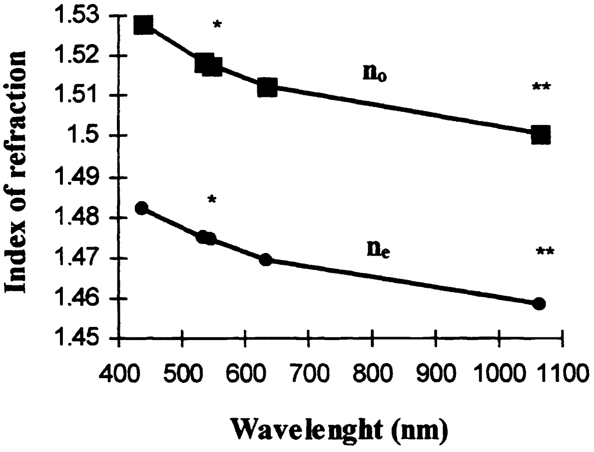


Fig.2 Dispersion of the index of refraction of the crystal $MgSO_3.6H_2O$

First experimenals values for Type I (O_1 O_{1}- E_2) and Type II (O_1 E_{1}- E_2) were obtained from direct measurements of SHG in unprocessed $MgSO_3.6H_2O$ bulk monocrystals in order to avoid possible mistakes from unaccurate crystal elements cutting. Type I phase matching is registered when the crystals are rotated around the Y axis (the laser beam is propagating in the (ZX) plane, with polarization along Y axis. For Type II phase matching the crystal orientation is the same, but the fundamental laser beam polarization is 45° with respect to the Y axis. Thus obtained phase matching angles are:

$$\Theta_{pm}\ (\text{I Type}) = 39°40' \pm 10',$$

$$\Theta_{pm}\ (\text{II Type}) = 61°10' \pm 10'$$

Intensities of second harmonic frequency, generated in $MgSO_3.6H_2O$ elements, cut at angles for Type I and Type II interactions were compared with the intensities of SHG in elements cut from DKDP and CDA monocrystals (Type I interaction). Typical phase matched curves obtained with $MgSO_3.6H_2O$ crystals are shown on Fig. 3 (a - for Type I interaction and b - for Type I I interaction).

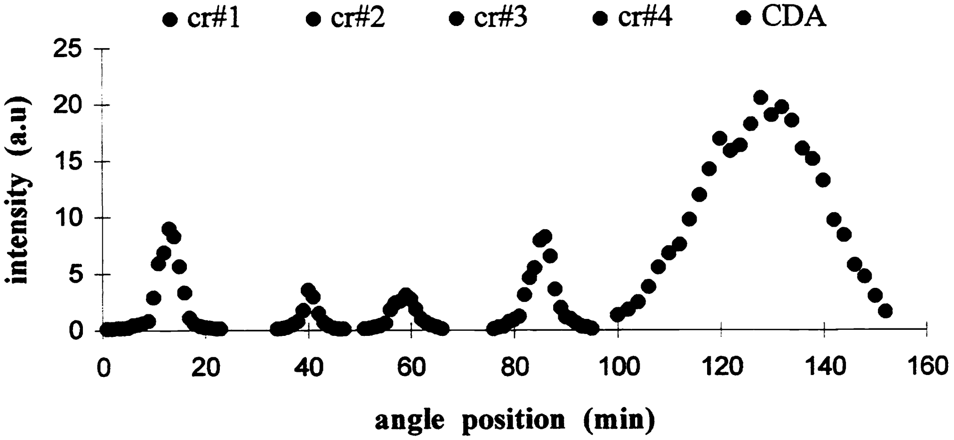
At low pump intensities and relatively short elements (as in our case) the SHG intensity is proportional to square of the effective nonlinearity and the square of the element length:

$$P_{SH} = \text{const.}\left(d_{\text{eff}}^{(2)}L\right)^2$$

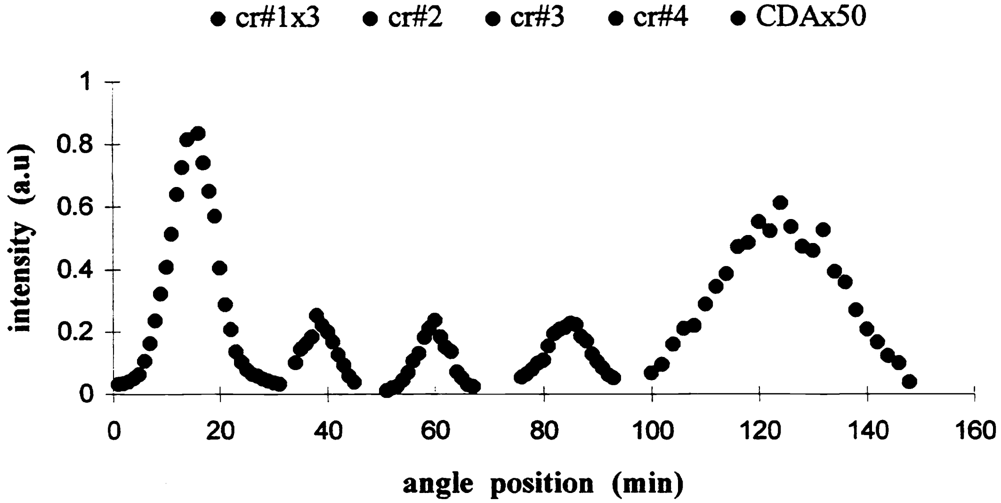
The expressions for $d_{\text{eff}}^{(2)}$ of $MgSO_3.6H_2O$, KDP and CDA are as follows:

$$d_{\text{eff}}^{(2)}(\text{Type I, } MgSO_3.6H_2O) = d_{15}\sin\theta + \cos\theta\left(d_{11}\cos 3\varphi - d_{22}\sin 3\varphi\right)$$

$$d_{\text{eff}}^{(2)}(\text{Type II, } MgSO_3.6H_2O) = \cos^2\theta\left(d_{11}\sin 3\varphi + d_{22}\cos 3\varphi\right)$$



(a)



(b)

Fig. 3 Phase matched curves for Type I (a) and Type II (b) interactions

$$d_{eff}^{(2)}(\text{Type I, DKDP and CDA}) = -d_{14}\sin\theta\sin 2\varphi$$

The first experimental comparison of SHG intensities in those four cases allows to estimate the values of the three components of second order susceptibility tensor of $MgSO_3.6H_2O$:

$\mathbf{d}_{11}^{(2)}$ ($MgSO_3.6H_2O$) = 1.1 $\mathbf{d}_{14}^{(2)}$ (KDP)

$\mathbf{d}_{22}^{(2)}$ ($MgSO_3.6H_2O$) = 0.5 $\mathbf{d}_{11}^{(2)}$ ($MgSO_3.6H_2O$)

$\mathbf{d}_{15}^{(2)}$ ($MgSO_3.6H_2O$) = 0.018 $\mathbf{d}_{11}^{(2)}$ ($MgSO_3.6H_2O$).

The estimated experimental accuracy is about 20 %.

4.DISCUSSION AND CONCLUSIONS

As seen from our first preliminary results $d_{eff}^{(2)}$ and respectively the second harmonic generation process efficiency in $MgSO_3.6H_2O$ are comparable to those of the crystals from the KDP group. That means that it is hard to expect $MgSO_3.6H_2O$ to be used as a very high efficiency frequency convertor. But it can be expected that this new nonlinear media may be quite useful in such applications as second order correlators for pico- and femto-second pulse duration measurements.

5.ACKNOWLEDGEMENTS

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