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SECOND HARMONIC GENERATION IN KDP CRYSTALS

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The possibility of doubling the frequency of light was pointed out theoretically as early as 1959 by TH. NEUGEBAUER [1]. The process consists of the annihilation of two photons with the simultaneous creation of a new photon with double frequency. Being a two photon process, its experimental observation requires a light intensity that classical light sources are not able to provide and it was only the advent of the high intensity laser that made it realisable [2]. The intensity and polarization of the light of double frequency (second harmonic) depend on the intensity and polarization of the fundamental beam and on the orientation of the applied crystal with respect to the incoming beam. Beside these dependences which are similar for either ideal or imperfect crystals, the outgoing intensity and thus the conversion efficiency i.e. the ratio of the outgoing and incoming intensities vary characteristically, depending on the quality of the crystal. The aim of the present paper is to develop a method based on the above mentioned relations for testing optical crystals grown at the Department of Experimental Physics, Technical University, Budapest.

Applied theoretical formulae

In the laser beam there is an extremely high number of photons present owing to which, instead of using quantum mechanics, the classical treatment of the electromagnetic field can be applied. When solving Maxwell's equations, if the nonlinear polarisation induced by intensive e.m. field is taken into consideration, for the intensity of the second harmonic light generated in the crystal the following formula holds [3]:

$$E_i^2(2\omega) = -rac{\mu}{arepsilon_i} \, \omega^2 (d_{ijk} E_j(\omega) E_k(\omega))^2 \cdot L^2 \; rac{\sin^2 rac{arDelta k \cdot L}{2}}{\left(rac{arDelta k \cdot L}{2}
ight)^2} \; .$$

Here E stands for the electric field strength, the lower indices refer to the usual rectangular coordinates and ω denotes angular frequency. μ is the magnetic permeability, which is taken for that of the vacuum as $\mu_r = 1$ for KDP [4]. L is the length of the crystal sample. ε_i is the dielectric constant and d_{ijk} is the nonlinear optical constant tensor of the crystal that has only 3 nonzero components for KDP, namely d_{14}, d_{25}, d_{36} with the restriction that $d_{14} = d_{25}$ [5]. The last factor in Eq. (1) describes the orientation dependence of the second harmonic intensity, as $\Delta k = 2k(\omega) - K(2\omega)$, where k and K are the wave vectors of the incoming and outcoming beam, respectively. The maximum in the second harmonic intensity (and in the conversion efficiency) is attained when $\Delta k = 0$ (phase-matching condition), which is fulfilled only when

$$\boldsymbol{n}_{\rm ord}(\omega) = \boldsymbol{n}_{\rm ext}(2\omega)\,,\tag{2}$$

i.e. the sample is oriented so that the ordinary and extraordinary refractive indices are equal [6]. The angle between the incoming laser beam at this orientation of the sample and the crystallographical axis z is denoted by Θ_M (phase-matching angle).

Experimental results and discussion

In our experiment the phenomenon of second harmonic generation has been observed in KDP crystal with a laser beam of a wavelength of 1060 nm. The scheme of the experimental set-up is shown in Fig. 1. The measurements have been carried out by means of a Nd-glass laser working in multimode regime, which has been Q-switched by a rotating prism to emit 15 nsec long pulses of a divergence of 10^{-2} rad.

The observed value of Θ_M , in good agreement with its theoretical value [7], is $42.1^{\circ} \pm 0.5^{\circ}$. The measured output intensity at 530 nm wavelength as a function of the angular deviation from the phasematching angle is plotted in Fig. 2. The theoretical curve [8] is also shown for comparison.



Fig. 1. Experimental set-up. KDP – crystal sample. F – filter, S – semi-transparent mirror, D – detector, P – plotter, M – monochromator, O – oscilloscope

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The dependence of the output intensity on the pumping intensity at Θ_M (i.e. at maximal output) has also been measured. The logarithms of the results so obtained are shown in Fig. 3. The slope of the straight line in Fig. 3. in accordance with Eq. (1) is 2. The highest efficiency amounted to 13.5% when the fundamental light intensity reached 22 MW \cdot cm⁻².

Finally, the polarization relations have been examined. The maximal intensity in the double frequency beam was observed when the polarization of the input light corresponded to that of the ordinary ray. When the arrangement was such, the second harmonic beam left the crystal as a linearly polarized extraordinary light ray. Whether it is true and if so to what extent, has been



Fig. 2. Second harmonic intensity as a function of the angular deviation from the phase-matching angle Θ_M



Fig. 3. Second harmonic intensity versus fundamental intensity in KDP



Fig. 4. Polarization of the second harmonic beam

tested too, with the aid of a Glan-Thompson prism as analyzer. The second harmonic beam intensity is plotted against orientation of the analyzer in Fig. 4. from which the degree of polarization amounts to 98.6%.

Our experimental results agree with the theoretical predictions about the intensity and polarization relations for second harmonic generation of light. Regarding the testing of the crystals it is deemed significant that in some cases a maximal efficiency of 13.5% has been achieved which means a load of 22 MWcm^{-2} without damage to the crystal. The results of the measurements regarding the angular dependence differ from the theoretical ones. This difference is probably due to the inhomogeneities grown into the crystal sample which make the equation $\Delta k = 0$ valid on the average only. The efficiency obtained for samples grown and oriented in the same way varies significantly (2-13%) with the result that the straight line in Fig. 3 shifts in parallel to itself.

On the basis of the experience obtained it is planned to establish a fast and reliable testing of the electrooptical crystals grown at the Department of Experimental Physics of the Technical University of Budapest.

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