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Phase-Matched Second Harmonic Generation in Urea

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From powder measurements the relatively high nonlinear optical susceptibility of urea is well known /1/. We are now able to show that efficient phase-matched frequency doubling is possible over a wide spectral region. The measurements were carried out using urea single crystals with a typical size of 5x10x10 mm^3. Urea crystallizes in the tetragonal class D_{2d} ($\bar{4}2m$) /2/ and is optically positive uniaxial. From symmetry considerations it follows that only the three components $\chi^1_{23} = \chi^2_{31} = \chi^3_{12}$ of the nonlinear optical susceptibility tensor are non-zero /3/; phase-matched (PM) frequency doubling of type II should be possible for the two equivalent components χ^1_{23} and χ^2_{31}.

The samples, cut at different angles versus the optical axis, were mounted on a goniometric mount where they could be rotated around two axis. The fundamental wave was obtained from a flash-lamp excited dye laser (Chromatix CMX 4) with a peak power of $\approx$ 5 kW, a repetition rate of 10 Hz, and a pulse length of 1 μs. The incident polarization could be rotated continuously by using a half-wave retardation plate. By rotating crystal and polarization all necessary configurations of input polarization to the crystal axis could be arranged. The generated second harmonic light was detected by a solar blind photomultiplier tube with a CsTe photocathode after removing the fundamental beam by means of optical filters.

The phase-match angles (inside the crystal with respect to the c-axis) measured in this arrangement are plotted in Fig. 1 for different wavelengths of the incident beam. Extrapolation yields a short wavelength limit of 476 nm, and no long wavelength limit up to the IR absorption bands of the crystal which were found to start at about 1.43 μm. Best efficiency (as phase matching is of type II) for frequency doubling is achieved for a PM angle of about 45^o, i.e. at a wavelength near 600 nm.

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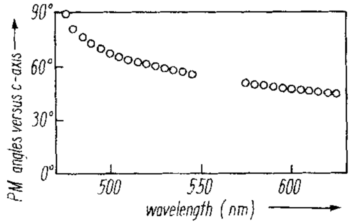

Fig. 1

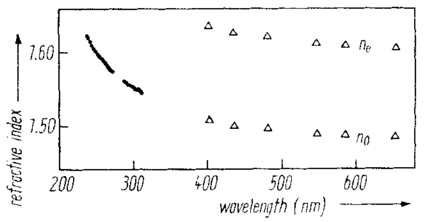

Fig. 2

Fig. 1. Phase-match angles for SHG in urea measured inside the crystal versus c-axis

Fig. 2. Refractive index of urea; Δ measured, ● calculated from the phase-match angles in Fig. 1

The refractive indices of urea in the region from 400 to 650 nm were measured by a conventional prism method (see triangles in Fig. 2). From these results and from the PM angles the index of the ordinary beam in the near ultraviolet region can be derived (see dots in Fig. 2). A sharp increase to short wavelengths is found which in spite of the large birefringence leads to the relatively near lying short wavelength limit.

The wavelength dependence of the ordinary beam can be approximated by a Sellmeier equation of the form

$$n^2 = A + B(\lambda^2 - C)^{-1}$$

with the coefficients A = 2.17, B = 0.014, and C = 0.028 (λ in μm).

To get an idea of the doubling efficiency, measurements of the SHG intensity were carried out at 600 nm. We found that, reduced to equal crystal sizes, the efficiency in urea is about 5 times greater than the efficiency in KDP, which means, that χ^1_{23} (urea) $\approx 3\ \chi^3_{12}$(KDP). This result deviates from earlier estimations for urea /1/ and similar materials /4/, where factors of about 25 and 100, respectively, were reported. This discrepancy may be in part due to the fact that these authors estimated the efficiency for a fundamental wavelength of 1064 nm.

In conclusion we can say that urea can be used as a nonlinear optical material in a wide wavelength region with rather good doubling efficiency. The nonlinear susceptibility exceeds that of KDP by about a factor of 3.

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