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OPTICAL MIXING IN TYPE I AND TYPE II KDP WITH PICOSECOND LASER PULSES*

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The frequencies produced in a KDP crystal by intense broadband 1.06 μm picosecond pulses in Type-I and Type-II processes are investigated as a function of crystal orientation about the degenerate phase-matching angle. The dispersive difference between the Type-I and Type-II process results in a tremendous difference in emission frequencies at a given crystal orientation angle from the degenerate phase matching position.

Non-linear optical effects induced by high power laser pulses can be used to generate optical waves in different frequency regions [1]. The birefringence of an anisotropic crystal is used to significantly convert particular frequencies by the technique of phase matching. This technique was first established by Giordmaine [2] with the efficient generation of second harmonic light in potassium dihydrogen phosphate (KDP). The spectral and temporal properties of the degenerate second harmonic phase matching peak has been investigated using picosecond laser pulses [3–5]. The purpose of this paper is to study the frequencies produced in KDP by intense broadband 1.06 μm picosecond pulses in Type-I and Type-II processes. The dispersive difference between the Type-I and Type-II processes results in a tremendous difference in the emission frequencies at a given crystal orientation angle from the degenerate phase matching angle.

In KDP and its isomorphs there are two types of phase matching commonly used for a three wave interaction – called type-I and type-II [6,7]. In type-I the pumping beams travel as ordinary waves. In type-II one pump beam propagates as an ordinary (O) wave and the other pump beam propagates as an extraordinary (E) beam. In both processes, the parametrically generated beam propagate as an E-wave. Recently, type-II phase-matching has become increasingly more popular than type-I phase-matching for second harmonic generation (SHG) because of its increased conversion efficiency and larger phase-matching acceptance angle.

In the experiment, a full train or an amplified single selected pulse from the emission of Nd: mode locked glass laser was used. The train consisted of 100 pulses peaked at ~1.06 μm width a maximum pulsewidth per pulse of 15 picoseconds and a spectral width, as measured on Kodak Z plates, of ~20 cm^{-1} about the central peak and a weaker background of ~250 cm^{-1} (approximately 2 1/2 times broader on the Stokes side). The characteristics of the selected single pulse is approximately half the above values. The energy of the train and amplified single pulse are 70 mJ and 30 mJ, respectively. The beam divergence is ~7 mrad. The train and single pulse pass through 18 inches of glass. The laser beam is passed through either a type-I or type-II KDP crystal of length 2.5 cm. The type-II crystal is placed in a cell filled with index matching fluid with AR coated windows. The crystals

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are rotated through an angular spread of ± 4 degrees about the second harmonic phase matching angle for the 1.06 beam. The emission from the crystals is displayed as a function of angle and wavelength by placing a 15.2 cm lens at the focal distance from a 2 cm high slit of a 1/2-m Jarrel–Ash spectrograph and 10.5 cm away from the KDP crystal. At a selected number of angles the spectral emission is measured. A 1-75 Corning filter plus various neutral density filters are placed at the slit. The spectra are displayed on Polaroid 57-type film.

Typical spectra obtained from KDP type-I and type-II at different crystal orientations displayed in fig. 1. The y direction corresponds to the angular emission, α, at a particular crystal orientation, θ, and the x direction corresponds to the emission wavelength. This results from placing the spectrograph at the focal plane of the lens. The phase matched wavelength is 0.5318 μm. The crystal is oriented at approximately $\Delta\theta = 0.46°$ off phase-matching for type I (fig. 1a) and $\Delta\theta = 4°$ for type II (fig. 1b). The wavelength emission along the x axis from KDP is plotted in fig. 2 as a function of crystal orientation relative to the c axis. The "new" spectral lines in both cases shift to the Stokes side of 0.53 μm and decreases rapidly with intensity with increasing angle. The intensity of the phase matched 0.53 μm line is $\sim 10^5$ times larger than the line at 0.55 μm and $\sim 10^7$ times larger than the line at 0.57 μm. The polarization of the "new" lines are the same as the 0.53 μm line. These tunable lines are ~60 Å wide. The "new" lines still appear if the intensity of the 1.06 μm beam is reduced a factor of 30 and does not appear if one only passes a 0.53 μm beam in this crystal – the latter observations rule out stimulated Raman scattering as the cause of the "new" spectral lines. In addition, when a narrow band filter of 100 Å wide at 1.06 μm was placed in front of the KDP the "new" spectral lines were not observed at any angle. Care was taken to make sure the intensity of the 1.06 μm beam did not damage the filter, and, the intensity of the SHG (0.53 μm line) was adjusted to a comparable intensity to the 0.53 μm line when the "new" spectral lines were present at any angle. The latter was checked by the use of a photomultiplier and film. In addition, in the type II crystal, a well defined absorption line centered at 5592 ± 16 Å appeared in the spectral emission as the orientation of the crystal was varied. This is illustrated in fig. 1c where the spectra was photographed at three different crystal orientations. In addition, in each of the three different exposures

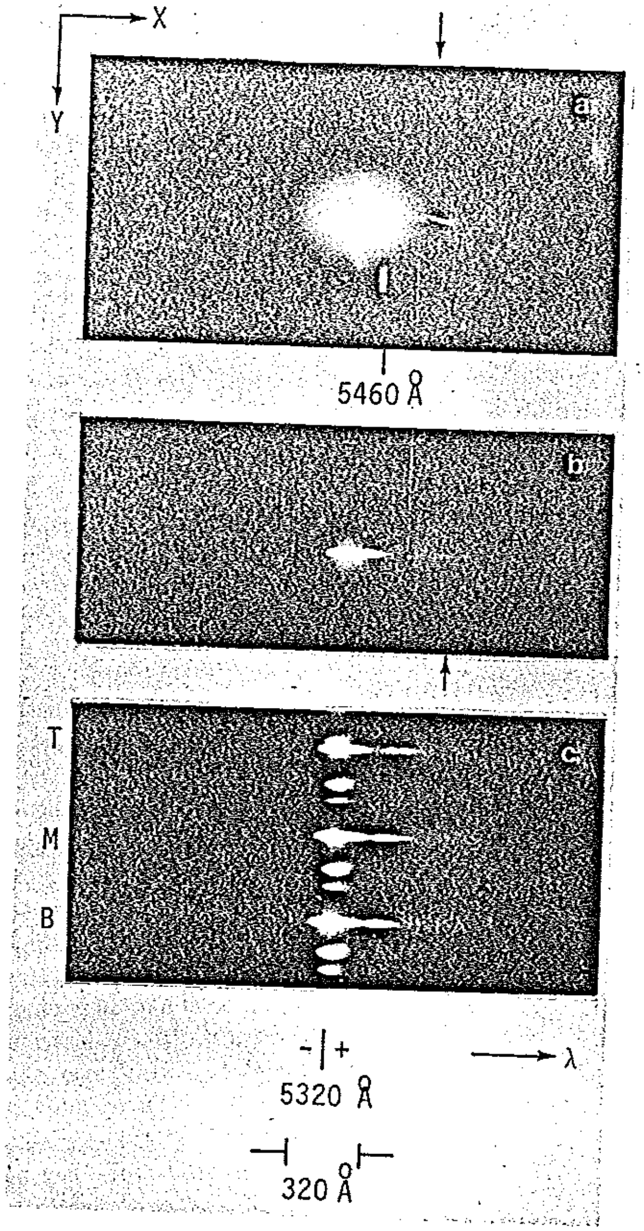

Fig. 1. Spectral emission from type I and II at different orientation of the crystal about the phase matching angle. (a) Type I, $\Delta\theta = 0.46°$ (notice the extra emission on the Stokes side of 5320 A; arrow pointing to its λ location). (b) Type II (notice the extra emission on the Stokes side of 5320 A; arrow pointing to its λ location). (c) Type II, top (T) $\Delta\theta = 1.8°$; middle (M) $\Delta\theta = 1.5°$; and bottom (B) $\Delta\theta = 1.1°$.

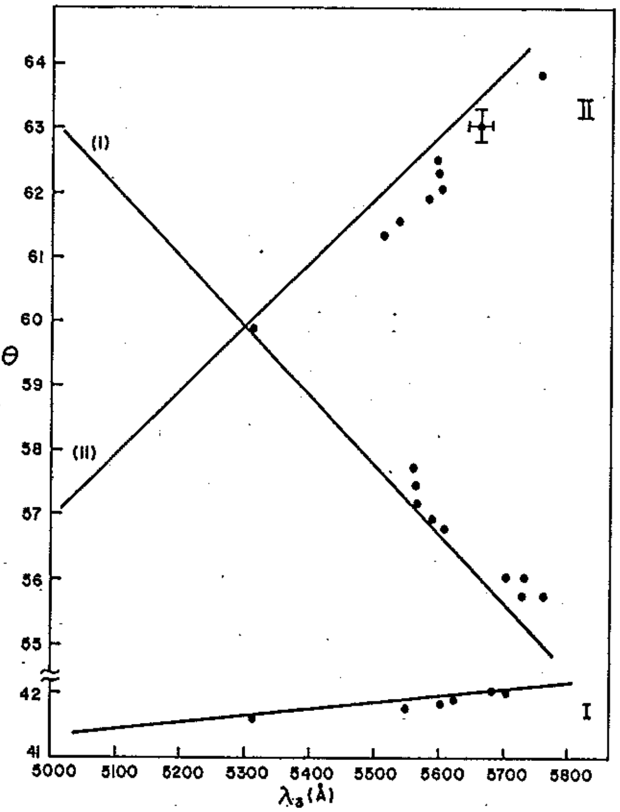

Fig. 2. A plot of the parametrically generated wavelengths versus phase matching angle in KDP type I and II processes. Dots are the experimental points. The indices of refraction were obtained from ref. [9]. The curves labeled by (i) and (ii) correspond to the solution of eqs. (3) and (4), respectively.

in fig. 1c there are two additional bands at approximately 0.532 μm which arise from the two non-collinear phase-matched cones.

To explain our results qualitatively we propose, for simplicity, a collinear three wave parametric interaction where the first pump wave is fixed at a peak wavelength, λ_1 of the laser emission, the second pump wave, λ_2, spans the spectral width of the spectral broaden laser pulse and the third parametrically generated wavelength λ_3, is determined by conservation of energy. This model has been previously proposed [8]. From Maxwell's equations it can be shown that this three wave interaction will produce significant radiation at λ_3, provided there is phase matching among the propagation vectors of the waves:

$$\Delta \bar{K} = \bar{K}_1 + \bar{K}_2 - \bar{K}_3 = 0. \qquad (1)$$

For a more quantitative theoretical treatment of this problem, an analysis similar to Glenn [5] should be used to describe the mixing of waves of different frequencies in a broad based continuum.

In the type I process there is one major phase-matched three wave process:

$$\Delta K_{\mathrm{I}} \doteq K^{\mathrm{O}}_{1.06} + K^{\mathrm{O}}_{\lambda_2} - K^{\mathrm{E}}_{\lambda_3}, \qquad (2)$$

where, as before, O, E stand for ordinary and extraordinary, respectively. In the type II process there are two major phase-matched three wave processes:

$$\Delta K_{\mathrm{II(i)}} = K^{\mathrm{O}}_{1.06} + K^{\mathrm{E}}_{\lambda_2} - K^{\mathrm{E}}_{\lambda_3}, \qquad (3)$$

and

$$\Delta K_{\mathrm{II(ii)}} \doteq K^{\mathrm{E}}_{1.06} + K^{\mathrm{O}}_{\lambda_2} - K^{\mathrm{E}}_{\lambda_3}. \qquad (4)$$

Theoretical plots of λ_3 versus the phase-matching angle θ, determined by setting $\Delta K = 0$ is shown in fig. 2 for Type I and Type II processes. The slope $|\Delta\theta/\Delta\lambda|$ for type II is ~0.01°/Å and for type I is ~0.006°/Å. Our experimental data plotted in fig. 2 is in reasonable agreement with the theoretical curves. The lack of data points on the antistokes side reflects the fact that the incoming beam is primarily broadened on the stokes side. The existence of the weak lines at ~ 5700 Å indicates a very weak spectral broaden background extending to ~ 2000 cm^{-1} on Stokes side of 1.06 μm that did not appear at Kodak Z plates. This background most likely arises from the passage of the 1.06 μm pulse through 18 inches of glass [10]. The smaller slope of the type I curve implies that the observation of the parametrically generated wave λ_3 will be more difficult to observe because it is always partially masked by the phasematched SHG due to the beam divergence. The observation of absorption lines in material using a continuum is not unprecedented [10] and is explained in terms of the inverse Raman effect. A strong Raman active [11] line at 914 cm^{-1} does lie near the observed frequency shift of ~940 cm^{-1}. This explanation is tentative since a direct light scattering should be performed to see if this is truly vibrational related.

In conclusion, we have observed light of different frequencies emitted upon the passage of broadband picosecond laser pulses through a KDP type-I and type-II crystal. The emission frequencies depend on crystal orientation with respect to the incoming beam. We have described this phenomena qualitatively as arising from three wave parametric generation. Our results clearly indicate that care must be taken in the utilization of Type-I and II KDP crystals as frequency doublers. If a broad band and divergent laser source is used for SHG such as the emission from a multistage picosecond laser amplifier system, parametric generated light will also exist in addition to the second harmonic component. This may have important consequences in the use of SHG pulses for laser induced fusion.

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