EFFECTS OF DISPERSION AND FOCUSING ON THE PRODUCTION OF OPTICAL HARMONICS

P. D. Maker, R. W. Terhune, M. Nisenoff, and C. M. Savage
Scientific Laboratory, Ford Motor Company, Dearborn, Michigan
(Received December 8, 1961)

Recently Franken et al.[1] demonstrated the generation of optical harmonics utilizing the nonlinearity in the electric susceptibility of piezoelectric crystals. Using a classical approach one can think of the laser red light generating a spatial arrangement of dipoles which in turn radiate blue light. For the case of a plane wave incident on a crystal of thickness x, the intensity of the blue light radiated in the forward direction is

$$S = 2\pi c P^2 (k_b/\Delta k)^2 \sin^2 \Delta k x, \quad \Delta k = |\vec{k}_b - 2\vec{k}_r|,$$

where $\vec{k}_r$ and $\vec{k}_b$ are the wave vectors for the red and blue lights, respectively, and P is the magnitude of the induced blue polarization. The $\sin^2\Delta kx$ term arises because of dephasing between red radiation (blue polarization) and blue radiation, due to dispersion of the crystal. This dephasing limits the effective crystal thickness. These dispersive effects have been demonstrated in quartz. A ruby laser beam was passed through an optically plane-parallel quartz platelet, the red light filtered out using a $CuSO_4$ solution filter and a grating monochrometer, and the blue light intensity measured photoelectrically. The sample was inclined to the beam by rotation about its z axis, thus increasing the optical thickness and generating the curve shown in Fig. 1. The spacing between successive maxima when reduced to thickness changes agreed well with the calculated value, 13.9 microns.

We were able to balance out the effect of dispersion in potassium dihydrogen phosphate (KDP). The matrix elements relating harmonic polarization to applied electric field, as limited by symmetry and conservation of electromagnetic energy, are for KDP:

$$P_x = aE_yE_z, \quad P_y = aE_zE_x, \quad P_z = aE_xE_y.$$

Thus ordinary exciting rays generate extraordinary harmonic rays. As the birefringence for KDP is greater than its dispersion, in certain orientations $\vec{k}_{be}$ exactly matches $2\vec{k}_{ro}$. This matching of wave vectors results in a 300-fold increase of blue light intensity. Figure 2 shows the θ and ϕ angular dependencies of this enhanced signal, together with a diagram indicating the locus of directions for which red and blue indices of refraction can be matched.

The above data on KDP were taken with the unfocused laser beam. Another large increase in blue light intensity was obtained by focussing. As

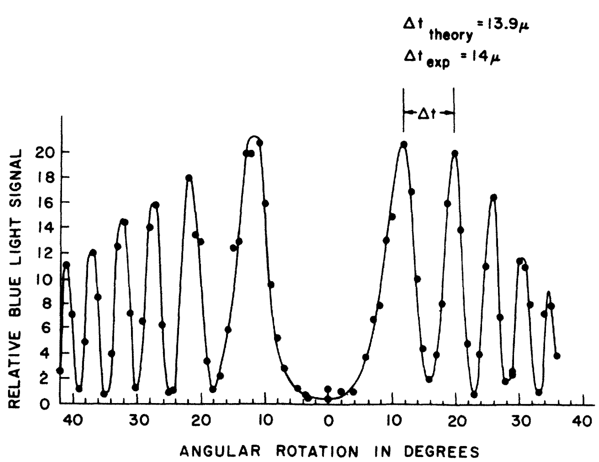

FIG. 1. Blue light generation vs inclination of 0.0308-in. thick quartz platelet to laser beam. Rotation axis normal to beam, parallel to crystal z axis. Red beam unfocused and polarized parallel to the z axis.

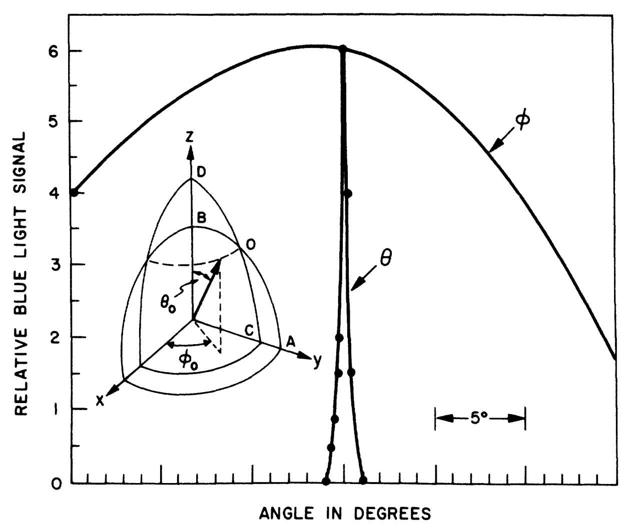

FIG. 2. Blue light intensity as a function of crystal orientation for KDP. Maximum output occurs at $\theta_0 = 52° \pm 2°$, $\phi_0 = 45°$. Laser beam collimated to within $\pm\frac{1}{4}°$. AOB is an arc on the index of refraction surface for red ordinary rays, COD for blue extraordinary rays.

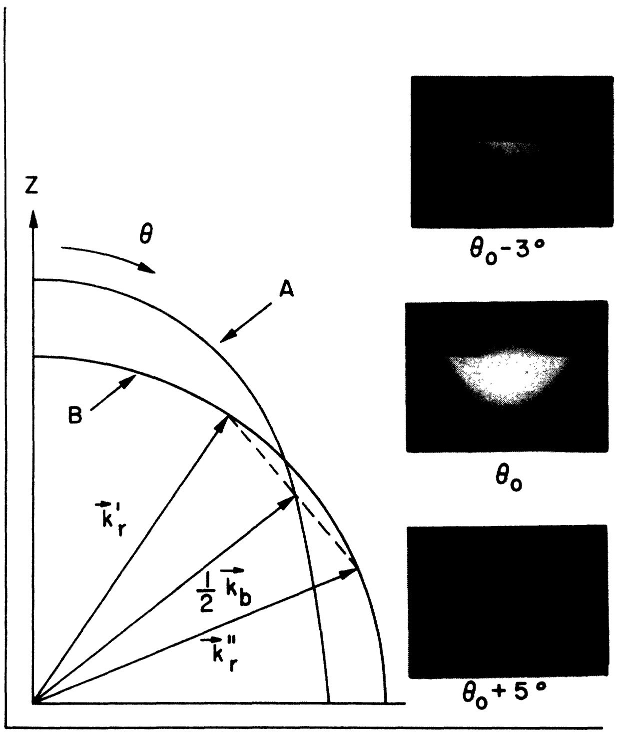

FIG. 3. Harmonic generation through mixing of divergent red rays in KDP near θ_0. The striking feature is that blue light can be radiated only into that fraction of the beam cone for which $\theta \geq \theta_0$. Diagram illustrates addition of two red wave vectors to produce a momentum-matched blue wave vector. A: blue extraordinary wave surface, half scale. B: red ordinary (spherical) wave surface. Photos show radiated blue light intensity patterns for different angles of incidence.

P is proportional to E_r^2, the total blue radiation varies inversely as the cross-sectional area of the laser beam. Also, the resultant weighting of the very small volume near the focus causes many of the thickness dependencies to vanish. Additionally, spatially separate red rays are brought together at the focus where they may beat to produce blue radiation. The polarization which results from the mixing of two divergent ordinary red rays with wave vectors $\vec{k}_{ro}'$ and $\vec{k}_{ro}''$ will propagate with wave vector $\vec{k}_{ro}'+\vec{k}_{ro}''$. Intense blue light will be generated in those directions (θ, ϕ) for which $\vec{k}_{be}(\theta, \phi) = \vec{k}_{ro}'(\theta+\Delta\theta, \phi+\Delta\phi) + \vec{k}_{ro}''(\theta-\Delta\theta, \phi-\Delta\phi)$, provided $\vec{k}_{ro}'(\theta+\Delta\theta, \phi+\Delta\phi)$ and $\vec{k}_{ro}''(\theta-\Delta\theta, \phi-\Delta\phi)$ are contained in the incident focused beam. An implied condition is that $|\vec{k}_{be}(\theta, \phi)| \leq |\vec{k}_{be}(\theta_0, \phi_0)| = 2|\vec{k}_{ro}|$ or that $\theta \geq \theta_0$. Shown in Fig. 3 are photographs of cross sections of the blue light beam. The sharp upper bounding line reflects the latter fact.

Observed blue light intensities, expressed as the number of red photons required to produce one blue photon, are for quartz: collimated beam, 5×10^{12}; focused beam using a one-inch focal length lens, 6×10^{10}; for KDP: collimated beam incident at (θ_0, ϕ_0) on a sample 1.5 mm thick, 3×10^9; focused beam using a one-inch focal length lens and incident at (θ_0, ϕ_0), 10^6. Rough calculations indicate the gain of 3000 upon focusing in KDP to be composed of a factor of 100 from beam diameter reduction and a factor of 30 due to mixing of diverging rays. These conversion ratios depend strongly upon the coherence nature of the particular laser employed and cannot be used to estimate P/E_r^2. It may be pointed out that since the laser beam is essentially unattenuated on a single passage through the KDP, an increase in blue light intensity of the order of 10^4 could be realized by placing the sample at the focus of an appropriate optical cavity.

We are indebted to Professor G. Weinreich for several stimulating discussions.

---

[1]P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, Phys. Rev. Letters 7, 118 (1961).

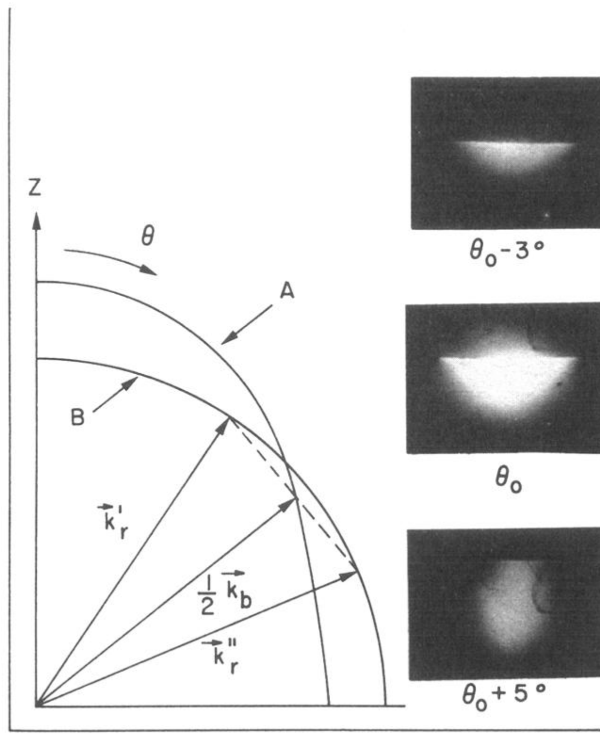

FIG. 3. Harmonic generation through mixing of divergent red rays in KDP near θ_0. The striking feature is that blue light can be radiated only into that fraction of the beam cone for which $\theta \geq \theta_0$. Diagram illustrates addition of two red wave vectors to produce a momentum-matched blue wave vector. A: blue extraordinary wave surface, half scale. B: red ordinary (spherical) wave surface. Photos show radiated blue light intensity patterns for different angles of incidence.