MIXING OF LIGHT BEAMS IN CRYSTALS

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This Letter reports the mixing of plane light waves having different directions of propagation and the attainment of coherence volumes of about 0.2 cm^3 in the production of second harmonic radiation. In these experiments a sizable intensity of second harmonic radiation is observed in potassium dihydrogen phosphate (KDP) without the use of focussed beams. This is one of a new class of optical experiments and techniques suggested by the work of Franken et al.[1] and made possible by maser sources of highly intense monochromatic light.

A plane light wave described by the form $\exp[i(\omega_1 t - \vec{k}_1 \cdot \vec{r})]$ launches in a suitable[1] nonlinear medium a travelling wave of polarization of the form $\exp[i(2\omega_1 t - 2\vec{k}_1 \cdot \vec{r})]$. This polarization can radiate a wave $\vec{k}_2$ at frequency $\omega_2 = 2\omega_1$ with maximum efficiency if the two remain in phase, that is, if $\vec{k}_2 = 2\vec{k}_1$. This condition implies that in the direction of $\vec{k}_1$ the phase velocity $v_1 = v_2$ or $n_1 = n_2$. Since almost all materials have normal dispersion in the optical region, the radiation will generally lag behind the polarization wave.

The phase velocities v_1 and v_2 may, however, be equal for certain directions in an anisotropic crystal. Consider a negative uniaxial crystal such as KDP having ordinary and extraordinary refractive indices $n^o = c/v^o$, $n^e = c/v^e$, respectively. If, as in KDP, $v_1{}^o < v_2{}^e$ the $O(\nu_1)$ and $E(\nu_2)$ wave normal surfaces intersect in a cone of half-angle $\psi_0 = \arcsin[(v_1{}^o)^2 - (v_2{}^o)^2]^{1/2}[(v_2{}^e)^2 - (v_2{}^o)^2]^{-1/2}$ centered on the optic axis. Similarly in a positive uniaxial crystal the $E(\nu_1)$ and $O(\nu_2)$ surfaces intersect if $v_1{}^e < v_2{}^o$.

From a single plane wave $\vec{k}_1$ completely in-phase harmonic radiation is produced in a uniaxial crystal only if $\vec{k}_1$ is in a direction ψ_0. This condition is the same as that for conservation of electromagnetic momentum in the harmonic generation process. The second harmonic power radiated per unit area from a column of length l under this condition is $S = 2\pi^3 l^2 p^2 \nu_2{}^2 (n_2 c)^{-1}$, where p is the amplitude of the ν_2 component of polarization.

More generally, two plane waves $\vec{k}_1$ and $\vec{k}_1{}'$ at frequency ν_1 can combine to produce a ν_2 wave $\vec{k}_2$ which is completely in phase with the polarization when

$$\vec{k}_1 + \vec{k}_1{}' = \vec{k}_2. \qquad (1)$$

When Eq. (1)[2] is satisfied, the entire irradiated crystal volume can radiate coherently, electromagnetic momentum is conserved, and optimum radiation efficiency is achieved. Consider the particular case in a negative uniaxial crystal of an O wave $\vec{k}_1$ at angle ψ_1 with the optic axis $\hat{z}$ mixing with a second O wave $\vec{k}_1{}'$ to form the harmonic E wave $\vec{k}_2$ emitted at an angle θ to $\vec{k}_1$. Equation (1) is satisfied if $\cos\theta = v_1/v_2(\theta)$. This transcendental equation has particularly simple solutions when $\psi_1 - \psi_0 = \Delta\psi$ is small, i.e., when the direction of $\vec{k}_1$ is close to the direction for which $v_1 = v_2$. Then Eq. (1) can be approximated by $\theta^2 = 2(v_2 - v_1)/v_2 = K(\Delta\psi - \theta\cos\alpha)$, where K denotes the constant $2[(v_2{}^e)^2 - (v_1{}^o)^2]^{1/2}[(v_1{}^o)^2 - (v_2{}^o)^2]^{1/2}(v_1{}^o)^{-2}$ and α is the angle between $\vec{k}_1\vec{k}_2$ and $\hat{z}\vec{k}_1$ planes. The solution of this quadratic equation in θ for a particular $\vec{k}_1$ represents a circular cone of wave vectors $\vec{k}_2$ of half-angle $\frac{1}{2}K(1 + 4\Delta\psi/K)^{1/2}$ centered about the direction in the $\vec{k}_1\hat{z}$ plane making an angle $\psi_1 + \frac{1}{2}K$ with the optic axis. No completely in-phase second harmonic generation is possible for $\psi_1 < \psi_0 - \frac{1}{4}K$. As ψ_1 increases from $\psi_0 - \frac{1}{4}K$, emission can occur on a cone of increasing radius which crosses the direction ψ_1 when $\psi_1 = \psi_0$.

These phenomena have been observed in KDP,[3] with the fundamental plane wave $\vec{k}_1$ provided by the intense collimated (unfocussed) beam of a ruby optical maser ($\lambda \cong 6940$ A). The second wave $\vec{k}_1{}'$ is provided by small-angle scattering from the main beam within and at the surface of the crystal. Symmetry considerations[1] show that in KDP fundamental O radiation gives rise to second harmonic E radiation ($\lambda \cong 3470$ A). The experimental arrangement is shown in Fig. 1. Since a more or less continuous angular distribution of scattered ν_1 light is present, ν_2 light is emitted over the entire cone of allowed directions θ described above.

Figure 2 shows photographs of the angular distribution of ν_2 radiation obtained with approximately normal incidence on a crystal plate of thickness 0.22 cm. The bright circular ring in the upper left photograph is the cone of harmonic emission described above. The bright spot within the ring is emission in the same direction as the maser beam. In contrast to the ring, this forward light is emitted with low efficiency since the condition $\vec{k}_2 = 2\vec{k}_1$ is not satisfied. The upper right photograph shows

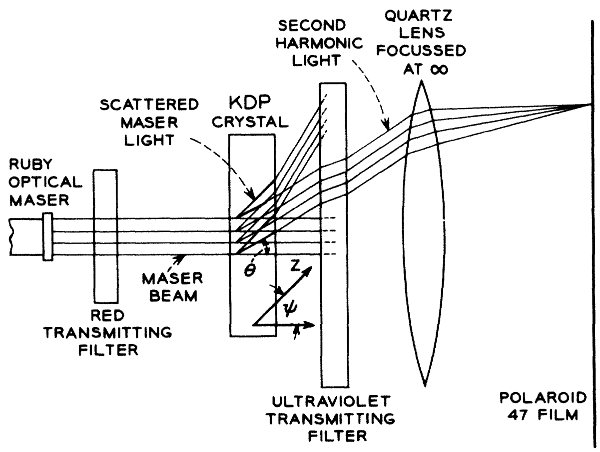

FIG. 1. Experimental arrangement. Focal length of lens 15.0 cm. The angle ψ_1 could be varied by rotating the crystal about an axis perpendicular to the page.

FIG. 2. Photographs of angular distribution of harmonic ν_2 emission. Above, $\psi_1 = 50.9°$; below, $\psi_1 = 49.6° \cong \psi_0$; the angular diameter of the ring in the above photographs is 3.9°. Each photograph was obtained with one flash (~1 joule) of the ruby maser.

the increase in intensity of the cone emission observed where the maser was flashed with a lightly ground transparent screen inserted in front of the KDP crystal to increase the light scattering. The lower photograph illustrates the large increase in intensity of the forward emission at $\psi_1 = \psi_0$. The maximum emitted power was of the order of 1 mw. The ring which is partly obscured in this overexposure passes through the direction of forward emission. The total emitted power dropped to half maximum intensity at a beam angle approximately 15′ from the angle ψ_0 of maximum emission. This angle corresponds to half the maser beam width. The angular aperture for completely in-phase emission was about 16′.

We have estimated the refractive indices by extrapolation from published data in the visible region to be $n_1^e = 1.466$, $n_2^e = 1.487$, $n_1^o = 1.506$, and $n_2^o = 1.534$. The observed values of ψ_0 and K were $49.8 \pm 1.0°$ and 0.061 ± 0.002 in good agreement with the predicted values of 50.4° and 0.060. Subsequent experiments with thicker crystals have demonstrated coherence lengths up to 1.0 cm.

In the two upper photographs arcs of additional rings can be seen with 16′ spacing. This emission is interpreted as radiation at successive angles for which $\vec{k}_1 + \vec{k}_1' - \vec{k}_2 = 3\pi/l, 5\pi/l, \cdots, (2n+1)\pi/l$. For these nonzero values of $\vec{k}_1 + \vec{k}_1' - \vec{k}_2$, the radiation and the polarization wave go out of phase by π in a distance $l/(2n+1)$, with consequent reduction in radiation efficiency.

The above techniques can also be applied to second harmonic generation in positive uniaxial and in biaxial piezoelectric crystals, and to coherent mixing of light of different frequencies.

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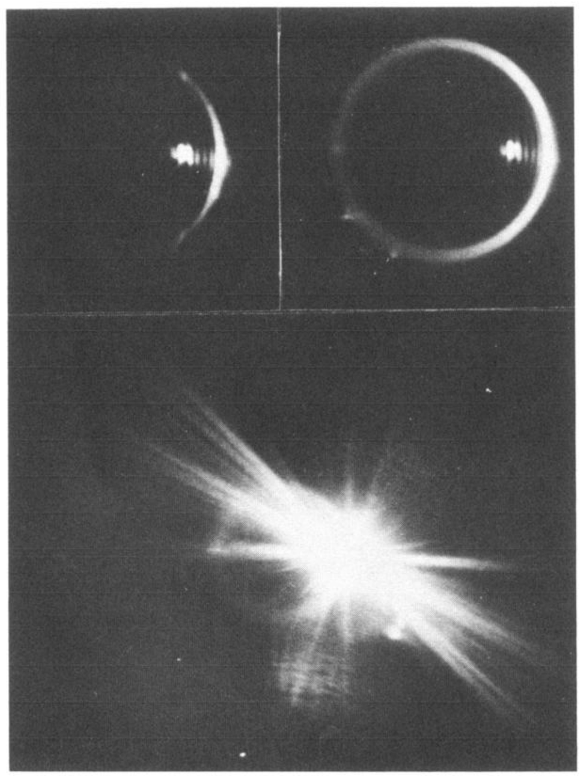
[1]P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, Phys. Rev. Letters 7, 118 (1961).

[2]Equation (1) has also been derived by P. K. Tien (unpublished).

[3]Kindly provided by W. L. Bond and R. P. Riesz.



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