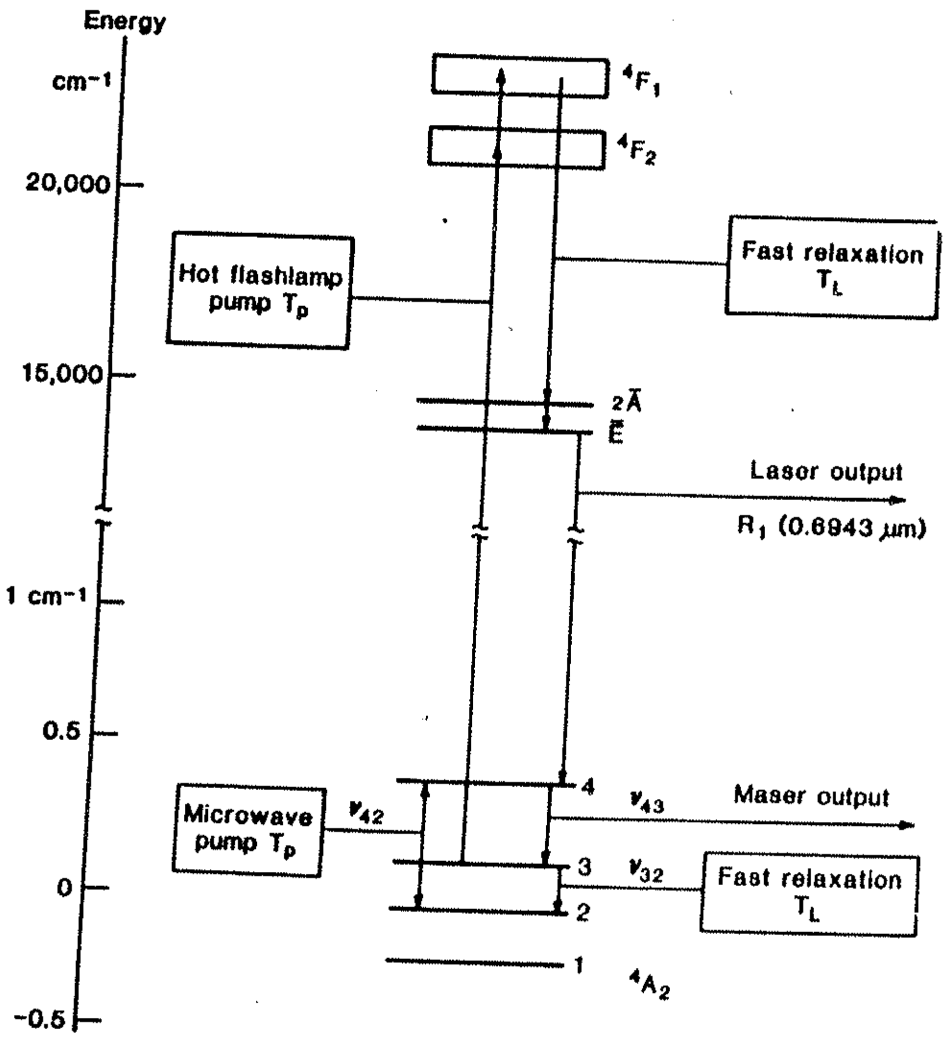
Maiman examines the first ruby laser, built at Hughes Research Laboratories (circa 1960).



Energy
cm⁻¹
20,000
15,000
1 cm⁻¹
0.5
0
-0.5
⁴F₁
⁴F₂
Hot flashlamp pump Tp
Fast relaxation TL
2Ā
Ē
Laser output
R₁ (0.6943 μm)
Microwave pump Tp
ν42
4
ν43
Maser output
3
ν32
Fast relaxation TL
2
1
⁴A₂

Maiman Ruby Laser is Nonlinear Optics

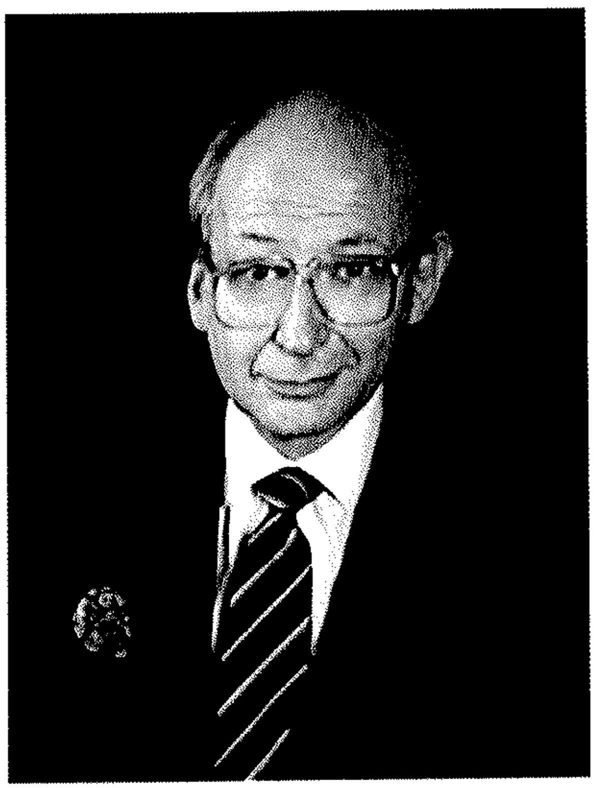
ω_L laser frequency

ω_p pump frequency in blue and green bands

$$P(\omega_L) = i\chi' E_L + i\chi^{(3)''} E(\omega_L) \sum_{\omega_p} |E(\omega_p)|^2$$

linear loss

$\chi^{(3)''} < 0$ for $\omega_p > \omega_L$



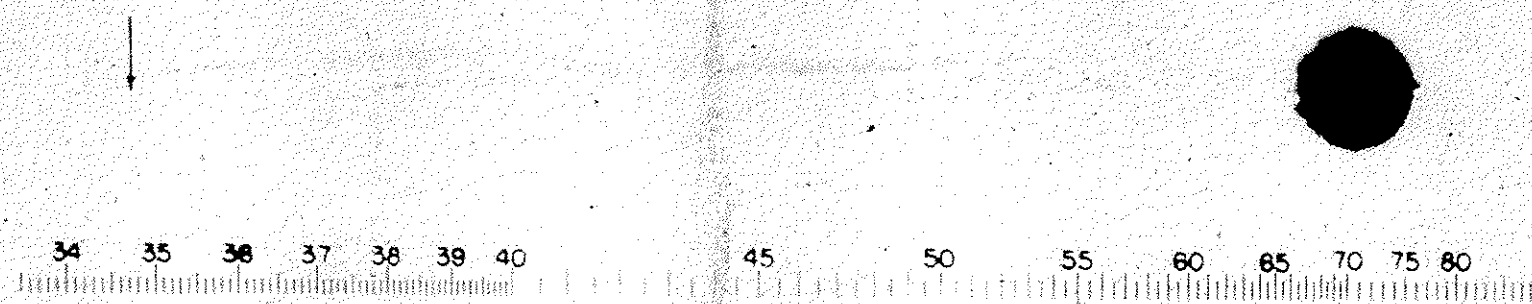


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

Interactions between Light Waves in a Nonlinear Dielectric

J. A. Armstrong, N. Bloembergen, J. Ducuing, P. S. Pershan

Phys. Rev. 127, 1918, 1962.

$$P_i = \chi_{ij} E_j + \chi^{(2)}_{ijk} E_j E_k + \chi^{(3)}_{ijkl} E_j E_k E_l + \cdots$$

Complex Nonlinear susceptibilities

$A_1\exp(i\omega_1 t)$, etc. The permutation symmetry relations make it possible to define a real constant

$$K=\frac{2\pi}{c^2}\hat{a}_3\cdot\chi(\omega_3):\hat{a}_1\hat{a}_2=\frac{2\pi}{c^2}\hat{a}_2\cdot\chi(\omega_2):\hat{a}_3\hat{a}_1$$
$$=\frac{2\pi}{c^2}\hat{a}_1\cdot\chi(\omega_1):\hat{a}_3\hat{a}_2$$

and the coupled amplitude equations become

$$\begin{aligned}dA_1^*/dz&=+i(\omega_1^2K/k_1\cos^2\alpha_1)A_3^*A_2e^{+i(\Delta kz+\Delta\phi)},\\ dA_2^*/dz&=+i(\omega_2^2K/k_2\cos^2\alpha_2)A_3^*A_1e^{+i(\Delta kz+\Delta\phi)},\\ dA_3/dz&=-i(\omega_3^2K/k_3\cos^2\alpha_3)A_1A_2e^{+i(\Delta kz+\Delta\phi)}.\end{aligned}\tag{4.9}$$

In a similar way one obtains the equations which describe the variations in amplitude and phase for a second harmonic interacting with the fundamental,

$$\begin{aligned}dA_1^*/dz&=+i(2\omega^2K/k_1\cos^2\alpha_1)A_2^*A_1e^{+i(2k_1-k_2)z},\\ dA_2/dz&=-i(4\omega^2K/k_2\cos^2\alpha_2)A_1^2e^{+i(2k_1-k_2)z}.\end{aligned}\tag{4.10}$$

These equations can be generalized to higher powers in the field amplitudes. The amplitude equations for the third harmonic are, for example,

$$\begin{aligned}dA_1^*/dz&=i(3\omega^2/k_1\cos^2\alpha_1)[CA_3^*A_1^2e^{i(3k_1-k_3)z}\\&\quad+C'A_1^*A_1A_1^*+C''A_1^*A_3A_3^*],\\ dA_3/dz&=-i(9\omega^2/k_3\cos^2\alpha_3)[CA_1^3e^{+i(3k_1-k_3)z}\\&\quad+3C''A_3A_1A_1^*+C'''A_3A_3A_3^*].\end{aligned}\tag{4.11}$$

The constant C is given by

$$\begin{aligned}C&=(2\pi/c^2)\hat{a}_3\cdot\chi'(\omega_3=\omega_1+\omega_1+\omega_1)\vdots\hat{a}_1\hat{a}_1\hat{a}_1\\&=(2\pi/c^2)\hat{a}_1\cdot\chi''(\omega_1=\omega_3-\omega_1-\omega_1)\vdots\hat{a}_3\hat{a}_1\hat{a}_1,\end{aligned}\tag{4.11a}$$

with the fourth-rank tensor χ' expressible in terms of the higher order nonlinear polarizability γ' given by Eq. (2.22). The quantities C', C'', and C''' can be referred in a similar way to the nonlinear polarizability. They occur in terms that are purely reactive in nature. It is clear from the notation that, e.g., the term $C''A_1^*A_3A_3^*=\partial A_1^*/\partial z$ corresponds to a partial contribution to the coherent scattering by a quantum process, in which photons at ω_1 and ω_3 get scattered simultaneously. There is no change of the power flow involved. In fact, these reactive terms represent a quadratic dc Kerr effect. The propagation constant for the wave at ω_1 changes in a similar way by a term proportional to $C^{dc}E_{dc}^2$ as by $C''A_3A_3^*+C'''A_3A_3^*$. This question will be discussed further in Sec. VII.

The third-harmonic situation is a special case of the interaction between four electromagnetic waves. If, for example, the frequency and momentum relationships

$$\omega_4+\omega_1=\omega_2+\omega_3,\quad \Delta k=+k_2+k_3-k_1-k_4$$

exist between the four waves, the coupled amplitude equations are

$$\begin{aligned}\frac{dA_1}{dz}&=-i\frac{\omega_1^2C}{k_1\cos^2\alpha_1}A_2A_3A_4^*e^{i\Delta kz}\\&\quad-\frac{i\omega_1^2}{k_1\cos^2\alpha_1}A_1\sum_{j=1}^4C_{1j}A_jA_j^*,\\ \frac{dA_2^*}{dz}&=+i\frac{\omega_2^2C}{k_2\cos^2\alpha_2}A_1^*A_3^*A_4^*e^{i\Delta kz}\\&\quad+\frac{i\omega_2^2}{k_2\cos^2\alpha_2}A_2^*\sum_{j=1}^4C_{2j}A_jA_j^*,\\ \frac{dA_3^*}{dz}&=+i\frac{\omega_3^2C}{k_3\cos^2\alpha_3}A_1^*A_2A_4^*e^{i\Delta kz}\\&\quad+\frac{i\omega_3^2}{k_3\cos^2\alpha_3}A_3^*\sum_{j=1}^4C_{3j}A_jA_j^*,\\ \frac{dA_4}{dz}&=-i\frac{\omega_4^2C}{k_4\cos^2\alpha_4}A_1^*A_2A_3e^{i\Delta kz}\\&\quad-\frac{i\omega_4}{k_4\cos^2\alpha_4}A_4\sum_{j=1}^4C_{4j}A_jA_j^*.\end{aligned}\tag{4.12}$$

The purely reactive, quadratic Kerr effect terms occur again on the right-hand side.

Some integrals of these complex amplitude equations can be obtained immediately by multiplying the equations by A_1^*, A_2, A_3, and A_4^*, respectively, and adding the complex conjugates. The right-hand sides become equal. Note that the component of the Poynting vector along the wave normal may be written as

$$\begin{aligned}|\mathbf{S}_1|\cos\alpha_1&=\frac{c\cos\alpha_1}{8\pi}|(\mathbf{E}_1\times\mathbf{H}_1^*+\mathbf{E}_1^*\times\mathbf{H}_1)|\\&=\frac{k_1c^2\cos^2\alpha_1}{4\pi\omega_1}A_1A_1^*,\end{aligned}\tag{4.13}$$

and similar relations at the other frequencies. In this manner, the Manley-Rowe relationships,[26] well known in the theory of parametric amplifiers[19] are obtained;

$$|S_1|\cos\alpha_1/\omega_1+|S_2|\cos\alpha_2/\omega_2,$$
$$|S_1|\cos\alpha_1/\omega_1+|S_3|\cos\alpha_3/\omega_3,$$

and

$$|S_1|\cos\alpha_1/\omega_1-|S_4|\cos\alpha_4/\omega_4\tag{4.14}$$

are "constants of the motion." The physical interpretation is that if the number of quanta passing through one cm^2 of the wave front per second increases by a certain amount in the wave at ω_1, the corresponding

[26] H. A. Haus, IRE Trans. on Microwave Theory and Tech. 6, 317 (1958).

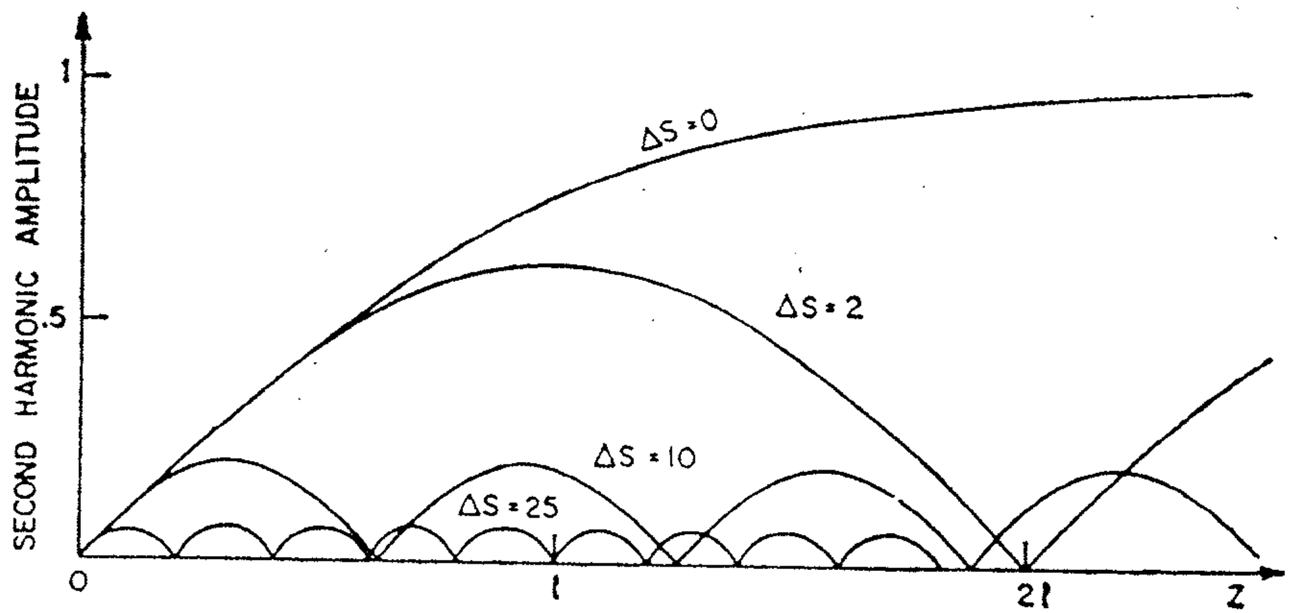
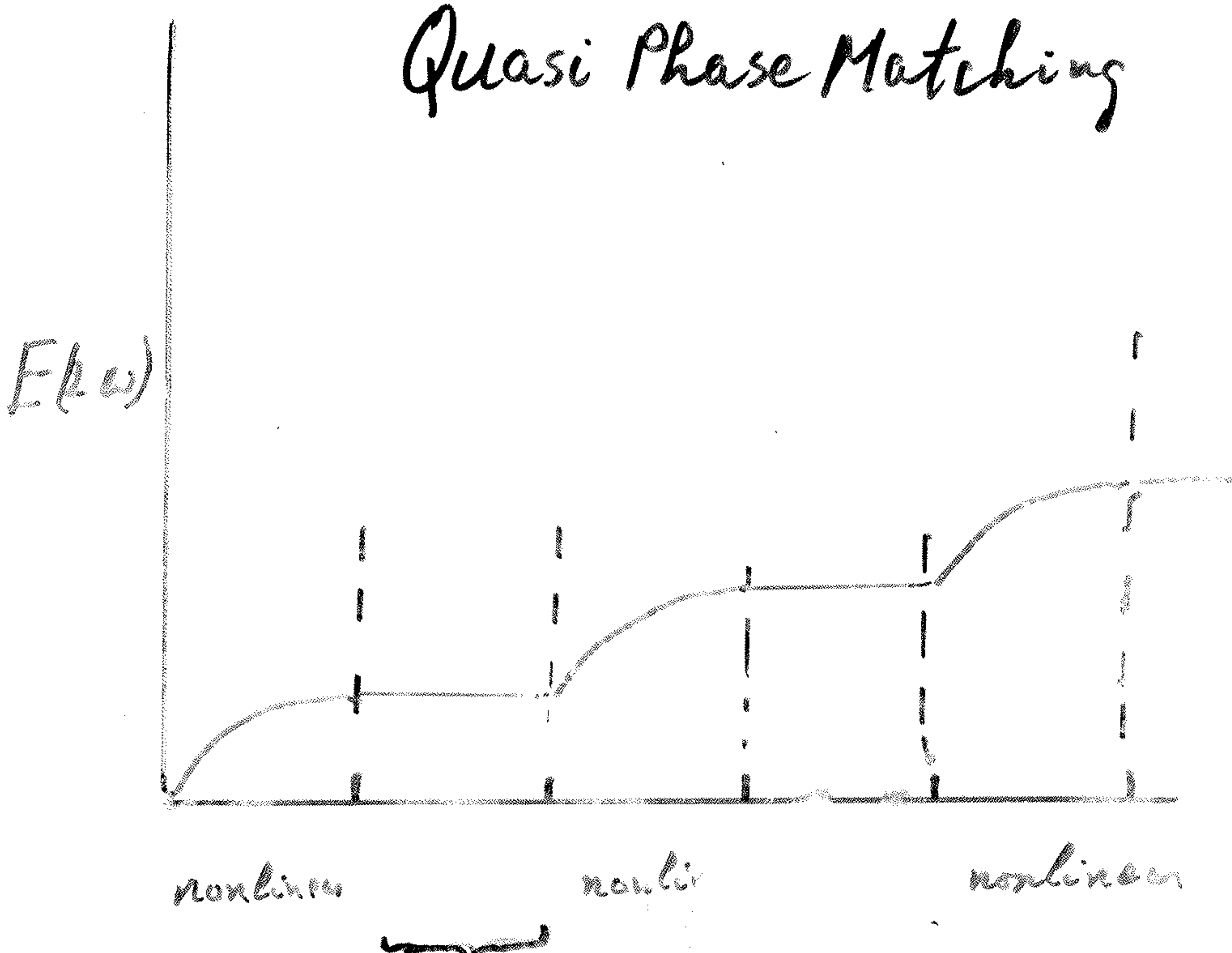


FIG. 5. The growth of the second-harmonic amplitude for varying degrees of phase mismatch.

Quasi Phase Matching



phase corr
in
linear dispersive layers

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 $\sin^2 \Delta k x$ 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 be .m was pass... 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:

$$P_x = aE_y E_z, \quad P_y = aE_z E_x, \quad P_z = aE_x E_y.$$

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

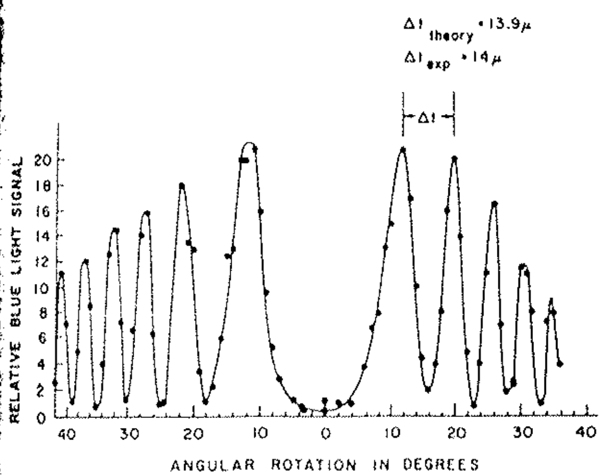


FIG. 1. Blue light generation vs inclination of 0.0308-in. thick quartz platelet to laser beam. Rotation axis normal to beam, parallel to crystal z axis. Red beam unfocused and polarized parallel to the z axis.

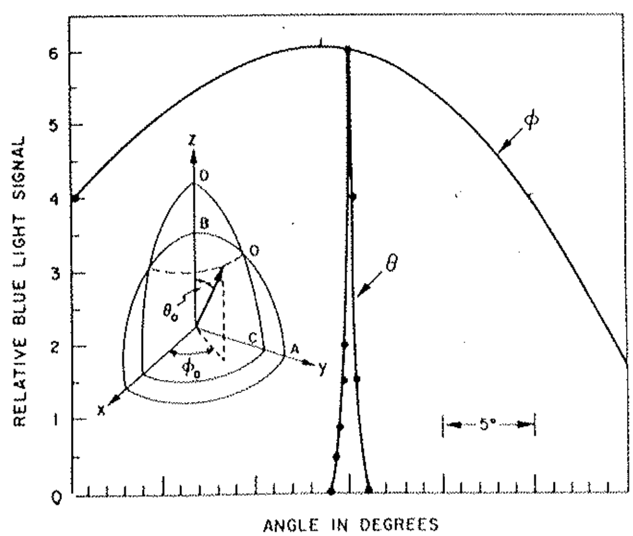


FIG. 2. Blue light intensity as a function of crystal orientation for KDP. Maximum output occurs at $\theta_0 = 52° \pm 2°$, $\phi_0 = 45°$. Laser beam collimated to within $\pm\frac{1}{4}°$. AOB is an arc on the index of refraction surface for red ordinary rays, COD for blue extraordinary rays.

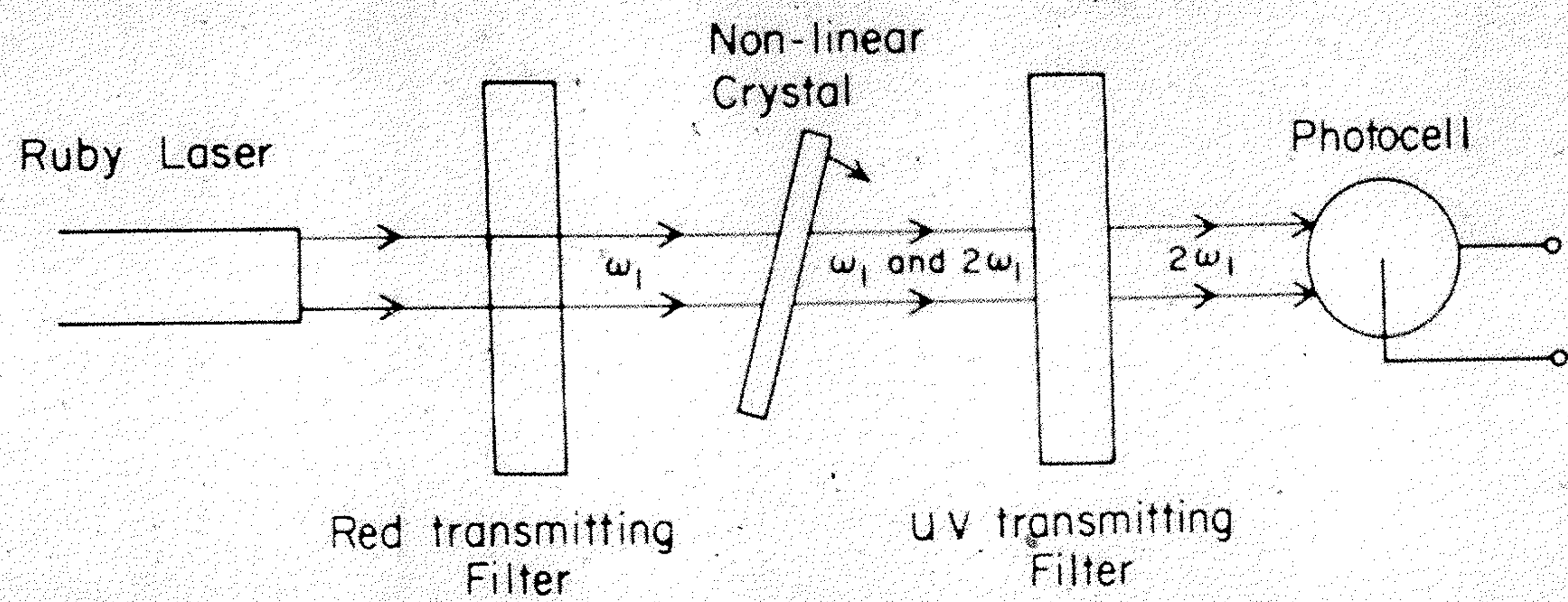
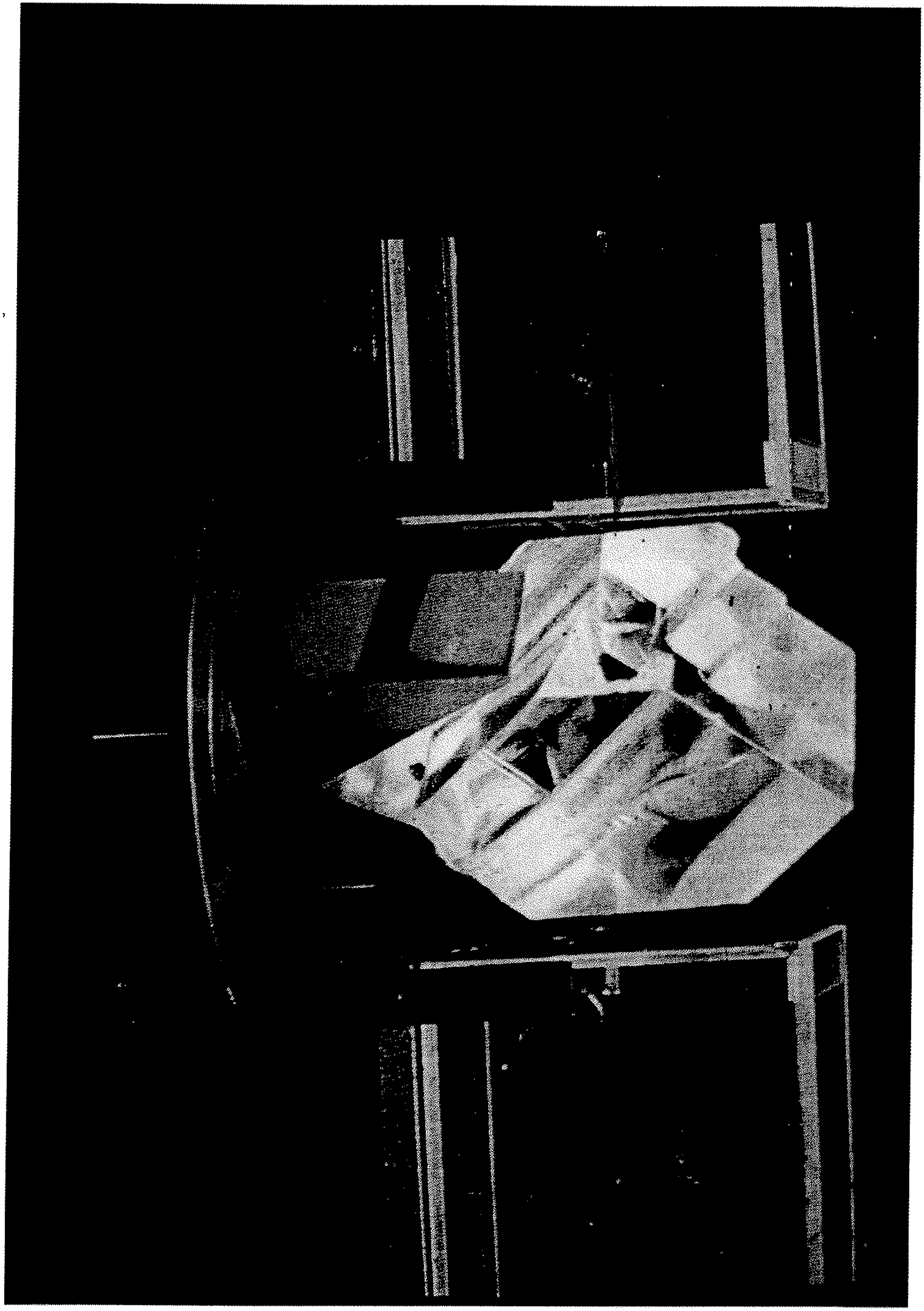


Fig. 3. Second harmonic generation of light.



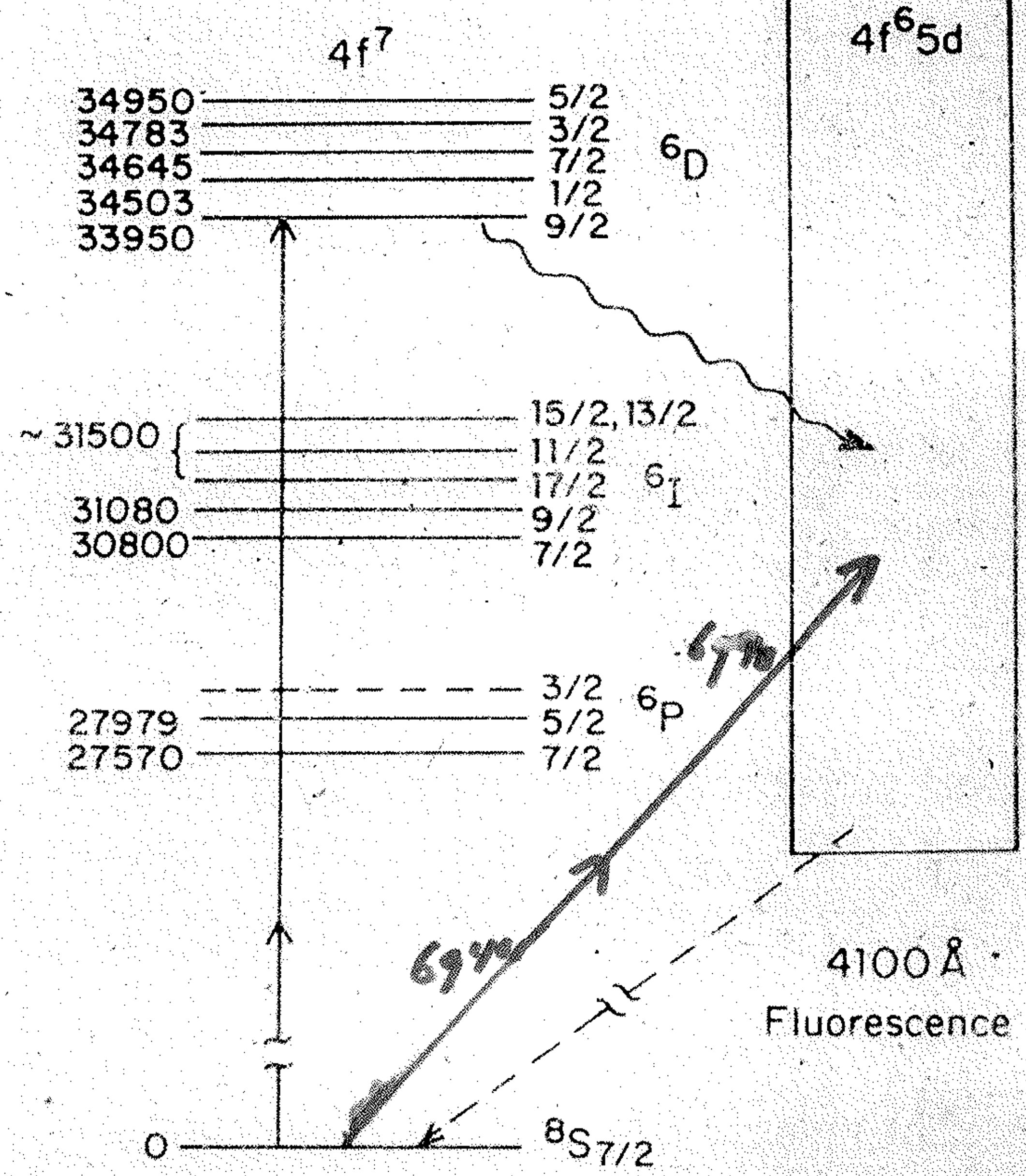


FIG. 1. Energy-level diagram for Eu^{2+}:CaF_2 showing two-photon excitation of $^6D_{9/2}$ and subsequent nonradiative and radiative relaxation. Left-hand column gives average energy of each J multiplet in cm^{-1}. Note the overlap of $4f^65d$ and $4f^7$ levels.

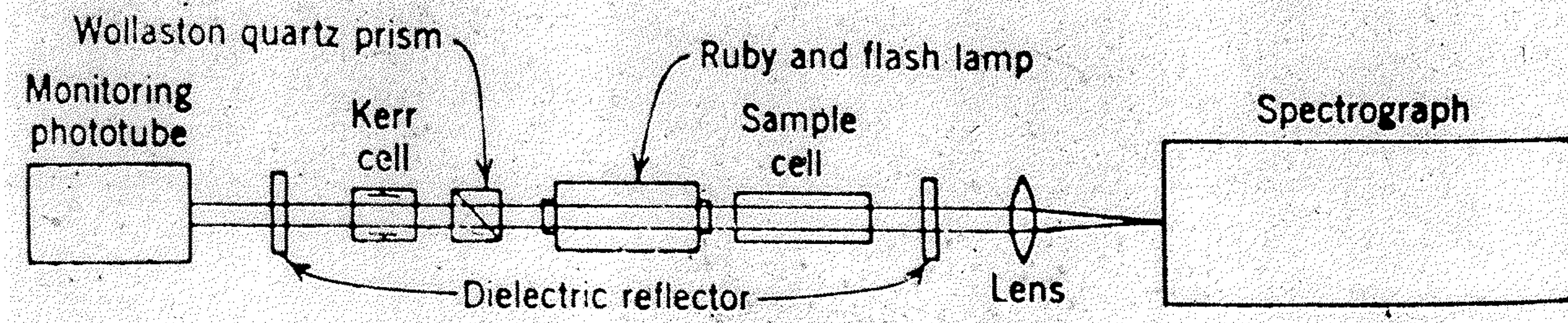


FIG. 2. The original Raman laser oscillator arrangement of Woodbury and Ng. Intense Stokes radiation corresponding to a dominant vibration of the fluid in the sample cell is seen. Without sample cell, Stokes radiation from nitrobenzene in the Kerr cell used for *Q*-switching may be observed. [After B. A. Langyel, *Introduction to Lasar Physics* (John Wiley & Sons, New York, 1966.)]

Н. БЛОМБЕРГЕН

НЕЛИНЕЙНАЯ ОПТИКА

GaAs
mirror
in water

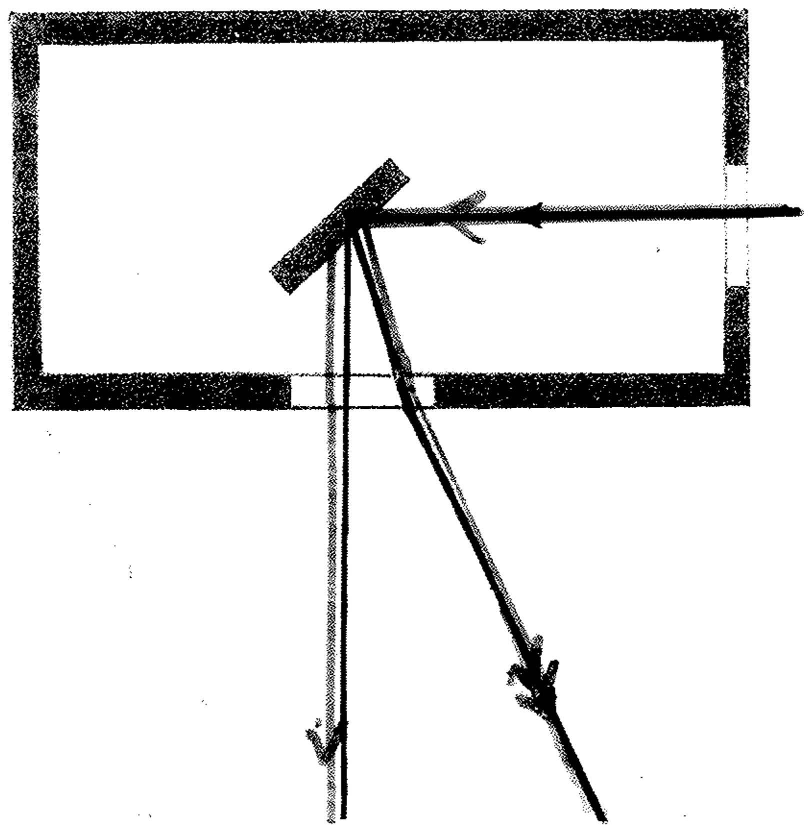


TABLE I. Historical dates of linear and nonlinear optical laws.

	Linear	Nonlinear
Law of reflection	1st century (Hero of Alexandria)	1962 (Bloembergen and Pershan)
Law of refraction	1621 (Snell)	1962 (Bloembergen and Pershan)
Intensity of reflected and refracted light	1823 (Fresnel)	1962 (Bloembergen and Pershan)
Conical refraction		
Theory	1833 (Hamilton)	1969 (Bloembergen and Shih)
Experiment	1833 (Lloyd)	1977 (Schell and Bloembergen)

ÉLECTRONIQUE QUANTIQUE

comptes-rendus de la
3e conférence internationale

QUANTUM ELECTRONICS

proceedings of
the third international congress

PARIS

P. GRIVET
Professeur à l'Université de Paris

N. BLOEMBERGEN
Professeur à l'Université de Harvard

2

DUNOD ÉDITEUR
PARIS

COLUMBIA UNIVERSITY PRESS
NEW YORK

1964