MEMORIES OF THE DISCOVERY OF OPTICAL HARMONICS, PETER FRANKEN, AND SOME THOUGHTS ABOUT THE FUTURE OF NONLINEAR OPTICS

By Alan E. Hill

Quantum Physics Institute, Texas A&M University, USA

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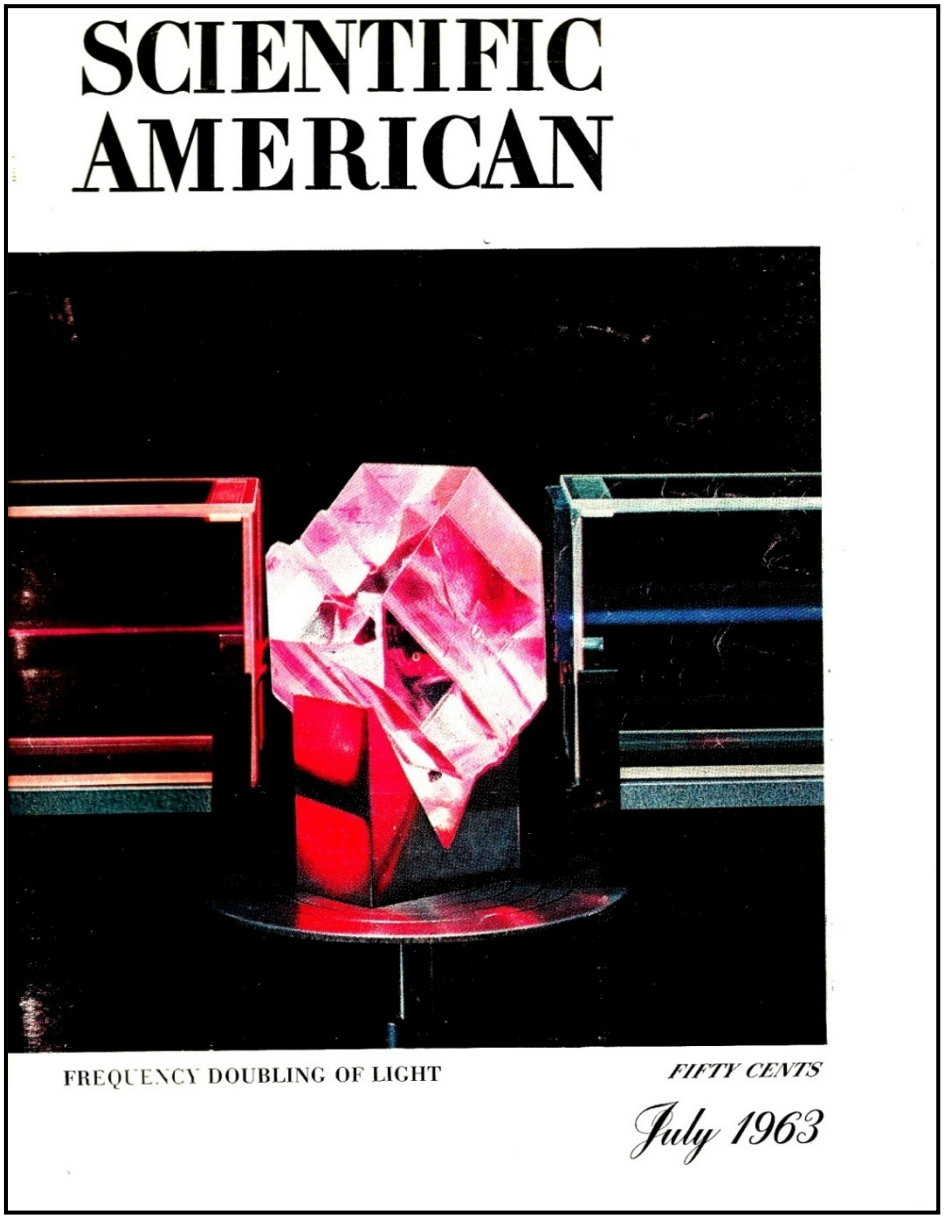
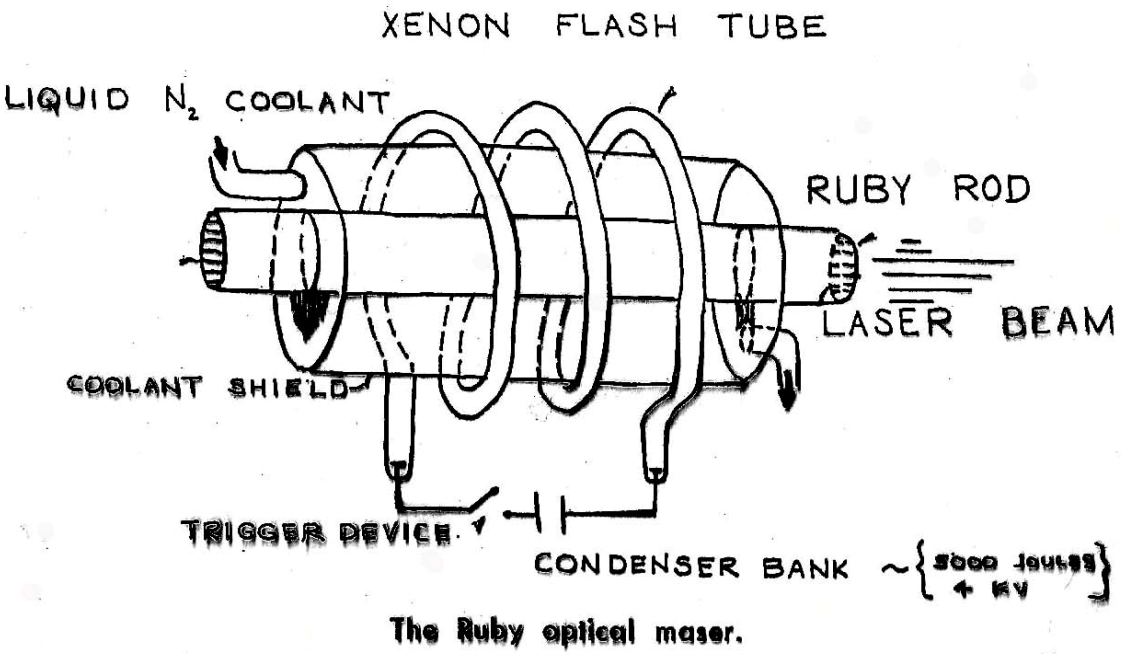
Nonlinear Optics International Conference: East-West Reunion
Suzdal, Russia



Advances in Optical Masers, by Arthur Shallow. Cover photo by Bob Terhune.

Peter Franken while at the University of Arizona

Maiman's "Optical Maser"



The Ruby optical maser.

DECEMBER, 1961

CHARACTERISTICS OF THE FIRST OPTICAL HARMONIC EVENT

RESULT:

CONVERSION EFFICIENCY	10^{-8}
PHOTON YIELD	$\sim 10^{11}$

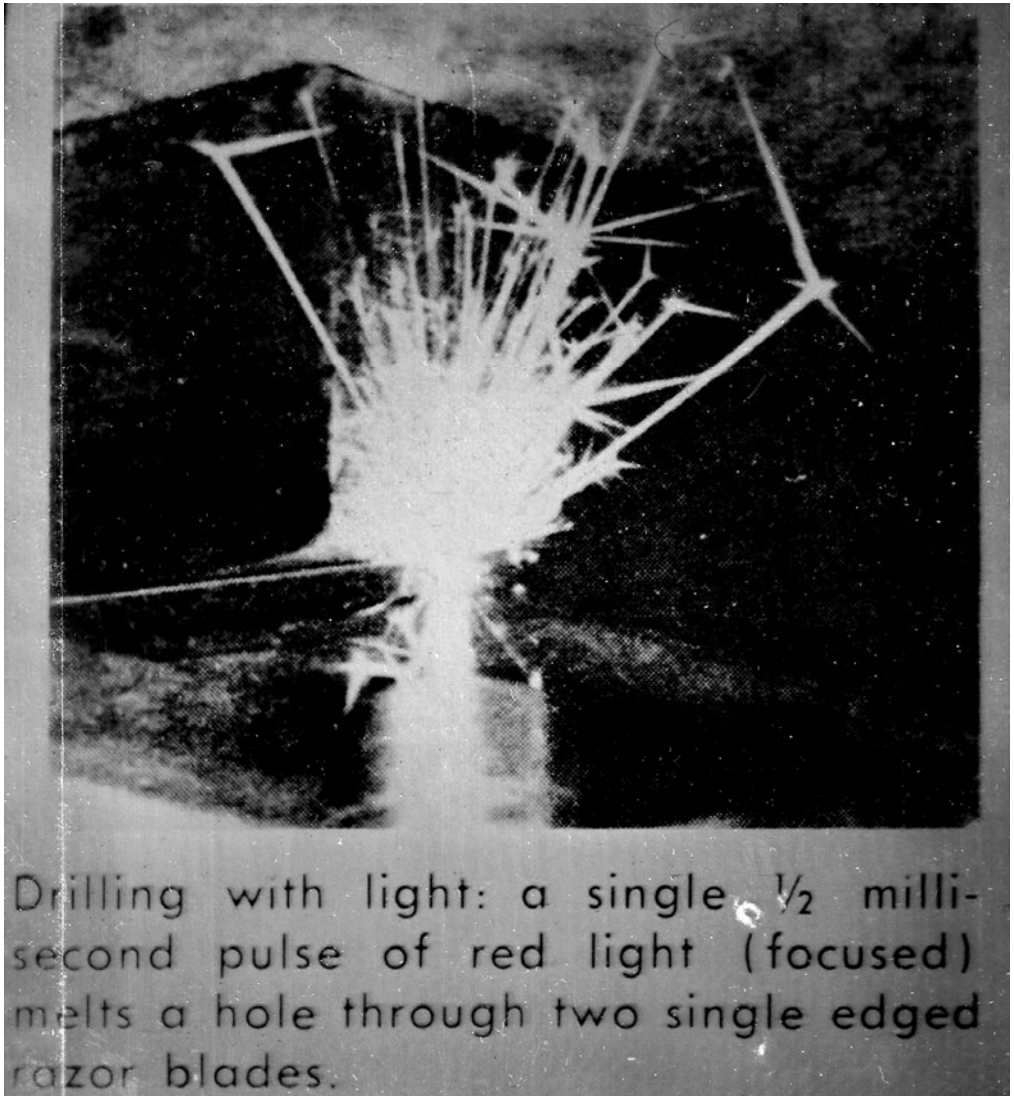
LASER PARAMETERS:

WAVELENGTH	6943 Å
ENERGY	3 JOULES
POWER	3 KW (1 MS PULSE)
	($\sim$30 MW FOCUSED)

HARMONIC GENERATOR PARAMETERS:

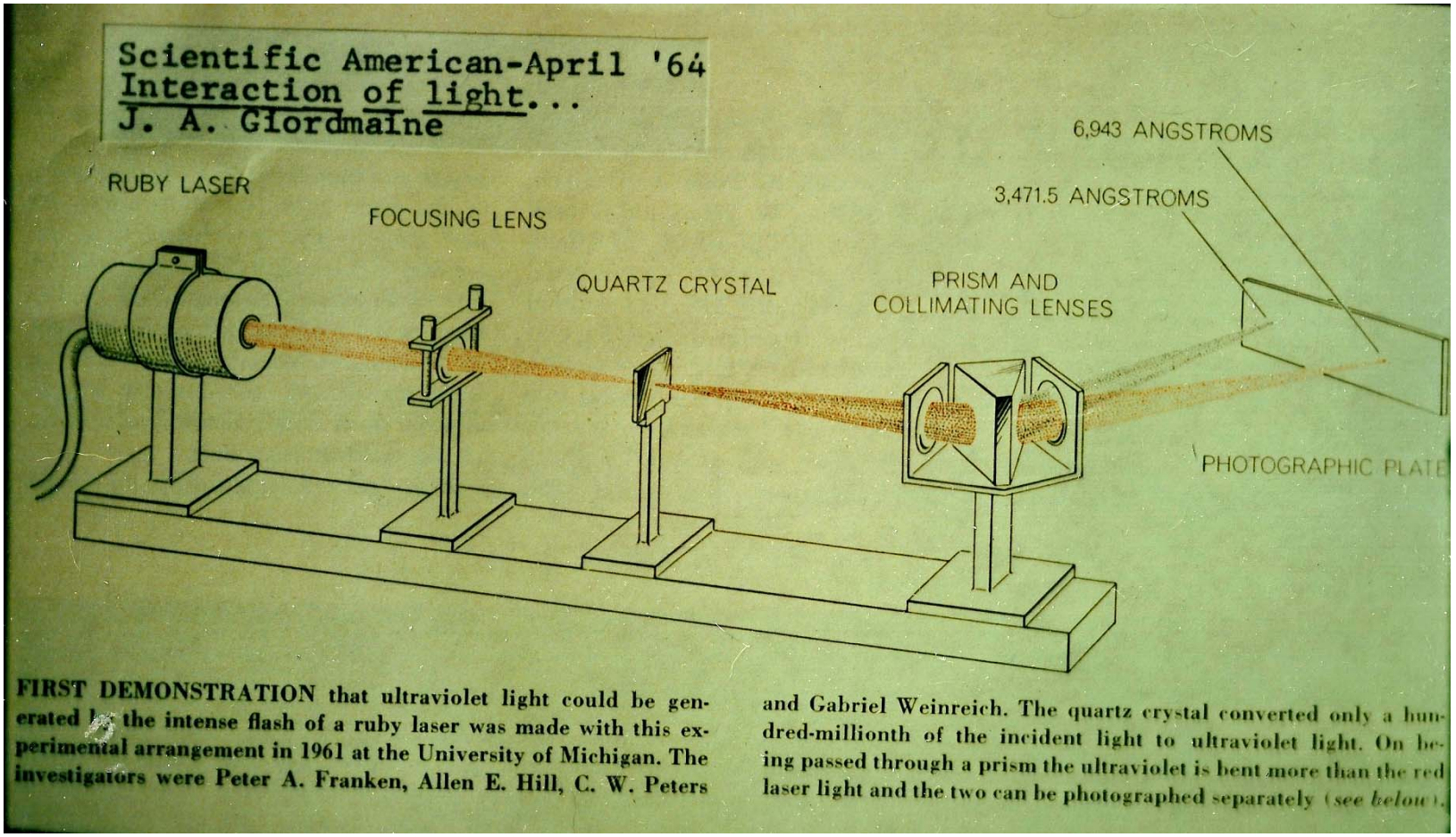
INDEX MATCHING	NONE
COHERENCE VOLUME	10^{-11} CM^2
	($\ell = 10^{-7}$ CM, Å = 10^{-4} CM^2)
FOCUSED $\vec{E}$ FIELD	$\sim 2 * 10^5$ V/CM

A “Two-Gillette Razorblade” Laser



Drilling with light: a single ½ millisecond pulse of red light (focused) melts a hole through two single edged razor blades.

First 2nd Harmonic Experimental Set-up



FIRST DEMONSTRATION that ultraviolet light could be generated by the intense flash of a ruby laser was made with this experimental arrangement in 1961 at the University of Michigan. The investigators were Peter A. Franken, Allen E. Hill, C. W. Peters and Gabriel Weinreich. The quartz crystal converted only a hundred-millionth of the incident light to ultraviolet light. On being passed through a prism the ultraviolet is bent more than the red laser light and the two can be photographed separately (*see below*).

First Optical Harmonics Publication

VOLUME 7, NUMBER 4 PHYSICAL REVIEW LETTERS AUGUST 15, 1961

GENERATION OF OPTICAL HARMONICS*

P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
The Harrison M. Randall Laboratory of Physics, The University of Michigan, Ann Arbor, Michigan
(Received July 21, 1961)

The development of pulsed ruby optical masers[1,2] has made possible the production of monochromatic (6943 A) light beams which, when focussed, exhibit electric fields of the order of 10^5 volts/cm. The possibility of exploiting this extraordinary intensity for the production of optical harmonics from suitable nonlinear materials is most appealing. In this Letter we present a brief discussion of the requisite analysis and a description of experiments in which we have observed the second harmonic (at ~3472 A) produced upon projection of an intense beam of 6943A light through crystalline quartz.

A suitable material for the production of optical harmonics must have a nonlinear dielectric coefficient and be transparent to both the fundamental optical frequency and the desired overtones. Since all dielectrics are nonlinear in high enough fields, this suggests the feasibility of utilizing materials such as quartz and glass. The dependence of polarization of a dielectric upon electric field E may be expressed schematically by

$$P=\chi E\left(1+\frac{E}{E_1}+\frac{E^2}{E_2^2}+\cdots\right), \tag{1}$$

where E_1, E_2 ... are of the order of magnitude of atomic electric fields (~10^8 esu). If E is sinusoidal in time, the presence in Eq. (1) of terms of quadratic or higher degree will result in P containing harmonics of the fundamental frequency. Direct-current polarizations should accompany the even harmonics.

Let $\vec{p}$ be that part of $\vec{P}$ which is quadratic in $\vec{E}$; that is, $\vec{p}$ is a linear function of the components of the symmetric tensor $\vec{E}\vec{E}$. The eighteen coefficients which occur in this function are subject to restrictions due to the point symmetry of the medium. These restrictions are, in fact, identical with those governing the piezoelectric coefficients. In particular, $\vec{p}$ necessarily vanishes in a material such as glass which is isotropic or contains a center of inversion. For crystalline quartz, however, there are two independent coefficients α and β in terms of which

$$\begin{aligned} p_x &= \alpha(E_x^2-E_y^2)+\beta E_y E_z, \\ p_y &= -\beta E_x E_z - 2\alpha E_x E_y, \\ p_z &= 0 \end{aligned} \tag{2}$$

Table I. The square of the total p perpendicular to the direction of propagation of light through crystalline quartz.

Direction of incident beam	The square of the total p perpendicular to direction of propagation
x ($E_x=0$)	$p_y^2+p_z^2=0$
y ($E_y=0$)	$p_z^2+p_x^2=\alpha^2E_x^4$
z ($E_z=0$)	$p_x^2+p_y^2=\alpha^2(E_x^2+E_y^2)^2$

(z is the threefold, or optic, axis; x a twofold axis). If a light beam traverses quartz in one of the three principal directions, Eqs. (2) predict the results summarized in Table I. The second-harmonic light should be absent in the first case, dependent upon incident polarization in the second case, and independent of this polarization in the third.

If an intense beam of monochromatic light is focussed into a region of volume V, there should occur an intensity I of second harmonic given (in Gaussian units) by

$$I\sim(\omega^4/c^3)(pv)^2(V/v), \tag{3}$$

where ω is the angular frequency of the second harmonic, c the velocity of light, and v an effective "volume of coherence"; that is, the size of a region within the sample in which there is phase coherence of the p excitation. (This volume may in practice be much smaller than V.) An estimate of v is governed by several considerations. For example, it is probably of no greater extent in the propagation direction than $\sim[n_2\times(n_2-n_1)^{-1}]\lambda_2$, where n_1 and n_2 are the indices of refraction for the fundamental and second harmonic frequencies, respectively, and λ_2 is the wavelength of the second harmonic. The lateral extent of this volume is determined in large part by the coherence characteristics of the optical maser. The situation for a maser of the gas discharge[3] type is clearly more favorable in this respect than that for the ruby device.[1,2] For a coherence volume of 10^{-11} cm^3, which we think may be realistic in our case, Eq. (1) indicates

U-514 1-2

The Missing Evidence: “Out, Out, Damn Spot!” (William Shakespeare)

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.

that second harmonic intensities as high as a fraction of a percent of the fundamental could be achieved.

In the experiments we have used a commercially available ruby optical maser[4] which produces approximately 3 joules of 6943A light in a one-millisecond pulse. This light is passed through a red filter for the elimination of the xenon flash background and is then brought to a focus inside a crystalline quartz sample. The emergent beam is analyzed by a quartz prism spectrometer equipped with red insensitive Eastman Type 103 spectrographic plates. A reproduction of the first plate in which there was an unambiguous indication of second harmonic (3472 A) is shown in Fig. 1. This plate was exposed to only one “shot” from the optical maser. We believe the following two facts, among others, rule out the possibility of artifact:

(1) The light at 3472 A disappears when the quartz is removed or is replaced by glass.

(2) The light at 3472 A exhibits the expected dependence on polarization and orientation summarized in Table I.

Considerations of the photographic image density and the efficiency of the optical system lead us to believe that the order of 10^{11} second harmonic photons were generated within the quartz sample per pulse.

The production of a second harmonic should be observable in isotropic materials such as glass if a strong bias field were applied to the sample. This bias could be oscillatory, thus producing sidebands on the fundamental frequency and the harmonics.

We would like to thank the staff of Trion Instruments, Inc., for their valuable and sustained cooperation in this work.

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*This work was supported in part by the U. S. Atomic Energy Commission.

[1]T. H. Maiman, Nature 187, 493 (1960).

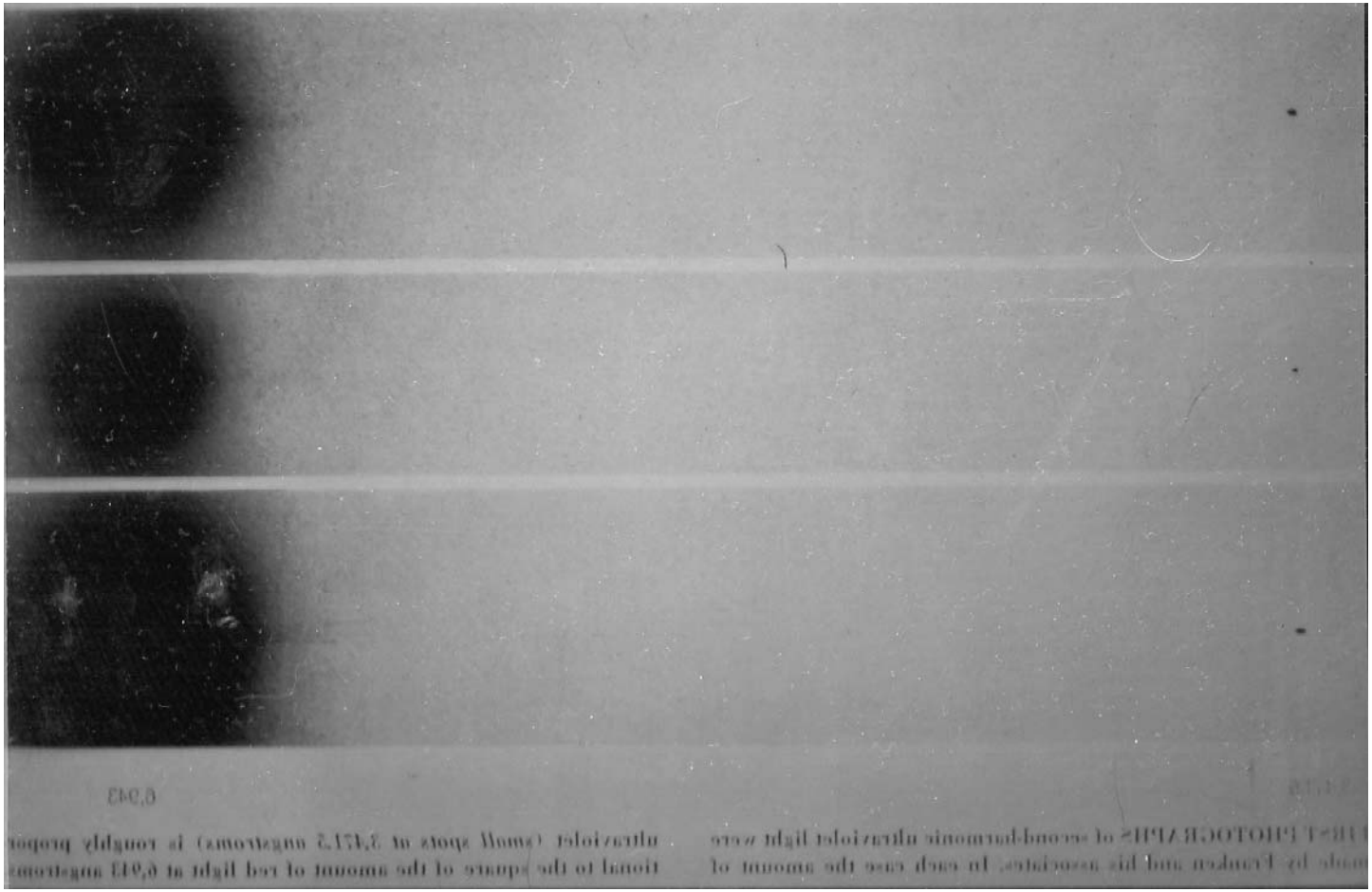
[2]R. J. Collins et al., Phys. Rev. Letters 5, 303 (1960).

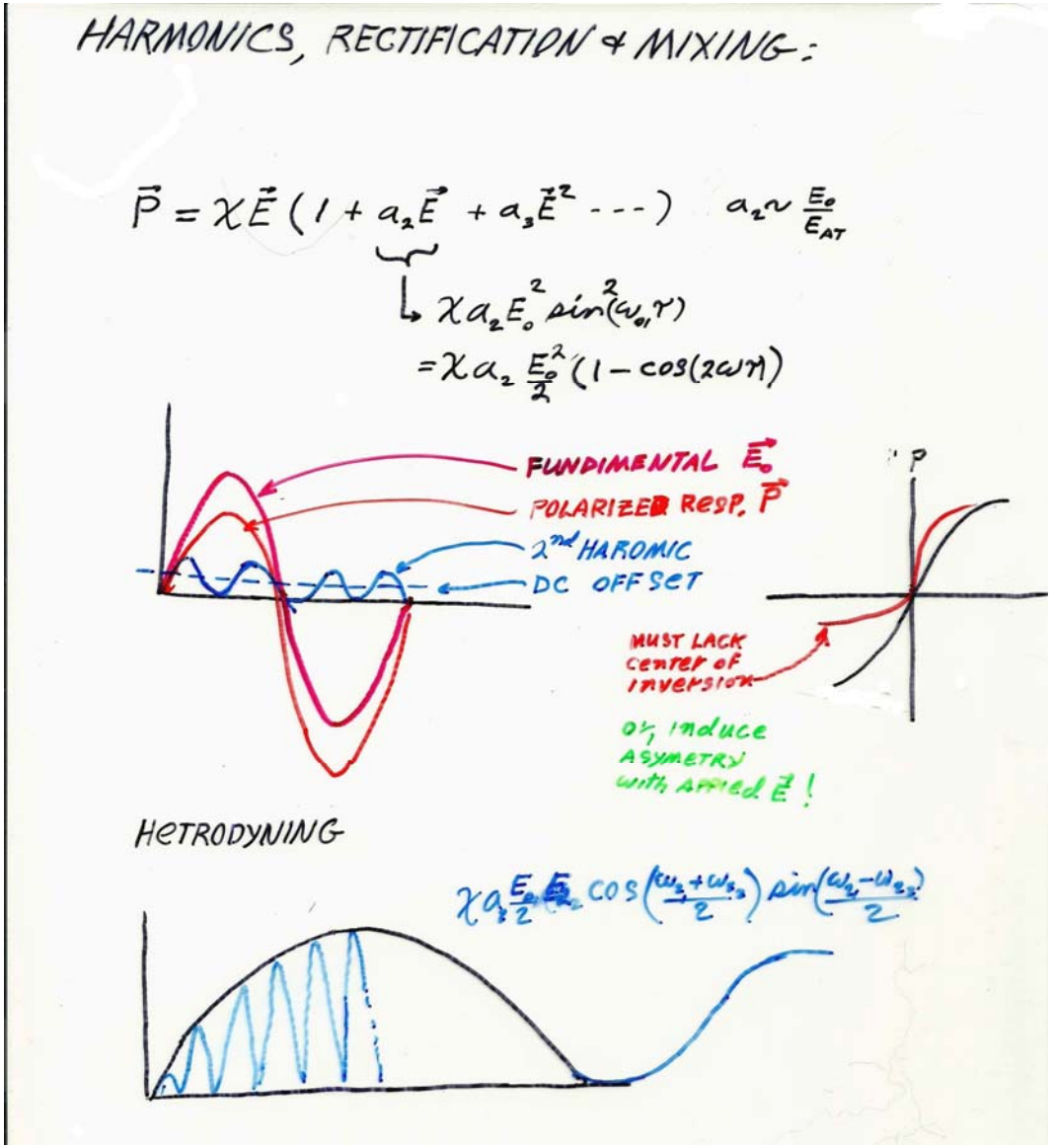
[3]A. Javan, W. R. Bennet, and D. R. Herriott, Phys. Rev. Letters 6, 106 (1961). Even though the intensity of the gas device is very low compared with ruby masers, the gain in coherence volume and the potential improvement of focussing suggest that the gas maser may be comparable or even superior as a source for optical harmonics.

[4]Trion Instruments, Inc., Model No. TO-3000.

Photo of Captured Harmonics Event

(exploded flash lamp to obtain clearly visible spots)





Early Presentation Viewgraph by Alan Hill

Optical Mixing Paper

VOLUME 8, NUMBER 1 PHYSICAL REVIEW LETTERS JANUARY 1, 1962

OPTICAL MIXING*

M. Bass, P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich
The Harrison M. Randall Laboratory of Physics, University of Michigan, Ann Arbor, Michigan
(Received December 4, 1961)

The presence of small quadratic terms in the optical polarizability of transparent piezoelectric materials has made possible the production of the optical second harmonic of the intense red light from a ruby laser.[1] These nonlinear terms should also permit the mixing of light from two different sources of different frequencies. The present Letter reports the observation of the sum frequency in the near ultraviolet of two ruby laser beams of different frequencies coincident simultaneously upon such a crystal.

In the experiment, the sources were Trion Instruments, Inc., pulsed ruby lasers—one operated at room temperature, the other at liquid nitrogen temperature. Abella and Cummins[2] have reported that there is a difference of about 10 A between the wavelengths emitted by ruby lasers at these two temperatures. The flash lamps of both units were triggered by the same voltage pulse, and it was observed that the start of laser action in one ruby was rarely delayed by as much as 100 μsec with respect to the other, as compared to a duration of laser action of about 500 μsec in each. The two laser beams were superimposed by means of a half-silvered mirror and the resultant beam was focussed by a 16-mm $f/1$ lens onto the front surface of a crystal of triglycine sulfate[3] about 3 mm thick. This crystal was placed at the entrance slit of a Hilger quartz prism spectrograph. The slit was set at approximately 25 microns in order to achieve the necessary resolution in the ultraviolet. It should be noted that this particular arrangement requires precise optical alignment: The primary beams must be coplanar, they must focus to the same spot on the crystal, and this focus must lie directly in front of the entrance slit of the spectrometer.

A magnified reproduction of the 3470A region of the most successful plate exposed to nine synchronized pulses from the two lasers is shown in Fig. 1. The line on the right is the second harmonic of the cold laser, the one on the left is the second harmonic of the warm one, and the middle one is the sum frequency of the two. The separation of the lines is 2.5 A, which is consistent with an original difference in the red of 10 A. The cold second harmonic is distinctly more intense than the warm one in accord with the primary intensities; the intensity of the sum frequency lies between these two. The "staggering" of the three lines in Fig. 1 is due to inevitable small departures from the alignment conditions noted above. Many repeated exposures with just one laser have yielded only a single line in the ultraviolet corresponding to its second harmonic.

FIG. 1. Magnified spectrum in the 3470A region made by nine successive synchronized shots of the warm and cold ruby lasers focussed on a triglycine sulfate crystal. The second harmonic of the cold laser is to the right, the second harmonic of the warm laser to the left, and the sum frequency between them. The distance between the lines on the plate is 0.004 inch, corresponding to 2.5 A.

It is a pleasure to acknowledge the technical assistance of Mr. William Fredrick and the continued help and cooperation afforded us by the staff of Trion Instruments. We would also like to thank Dr. J. Giordmaine of the Bell Telephone Laboratories for several valuable discussions.

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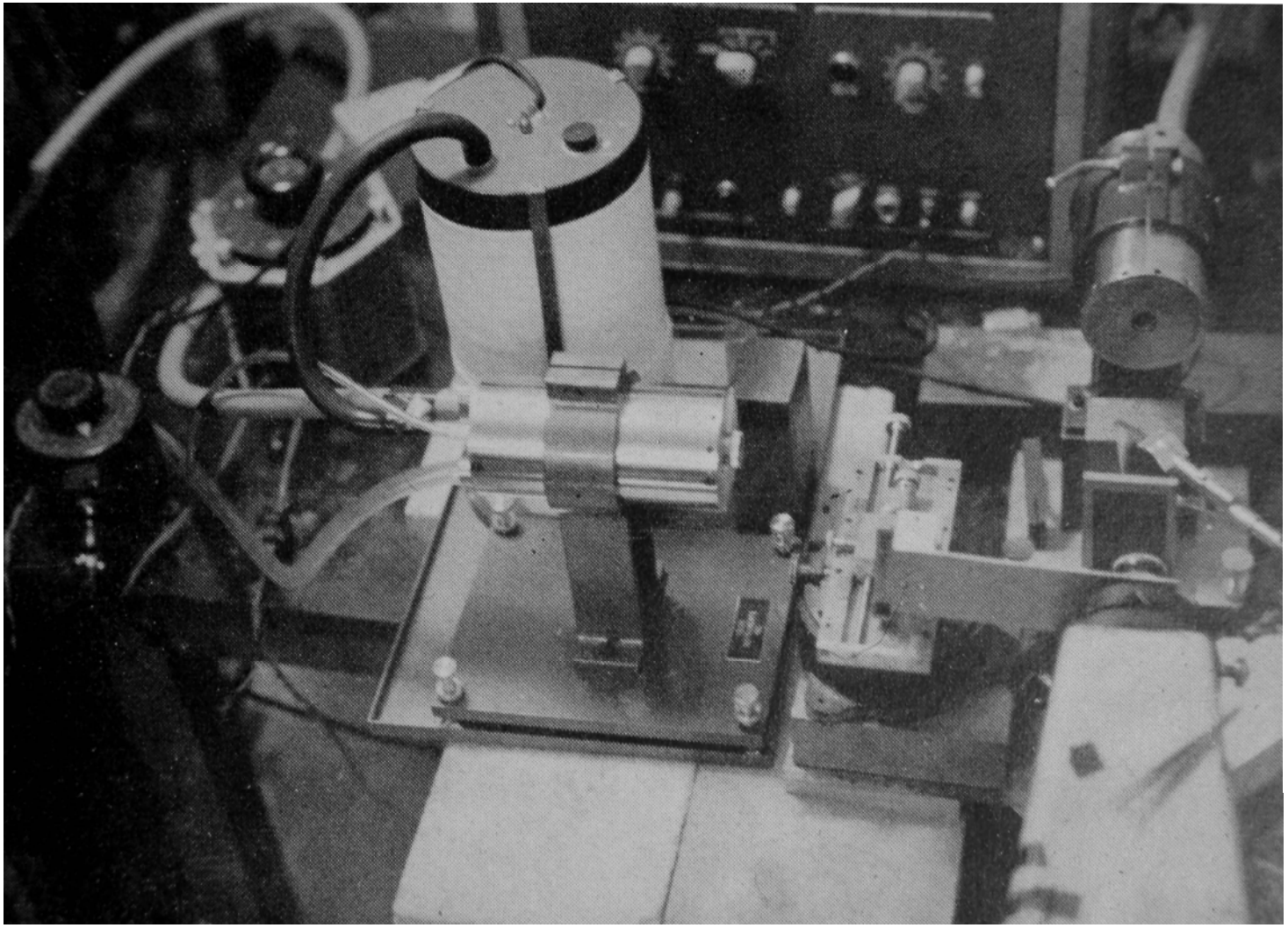
*This work was supported in part by the U. S. Atomic Energy Commission.

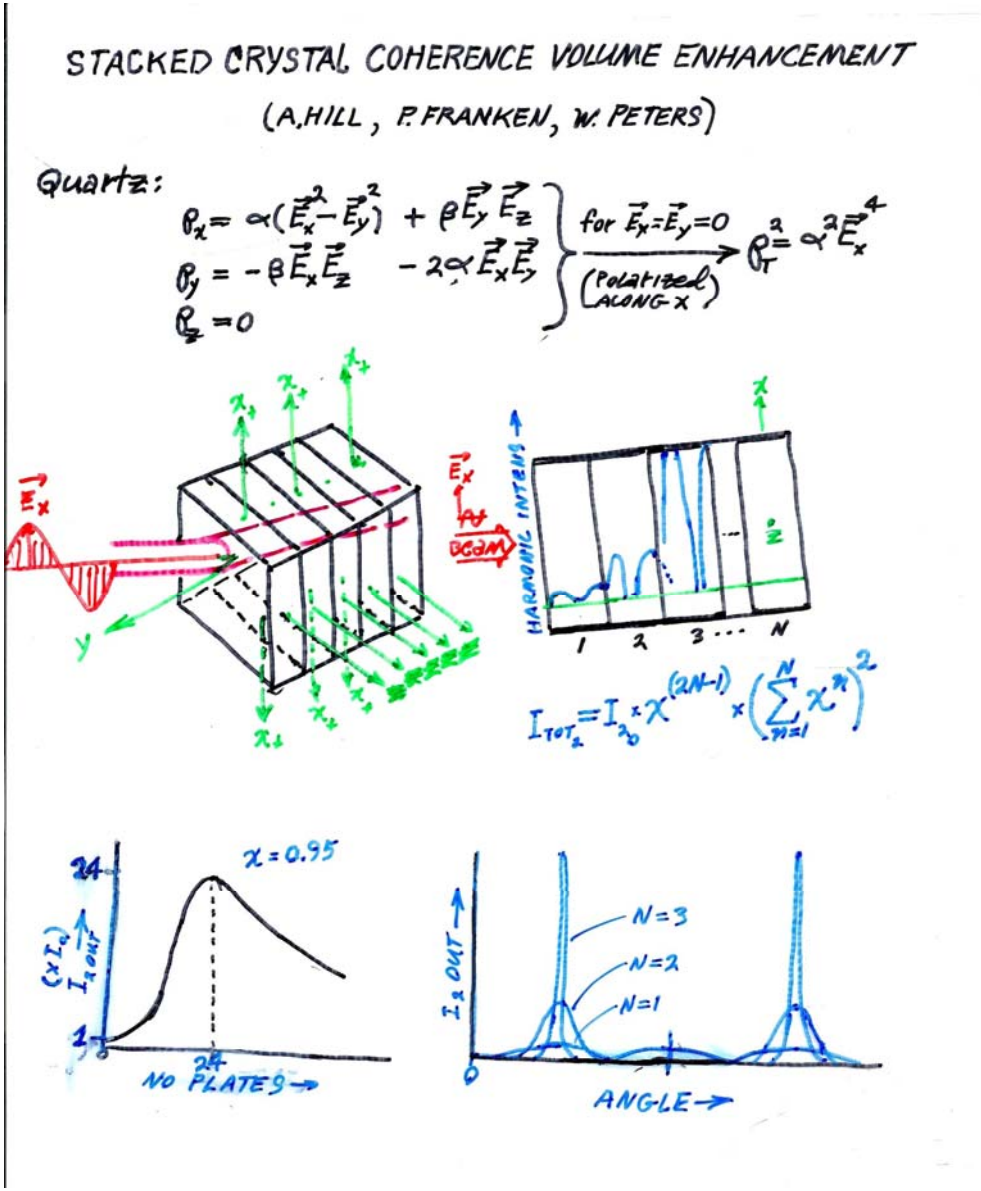
[1] P. A. Franken, A. E. Hill, C. W. Peters, and G. Weinreich, Phys. Rev. Letters 7, 118 (1961).

[2] I. D. Abella and H. Z. Cummins, J. Appl. Phys. 32, 1177 (1961).

[3] Triglycine sulfate was used because in our apparatus it has appeared to be more effective than many other crystals for the production of optical harmonics. It is interesting to note that this crystal does not yield the second harmonic when it is at a temperature above its Curie point (~50°C), due to the change in its crystal symmetry at the Curie point as reported by S. Hoshino, Y. Okaya, and R. Pepinsky, Phys. Rev. 115, 323 (1959).

Photo of Optical Mixing Apparatus





Early Presentation Viewgraph by A. Hill

*Note: The only publication of this work was a reference in J. A. Giordmaine's 1964 *Scientific American* article

2nd Harmonic Intensity From Quartz vs. Incident Angle

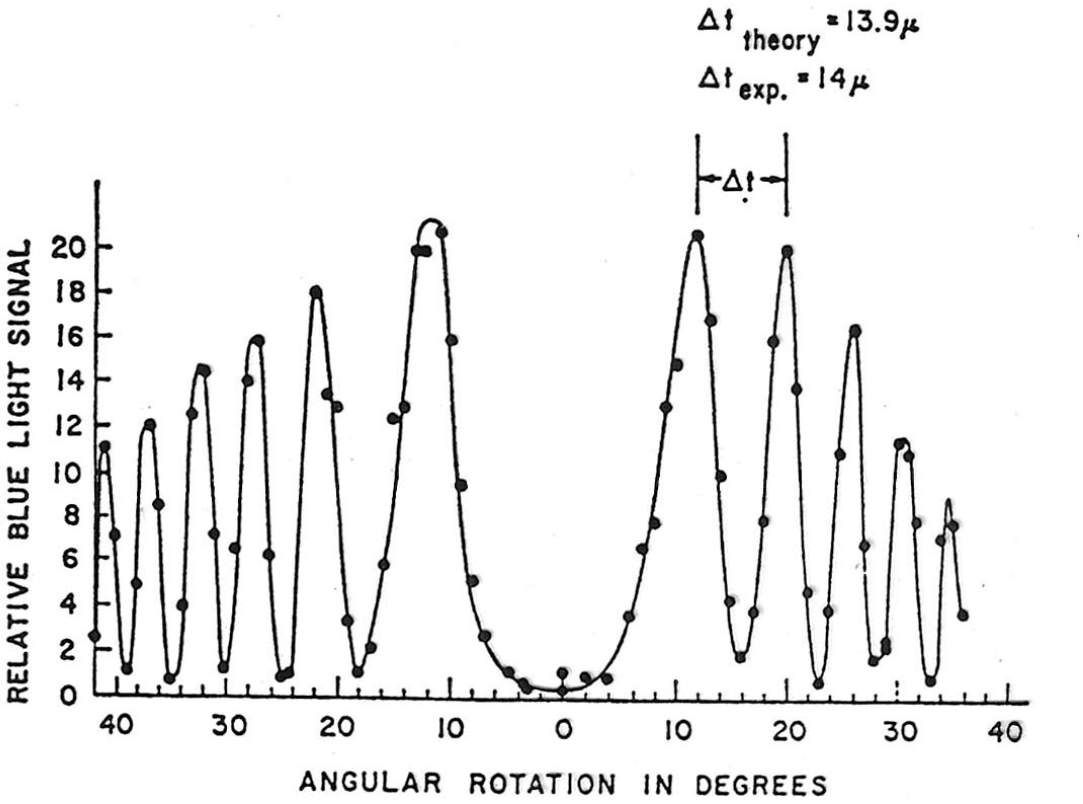
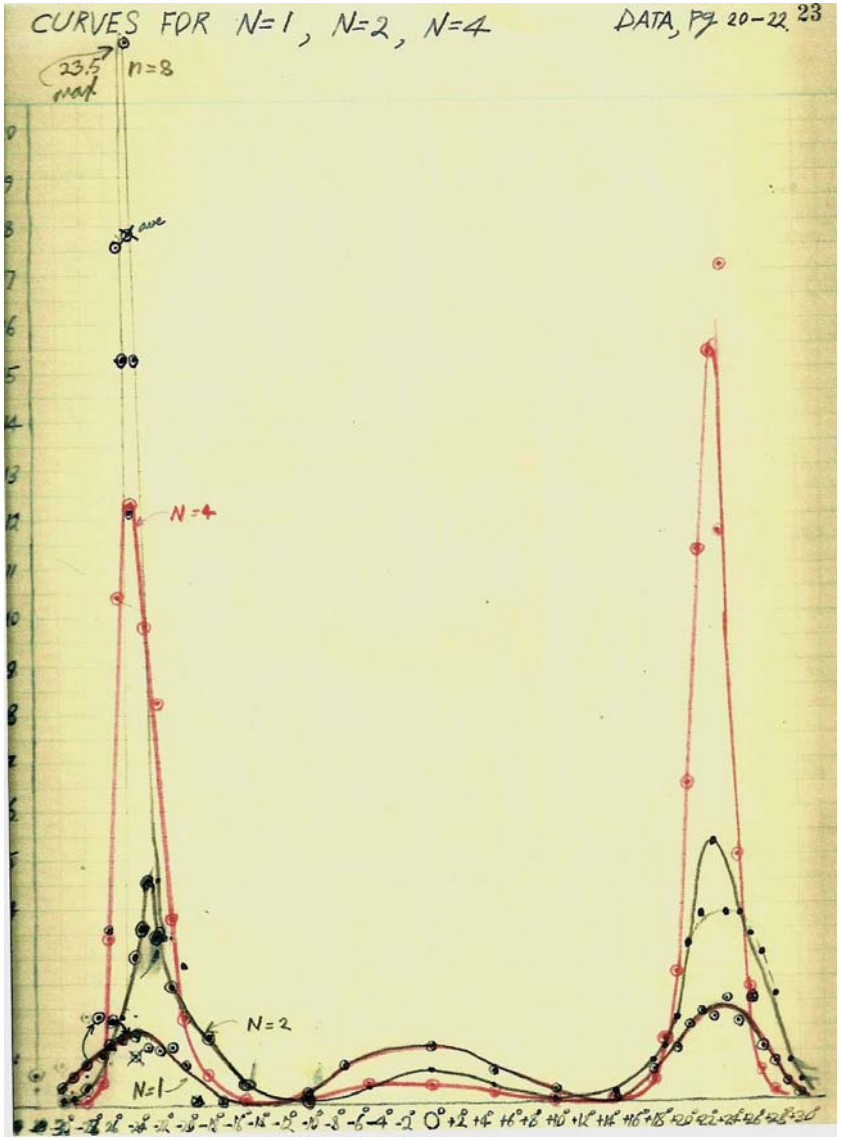


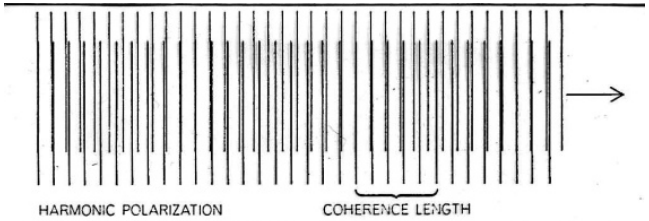
FIG. 3. Second-harmonic generation vs inclination of 0.0308-in.-thick quartz platelet to laser beam. Rotation axis normal to beam, parallel to crystal z axis. Laser beam unfocused and polarized parallel to the x axis. [Figure reproduced from reference (M62).]

2nd Harmonic Coherence Volume Enhancement vs. Angle of Stacked Quartz Plates

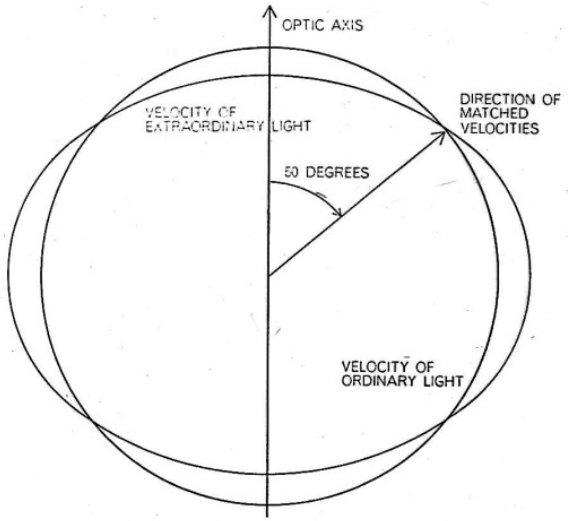


Taken from A. Hill's Notebook

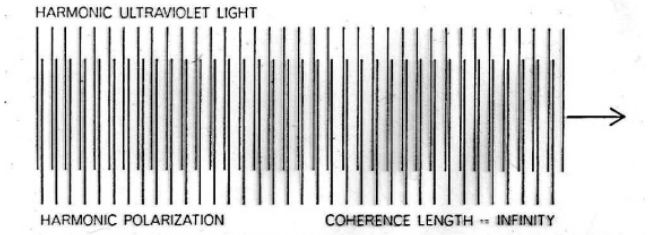
Index Matching Method for Coherence Volume Enhancement (Terhune et al.)



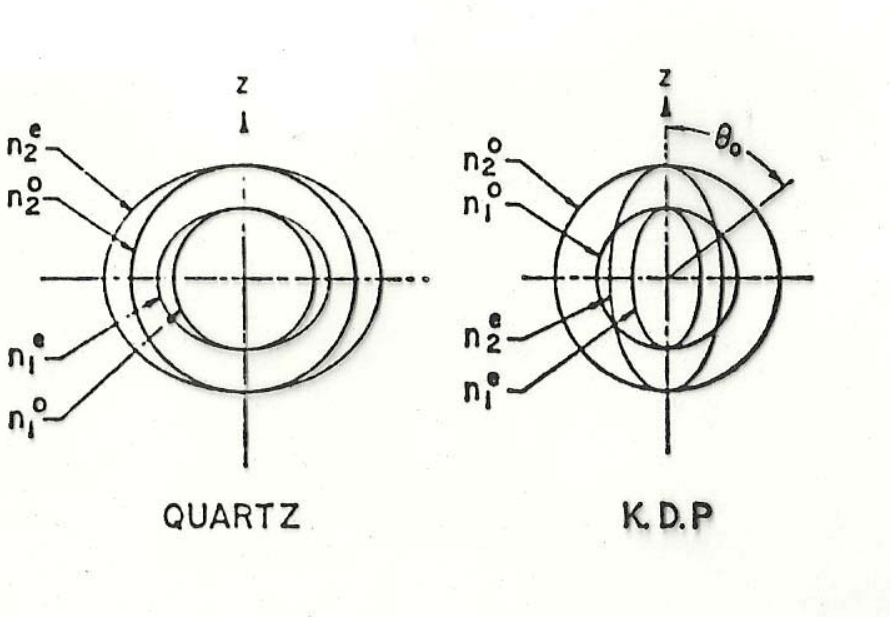
DESTRUCTIVE WAVE INTERFERENCE occurs in a crystal because the polarization wave (*black*) normally travels a little faster than its harmonic radiation (*color*). The polarization and its radiation get completely out of step in a distance called the coherence length.



INTERFERENCE CAN BE OVERCOME by generating harmonic light in a crystal that exhibits double refraction such as KDP. The black circle represents the velocity of ruby laser light of 6,943 angstroms polarized perpendicularly to the page. The colored oval represents the velocity of second-harmonic light of 3,471.5 angstroms polarized in the plane of the page. At 50 degrees to the optic axis both waves travel at the same velocity.

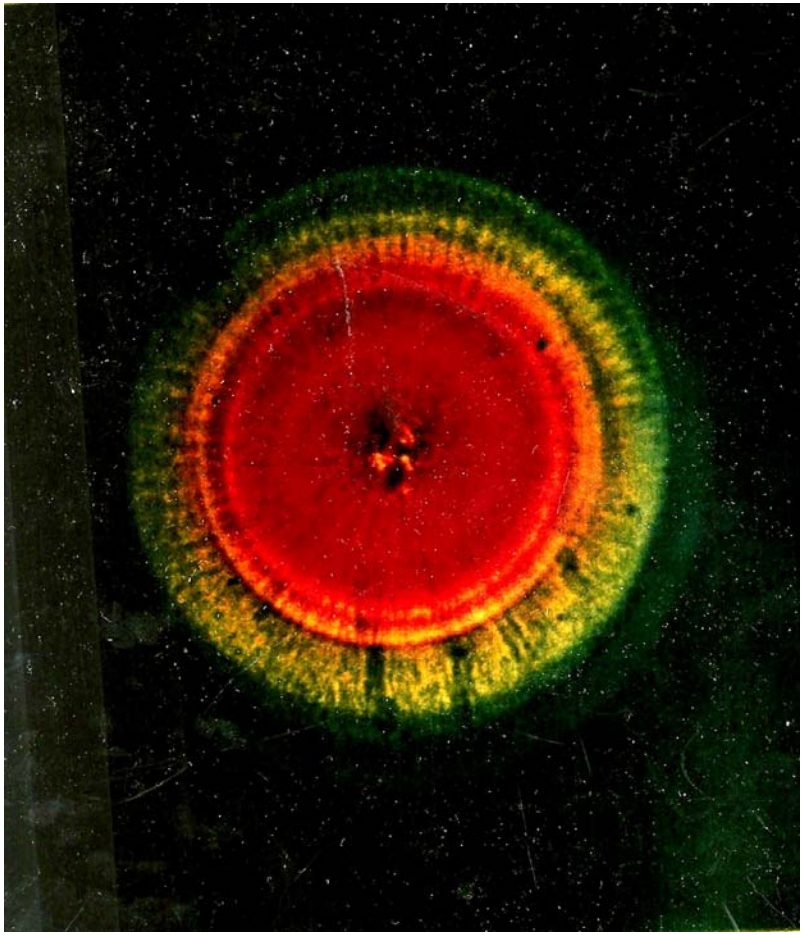


ELIMINATION OF WAVE INTERFERENCE is achieved by sending a ruby laser beam into a crystal of KDP at 50 degrees to the optic axis. Now the harmonic polarization wave (*black*) and the harmonic ultraviolet light it radiates (*color*) stay in step indefinitely.



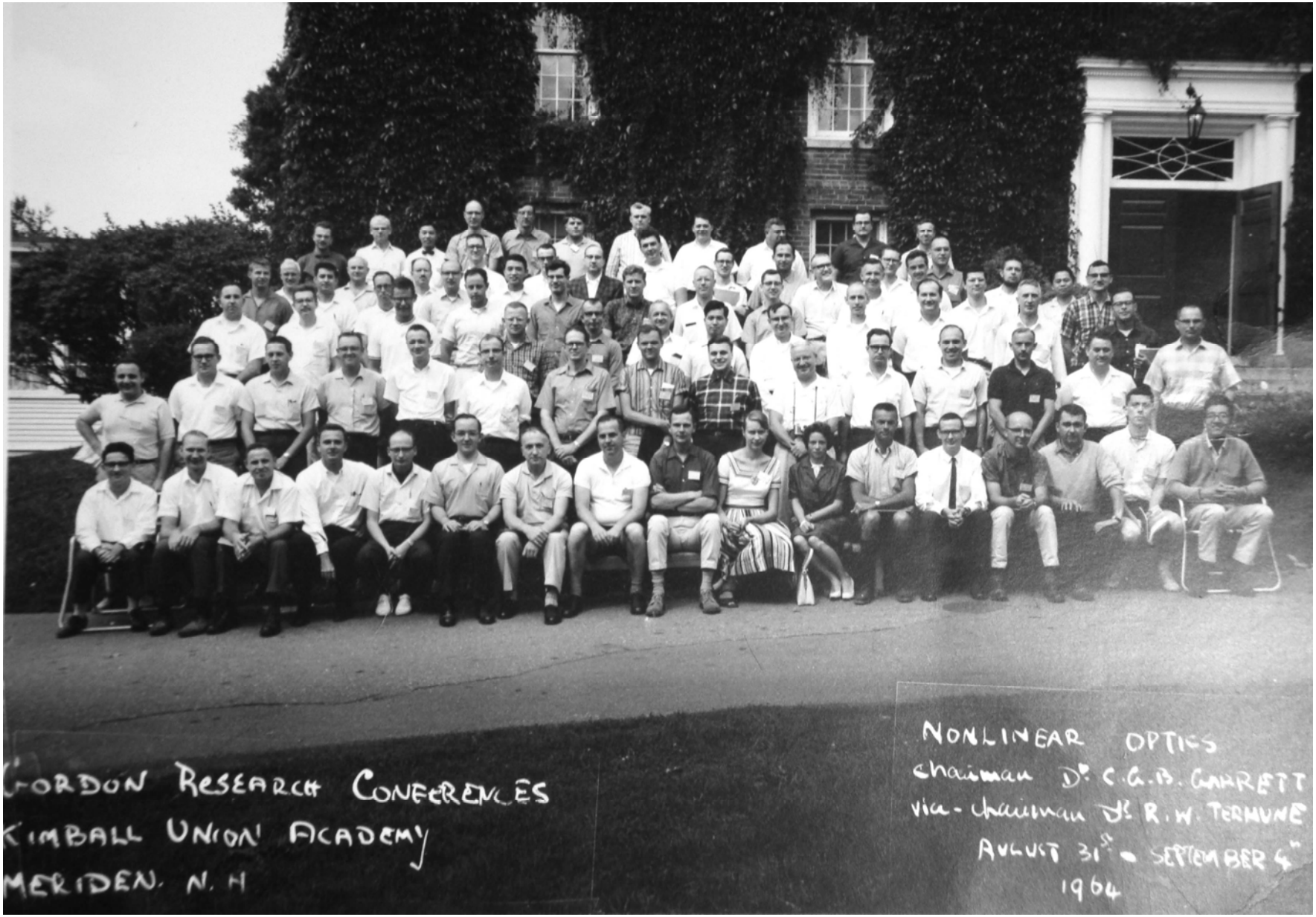
Refractive index surfaces for quartz and KDP. The subscripts 1 and 2 refer to radiation at the Ruby-laser frequency and the second harmonic, Respectively. The superscripts *o* and *e* refer to Ordinary and extraordinary rays, respectively. The figures are exaggerated for purposes of Illustration.

Stimulated Raman Effect



The Stimulated Raman Effect is seen when the output of a giant-pulse laser is focused in benzene. The vibration frequency of the benzene molecule adds to the laser frequency, yielding the colored rings. *Scientific American*, July 1963; article by A. Shallow; photo by R. W. Terhune.

FIRST NONLINEAR OPTICS CONFERENCE: Gordon Res. Conf., Aug. 31-Sept. 4th, 1964:



Optical Harmonics Created in a Vacuum

A Crazy 50-Year Old Idea:

- Since ultra-strong E-fields of a laser beam had to be demonstrated to stress the electrical polarization of a dielectric media thereby producing a harmonic component of that beam,
- And, since a "vacuum" possesses physical characteristics analogous to a media, might it likewise be stressed by a laser, thereby producing harmonic components of the laser beam.

Physical Characteristics of a Vacuum Analogous to Matter:

	Vacuum	Dielectric Media
Electric permittivity	ϵ_o	$\epsilon = \epsilon_o\epsilon_r$
Magnetic permeability	μ_o	$\mu = \mu_o\mu_r$
Wave velocity	$c = \sqrt{1/\mu_o\epsilon_o}$ = 3 x 10^8 m/sec	$c = \sqrt{1/\mu_r\epsilon_r}$
Electrical impedance	$Z = \sqrt{\frac{\mu_o}{\epsilon_o}} = \frac{E}{H}$ = 376 ohms	$Z = \sqrt{\frac{\mu_r}{\epsilon_r}} = \sqrt{\frac{L}{C}}$

Another striking analogy is the velocity of acoustic waves in a solid (or gas) established by the relationship $V = \frac{1}{\sqrt{\rho K_S}}$, which is of the same form as C, and once more set by the physical constraints that define the media (ρ = density, K_s = isotropic compressibility).

Apparent Difficulty: Outcome of the Michelson-Morley Experiment

- In light of the Michelson-Morley experiment, everyone became convinced that there was no ether; hence, vacuum was simply empty space.
  ** Ether: The medium supposed by the ancients to fill the regions of space.*

- Yet, the foregoing physical analogues attest to the premise that the vacuum is anything but a void and susceptible to non-linear polarization by a sufficiently strong field of a high-power laser.

- 50 years ago Gabriel Weinrich took my suggestion seriously, but pointed out that e^+e^- production would probably be involved.

Physical Parameters Needed For Optical Harmonic Production in a Vacuum

- Threshold energy for e^+e^- pair production as a single reaction = 2 x 55 mev.

- Now lasers are pushing towards the energy levels required for e^+e^- pair production!

- QED theory predicts quantum fluctuations; i.e., constant flow of energy in and out of a vacuum, which is needed for e^+e^- pair production and optical harmonics.

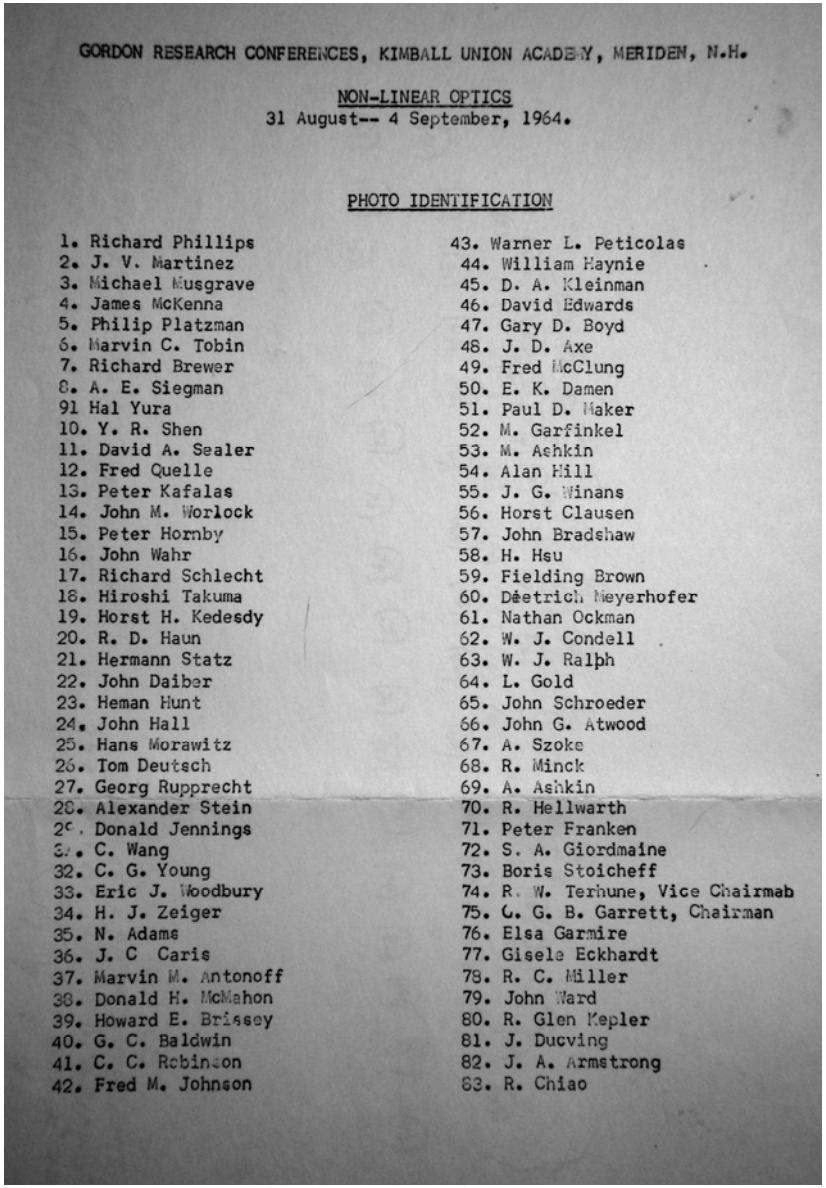
- An 1.3×10^{16} volts/cm optical-electric field is needed to produce e^+e^- pair from a single event. In turn, 2.3×10^{29} watts/cm^2 of laser power is required (called the "Schwinger Intensity")

- However, Gerard Mourou et al. have suggested numerous mechanisms whereby harmonics might be observed at much lower laser powers – perhaps ~10^{27} watts/cm^2 and possibly 10^{26} watts/cm^2.

- This is seemingly within reach at the ELI facility by means of combining many lasers.

- Engineering challenges of delivering all that energy within the necessary point in time and space are truly formidable!

For Reference: List of Attendees, 1964 Gordon Research Conference



GORDON RESEARCH CONFERENCES, KIMBALL UNION ACADEMY, MERIDEN, N.H.

NON-LINEAR OPTICS
31 August-- 4 September, 1964.

PHOTO IDENTIFICATION

1. Richard Phillips
2. J. V. Martinez
3. Michael Musgrave
4. James McKenna
5. Philip Platzman
6. Marvin C. Tobin
7. Richard Brewer
8. A. E. Siegman
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10. Y. R. Shen
11. David A. Sealer
12. Fred Quelle
13. Peter Kafalas
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16. John Wahr
17. Richard Schlecht
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59. Fielding Brown
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66. John G. Atwood
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69. A. Ashkin
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71. Peter Franken
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73. Boris Stoicheff
74. R. W. Terhune, Vice Chairmab
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76. Elsa Garmire
77. Gisele Eckhardt
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79. John Ward
80. R. Glen Kepler
81. J. Ducving
82. J. A. Armstrong
83. R. Chiao