

phases of the q pump modes. α_s is the single pass power loss of the signal modes, and all idler modes are assumed to have the same single pass loss α_i . κ is the parametric coupling constant, ν_s is the signal frequency, and ν_i is the mean frequency of the idler modes.

For calculating threshold of the system we assume that the relative mode phases ($\varphi_{pq} - \varphi_{iq} - \varphi_s$) are all equal to 90° . We note that although the phases of the pump modes are fixed, the phases of the idler modes are free to adjust to achieve this condition. Assuming all modes to vary as e^{st} and evaluating the resulting $(q + 1)$ th order determinant, we find the threshold condition for parametric oscillation to be given by

$$\sum_q E_{pq}^2 > \frac{\alpha_s \alpha_i}{\nu_s \nu_i \kappa^2}. \quad (2)$$

This perhaps surprising result states that the details of the distribution of pump power between the q pump modes are irrelevant to threshold. In particular, the multimode system described here will have an identical threshold to that of a single pump mode coupled to single idler and signal modes.¹ The result further implies that even if the pump modes are competing and have changing relative amplitudes, if the total pump power is constant, the signal power will be constant. In a system of this type it will probably be advantageous to phase-lock the pump laser, thus insuring that the pump modes are actually equally spaced in frequency and eliminating the effect of atomic mode pulling.

ACKNOWLEDGMENT

The author gratefully acknowledges helpful discussions with M. K. Oshman and A. E. Siegman.

S. E. HARRIS
Dept. of Elec. Engrg.
Stanford University
Stanford, Calif.

¹ R. H. Kingston, "Parametric amplification and oscillation at optical frequencies," *Proc. IRE*, vol. 50, p. 472, April 1962.

Electrooptic Effect in Trigonal Selenium at 10.6 μm

Recent interest in the nonlinear optical behavior of selenium in the infrared [1], [2] has led us to consider its electrooptic properties in this region. We have observed the electrooptic effect in crystalline selenium (32 point-group symmetry) for 10.6 μm radiation, and measured its value to be $r_{11} \sim 2.5 \times 10^{-10}$ cm/V. Trigonal selenium, which is a member of group VIB of the periodic table, is an elemental semiconductor with a bandgap at ~ 8000 Å. It is uniaxial and piezoelectric, and appears to be the first elemental crystal in which the linear electrooptic effect has been measured. As expected from its large index of refraction [3], the electrooptic coefficient of selenium was found to be relatively high (considering class 32 crystals, it is the highest observed to date). Our measurements are in qualitative agreement with Miller's phenomenological theory of second harmonic generation and electrooptic effect [4].

The selenium crystal used in these experiments was cut from a 6 cm long by 1 cm diameter boule which was grown from the melt at high pressures at the Westinghouse Research Laboratory [5]. The sample, a rectangular bar 4 mm by 8.5 mm by 6 mm in the a , x_2 , and c directions, respectively, had silver-gold microalloyed contacts

evaporated on the polished a faces. The dark, dc resistivity of the sample at room temperature was 0.3×10^6 $\Omega\text{-cm}$ when measured across the evaporated electrodes [6]. Voltage pulses of 4 kV magnitude and 1-3 μs duration, obtained from a Velonex Model 350 High Power Pulse Generator, were applied across the contacts, producing an electric field of 10 kV/cm in the sample. The pulse repetition rate was 300 Hz. In performing an experiment, approximately one watt of linearly polarized 10.6 μm radiation, from a cw CO_2 laser, was perpendicularly incident on the polished c face of the Se crystal. The electric field vector was at an angle of 45° to the a $\langle 11\bar{2}0 \rangle$ direction. The emerging radiation was then passed through a Perkin-Elmer wire-grid infrared polarizer in the uncrossed position, and onto a Ge: Cu detector. To measure the electrooptic coefficient r_{11} , the polarizer was rotated until a null was observed, at which point the electric field was applied to the crystal, and the response of the detector compared with that obtained when the polarizer was in the uncrossed position. Because pulses as short as 1 μs were used, the measurement above represents the primary or "zero-strain" electrooptic effect [7].

With the experimental configuration described above, the retardation Γ (in radians) is given by [8] $\Gamma = 2\pi d n_0^3 r_{11} E_0 / \lambda$ where d is the length of the light path in the sample, n_0 is the index of refraction of the ordinary ray, r_{11} is the electrooptic coefficient, E_0 is the applied electric field, and λ is the free-space wavelength of the incident radiation. With the sample placed between crossed polarizers, the retardation Γ is determined by a measurement of the ratio I/I_0 , where I is the shuttered intensity when the electric field is applied, and I_0 is the intensity of the radiation arriving at the detector through the uncrossed polarizer. From these experiments, the electrooptic coefficient for Se was determined to be $r_{11} \sim 2.5 \times 10^{-10}$ cm/V. This value is considered reliable only to within the factor of 2, owing largely to the low-angle boundaries in the crystalline material [5]. Second harmonic generation (SHG) data for selenium has suffered from a similar lack of accuracy [2], since it has been difficult to obtain good crystals of selenium heretofore. New methods of selenium crystal growth, which have recently been reported, however, promise improved materials in the future [9].

The observed electrooptic coefficient is in agreement with Miller's phenomenological theory of optical harmonic generation, optical rectification, and linear electrooptic effect. Thus the tensor element discussed by Miller, calculated from the measured electrooptic coefficient r_{11} , has a value $\delta_{11}^o(\text{Se}) / (4\pi)^3 \sim 0.1 \times 10^{-9}$ esu, which is comparable in size with the value for quartz which is $\delta_{11}^o(\text{quartz}) / (4\pi)^3 \simeq 0.25 \times 10^{-9}$ esu (quartz is also a piezoelectric crystal of 32 point-group symmetry), as well as with the values for other crystals measured to date [4]. Qualitative agreement of the kind discussed by Miller is also seen between the SHG tensor element for Se which is given by Patel [2] as $\delta_{11}^{2\omega}(\text{Se}) / (4\pi)^3 \sim 0.5 \times 10^{-9}$ esu, and the value of $\delta_{11}^o(\text{Se}) / (4\pi)^3$ given above.

In spite of its high electrooptic coefficient, the following factors prevent selenium from being an efficient electrooptic modulator in the infrared. It has a low transmission (the transmission of the 6 mm sample employed in the above experiments was of the order of a few percent), and a low melting point (220°C), so that moderate laser powers (of several watts) cause the sample to melt through absorption. Furthermore, the material is uniaxial, exhibiting a very large birefringence ($n_o = 2.78 \pm .02$; $n_e = 3.58 \pm .02$) [3]. This gives rise to a large background retardation for light travelling nonparallel to the optic axis, and thus makes crystal alignment difficult (if low leakage intensity is desired), as well as seriously limiting angular apertures. With the experimental configuration used, a measurement of r_{41} (the only other nonzero electrooptic coefficient for crystals of class 32), although possible, would be technically difficult since it would require accurate measurements with the incident radiation impinging on the crystal at an angle. As with quartz [4], however, r_{41} is expected to be of the same order of magnitude as r_{11} .

In conclusion, a relatively strong electrooptic effect has been found for crystalline selenium. The electrooptic coefficient r_{11} (Se) $\sim 2.5 \times 10^{-10}$ cm/V is to be compared with the coefficient for quartz [8], r_{11} (quartz) = 0.47×10^{-10} cm/V, and the coefficient for GaAs [10], [11], r_{41} (GaAs) = 1.6×10^{-10} cm/V. The parameter n_{3p}^2 (which is an indicator of the modulation efficiency) is comparable for Se and for GaAs. Because of the low resistivity and low transmission of available Se crystals, GaAs appears to be a more practical material from a device point of view. It should be noted that since selenium has a high nonlinear optical coefficient [2], the electrooptic effect (change in refractive index with applied electric field) may be useful in the tuning of an infrared parametric oscillator.

ACKNOWLEDGMENT

We should like to thank A. J. Strauss for information regarding the selenium crystal and F. D. Carroll for capable technical assistance. We are grateful to R. H. Kingston for valuable discussions.

M. C. TEICH

T. KAPLAN

M.I.T. Lincoln Laboratory¹
Lexington, Mass.

REFERENCES

- [1] C. K. N. Patel, "Efficient phase-matched harmonic generation in tellurium with a CO₂ laser at 10.6 μ ," *Phys. Rev. Letters*, vol. 15, pp. 1027-1030, December 1965.
- [2] —, "Optical harmonic generation in the infrared using a CO₂ laser," *Phys. Rev. Letters*, vol. 16, pp. 613-616, April 1966.
- [3] R. S. Caldwell and H. Y. Fan, "Optical properties of tellurium and selenium," *Phys. Rev.*, vol. 114, pp. 664-675, May 1959. Note that the value of n_e for selenium is misprinted in reference 2.
- [4] R. C. Miller, "Optical second harmonic generation in piezoelectric crystals," *Appl. Phys. Letters*, vol. 5, pp. 17-19, July 1964.
- [5] D. E. Harrison and W. A. Tiller, "Growth of large single crystals of hexagonal selenium from the melt at high pressures," *J. Appl. Phys.*, vol. 36, pp. 1680-1683, May 1965.
- [6] The resistivity of selenium is a strong function of several variables including temperature, incident radiation flux, and the magnitude and direction of the applied electric field. See H. Gobrecht and H. Hamisch, "Zur Feldstärkabhängigkeit der elektrischen Leitfähigkeit von Seleneikristallen," *Z. Physik*, vol. 148, pp. 218-232, 1957. Also, H. W. Henkels, "Electrical properties of selenium: I. Single crystals," *J. Appl. Phys.*, vol. 22, pp. 916-925, July 1951.
- [7] R. O. Carpenter, "The electro-optic effect in uniaxial crystals of the dihydrogen phosphate type. III. Measurement of coefficients," *J. Opt. Soc. Am.*, vol. 40, pp. 225-229, April 1950.

¹ Operated with support from the U. S. Air Force.

- [8] T. R. Sliker, "Linear electro-optic effects in class 32, 6, 3m, and 43m crystals," *J. Opt. Soc. Am.*, vol. 54, pp. 1348-1351, November 1964.
- [9] R. C. Keezer, C. Wood, and J. W. Moody, "The effect of impurities on the single crystal growth of hexagonal selenium," paper M-III-3 presented at 1966 Internat'l Conf. on Crystal Growth, *J. Phys. Chem. Solids*, to be published.
- [10] A. Yariv, C. A. Mead, and J. V. Parker, "GaAs as an infrared electrooptic modulation material," *IEEE J. Quantum Electronics*, vol. QE-2, pp. 243-245, August 1966.
- [11] L. Ho and C. F. Buhrer, "Electro-optic effect of gallium arsenide," *Appl. Optics*, vol. 2, pp. 647-648, June 1963.

Notes and Lines

New Laser Wavelengths in Krypton

We have observed two new laser wavelengths in a pulsed krypton discharge. The laser was 5 mm diameter by 1 meter long, and employed a hot oxide cathode. Pulse currents of 500-1000 amperes were obtained by capacitive discharge. The measured wavelengths were $6072 \pm 1 \text{ \AA}$ and $6417 \pm 1 \text{ \AA}$. We have assigned the 6417 line to the 6416.61 \AA [$5p' \ ^2P_{3/2}^0 \rightarrow 4d \ ^2P_{3/2}$] transition in krypton II. There is no electric dipole transition between tabulated energy levels in krypton that falls within our experimental error for the 6072 \AA line.

T. H. E. COTTRELL
D. C. SINCLAIR
J. M. FORSYTH
Institute of Optics
University of Rochester
Rochester, N. Y.

Manuscript received July 19, 1966. This work was supported by the Night Vision Laboratory, Ft. Belvoir, Va.