OPTICAL RECTIFICATION*

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We wish to report the observation of a dc polarization accompanying the passage of an intense ruby laser beam through certain crystals. This nonlinear optical effect is particularly interesting because it is the first which can be predicted quantitatively from the crystals' known optical properties.[1]

For a qualitative understanding of this effect consider a crystal whose symmetry is sufficiently low to provide a preferred internal direction, so that the polarization induced by an applied electric field need not reverse exactly when the field is reversed. If the applied electric field varies sinusoidally in time, then a net time average polarization will be developed, in analogy with the dc currents in ordinary electronic rectifiers.

Available optical electric field strengths, even with laser sources, are small compared with characteristic crystalline fields. This suggests an expansion of the total polarization in powers of the field. It can be readily verified that the quadratic term is the lowest which can yield a dc contribution. The most general quadratic relation between this dc polarization, p^0, and the amplitude E^ω of the applied optical electric field at frequency ω has the form

$$p_i^0 = \sum_{jk} \chi_{ijk}^0 E_j^\omega E_k^\omega, \qquad (1)$$

where χ_{ijk}^0 are coefficients characteristic of the particular crystal and the superscripts serve to indicate the relevant frequencies. These coefficients are subject to restrictive relations which follow from the point symmetry of the crystal. In particular, they must all vanish for crystals which possess a center of inversion.

A formal description similar to Eq. (1) may also be applied to the well-known linear electro-optic effect in which the optical polarizability of certain crystals is modified by the application of a strong electric field at "zero" frequency:

$$p_i^\omega = \sum_{jk} \chi_{ijk}^\omega E_j^0 E_k^\omega. \qquad (2)$$

In this expression p_i^ω denotes the change in the optical polarization in the i direction due to the components E_j^0 of the applied dc field. From a quantum mechanical formulation Armstrong et al.[1] have predicted a quantitative relationship between the coefficients descriptive of these two effects:

$$\chi_{ijk}^0 = \chi_{jik}^\omega. \qquad (3)$$

The present experiments were performed with potassium dihydrogen phosphate (KDP) and potassium dideuterium phosphate (KD_dP) because these crystals exhibit a large linear electro-optic effect, the experimental data for which[2] may be used with the indices of refraction and Eq. (3) to predict[3] the magnitude of the dc-effect coefficients χ_{ijk}^0. These are listed in Table I.

A high-power ruby laser[4] provided a linearly polarized (6943Å) light beam of one megawatt intensity in pulses of approximately 10^{-7} second duration. The crystals of KDP and KD_dP were prepared in rectangular blocks of approximately 1-cm dimensions with highly polished surfaces. The z axis (optic axis) was normal to two faces and the x and y axes intersected the other faces at 45°. In the experiments the crystals were mounted between two polished brass plates per-

Table I. dc effect coefficients predicted for potassium dihydrogen phosphate (KDP) and potassium dideuterium phosphate (KD_dP) from the available electro-optic data.[a]

(KDP)	$\alpha_H = (\chi_{xyz}^0 + \chi_{xzy}^0) = (\chi_{yxz}^0 + \chi_{yzx}^0) = -11 \times 10^{-8}$ esu
(KDP)	$\beta_H = (\chi_{zxy}^0 + \chi_{zyx}^0) = 13 \times 10^{-8}$ esu
(KD_dP)	$\alpha_D = (\chi_{xyz}^0 + \chi_{xzy}^0) = (\chi_{yxz}^0 + \chi_{yzx}^0) =$ (no data available)
(KD_dP)	$\beta_D = (\chi_{zxy}^0 + \chi_{zyx}^0) = 28 \times 10^{-8}$ esu

[a]See reference 2.

pendicular to the z axis and separated from the crystal by a gap of ~0.002 inch. This gap and the high polish of the plates and crystal minimized spurious effects due to scattered light and consequent pyroelectric phenomena. The laser beam was transmitted through the crystal normal to one of the faces which contains the optic axis.

The crystal and plate assembly formed a capacitor across which potentials appeared due to the laser-induced dc polarization in the crystal. This capacitor was coupled to a cathode follower, whose output was amplified with a wide-band system and subsequently displayed on an oscilloscope. The laser-beam intensity was monitored by a fast photocell after its passage through the crystal.

From Eq. (1) and Table I, one can predict the ratio of the observed dc voltage V_{dc} to the laser-beam intensity I in the present geometry:

$$V_{dc}/I = K\beta \sin^2\theta, \quad (4)$$

where θ is the angle between the optical electric field and the crystal z axis, β is either β_H or β_D of Table I, and K is a constant which depends on geometrical factors and circuit parameters.

Figure 1(a) is a photograph of a dual-beam oscilloscope trace in which the upper beam shows the intensity of the laser light and the lower shows the voltage V_{dc} from a KDP crystal. For this trace the angle θ was 90°. Fig. 1(b) was taken under the same circumstances but with the angle θ set at 0°. Attention is called to the change in baseline which occurs at the time of the laser pulse. We believe this to be due to a residual pyroelectric effect because it is independent of the angle θ and has a long decay time. The delay between the upper and lower traces is due to a 3×10^{-8}-second delay in the amplifier system. The amplitude of the dc effect shown in Fig. 1(a) is 200 microvolts. Signals in excess of one millivolt have been achieved in crystals large enough to receive the full laser beam.

In Fig. 2 the observed variation of the quantity V_{dc}/I with respect to θ is shown and compared with the function $\sin^2\theta$ predicted by Eq. (4).

We have determined the ratio of the β coefficients in KD_dP and KDP to be

$$\beta_D/\beta_H = 2.1 \pm 0.3,$$

which is to be compared with the ratio of 2.2 predicted from the data of Table I. The absolute measured value of β_H is estimated to be 5×10^{-8} esu with an uncertainty of a factor of 3 due primarily to the difficulties in obtaining an absolute calorimetric calibration of the laser intensity.

Further theoretical studies[3] indicate that there may be crystals for which the $\chi_{ijk}{}^0$ coefficients are considerably larger than for KDP. In particular, one is hopeful about crystals which ex-

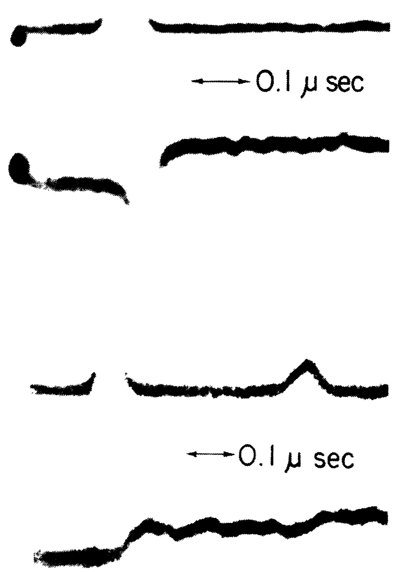

FIG. 1. Photographs of a dual-beam oscilloscope trace in which the upper beam shows the intensity of the laser light and the lower shows the signal from a KDP crystal; time increasing to the right. Fig. 1(a), $\theta = 90°$; Fig. 1(b), $\theta = 0°$.

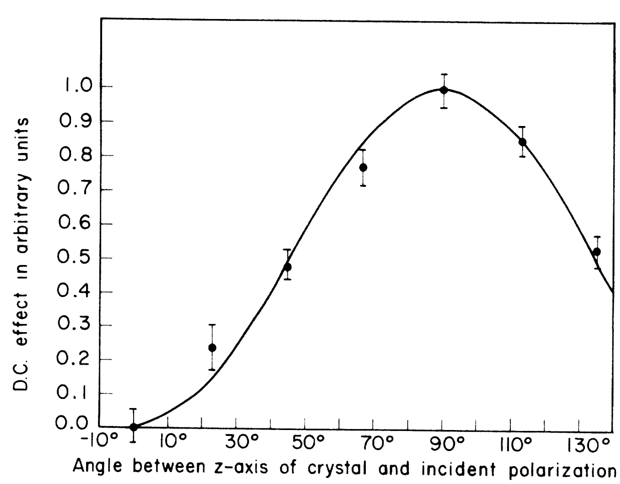

FIG. 2. The variation with angle θ of the measured ratio of dc voltage to red-light intensity, normalized at $\theta = 90°$. The solid curve is the function $\sin^2\theta$.

hibit absorption bands far closer to the laser frequency. We have performed some initial experiments with CdS which are inconclusive at the present time due to the large pyroelectric and photoconductive effects in this material.

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[1]J. A. Armstrong, N. Bloembergen, J. Ducuing, and P. S. Pershan, Phys. Rev. 127, 1918 (1962).

[2]American Institute of Physics Handbook. These data are for the "unclamped" electro-optic coefficients. The "clamped" coefficients are relevant to the present experiments due to the short pulse duration and may be as much as 10% smaller. H. Jaffe (private communication).

[3]P. A. Franken and J. F. Ward (to be published).

[4]Trion Instruments Model No. LS-2 with MH1 OPTUL attachment.

EVIDENCE FOR THE NEGATIVE SURFACE ENERGY MODELS OF SUPERCONDUCTIVITY IN Nb_3Sn, Nb_3Al, V_3Ga, AND V_3Si

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Large specimens (diameter greater than about 1 mm) of natural high-field superconductors,[1,2] such as Nb_3Sn, and synthetic high-field superconductors[3] (interconnected 30Å mercury filaments imbedded in Vycor glass) exhibit nearly identical magnetization loops. These loops, shown in somewhat idealized form in Fig. 1, have the following distinguishing features:

(1) The applied magnetic field H_a penetrates to the center of the specimen when it reaches the magnitude $4\pi J_I R/10$, where J_I is the superconducting current density in A/cm^2 induced to flow within the bulk of the specimen by the applied magnetic field, and R is the radius (cm) of the cylindrical specimen. In Fig. 1, J_I is assumed to be a constant independent of magnetic field up to the critical field.

(2) For applied magnetic fields $>4\pi J_I R/10$, but less than the critical field, the magnetization is given by

$$-4\pi M = 4\pi J_I R/30. \quad (1)$$

(3) The magnetization loop is symmetric about the horizontal axis.

It is interesting to note parenthetically that these features imply the existence of concentric superconducting current-carrying loops which are activated by the applied magnetic field.

If high-field superconductivity in the natural high-field superconductor originates solely from the presence of physically real superconducting filaments, as is the case for the synthetic high-field superconductor, then the magnetization loops of the natural high-field superconductor must close symmetrically about the horizontal axis with decreasing specimen size in accordance with

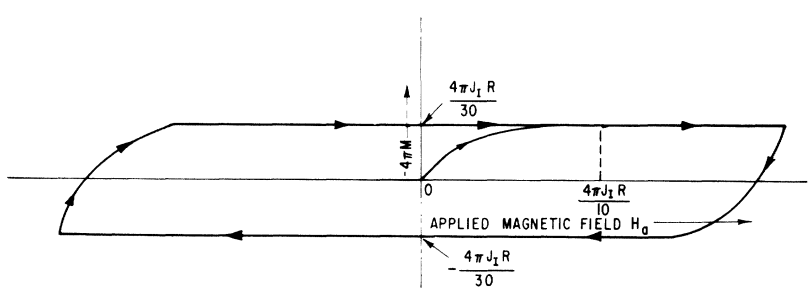
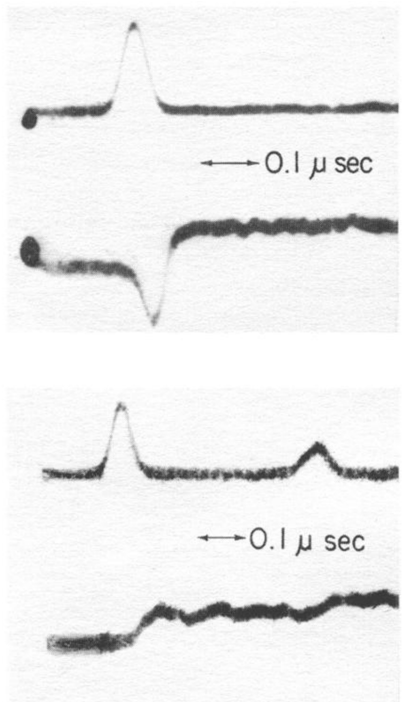


FIG. 1. Magnetization loop of large cylindrical specimens of both natural and synthetic high-field superconductors. J_I is the superconducting current density in A/cm^2 induced to flow within the bulk of the specimen by the applied magnetic field, and R is the radius (cm).



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