requires a field of 4.8×10^5 G according to (9). Fields of this strength are presently unavailable. Therefore, an increase in accuracy of measurement is required to make the effect observable. For transition metals with their high paramagnetic susceptibilities we expect a much larger effect, however. Owing to their nonspherical Fermi surfaces, the effect will depend on crystal orientation.

As opposed to the other effects calculated previously,[1] Eq. (7) shows that the effect considered here is independent of the direction of propagation of sound wave.

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Structure of Nonlinear Optical Phenomena in Potassium Dihydrogen Phosphate*

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Previously available data on nonlinear optical phenomena in potassium dihydrogen phosphate (KDP) are interpreted together with new data on the linear electrooptic effect. It is shown that second harmonic generation is dominated by energy levels in the ultraviolet ("electronic" levels) whereas the dc and linear electrooptic effects may have contributions due to processes simultaneously dependent on ultraviolet and infrared ("ionic") levels. The contribution of this "electronic–ionic" process to the dc and linear electrooptic effects is less than 50% and may indeed be negligible. The close relationship between the dc and linear electrooptic effects is reexamined and shown to be in better agreement with experiment than previously reported. The limitations and implications of Kleinman's symmetry condition are discussed in the light of recent experimental data.

I. INTRODUCTION

POTASSIUM dihydrogen phosphate (KDP) has been carefully studied in recent experiments with optical second harmonic generation[1,2] and optical rectification (the dc effect).[3] In this work laser sources were required because of the relative minuteness of these nonlinear phenomena. The linear electrooptic effect (or Pockels) effect has also been measured in KDP,[4] by techniques which utilize conventional light sources. The magnitudes of all of these phenomena in KDP are amongst the largest observed with any crystal.

The purpose of the present paper is to interpret the previously available data on nonlinear optical phenomena in KDP together with new data on the wavelength dependence of the linear electrooptic effect. It is shown that the second harmonic effect is dominated by energy levels in the ultraviolet ("electronic" levels) whereas the dc and the linear electrooptic effects may have contributions due to processes simultaneously dependent on ultraviolet and infrared ("ionic") levels. Uncertainties in the experimental data make it possible to say only that the contribution of this "electronic-ionic" process to the dc and linear electrooptic effects is less than 50% and may indeed be negligible. The remainder is due to the "electronic-electronic" process which dominates the second harmonic effect.

An important relationship between the dc and linear electrooptic effects was recognized by Armstrong *et al.*[5] in one of the first detailed theoretical discussions of nonlinear optical phenomena. In the present paper we reexamine this relationship and find that there is even better agreement between theory and experiment than was previously reported.[3] Finally, a symmetry condition first proposed by Kleinman[6] is discussed in terms of the experimental data and the present analysis.

II. SUMMARY OF EXPERIMENTAL DATA

Three phenomena are examined in the present paper.

(1) The generation of optical second harmonic.

(2) The dc effect, which is the production of a steady polarization in the crystal by the action of an intense optical electric field.

(3) The linear electrooptic effect, which is the modification of the refractive indices by a low-frequency (or dc) electric field.

Each of these effects represents an extra polarization produced in the crystal which is described by a third-rank tensor operating on a bilinear function of the electric field amplitudes. The formulation is summarized in Table I.

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

[1] A. Ashkin, G. D. Boyd, and J. M. Dziedzic, Phys. Rev. Letters **11**, 14 (1963).

[2] R. C. Miller, Phys. Rev. **131**, 95 (1963).

[3] M. Bass, P. A. Franken, J. F. Ward, and G. Weinreich, Phys. Rev. Letters **9**, 446 (1962).

[4] R. O'B. Carpenter, J. Am. Opt. Soc. **40**, 225 (1950).

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

[6] D. A. Kleinman, Phys. Rev. **126**, 1977 (1962).

The most general (3×3×3) third-rank tensor has 27 elements. In the present case the KDP crystal symmetry (V_d) reduces the number of independent, nonzero elements to three:

$$X_{xyz}=X_{yxz};\quad X_{yzx}=X_{xzy};\quad X_{zxy}=X_{zyx}. \tag{1}$$

Table II shows the combinations of coefficients which may be measured together with their equivalents when Eq. (1) is used to exclude those having a noncyclic arrangement of indices. A second harmonic or dc experiment necessarily measures the sum of two coefficients because the interchange of the indices j and k, referring to fields at the same frequency, is not physically significant. It is evident that there are only two independent measurable quantities for each of these effects. The same arguments do not apply to the linear electrooptic effect and it might appear that, by suitable choice of geometry, three independent measurements may be made. However, the quantum mechanical formulation of Sec. III (or a thermodynamic argument) shows that $X_{xyz}{}^{\omega}\equiv X_{zxy}{}^{\omega}$, and therefore there are only two independent, measurable quantities.

1. Second Harmonic Data

Ashkin *et al.*[1] have recently measured one of the second harmonic coefficients in KDP using a cw gas-laser source at 11 526 Å.

$$\tfrac{1}{2}|d_{36}|=|X_{zxy}{}^{2\omega}|=(3\pm1)\times10^{-9}\text{ esu}. \tag{2}$$

This determination is an order of magnitude more precise than previous measurements,[7] which have utilized pulsed ruby laser sources. Since this coefficient is not expected to vary more than a few percent between 12 000 and 7000 Å we adopt the Ashkin *et al.* value throughout this region.

Miller[2] has measured the ratio of the two independent elements of the second harmonic tensor (ruby laser as fundamental) and finds them to be equal within 5%:

$$d_{14}/d_{36}-1=[(X_{xyz}{}^{2\omega}+X_{yzx}{}^{2\omega})/(2X_{zxy}{}^{2\omega})]-1\leqslant0.05. \tag{3}$$

2. dc Effect

Bass *et al.*[3] have measured the magnitude of one of the dc coefficients in KDP, with an accuracy of a factor of three

$$|d_{36}{}^{0}|=|2X_{zxy}{}^{0}|=5\times10^{-8}\text{ esu}. \tag{4}$$

3. Linear Electrooptic Effect

Carpenter[4] has measured the magnitude of the linear electrooptic effect coefficient at 5560 Å:

$$\begin{aligned} r_{63}&=(4\pi/n_0{}^4)X_{yzx}{}^{\omega}=(3.15\pm0.07)\times10^{-7}\text{ esu},\\ r_{41}&=(4\pi/n_0{}^2n_e{}^2)X_{xyz}{}^{\omega}=(2.58\pm0.05)\times10^{-7}\text{ esu}, \end{aligned} \tag{5}$$

where n_0 and n_e are the ordinary and extraordinary refractive indices, respectively. The relationship between the observed quantities r and the tensor elements X^{ω} is treated in Appendix I. Whereas the sign of either coefficient is a matter of convention, the sign of the ratio is of physical significance and has been tentatively established as positive in recent experiments by Billings and Ploss.[8]

TABLE I. Equations descriptive of the effects discussed in this paper. The X are third-rank tensors and the convention of summation over repeated indices is adopted. The subscripts refer to Cartesian vector components and the superscripts serve to indicate relevant frequencies.

Effect	Descriptive equation
Second harmonic production	$p_i{}^{2\omega}=X_{ijk}{}^{2\omega}E_j{}^{\omega}E_k{}^{\omega}$
dc effect	$p_i{}^{0}=X_{ijk}{}^{0}E_j{}^{\omega}E_k{}^{\omega}$
Linear electrooptic effect	$p_i{}^{\omega}=X_{ijk}{}^{\omega}E_j{}^{0}E_k{}^{\omega}$

In the present paper we present new experimental data on the linear electrooptic effect in the form of the variation with wavelength λ of the voltage $V_{\lambda/2}$ required to produce halfwave retardation in the crystal. The relationship between these quantities and the tensor elements X^{ω} is, as shown in Appendix A

$$(\lambda/V_{\lambda/2})=(8\pi/n_0)X_{yzx}{}^{\omega}. \tag{6}$$

The data are shown in Fig. 1, and the experimental details are given in Appendix B.

III. QUANTUM MECHANICAL EXPRESSIONS FOR THE TENSOR ELEMENTS

The first quantum mechanical treatment of these nonlinear optical processes was developed by Armstrong *et al.*[5] This work is reviewed in Ref. 7, where the approach of the present paper is developed. The assumptions of the theory are that long-range effects, or correlation phenomena between distant lattice cells, can be ignored and that electric dipole approximation to the electromagnetic interaction is valid. The subscript g is used to denote the ground state of the particular lattice cell under study, and the subscripts n and n' are reserved

TABLE II. Measurable combinations of coefficients together with their equivalents when Eq. (1) is used to exclude those having a noncyclic arrangement of indices.

Measurable combinations of coefficients	Equivalent
$\frac{1}{2}(X_{zxy}{}^{2\omega}+X_{zyx}{}^{2\omega})$	$X_{zxy}{}^{2\omega}$
$\frac{1}{2}(X_{xyz}{}^{2\omega}+X_{xzy}{}^{2\omega})$ or $\frac{1}{2}(X_{yzx}{}^{2\omega}+X_{yxz}{}^{2\omega})$	$\frac{1}{2}(X_{xyz}{}^{2\omega}+X_{yzx}{}^{2\omega})$
$\frac{1}{2}(X_{zxy}{}^{0}+X_{zyx}{}^{0})$	$X_{zxy}{}^{0}$
$\frac{1}{2}(X_{xyz}{}^{0}+X_{xzy}{}^{0})$ or $\frac{1}{2}(X_{yzx}{}^{0}+X_{yxz}{}^{0})$	$\frac{1}{2}(X_{xyz}{}^{0}+X_{yzx}{}^{0})$
$(X_{xzy}{}^{\omega}\cdot X_{yzx}{}^{\omega})^{1/2}$	$X_{yzx}{}^{\omega}$
$X_{yxz}{}^{\omega}$ or $X_{zxy}{}^{\omega}$ or $X_{zyx}{}^{\omega}$ or $X_{xyz}{}^{\omega}$	$X_{xyz}{}^{\omega}$ or $X_{zxy}{}^{\omega}$

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[7] For a review of nonlinear optical phenomena and references see P. A. Franken and J. F. Ward, Rev. Mod. Phys. **35**, 23 (1963).

[8] B. H. Billings and R. Ploss (to be published). We are grateful to these authors for their communication of this result prior to publication.

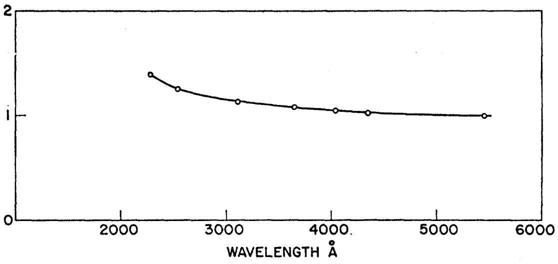

FIG. 1. Experimental wavelength dependence of $(\lambda/V_{\lambda/2})$ normalized to 1 at $\lambda=5461$ Å.

for excited states. The second harmonic tensor elements are:

$$\begin{aligned}
&\tfrac{1}{2}(X_{ijk}{}^{2\omega}+X_{ikj}{}^{2\omega})\\
&\quad=e^3/4\hbar^2S^{2\omega}\sum_{nn'}\{[\langle r_j\rangle_{ng}\langle r_k\rangle_{n'g}\langle r_i\rangle_{nn'}\\
&\qquad+\langle r_k\rangle_{ng}\langle r_j\rangle_{n'g}\langle r_i\rangle_{nn'}]A_{nn'}\\
&\qquad+[\langle r_i\rangle_{ng}\langle r_j\rangle_{n'g}\langle r_k\rangle_{nn'}+\langle r_i\rangle_{ng}\langle r_k\rangle_{n'g}\langle r_j\rangle_{nn'}]B_{nn'}\\
&\qquad+[\langle r_j\rangle_{ng}\langle r_i\rangle_{n'g}\langle r_k\rangle_{nn'}+\langle r_k\rangle_{ng}\langle r_i\rangle_{n'g}\langle r_j\rangle_{nn'}]B'_{nn'}\}\,,
\end{aligned}\tag{7}$$

where, for example, $\langle r_i\rangle_{nn'}$ is the matrix element of x, y, or z between the states n and n'. The $S^{2\omega}$ is included to relate microscopic and macroscopic field quantities. This factor and similar factors S^0 and S^ω, which serve the same function in the dc and linear electrooptic effect expressions, will be discussed at the end of this section. The frequency-dependent terms $A_{nn'}$, $B_{nn'}$, and $B'_{nn'}$ are:

$$\begin{aligned}
A_{nn'}&=-\frac{1}{2}\left[\frac{1}{(\omega_{ng}+\omega)(\omega_{n'g}-\omega)}+\frac{1}{(\omega_{ng}-\omega)(\omega_{n'g}+\omega)}\right],\\
B_{nn'}&=-\frac{1}{2}\left[\frac{1}{(\omega_{ng}+2\omega)(\omega_{n'g}+\omega)}+\frac{1}{(\omega_{ng}-2\omega)(\omega_{n'g}-\omega)}\right],\\
B'_{nn'}&=-\frac{1}{2}\left[\frac{1}{(\omega_{ng}+\omega)(\omega_{n'g}+2\omega)}+\frac{1}{(\omega_{ng}-\omega)(\omega_{n'g}-2\omega)}\right],
\end{aligned}\tag{8}$$

where, for example, ω_{ng} denotes the difference in energy between the states n and g in units of angular frequency, and ω is the angular frequency of the optical electric field.

The dc tensor elements are:

$$\begin{aligned}
&\tfrac{1}{2}(X_{ijk}{}^{0}+X_{ikj}{}^{0})\\
&\quad=(e^3/4\hbar^2)S^{0}\sum_{nn'}\{[\langle r_j\rangle_{ng}\langle r_k\rangle_{n'g}\langle r_i\rangle_{nn'}\\
&\qquad+\langle r_k\rangle_{ng}\langle r_j\rangle_{n'g}\langle r_i\rangle_{nn'}]C_{nn'}\\
&\qquad+[\langle r_i\rangle_{ng}\langle r_j\rangle_{n'g}\langle r_k\rangle_{nn'}+\langle r_i\rangle_{ng}\langle r_k\rangle_{n'g}\langle r_j\rangle_{nn'}]D_{nn'}\\
&\qquad+[\langle r_j\rangle_{ng}\langle r_i\rangle_{n'g}\langle r_k\rangle_{nn'}+\langle r_k\rangle_{ng}\langle r_i\rangle_{n'g}\langle r_j\rangle_{nn'}]D'_{nn'}\}\,,
\end{aligned}\tag{9}$$

where the frequency dependent terms $C_{nn'}$, $D_{nn'}$, and $D'_{nn'}$ are:

$$\begin{aligned}
C_{nn'}&=\frac{1}{2}\left[\frac{1}{(\omega_{ng}+\omega)(\omega_{n'g}+\omega)}+\frac{1}{(\omega_{ng}-\omega)(\omega_{n'g}-\omega)}\right],\\
D_{nn'}&=\frac{1}{2}\left[\frac{1}{\omega_{ng}(\omega_{n'g}+\omega)}+\frac{1}{\omega_{ng}(\omega_{n'g}-\omega)}\right],\\
D'_{nn'}&=\frac{1}{2}\left[\frac{1}{(\omega_{ng}+\omega)\omega_{n'g}}+\frac{1}{(\omega_{ng}-\omega)\omega_{n'g}}\right].
\end{aligned}\tag{10}$$

The linear electrooptic effect was studied in Ref. 7, following the initial work of Armstrong *et al.*[5] as a limiting case of a difference frequency process. Unfortunately, this method is prone to numerical errors owing to possible ambiguities in the appropriate frequency limit. A more direct technique is to pursue a second-order perturbation treatment in the style of Sec. IV.1, Ref. 7, using a Hamiltonian that contains an optical and dc field explicitly. This calculation yields directly the linear electrooptic tensor elements:

$$\begin{aligned}
&X_{ijk}{}^{\omega}=\tfrac{1}{2}(X_{ijk}{}^{\omega}+X_{kji}{}^{\omega})\\
&\quad=(e^3/\hbar^2)S^{\omega}\sum_{nn'}\{[\langle r_i\rangle_{ng}\langle r_k\rangle_{n'g}\langle r_j\rangle_{nn'}\\
&\qquad+\langle r_k\rangle_{ng}\langle r_i\rangle_{n'g}\langle r_j\rangle_{nn'}]C_{nn'}\\
&\qquad+[\langle r_j\rangle_{ng}\langle r_k\rangle_{n'g}\langle r_i\rangle_{nn'}+\langle r_j\rangle_{ng}\langle r_i\rangle_{n'g}\langle r_k\rangle_{nn'}]D_{nn'}\\
&\qquad+[\langle r_i\rangle_{ng}\langle r_j\rangle_{n'g}\langle r_k\rangle_{nn'}+\langle r_k\rangle_{ng}\langle r_j\rangle_{n'g}\langle r_i\rangle_{nn'}]D'_{nn'}\}\,,
\end{aligned}\tag{11}$$

where the frequency-dependent terms $C_{nn'}$, $D_{nn'}$, and $D'_{nn'}$ are defined in Eq. (10). One should note the symmetry between the first and last suffices in this linear electrooptic tensor, in contrast to the symmetry between the last two suffices of both the dc and second harmonic tensors. This symmetry, together with the crystal symmetry conditions of Eq. (1) establishes the contention of Sec. I that $X_{xyz}\equiv X_{zxy}$.

The factors S which interelate the microscopic and macroscopic analyses are discussed by Armstrong *et al.*[5] If the Lorentz factor[9] is used to relate applied to local electric fields in a medium with dielectric constant ϵ (at frequencies indicated by superscripts) and number density of microscopic systems N, the factors S are:

$$S^{2\omega}=\left(\frac{\epsilon^\omega+2}{3}\right)^2\left(\frac{\epsilon^{2\omega}+2}{3}\right)N,\tag{12}$$

$$S^0=S^\omega=\left(\frac{\epsilon^\omega+2}{3}\right)^2\left(\frac{\epsilon^0+2}{3}\right)N.\tag{13}$$

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[9] H. A. Lorentz, *Theory of Electrons* (B. G. Teubner, Leipzig, 1909), pp. 138, 306.

This analysis has two limitations. Firstly, it only applies to media with isotropic dielectric constants. Secondly, the local field so derived from the applied field is the *average* over the unit cell whereas it is the local field *at the site of the nonlinear interaction which is required.* This difference may be appreciable if the dielectric constant is large.[10] Equation (12) should give a good approximation to $S^{2\omega}$ as $\epsilon^{2\omega}$ and ϵ^{ω} are almost isotropic and small. However, the limitations mentioned above make Eq. (13) a crude estimate for S^0 and S^{ω} as ϵ^0 is both very anisotropic and large ($\epsilon_x{}^0=42$, $\epsilon_z{}^0=21$). For the evaluation of S^{ω} in subsequent sections, we use Eq. (13) with an assumed effective value for ϵ^0:

$$\epsilon^0=(31\pm10). \tag{14}$$

We think it unlikely that the correct effective value lies outside these limits.

IV. RELATIONSHIP BETWEEN THE DC AND LINEAR ELECTROOPTIC EFFECTS

Inspection of Eqs. (9) and (11) yields immediately the following important relationship between the dc and linear electrooptic tensor elements:

$$\tfrac{1}{2}(X_{ijk}{}^0+X_{ikj}{}^0)=\tfrac{1}{4}X_{jik}{}^{\omega}. \tag{15}$$

This is a special case of the general permutation symmetries developed by Armstrong *et al.*[5] Equation (15) permits a quantitative prediction of the magnitude of the dc effect in KDP from the available numerical data on the linear electrooptic effect [Eq. (5)]:

$$(2X_{zxy}{}^0)_{\text{predicted}}=-(n_0{}^4/8\pi)r_{63}=-6.5\times10^{-8}\text{ esu}. \tag{16}$$

This is seen to be in excellent agreement with the experimental magnitude of 5×10^{-8} esu quoted by Bass *et al.*[3] [The predicted value of the dc effect was incorrectly given as 13×10^{-8} esu in both Ref. 3 and in Ref. 7, Eq. (V.4).]

V. INTERRELATION OF THE NONLINEAR EFFECTS

In this section we wish to examine the available experimental data with a view to eliciting the role played by the ultraviolet and infrared bands in KDP. Since the intimate relationship between the dc and linear electrooptic effect shown in Eq. (15) appears to be confirmed by experiment, it remains to relate either effect to the second harmonic effect.

In order to facilitate the discussion we shall now consider that the quantum mechanical expressions for the tensor elements [Eqs. (7), (9), and (11)] are each dominated by a single term in the summation involving a particular pair of energy levels n and n'. This assumption provides considerable simplicity and, we believe, still permits a meaningful examination of the possible processes. For convenience we introduce the dimensionless parameters ρ, ρ' and σ which are defined by the equations:

$$\begin{aligned}\omega_{ng}&=\rho\omega_0,\\ \omega_{n'g}&=\rho'\omega_0,\\ \omega&=\sigma\omega_0\end{aligned} \tag{17}$$

ω_0 corresponds to a wavelength in the ultraviolet, and ω is the applied optical frequency.

We wish to examine three distinct cases, characterized by the energies of the levels n and n'

Case (i)	"electronic-electronic"	$\rho\simeq\rho'\simeq1$,
Case (ii)	"ionic-ionic"	$\rho\simeq\rho'\ll1$,
Case (iii)	"electronic-ionic"	$\rho\simeq1$; $\rho'\ll1$.

By "electronic" and "ionic" we mean energy levels with transition wavelengths shorter and longer, respectively, than the limits of the region of optical transparency. KDP exhibits greater than 50% transmission (2 mm plate) for the wavelength range 2100 to 13 000 Å.

Sensible estimates of ω_0 and the values of the parameters ρ and ρ' for the ionic levels may be based on extrapolations of refractive index data that seek to locate the positions of dominant optical levels. Fitting a one-level Clausius-Mossotti equation to the refractive index data for KDP[11] yields a value for ω_0 corresponding to 870 Å. An extension of the formula to two levels indicates an infrared absorption level on the long wavelength side of 50 000 Å. It is possible that fitting the data to a three-level formula would show that the one level at 870 Å is actually an average of two distinctly spaced levels. In spite of the vagueness of these arguments, useful progress may be made with the plausible assumption that $2\pi c/\omega_0=900\pm200$ Å and that the values of ρ and ρ' for the ionic levels are of order 10^{-2} or less. Since ruby laser radiation was employed in obtaining the experimental data under discussion we set $\sigma=1/8$.

In Table III we list the approximate values, in units

TABLE III. Relative magnitudes of frequency factors in units of $\omega_0{}^{-2}$ for the three cases (i) "electronic-electronic" (ii) "ionic-ionic" (iii) "electronic-ionic." σ, ρ, ρ', are defined in Eq. (17). For ruby laser experiments $\sigma^2\simeq0.015$. The last column gives the magnitudes of the frequency factors for Case (iii) and $\rho'=10^{-2}$.

	Case (i)	Case (ii)	Case (iii)	
$A_{nn'}$	$-1-\sigma^2$	$+\sigma^{-2}$	ρ'/σ^2-1	-0.3
$B_{nn'}$	$-1-7\sigma^2$	$-\frac{1}{2}\sigma^{-2}$	ρ'/σ^2+2	$+2.7$
$B'_{nn'}$	$-1-7\sigma^2$	$-\frac{1}{2}\sigma^{-2}$	$(\frac{1}{4})(\rho'/\sigma^2)+\frac{1}{2}$	$+0.7$
$C_{nn'}$	$+1+3\sigma^2$	$+\sigma^{-2}$	$-\rho'/\sigma^2-1$	-1.7
$D_{nn'}$	$+1+\sigma^2$	$-(\rho'/\rho)\sigma^{-2}$	$-\rho'/\sigma^2$	-0.7
$D'_{nn'}$	$+1+\sigma^2$	$-(\rho/\rho')\sigma^{-2}$	$1/\rho'$	$+100$

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[10] For a general discussion see C. J. F. Böttcher, *Theory of Electric Polarization* (Elsevier Publishing Company, Inc., New York, 1952), Sec. 33. For specific discussion of KDP see J. C. Slater, J. Chem. Phys. **9**, 16 (1941).

[11] *The International Critical Tables* (Maple Press, York, Pennsylvania, 1930), Vol. 7, p. 27 gives refractive index data in the wavelength range 4860–6560 Å. J. L. Dennis and R. H. Kingston (to be published) have recently measured the extraordinary refractive index in the ultraviolet to 2537 Å and we are grateful to these authors for communicating their results prior to publication.

of $1/\omega_0^2$, of the frequency dependent terms $A_{nn'}$, $B_{nn'}$, $B'_{nn'}$, $C_{nn'}$, $D_{nn'}$, and $D'_{nn'}$ for each of the three cases. The $A_{nn'}$, $B_{nn'}$, and $B'_{nn'}$ terms are appropriate to the second harmonic tensor [Eqs. (7) and (8)] whereas the $C_{nn'}$, $D_{nn'}$, and $D'_{nn'}$, characterize both the dc and linear electrooptic effects [Eqs. (9), (10), and (11)]. Approximate numerical values for Case (iii) are given in the last column.

The experimental results are now described in this simplified quantum mechanical formulation.

1. Wavelength Dependence of the Linear Electrooptic Effect

From Eqs. (6) and (11) the wavelength dependence of the linear electrooptic effect may be expressed as

$$(\lambda/V_{\lambda/2}) \propto [(n^2+2)^2/n]\{\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'}C_{nn'} + \langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'}D_{nn'} + \langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'}D'_{nn'}\}. \quad (18)$$

Figure 2 shows the calculated wavelength dependence of $c_{nn'}$, $d_{nn'}$, and $d'_{nn'}$, which are $(n^2+2)^2/n$ times $C_{nn'}$, $D_{nn'}$, and $D'_{nn'}$, respectively, for each of the three cases (i), (ii), and (iii) indicated by superscripts. The figure includes the experimental wavelength dependence of $\lambda/V_{\lambda/2}$ taken from Fig. 1. It is clear that *the experimental data do not correspond to a Case (ii) process* but rather seem to be dominated by significant Case (i) and Case (iii) contributions. It is not possible to draw useful conclusions about the relative magnitudes of the Case (i) and Case (iii) contributions as this assignment is critically dependent on the particular choice of value for ω_0.

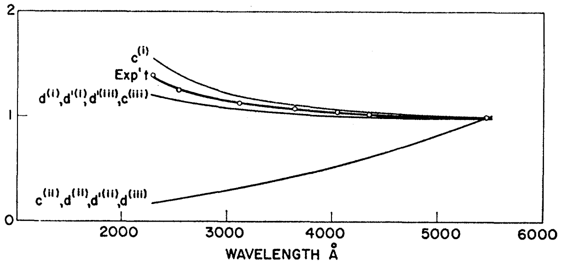

FIG. 2. Calculated wavelength dependence of c, d, and d' which are $(n^2+2)^2/n$ times $C_{nn'}$, $D_{nn'}$, and $D'_{nn'}$ respectively, for each of the three cases (i), (ii), and (iii) indicated by superscripts. Any combinations of these represents a possible wavelength dependence for $\lambda/V_{\lambda/2}$. Experimental data from Fig. 1 are included for comparison. All curves are normalized to 1 at $\lambda = 5461$ Å. (Only $c^{(iii)}$ is sensitive to the choice of value for ρ' and it is given here for $\rho'=0$. For larger ρ', $c^{(iii)}$ is depressed and reached approximately halfway between the two lower curves for $\rho'=10^{-2}$. This does not affect the conclusions drawn in the text.)

2. Ratio of Second Harmonic Coefficients

From Eqs. (3) and (7) the ratio of second harmonic coefficients may be expressed as

$$|(d_{14}/d_{36})-1| \equiv q = |[\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'}(\tfrac{1}{2}B_{nn'}+\tfrac{1}{2}B'_{nn'}-A_{nn'}) + \langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'}(\tfrac{1}{2}A_{nn'}+\tfrac{1}{2}B'_{nn'}-B_{nn'}) + \langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'}(\tfrac{1}{2}A_{nn'}+\tfrac{1}{2}B_{nn'}-B'_{nn'})] / [\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'}A_{nn'} + \langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'}B_{nn'} + \langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'}B'_{nn'}]|. \quad (19)$$

The experimental upper limit for q [Eq. 3] is 0.05. The approximate equality of the coefficients is seen to be satisfied by Case (i) ($A_{nn'} \simeq B_{nn'} \simeq B'_{nn'}$, see Table III) for arbitrary values of the matrix elements. For Cases (ii) and (iii) the approximate equality still holds for suitable magnitudes of the matrix element products. However for Case (ii) ($A_{nn'} = -2B_{nn'} = -2B'_{nn'}$), this requires that $(\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'} + \langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'} - 2\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'}) = 0$ which in turn, makes both coefficients zero in this approximation. As the coefficients in KDP are among the largest found in any crystal, it seems unlikely that they should arise from intrinsically enormous coefficients fortuitously reduced by this situation. We therefore *exclude Case (ii) as a dominant process for the second harmonic effect.* Case (iii) is not similarly excluded.

3. Ratio of the Linear Electrooptic Coefficients

From Eqs. (5) and (11) the ratio of the linear electrooptic coefficients may be expressed as

$$(r_{41}/r_{63})(S_{63}^{\omega}/S_{41}^{\omega}) \equiv k = [\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'}\tfrac{1}{2}(D_{nn'}+D'_{nn'}) + \langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'}\tfrac{1}{2}(C_{nn'}+D'_{nn'}) + \langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'}\tfrac{1}{2}(C_{nn'}+D_{nn'})] / [\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'}C_{nn'} + \langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'}D_{nn'} + \langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'}D'_{nn'}], \quad k = \frac{2.6\times10^{-7}}{3.2\times10^{-7}}\frac{S_{63}^{\omega}}{S_{41}^{\omega}}. \quad (20)$$

The factors S^{ω} are not necessarily identical in the anisotropic crystal but must be approximately so and therefore k must be approximately $+1$. Reference to Table III shows that for Case (i) all six frequency factors appearing in Eq. (20) are approximately equal and therefore the predicted ratio must be $+1$ for this case. Dominance of Case (iii) could predict essentially any ratio including ±1. Therefore the importance of the recent experimental determination of the sign of the ratio as positive (see Sec. IIc) is that it fails to preclude Case (i). The situation is still that *the linear electrooptic effect is dominated by an as yet unspecified mixture of Case (i) and Case (iii) processes.*

4. Ratio of the Linear Electrooptic and Second Harmonic Coefficients

From Eqs. (2), (5), (7), and (11) the ratio of the linear electrooptic and second harmonic coefficients may be expressed as

$$\left|\frac{r_{63}n_0{}^4S^{2\omega}}{d_{36}32\pi S^{\omega}}\right|\equiv m=\left|\frac{\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'}C_{nn'}+\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'}D_{nn'}+\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'}D'_{nn'}}{\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'}A_{nn'}+\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'}B_{nn'}+\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'}B'_{nn'}}\right|,\quad m=1.4\pm0.6. \tag{21}$$

The range includes the experimental errors in measuring d_{36} and the uncertainty in the factor S^{ω}. The range of uncertainty precludes any very definite prediction from this datum alone. We note that Case (i) requires the ratio to be approximately +1. For Case (iii), however, $D'_{nn'}$ is large compared with all other frequency factors. This would result in a larger Case (iii) contribution to the linear electrooptic effect than to the second harmonic effect *unless* there is a "conspiracy" among the matrix elements [e.g., Case (iii) $\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'}\ll\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'}$ or $\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'}$]. The analysis of the next section circumvents the necessity for assumptions about the magnitudes of matrix elements.

5. Relative Magnitudes of Case (i) and Case (iii) Contributions to the Linear Electrooptic and Second Harmonic Effects

The present conclusion is that the linear electrooptic and second harmonic effects may have significant contributions from Case (i) and Case (iii) processes but not from Case (ii) processes. We now investigate the relative contributions of Case (i) and Case (iii) processes to each of the two effects. It is convenient to introduce the parameters α and β:

$\alpha\equiv$[Case (iii) contribution to d_{36}]/[Case (i) contribution to d_{36}]

$$=\frac{A_{nn'}{}^{(iii)}(\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'})^{(iii)}+B_{nn'}{}^{(iii)}(\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(iii)}+B'_{nn'}{}^{(iii)}(\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(iii)}}{A_{nn'}{}^{(i)}(\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'})^{(i)}+B_{nn'}{}^{(i)}(\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(i)}+B'_{nn'}{}^{(i)}(\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(i)}}, \tag{22}$$

$\beta\equiv$[Case (iii) contribution to r_{63}]/[Case (i) contribution to r_{63}]

$$=\frac{C_{nn'}{}^{(iii)}(\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'})^{(iii)}+D_{nn'}{}^{(iii)}(\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(iii)}+D'_{nn'}{}^{(iii)}(\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(iii)}}{C_{nn'}{}^{(i)}(\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z'\rangle_{nn'})^{(i)}+D_{nn'}{}^{(i)}(\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(i)}+D'_{nn'}{}^{(i)}(\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(i)}}. \tag{23}$$

In these equations the two processes are distinguished by the superscripts (*i*) and (*iii*). The frequency factors may be expressed in terms of ρ' and σ from Table III, and the small terms in σ^2 ignored for the Case (i) factors. Equations (22) and (23) can then be rewritten in an algebraic form more convenient for analysis:

$$\begin{aligned}&[\rho'/\sigma^2-1](\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'})^{(iii)}+[\rho'/\sigma^2+2](\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(iii)}+[\rho'/4\sigma^2+1/2](\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(iii)}\\&\qquad+\alpha[(\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'})^{(i)}+(\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(i)}+(\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(i)}]=0,\end{aligned} \tag{22'}$$

$$\begin{aligned}&[\rho'/\sigma^2+1](\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'})^{(iii)}+[\rho'/\sigma^2](\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(iii)}+[-1/\rho'](\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(iii)}\\&\qquad+\beta[(\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'})^{(i)}+(\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(i)}+(\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(i)}]=0.\end{aligned} \tag{23'}$$

Equations (19), (20), and (21) represent the three available experimental data. Manipulations similar to the above yield:

$$\begin{aligned}&[18-3\rho'/\sigma^2+8q-8q\rho'/\sigma^2](\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'})^{(iii)}\\&\quad+[-18-3\rho'/\sigma^2-16q-8q\rho'/\sigma^2](\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(iii)}+[6\rho'/\sigma^2-4q-2q\rho'/\sigma^2](\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(iii)}\\&\qquad+8q[(\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'}{}^{(i)}+(\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(i)}+(\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(i)}]=0,\end{aligned} \tag{19'}$$

$$\begin{aligned}&[\tfrac{1}{2}\rho'-\rho'/2\sigma^2+k+k\rho'/\sigma^2](\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'})^{(iii)}\\&\quad+[\tfrac{1}{2}\rho'-\tfrac{1}{2}-\rho'/2\sigma^2+k\rho'/\sigma^2](\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(iii)}+[-k/\rho'-\tfrac{1}{2}-\rho'/\sigma^2](\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(iii)}\\&\qquad+[-k+1][(\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'})^{(i)}+(\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(i)}+(\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(i)}]=0,\end{aligned} \tag{20'}$$

$$\begin{aligned}&[-1-\rho'/\sigma^2-m+m\rho'/\sigma^2](\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'})^{(iii)}\\&\quad+[-\rho'/\sigma^2+2m+m\rho'/\sigma^2](\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(iii)}+[1/\rho'+m/2+m\rho'/\sigma^2](\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(iii)}\\&\qquad+[1-m][(\langle x\rangle_{ng}\langle y\rangle_{n'g}\langle z\rangle_{nn'})^{(i)}+(\langle z\rangle_{ng}\langle x\rangle_{n'g}\langle y\rangle_{nn'})^{(i)}+(\langle y\rangle_{ng}\langle z\rangle_{n'g}\langle x\rangle_{nn'})^{(i)}]=0.\end{aligned} \tag{21'}$$

In the preceding five equations ρ', σ^2, k, m, q are treated as known parameters. There remain five unknowns: α and β which are of particular interest here and three others, each of which is a Case (i) matrix element triple product divided by the sum of three Case (iii) matrix element triple products. Thus it is possible, although tedious, to solve for α and β. The leading terms of the solution are:

$$\alpha=2\rho'(1-m)-\tfrac{2}{3}q-\rho'(k-1) \tag{24}$$

$$\beta=-(1-m)-\tfrac{2}{3}q-\rho'(k-1). \tag{25}$$

Numerical restrictions on the experimental parameters are:

- $\rho'\leqslant 0.01$ (ratio of infrared to ultraviolet absorption band frequencies)
- $m=$ 0.8 to 2.0 [ratio of electrooptic and second harmonic effects, Eq. (21)]
- $q\leqslant 0.05$ [relative inequality of the second harmonic coefficients, Eq. (19)]
- $k\simeq 1$ [ratio of the two linear electrooptic coefficients, Eq. (20)].

Inspection of Eq. (24) reveals that α cannot be larger than a few percent. Thus the second harmonic effect is dominated by "electronic-electronic" [Case (i)] processes, with no more than a few percent contribution from "electronic-ionic" [Case (iii)] processes.

Inspection of Eq. (25) reveals that β can be as large as 1.0. Thus the Case (iii) contribution to the linear electrooptic effect would lie in the range of 0 to 50%. This assignment could be sharpened considerably with an improvement in the evaluation of the shielding factors S^ω, together with an even more accurate experimental value for d_{36}.

An analysis of the d_{14} and r_{41} coefficients, similar to the analysis of d_{36} and r_{63} given in this section, yields essentially the same conclusions.

6. Kleinman's Symmetry Condition

Kleinman has discussed an additional symmetry restriction on the nonlinear coefficients[6] which should be operative for "electronic" processes in a frequency range where absorption and dispersion are negligible. In particular, for KDP, this symmetry restriction requires that the second harmonic coefficients d_{14} and d_{36} be equal. The analytic part of Eq. (19) demonstrates this equality in the present formalism for Case (i) ("electronic-electronic") processes, even for arbitrary values of the matrix elements, *provided* the frequency factors are equal. This condition is that σ should be small which requires distant absorption bands which, in turn, is equivalent to the condition of negligible absorption and dispersion.

The extent to which Kleinman's symmetry condition is relaxed when dispersion is present (σ not negligible depends both on the relative magnitudes of the matrix elements appearing in Eq. (19), and on which process [Case (i), (ii), or (iii)] is dominant. If the matrix element triple products are equal then the symmetry condition is exactly obeyed for each of the three cases regardless of the value of σ. This insensitivity to dispersion was not appreciated in the earlier discussion of Franken and Ward.[7] In addition, the possible validity of this symmetry condition even for "ionic-ionic" and "ionic-electronic" processes was not recognized in the original discussion by Kleinman.[6] That is, the fact that the two coefficients are nearly equal does not in itself preclude the possibility that "electronic-ionic" [Case (iii)] processes are operative in second harmonic generation. [Case (ii) processes can be excluded when $d_{14}=d_{36}$, as is shown in Sec. V.2.] However, if the matrix element products do in fact differ appreciably then the experimentally verified equality of the coefficients[2] does imply a dominant contribution from Case (i) processes. [This follows directly from inspection of Eq. (19) and Table III.] In the absence of an *a priori* knowledge of the matrix elements we cannot deduce from the experimentally established equality of the coefficients precisely what role is played by ionic processes. The analysis of Sec. V.5, with its cognizance of other data, is required to establish the predominance of "electronic-electronic" processes in second harmonic generation.

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APPENDIX A

Linear Electrooptic Effect

The linear electrooptic effect is usually described[12] in terms of the coefficients r_{ij} defined by

$$\Delta\left(\frac{1}{n^2}\right)_i=\sum_{j=1}^{3} r_{ij}E_j^0, \tag{A1}$$

where i is xx, yy, zz, yz, zx, or xy and j is x, y, or z.

The analysis is in terms of the Fresnel ellipsoid. It is instructive to derive directly the relation between the experimentally measured quantities and the X^ω because attempts at translation between the two descriptions can lead to confusion.

Maxwell's equations for the nonmagnetic insulator are

$$\begin{aligned}\nabla\times\mathbf{H}&=(1/c)\boldsymbol{\varepsilon}\dot{\mathbf{E}}\\ \nabla\times\mathbf{E}&=-(1/c)\dot{\mathbf{H}}\end{aligned} \tag{A2}$$

giving a wave equation for electric field in the crystal

$$\nabla\times\nabla\times\mathbf{E}=-(1/c^2)\boldsymbol{\varepsilon}\ddot{\mathbf{E}} \tag{A3}$$

and for a Fourier component at frequency ω

$$\nabla\times\nabla\times\mathbf{E}_{(r)}{}^{\omega}-(\omega^2/c^2)\boldsymbol{\varepsilon}\mathbf{E}_{(r)}=0. \tag{A4}$$

The solution expressed in terms of a refractive index n and a direction of propagation $\mathbf{s}$, is of the form

$$\mathbf{E}_{(r)}{}^{\omega}=\mathbf{E}^{\omega}\exp[i(\omega/c)n\mathbf{s}\cdot\mathbf{r}]. \tag{A5}$$

An equation determining two values for n and the corresponding eigenvectors $\mathbf{E}$ is obtained by putting this solution back into the wave equation

$$[n^2(1-\mathbf{ss})-\boldsymbol{\varepsilon}]\mathbf{E}^{\omega}=0. \tag{A6}$$

The dielectric tensor as modified by the linear electrooptic effect in KDP is

$$\boldsymbol{\varepsilon}=\begin{bmatrix} n_0{}^2 & 4\pi X_{yzx}E_z{}^0 & 4\pi X_{xyz}E_y{}^0 \\ 4\pi X_{yzx}E_z{}^0 & n_0{}^2 & 4\pi X_{xyz}E_x{}^0 \\ 4\pi X_{xyz}E_y{}^0 & 4\pi X_{xyz}E_x{}^0 & n_e{}^2 \end{bmatrix}. \tag{A7}$$

For the situation of the present experiments and in Carpenter's experiments to measure $r_{63}[E_x{}^0=E_y{}^0=0;$ $E_z{}^0\neq 0;$ $s=(0,0,1)]$, we find that the refractive indices are

$$\begin{aligned} n_+&=n_0+\tfrac{1}{2}(4\pi X_{yzx}{}^{\omega}/n_0)\\ n_-&=n_0-\tfrac{1}{2}(4\pi X_{yzx}{}^{\omega}/n_0) \end{aligned} \tag{A8}$$

and the corresponding eigenvectors of $\mathbf{E}^{\omega}$ are

$$\begin{aligned} \mathbf{E}_+{}^{\omega}&=(1/\sqrt{2})(1,1,0)\,|E^{\omega}|\\ \mathbf{E}_-{}^{\omega}&=(1/\sqrt{2})(-1,1,0)\,|E^{\omega}| \end{aligned} \tag{A9}$$

and by comparison with Carpenter

$$X_{yzx}{}^{\omega}=-r_{63}n_0{}^4/4\pi. \tag{A10}$$

For the present experiment, the KDP crystal is placed between a polarizer and an analyzer both having transmitting axes parallel to (1,1,0). Defining the fractional modulation M as the ratio of the transmission with a potential V_z across the crystal to the transmission when $V_z=0$, and defining the half-wave voltage $V_{\lambda/2}$ as that voltage which produces zero transmission, we have the relations

$$\left(\frac{\lambda}{V_{\lambda/2}}\right)=\frac{2}{\pi}\,\frac{\lambda\cos^{-1}(M)^{1/2}}{V_z}=\frac{8\pi X_{yzx}}{n^0}. \tag{A11}$$

APPENDIX B

Measurement of the Wavelength Dependence of the Linear Electrooptic Effect

The experimental arrangement is shown in Fig. 3. The light source is a microwave excited mercury or cadmium discharge in a quartz tube and the required wavelength is isolated with a small grating monochromator. The light propagates along the z axis of a ~1 mm thick KDP crystal (Baird Atomic) placed between uncrossed polarizers which transmit light polarized parallel to the x axis of the crystal. Gold grids (holes ~1 mm square) deposited on the z faces of the crystal serve as electrodes for the application of a 60 cps electric field to the crystal in the z direction. The angular aperture for the light propagating through the crystal is limited to 1°. The light which is modulated at 120 cps is detected with a photomultiplier and displayed on an oscilloscope.

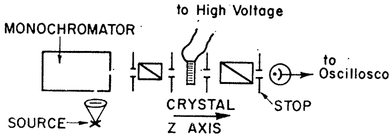

FIG. 3. Experimental arrangement for measuring the wavelength dependence of the linear electrooptic coefficient $r_{63}(X_{yzx})$.

The amplitude of the 60-cps voltage across the crystal which produces a certain convenient depth of modulation (40%) is a measure of the electrooptic coefficient. The required applied voltage at $\lambda=5461$ Å was found to be ~25% larger than that predicted from Carpenter's data. However, we do not claim an *absolute* accuracy better than 25%. Absolute accuracy is limited by the variation of the modulating field in the xy plane and the uncertainty in the degree of clamping introduced by the crystal mount. Neither of these factors affects the determination of the variation of the coefficient with wavelength, which is of particular interest here. In Fig. 1 are plotted experimental values for $\lambda/V_{\lambda/2}$ against λ normalized to $\lambda/V_{\lambda/2}=1$ at $\lambda=5461$ Å. We consider this relative data reliable to within 2%.

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[12] See, for example, *American Institute of Physics Handbook*, edited by D. Gray (McGraw-Hill Book Company, Inc., New York, 1957).