Magnitude of Electrogyratory Effects

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First-order electrogyratory effects can be expressed in terms of a nonlinear gyratory susceptibility χ_g^{NL} that is defined analogously to the conventional nonlinear optical susceptibility. It is shown that χ_g^{NL} can be represented as $\Delta_g(\omega)[\chi(\omega)]^2\chi(0)$, where $\chi(\omega)$ and $\chi(0)$ are linear susceptibilities, and where $\Delta_g(\omega)$ is expected to remain fairly constant for a wide variety of materials. This relationship is formally identical to that commonly used to estimate the conventional nonlinear optical susceptibility. The ratio between $\Delta_g(\omega)$ and the quantity $\Delta(\omega)$ that describes conventional optical nonlinearities is approximately the ratio of the natural gyration G to the refractive power n^2-1. Values of $\Delta_g(\omega)$ inferred from previously reported measurements of the magnitude of electrogyratory effects are from two to nearly five orders of magnitude larger than the predicted value. It is shown that the observations in these instances can be explained solely in terms of conventional electro-optic (birefringence) effects and therefore have no bearing on the magnitude of electrogyratory effects. A method is described that permits the direct observation of electrogyratory effects in the presence of considerably larger electro-optic birefringence effects. The method is applied to the measurement of electrogyration in bismuth germanium oxide $Bi_{12}GeO_{20}$. Even though the sensitivity of the method is limited by indeterminate amounts of stray birefringence in the optical system, the upper limit obtained for the magnitude of the effect is considerably smaller than the previously reported value.

I. INTRODUCTION

For a given propagation direction of light the refractive indices n of a medium exhibiting gyration (rotation of the plane of polarized light) are the positive solutions to the equation[1]

$$(n^2-n'^2)(n^2-n''^2)=(g_{ij}l_il_j)^2\equiv G^2, \tag{1}$$

where n' and n'' are the refractive indices that would obtain in the absence of gyration, l_i and l_j are direction cosines of the propagation vector and the quantities g_{ij} are elements of the gyration tensor of the medium. [The convention of summing over repeated indices is observed in Eq. (1) and elsewhere in this report.] The quantities g_{ij} may be altered by subjecting the medium to a low-frequency electric field having components E_k, etc., as expressed by Eq. (2):

$$g_{ij}=g_{ij}^{(0)}+\gamma_{ijk}E_k+\beta_{ijkl}E_kE_l+\cdots. \tag{2}$$

The point-group symmetries of the medium that permit the tensor coefficients γ_{ijk} and β_{ijkl} to be nonvanishing, and other aspects of these effects have been discussed by Zheludhev and others.[2-6] The theoretical treatments[4-6] do not provide a direct estimate of the magnitude of these effects; an attempt is made here to do so. The estimate obtained is found to be considerably smaller than the magnitude implied by some experiments. These experiments are examined critically, and it is concluded that they may not in fact represent observations of electrogyratory effects. A determination by a new method is made of the magnitude of the electrogyratory effect in bismuth germanium oxide, $Bi_{12}GeO_{20}$. The effect is found to be substantially smaller than what has previously been reported.

II. ESTIMATION OF MAGNITUDE OF ELECTROGYRATORY EFFECTS

The approach pursued to estimate the magnitude of these effects is to consider the way optical gyration arises in a medium, and the way this gyration is changed by subjecting the medium to an electric field.

For isotropic and cubic media the nonvanishing components of the gyration tensor are $g_{11}=g_{22}=g_{33}\equiv G$.[1] For such media $n'=n''$, so in accord with Eq. (1), G is equal in magnitude to one-half the difference between the squares of the refractive indices for left- and right-circularly-polarized light. If the Lorentz effective-field approximation is assumed valid, then the gyration in this case can be expressed[7,8] as

$$G=\frac{16\pi\nu Nn'(n'^2-2)}{9h}\sum_{b\neq 0}\frac{\mathrm{Im}(\mu_{0b}\cdot m_{b0})}{\nu_{b0}^2-\nu^2}, \tag{3}$$

where ν is the optical frequency, N is the number of structural units (assumed to be in the ground state 0) per unit volume, μ_{ab} and m_{ab} are, respectively, the matrix elements for the electric and magnetic dipole moment operators between states a and b of the structural unit, ν_{ab} is the energy difference, expressed in units of frequency, between states a and b, and h is Planck's constant.

The specific values of the matrix elements in Eq. (3) depend on the microscopic model that is assumed. Several different models that give rise to optical gyration have been investigated, both quantum mechanically, as by Eq. (3), and clas-

sically. Among these models are the disymmetric one-electron oscillator,[8] coupled anisotropic oscillators,[9] and the twisted anisotropic susceptor.[10] The presence of magnetic moment matrix elements m_{b0} in Eq. (3), even when the electric dipole approximation is made for the interaction of radiation with material media, can be considered to arise from the relationship between curl $\vec{E}$ and $\vec{B}$ implied by Maxwell's equations. Curl $\vec{E}$ is a measure of the spatial variation of the electric field. The fundamental cause of optical gyration is, in fact, the variation of the phase of the radiation field over the dimensions of the structural units being considered. For this reason the magnitudes of the gyratory coefficients calculated for the various models cited above are usually smaller than the refractive power, $n'^2 - 1$, by a factor that approximates in magnitude the ratio of atomic or molecular dimensions to the wavelength of light. Observed values for visible wavelengths of $G/(n^2 - 1)$ for a number of common gyratory solids[11] and for the strongly gyratory material $AgGa_2S_4$[12] range between 1.0×10^{-5} and 6.3×10^{-4}, more or less in agreement with the predicted magnitudes. The extraordinarily large gyratory coefficients observed in the cholesteric mesophases are due to the fact that the pitch of the cholesteric helix is much greater than ordinary atomic or molecular dimensions.[13]

The parameters such as interatomic distances, resonant frequencies, polarizabilities, etc., that characterize the various models that exhibit optical gyration will, in general, be modified when the models are subjected to a low-frequency electric field. The extent to which these perturbations occur to first order in the applied field will govern the magnitude of the electrogyratory tensor coefficients γ_{ijk}. In estimating the magnitude of these coefficients it is instructive to consider as an example the coupled oscillator model investigated by Chandrasekhar.[14] The structural unit in this model consists of two atoms at coordinates $(l, 0, 0)$ and $(0, l, d)$. The first atom is polarizable only along the x direction, and the second only along the y direction. Dipolar coupling exists between the atoms. For light of free-space wavelength λ propagating along the z direction, and for $d \gg l$, the gyration exhibited by this model is

$$G \sim 8\pi^2 n'(n'^2+2)^2 N\alpha^2 l^2/3\lambda d^4 , \tag{4}$$

where α is the polarizability of each atom. The electrogyratory coefficient γ can be written

$$\gamma = \frac{dG}{dE} = \frac{\partial G}{\partial d}\frac{\partial d}{\partial E} + \frac{\partial G}{\partial \alpha}\frac{\partial \alpha}{\partial E} + \cdots$$

$$= -\frac{4G}{d}\frac{\partial d}{\partial E} + \frac{2G}{\alpha}\frac{\partial \alpha}{\partial E} + \cdots . \tag{5}$$

The first term, $-(4G/d)(\partial d/\partial E)$, can be expressed as $-(4G/d)e\beta(k_2^{-1} - k_1^{-1})$, where β is the factor relating the effective field to the macroscopic field E, and k_1 and k_2 are stiffness constants appropriate to the two atoms. If k_1 and k_2 differ appreciably from one another, then the first term can be expressed as

$$\frac{\partial G}{\partial d}\frac{\partial d}{\partial E} = (\pm)\frac{4G\beta a_1^3}{ed} , \tag{6}$$

where a_1 is a length whose order of magnitude is of atomic dimensions. The magnitude of the second term in Eq. (5), which contains the dependence of the polarizability upon the applied field, can be estimated from the same anharmonic-oscillator model[15] used to treat conventional electro-optic and nonlinear optical effects. With this model, the second term is given by

$$\frac{\partial G}{\partial \alpha}\frac{\partial \alpha}{\partial E} = \frac{2G}{\alpha N}\left(\frac{n'^2+2}{3}\right)^{-1}\chi^{NL}$$

$$= -\left(\frac{n'^2+2}{3}\right)^{-1}\frac{(n'^2-1)^2\beta v G}{4\pi^2 N^2 e k \alpha} \tag{7}$$

In Eq. (7), $[\frac{1}{3}(n'^2+2)]^{-1}$ is the effective field parameter, analogous to β, that obtains at optical frequencies, χ^{NL} is the appropriate nonlinear susceptibility, and k and v are, respectively, the linear (harmonic) and quadratic (anharmonic) force constants of the oscillator. If it is assumed[15] that the magnitude of v is such that the linear and nonlinear forces are equal for a displacement a_2 of the order of atomic dimensions, then v can be replaced by k/a_2. From the relation

$$(n'^2 - 1)/(n'^2 + 2) = \tfrac{4}{3}\pi N\alpha \tag{8}$$

and the substitution $N = a_0^{-3}$ it follows that

$$\frac{\partial G}{\partial \alpha}\frac{\partial \alpha}{\partial E} = -\frac{G(n'^2-1)\beta a_0^3}{\pi e a_2} . \tag{9}$$

Since a_0, a_1, a_2, and d are all lengths of the order of atomic dimensions, it is apparent that contributions to the electrogyratory coefficient of the types represented by Eqs. (6) and (9) can be comparable to one another in magnitude. These contributions may in fact be of opposite sign and partially cancel one another; it is our present intention, however, to demonstrate how large the effect can be.

It is possible to define a nonlinear gyratory susceptibility χ_g^{NL} that is related to γ in a manner analogous to the way the nonlinear susceptibility $\chi^{NL}(\omega, \omega, 0)$ is related to the conventional electro-optic effect:

$$\chi_g^{NL} = \gamma/4\pi. \tag{10}$$

If the approximation is made that γ can be represented by just the contribution expressed in Eq.

(9), it follows that

$$\chi_{\mathcal{E}}^{\mathrm{NL}} = -\frac{2G}{n^2-1}\left[\frac{2a_0^3\beta}{ea_2\chi(0)}\right]\chi^2(\omega)\chi(0), \qquad (11)$$

where $\chi(\omega) = (n^2-1)/4\pi$, $\chi(0) = (\epsilon-1)/4\pi$, and ϵ is the low-frequency dielectric constant. The expres sion in brackets in Eq. (11) is precisely the value dictated by the anharmonic-oscillator model[15] of the constant $\Delta(\omega)$, first applied by Miller[16] to describe the electro-optic properties of a wide variety of materials.

The first-order electrogyratory effect is permitted[2] in certain point groups where the gyration G vanishes; indeed, the model discussed here could have consisted of two pairs of anharmonic osicllators with cancelling contributions to the gyration in the absence of an electric field, but with reenforcing contributions to the electrogyratory effect. Equation (11) should therefore not be constructed to imply that the electrogyratory effect vanishes for crystals with zero or vanishingly small values of the gyration. Values of the quantity $G/(n^2-1)$ for individual crystals should thus not be used in the equation. Instead it seems more appropriate to use an average value for $G/(n^2-1)$, whose magnitude is approximately constant for many crystals. If this is done, Eq. (11) can be represented as

$$\chi_{\mathcal{E}}^{\mathrm{NL}} = \Delta_{\mathcal{E}}(\omega)\chi^2(\omega)\chi(0), \qquad (12)$$

where $\Delta_{\mathcal{E}}(\omega)$ is smaller than Miller's $\Delta(\omega)$ by about the same factor (the ratio of atomic dimensions to the optical wavelength) that related natural gyration G to refractive power n^2-1. The value of $\Delta(\omega)$ is near 3×10^{-6} esu.[17] If a representative value for $G/(n'^2-1)$ of 0.8×10^{-4} is assumed, then it follows from Eqs. (11) and (12) that $\Delta_{\mathcal{E}}(\omega)$ should be of the order of 5×10^{-10} esu.

III. COMPARISON WITH PREVIOUSLY REPORTED OBSERVATIONS

Experimental observation of the first-order electrogyratory effect, corresponding to the tensor coefficients γ_{ijk}, has been reported for quartz,[18] and for the isostructural compounds $Bi_{12}GeO_{20}$ and $Bi_{12}SiO_{20}$.[19–22] The reported value[18] for γ_{11} (condensed subscript notation) in quartz is $(12.7\pm1.4)\times10^{-7}$ esu for $\lambda = 461$ nm. Since for quartz $\epsilon_1 = 4.5$ and $n_1' = 1.5519$, the corresponding value of $\Delta_{\mathcal{E}}(\omega)$ is 2.9×10^{-5} esu. This is almost five orders of magnitude larger than the theoretical estimate made above! The electrogyratory coefficients for $Bi_{12}GeO_{20}$ and $Bi_{12}SiO_{20}$ have not been reported explicitly. However, from the observed values for the refractive index and gyration[19] ($n = 2.55$, $G = 28.3\times10^{-5}$ at $\lambda = 510$ nm), the dielectric constant[23] ($\epsilon = 38$) and the statement[21] that an applied field of about 10 kV/cm changes the optical activity by about 5%, it can be inferred that for these compounds $\Delta_{\mathcal{E}}(\omega)$ is approximately 6×10^{-6} esu. Although this is much smaller than the value of $\Delta_{\mathcal{E}}(\omega)$ obtained for quartz, it is still more than two orders of magnitude larger than the theoretical estimate.

We have attempted without success to find mechanisms different from those discussed above that might account for such large observed values of $\Delta_{\mathcal{E}}(\omega)$. It was decided therefore to examine more closely the data on which the reported observations of electrogyratory effects are based. The value reported for γ_{11} in quartz[18] is obtained by determining the field dependence of the gyratory coefficient g_{11}. The gyratory coefficient was reportedly measured for light propagated parallel to the x axis by the method of Konstantinova *et al.*[24] In this method the light prior to transmission through the crystal is plane-polarized parallel to one of the vibration directions (in this case, the optic axis) of the crystal. Passage through the crystal imparts to the light a slight degree of ellipticity. As the wavelength of the incident light is varied, the orientation of the major axis of this ellipse varies symmetrically about the direction of polarization of the incident light. The gyratory coefficient g_{11} is determined from the amplitude of this oscillation. When an electric field is applied (parallel to the x axis) the electrogyratory effect should change the amplitude of this oscillation. What is actually reported,[18] however, and construed to be the manifestation of an electrogyratory effect, is a shift or biasing of the entire oscillatory function without change of amplitude.

Such a shift is known[24] to arise from misorientation of the polarization direction of the incident light with respect to the vibration directions of the crystal. The vibration directions of quartz are, in fact, changed by an electric field applied in the reported direction, due to the conventional electro-optic effects involving the coefficient r_{41}. The calculated value of this shift, which arises from a mechanism entirely independent of electrogyratory effects, is in reasonable agreement with the observed value.

Observation of a first-order field-induced shift with two identical crossed quartz plates is also reported.[18] It can be shown that the only way such a shift can arise when the field is applied to both plates is for the fields to be of opposite sense in the two plates. Once again, the data can reasonably be explained solely on the basis of conventional electro-optic effects. It is concluded that the reported observations on quartz do not really establish the magnitude of the electrogyratory effects in this material.[25]

The results[19–21] on $Bi_{12}GeO_{20}$ and $Bi_{12}SiO_{20}$ are difficult to assess because neither the experimen-

tal configuration nor the method employed for interpretation of the data are described as explicitly as might be desired. Phenomenological considerations[2] make it doubtful whether the electrogyratory effect should, in fact, be observable under the conditions reported. A possible explanation of these results is that the effects of reflection at the interfaces of the crystal were not properly taken into account.

The following considerations explain how the neglect of reflections can give rise to an apparent electrogyratory effect with magnitude comparable to what is reported for $Bi_{12}GeO_{20}$ and $Bi_{12}SiO_{20}$. The transmission of light through a medium exhibiting gyration and birefringence can be expressed in terms of the eigenfunctions

$$\vec{\psi}_1 = \frac{1}{\sqrt{2}}\left[i(1+q)^{1/2}\hat{x} + (1-q)^{1/2}\hat{y}\right], \tag{13a}$$

$$\vec{\psi}_2 = \frac{1}{\sqrt{2}}\left[-i(1-q)^{1/2}\hat{x} + (1+q)^{1/2}\hat{y}\right], \tag{13b}$$

where $\hat{x}$ and $\hat{y}$ are unit vectors parallel, respectively, to the characteristic directions of the refractive components n' and n'' in Eq. (1). In Eqs. (13), $q = \delta/\Delta$, where $\delta = 2\pi(n' - n'')/\lambda$ is the phase difference per unit length between the two eigenfunctions that would arise solely from the birefringence, $\Delta = (\delta^2 + 4\rho^2)^{1/2}$ is the total phase difference per unit length, $\rho \sim \pi g/\bar{n}\lambda$ is the rotation per unit length (rotatory power), and $\bar{n}$ is the mean refractive index. If normally incident light of a given polarization state can be represented prior to passage through the medium as $A_i\vec{\psi}_i$, then when reflections at the interfaces are neglected, the polarization state after passage can be represented as $a_{ij}A_j\vec{\psi}_i (i, j = 1, 2)$, where

$$a_{11} = 1/\mu\phi, \quad a_{12} = a_{21} = 0, \quad a_{22} = \phi/\mu. \tag{14}$$

In Eqs. (14), $\mu = e^{2\pi i\bar{n}z/\lambda}$ and $\phi = e^{i\Delta z/2}$, where z is the thickness of the medium. When reflections at the interfaces are taken into account, the polarization state after passage is a different linear combination $b_{ij}A_j\vec{\psi}_i$ of the eigenfunctions. It can be inferred from the continuity[26] of the tangential components of the electric and magnetic fields across the interfaces that the matrix elements b_{ij} are given by

$$b_{11} = 4\phi\mu\{\mu^2 N(\bar{n}K - \eta^2 rL) + M[\bar{n}(1-q^2)L - q^2\phi^2(\bar{n}+r)M]\}/D, \tag{15a}$$

$$b_{12} = 4\phi\mu Mq(1-q^2)^{1/2}[\bar{n}L - \phi^2\mu^2 rK + \phi^2(\bar{n}+r)M]/D, \tag{15b}$$

$$b_{21} = 4\phi\mu Lq(1-q^2)^{1/2}[\mu^2 r\bar{n} - \phi^2\bar{n}M - (\bar{n}-r)L]/D, \tag{15c}$$

$$b_{22} = 4\phi\mu\{\phi^2\mu^2 K(\bar{n}N - q^2 rM) + L[\phi^2\bar{n}(1-q^2)M - q^2(\bar{n}-r)L]\}/D, \tag{15d}$$

where

$$D = \kappa[\phi^2(\mu^2 KN + LM)^2 - q^2\mu^2(\phi^2 KM + LN)^2]. \tag{16}$$

In Eqs. (15) and (16), $\kappa = e^{2\pi i z/\lambda}$, $r = \lambda\Delta/4\pi$, $K = 1 + \bar{n} + r$, $L = -1 + \bar{n} + r$, $M = 1 - \bar{n} + r$, and $N = 1 + \bar{n} - r$.

The intensity of polarized light transmitted through a $Bi_{12}GeO_{20}$ crystal in combination with a coupled birefringence compensator and analyzer was calculated as a function of the compensator setting, the azimuth setting of the analyzer, and the dc voltage applied to the crystal. The conditions assumed were identical to those reported,[19,20] and Eqs. (15) and (16), which take internal reflections into account, were employed. The calculations were made for a wavelength of 6660 Å, at which the coefficient $n_0^3 r_{41}$ describing the conventional electro-optic effect has a value[19] of 1.6×10^{-6} cm/statvolt, and the optical rotatory power has a value[27] of 19°/mm. Of greatest importance in these considerations is that *the electrogyratory effect was assumed to be negligibly small*. It was also assumed that that the incident light was sufficiently incoherent and/or that the crystal surfaces were sufficiently nonplanar that averaging could be performed over the variations in the intensity due to interference ("Fabry-Perot modes"). [In the only treatment[24] of internal reflections in birefringent gyratory media published to the author's knowledge, this averaging is performed by setting to zero all terms that vary rapidly with respect to changes in the optical frequency. However, the correct average over ω of, for example, $1/(a + b\cos\omega)$ is not $1/a$, but rather $(a^2 - b^2)^{-1/2}$.] Values of the compensator setting and analyzer azimuth setting that minimized the calculated intensity were then found for each applied dc voltage.

Equations (14), which neglect the effect of internal reflections were then used to calculate from the values of these settings the rotatory power ρ and birefringence parameter δ of the crystal. This is the way these parameters are most commonly inferred. As shown in Fig. 1 the values of ρ and δ calculated in this manner differ appreciably from the true values, which would have to be calculated taking the effects of reflection into account. At 2-kV applied voltage, the apparent value of ρ is about 2.6% greater than the value with no voltage applied. This apparent change in the rotatory power is comparable in magnitude with what has been observed[19-21] at the same applied voltage and ascribed to the electrogyratory effect.

IV. DIRECT OBSERVATION OF ELECTROGYRATION IN PRESENCE OF OTHER ELECTRO-OPTIC EFFECTS

The method described below for measuring electrogyratory effects in crystals having no natural birefringence avoids the difficulties encountered by

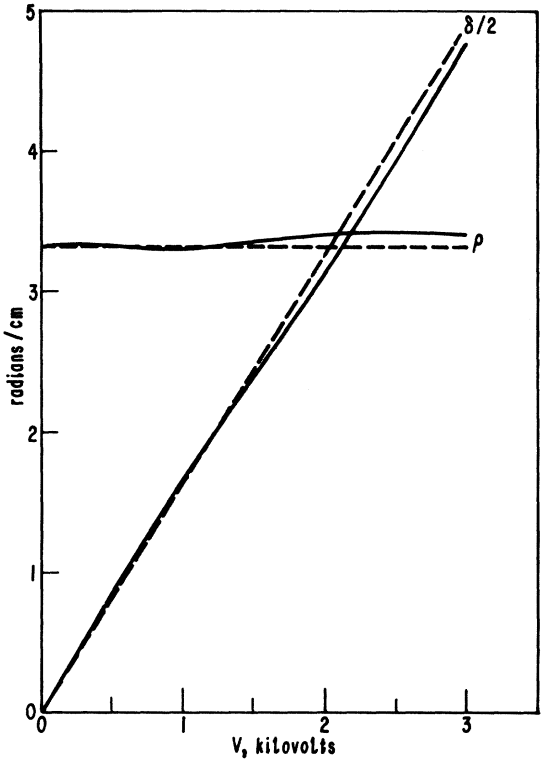

FIG. 1. Values of the birefringence parameter $\delta/2$ and the rotatory power ρ as functions of applied voltage. Dashed lines show true values; solid lines show values calculated by neglecting the effects of reflection.

other investigators in separating changes in the birefringence from changes in the gyration. A crystal is situated between two polarizers oriented, respectively, to $\hat{x}$ by angles θ and η. A low-frequency ac electric field $E_0\cos\omega t$ is applied to the crystal. The transmitted intensity I can in general be expressed as

$$I = I_0\left[S + E_0\cos\omega t\left(T\frac{\partial\delta}{\partial E} + U\frac{\partial\rho}{\partial E}\right) + O(E_0^2)\right]. \quad (17)$$

The functions S, T, and U shall be specified below. In Eq. (17), $\partial\delta/\partial E$ is the coefficient that represents the magnitude of the conventional electro-optic effect for the propagation direction, field direction, and optical wavelength appropriate to the observation, $\partial\rho/\partial E$ is the analogous coefficient for the electrogyratory effect, and $O(E_0^2)$ represents terms of second or higher order in E_0. If it is assumed that averaging can be performed as before over the variations in intensity due to interference then is follows from Eqs. (15) and (16) (which take reflections into account) that the functions S, T, and U are given by

$$S = \frac{8\bar{n}^3\cos^2(\theta-\rho z-\eta)}{4\bar{n}^2+4\bar{n}^4-g^2}, \quad (18)$$

$$T = \frac{\lambda(1-\bar{n}^2)\cos(\theta-\rho z-\eta)\cos(\theta-\rho z+\eta)}{2\pi(1+\bar{n}^2)^2} + O(\lambda g), \quad (19)$$

$$U = \frac{4\bar{n}z\cos(\theta-\rho z-\eta)\sin(\theta-\rho z-\eta)}{1+\bar{n}^2} + O(\lambda g). \quad (20)$$

The coefficient T represents modulation of the transmitted intensity that can be considered to arise from the effects of electroreflectance. For typical values of λ, $\bar{n}$, and z this coefficient is of the order of 10^{-6} as large as U. In our experiments this term accounted for less than 1% of the observed ac signal. If the coefficient T can be neglected, then the ac-modulated component of the transmitted intensity for this configuration represents a measure of the electrogyratory effect that is essentially free of "cross talk" due to the conventional electro-optic effect. The transmitted intensity can be used to activate a photodetector. It is convenient to measure simultaneously the dc and ac components of the photodetector output and plot one against the other for various angles η of the output polarizer. Equations (18) and (20) imply that this plot should be an ellipse having the position and orientation shown by the dashed curve in Fig. 2. The value of $\partial\rho/\partial E$ can, in principle, be inferred from a knowledge of E_0, $\bar{n}$, and the axial ratio of the ellipse.

V. OBSERVATIONS ON BISMUTH GERMANIUM OXIDE, $Bi_{12}GeO_{20}$

The method described in Sec. IV was used to estimate the magnitude of the electrogyratory effect in bismuth germanium oxide, $Bi_{12}GeO_{20}$, for the same directions of optical propagation and applied electric field that were used in the previously reported measurements.[19–21] A 0.3-cm cube of $Bi_{12}GeO_{20}$ having high optical quality[28] with faces oriented perpendicular to the [110], [1$\bar{1}$0], and

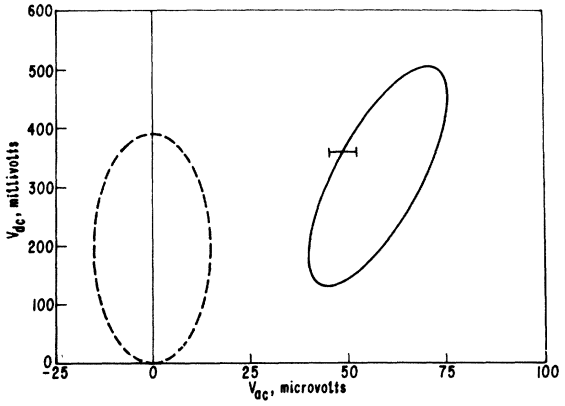

FIG. 2. Predicted (dashed) and observed (solid) plots of V_{ac} vs V_{dc} for bismuth germanium oxide, $Bi_{12}GeO_{20}$.

[001] crystallographic directions was used. Monochromatized light with $\lambda = 600$ nm and a spectral width of approximately 10 nm was propagated along $[1\bar{1}0]$. The quality of the crystal and of the rest of the optical system was such that no birefringence due to strain could be detected visually. When the polarizers were set for minimum transmission the 600-nm light was effectively extinguished, and only green light, due to leakage through the monochromator, could be seen. (Because the optical rotatory power varies appreciably with wavelength, the polarizer setting that extinguishes light at 600 nm does not extinguish light at shorter wavelengths.) The crystal surfaces perpendicular to [110] were electroded with a liquid gallium-indium alloy. For most of the measurements 250 V rms at 1000 Hz was applied to the electrodes. Other details of the experimental apparatus are described elsewhere.[29]

Values of the ac photodetector output V_{ac} and the dc output V_{dc} were measured for twenty different orientations of the output polarizer. If was found that within the limits of experimental error, V_{ac} was proportional to the first power of the voltage applied to the crystal. (The method employed here[29] discriminates intrinsically against even-order electro-optic effects; odd-order effects of order higher than first are apparently negligibly small.) The principal errors are in the measurement of V_{ac} and are governed mainly by the intensity of the transmitted light and the noise bandwidth of the ac detector. The solid ellipse shown on Fig. 2 was obtained by least-square analysis of the observed data. The error flag indicates approximately the standard deviation for each measurements of V_{ac}. (The signals that are observed represent small optical effects. If the optical system had been arranged to detect the conventional electro-optic effect, the indicated error would correspond to an uncertainty in retardation of less than $3\times10^{-6}\lambda$.)

The observed plot differs from the predicted plot in three respects: The minimum value of V_{dc} is greater than zero, the ellipse is not centered about the $V_{ac}=0$ axis, and the axes of the ellipse are tilted with respect to the coordinate axes. These differences can be interpreted as follows. The failure of V_{dc} to assume a minimum value close to zero is due to the presence in the system of stray light of appreciable intensity. No special precautions were taken to exclude ambient light from the optical system. A major source of stray light appears to be the previously mentioned leakage through the monochromator. Failure of the ellipse to be centered about the $V_{ac}=0$ axis implies that even when averaging is performed over all orientations of the output polarizer there is a net ac modulation. This can arise only if there is modulation of the intensity of the light transmitted through the crystal, in addition to modulation of the polarization state. The effect associated with the coefficient T in Eq. (17) leads to such intensity modulation and to a tilting of the ellipse axes, but the magnitude of the effect seems to be too small to explain the observations. The first-order electroabsorption effect that can occur[30] in noncentrosymmetric crystal can be shown to give rise to an intensity modulation of the proper magnitude. The principal contribution arises from stray light at wavelengths near the fundamental absorption edge of the crystal. Because the amplitude of this modulated component changes as the output polarizer is rotated, a tipping of the ellipse axes will result. Electroabsorption effects can also contribute to the width of the ellipse along the V_{ac} axis.

If it is assumed that the width along the V_{ac} axis of the observed ellipse is due to the electrogyratory effect, then the value of the effect calculated from Eqs. (17), (18), and (20) is $\partial\rho/\partial E = 4.1\times10^{-5}$ rad/statvolt. The previously reported observations[19-21] correspond to a value for $\partial\rho/\partial E$ of about 6.3×10^{-3} rad/statvolt. Although these results indicate strongly that the effect is a good deal smaller than previously reported, they cannot be construed to represent an accurate determination of the magnitude of the effect. Stray birefringence in the crystal or the optical system could not be perceived visually, but its effects at the low level of signals encountered in these measurements cannot be ignored. Stray birefringence can cause the conventional electro-optic effect to generate a signal that is indistinguishable from what would arise via the electrogyratory effect. It can be shown that even if the electrogyratory effect were absent a suitably oriented stray birefringence corresponding to a retardation of less than 0.001λ (about an order of magnitude smaller than what can be perceived visually) could account for the width along the V_{ac} axis and the inclination of the observed ellipse. In the unlikely circumstance that the effects due tl electrogyration and stray birefringence were opposite in sign and almost exactly equal in magnitude the coefficient $\partial\rho/\partial E$ could be as much as an order of magnitude larger than the value stated above. Even this is about 15 times smaller than the value previously reported. It is believed that the effects of reflection, as discussed in this report, are probably responsible for the main part of the latter value.

VI. CONCLUSIONS

Electrogyratory effects are estimated for visible wavelengths to be about four orders of magnitude smaller than conventional electro-optic (birefringence) effects. Thus in attempting to measure electrogyratory effects, the investigator is often beset with the problem of separating a weak

effect from a similar, but much stronger effect. The optical behavior of media that simultaneously exhibit gyration and birefringence is perhaps more complicated than is sometimes appreciated, and a small error in the way the separation is made can lead to an overwhelming error in the estimated magnitude of the electrogyratory effect.

For optical propagation directions along which natural birefringence is absent, an ac measurement technique can permit direct observation of electrogyration with a considerable degree of isolation from interference due to electro-optic birefringence effects. The degree of isolation, and hence the sensitivity of the method, is limited by small, indeterminate amounts of stray birefringence in the system. It is suspected that stray birefringence will similarly hamper the sensitivity of other methods of measurement when electro-optic birefringence effects are present. In order to achieve enough sensititivy to permit the accurate determination of electrogyratory coefficients it will probably be necessary to follow the suggestion[6] that measurements be made for those directions of optical propagation and applied electric field for which electrogyratory effects exist, but electro-optic birefringence effects vanish.

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