# MCCOY, BAROUCH, AND ABRAHAM

 $-7\alpha^{2}e^{2i\phi}(1-\alpha^{2}e^{2i\phi})^{-1}-(1-e^{-2i\phi})^{-1}-\frac{5}{2}]\} + (\alpha^{-1}-\alpha)^{-2}\left[(\alpha^{-1}e^{-i\phi}-\alpha e^{i\phi})/(\alpha e^{-i\phi}-\alpha^{-1}e^{i\phi})\right]^{1/2}$ 

 $\times \left\{ 1 + (2R)^{-1} \left[ 2(e^{-2i\psi} - 1)^{-1} - 7\alpha^2(1 - \alpha^2)^{-1} - \frac{1}{2}(\alpha^{-2}e^{2i\psi} - 1)^{-1} + \frac{3}{2}(\alpha^{-2}e^{-2i\psi} - 1)^{-1} - 2 \right] \right\} \right].$ 

PHYSICAL REVIEW A

VOLUME 4, NUMBER 6

DECEMBER 1971

# Origin of the Nonlinear Refractive Index of Liquid CCl<sub>4</sub><sup>+</sup>

R. W. Hellwarth,\* Adelbert Owyoung,<sup>‡</sup> and Nicholas George California Institute of Technology, Pasadena, California 91109 (Received 26 July 1971)

We report here the first determination for a simple liquid (specifically liquid  $CCl_4$ ) of the fraction of its Kerr effect that arises from the (nearly instantaneous) nonlinear response of its electronic currents, and hence would exist even if the nuclei were frozen in position. To do this, we have remeasured the power dependence of the rotation of the polarization ellipse of a monochromatic beam in  $CCl_4$  with greatly improved accuracy ( $\pm 10\%$  absolute) using a single-Gaussian-mode ruby (giant pulse) laser. We then compare the results of this ellipse rotation measurement with existing Kerr data, and, using a general relation between the relative electronic contributions to both effects which we demonstrate, we show that  $(54 \pm 16)\%$  of the Kerr effect in  $CCl_4$  arises from nonlinear electronic response. The method should be useful for any isotropic material.

## I. INTRODUCTION

The second-order nonlinear electric susceptibility tensor  $\chi^{(2)}$  of a material manifests itself in a variety of commonly observed effects, such as electric-field-induced birefringence (Kerr effect). The physical mechanisms which can contribute to this nonlinear susceptibility at frequencies well below any electronic absorption frequency are of two distinct types, and contribute additively to  $\chi^{(2)}$ . An "electronic" contribution arises from a nonlinear distortion of the electron orbits around the nuclei, considered to be fixed in an average or typical arrangement.<sup>1</sup> This contribution would be observable, in principle, within a few electronic cycles (~ $10^{-16}$ sec) after sudden application of a strong electric field, and is independent of temperature at constant density. The second, or "nuclear," contribution arises from an electric-fieldinduced change in the motions of nuclei; in the presence of these changed motions the electronic currents respond linearly to the impressed electric fields.<sup>2</sup> This nuclear contribution could be observed after the sudden impression of a field only following a time lapse of the order of the time  $(\sim 10^{-12} \text{sec})$  required for a thermal nucleus to move a typical internuclear distance or execute a vibrational cycle. These nuclear contributions are generally temperature-dependent at constant densitv.

Owing mainly to the fact that observed temperature dependences of the Kerr effect (at constant pressure) are generally too large to be consistent with the former electronic mechanism, it has usually been assumed that the latter nuclear contributions dominate in liquids.<sup>2</sup> However recent measurements of this temperature dependence in various simple liquids (composed of electronically saturated, nonassociating, electrically isotropic and neutral atoms or molecules) have yielded dependencies sometimes small enough to be consistent with electronic mechanisms being important.<sup>3</sup> This is especially evident in the case of the commonest and most widely studied of simple liquids: carbon tetrachloride  $(CC1_4)$ .<sup>3</sup> In this liquid, measured light-scattering intensities are more consistent with its Kerr constant being partly electronic than not.<sup>4</sup> Furthermore, recent measurements of the purely electronic nonlinearity in  $CC1_4$  gas, <sup>5-7</sup> when extrapolated to liquid density by unreliable theory, also suggest that the two classes of mechanisms may contribute comparable amounts to the Kerr effect in  $CC1_4$ . Since so much of the interpretation of various data on CC14 depends on the relative importance of these mechanisms, we have undertaken a more direct measurement of the electronic fraction of the room-temperature Kerr constant of liquid CCl<sub>4</sub> using a novel technique (discussed in Sec. II) which we have employed previously to answer similar questions about nonlinear optical effects in glasses.<sup>8</sup>

We find this fraction to be  $(54 \pm 16)\%$  by comparing our measurements (described in Sec. III) of the power-dependent rotation of the polarization ellipse of a monochromatic beam in CCl<sub>4</sub> with previous measurements of its Kerr effect, all in the light of a general relation between the two effects derived in Sec. II. Our value for the ellipse rotation is roughly 50% higher than that measured previously by Wang<sup>9</sup> and over five times that reported by Maker *et al.*<sup>10</sup> in their original prediction and first measurements of this effect. Unlike these earlier investigations, we have been able to employ new techniques to obtain a single-Gaussian-mode laser beam, a fact which we believe is mainly responsible for the discrepancies.

In our concluding Sec. IV, we discuss various alternative methods of separating electronic from nuclear contributions to the nonlinear polarization (third order in the electric field), including the extrapolation to liquid densities of recent hyperpolarizability measurements on vapors and absolute measurements of light scattering intensities. We argue that our technique of comparing Kerr and ellipse-rotation data offers the simplest and most accurate method presently available for distinguishing these mechanisms in isotropic media.

# II. RELATION BETWEEN ELLIPSE-ROTATION AND KERR EFFECTS

We outline here how the two classes of mechanisms determine the ellipse-rotation and Kerr effects in such a way as to allow their unique separation by measuring the two effects. More details of this and related relations are given elsewhere.<sup>8,11</sup>

We start from the fact that the nonlinear polarization density  $\vec{\mathbf{P}}^{\text{NL}}(\vec{\mathbf{r}},t)$ , third order in the electric field, may be separated into two parts. First, there is an electronic part  $\vec{\mathbf{P}}_e(\vec{\mathbf{r}},t)$  which results from a distortion of the electron orbits about the nuclei, considered fixed in a typical spatial configuration. This polarization responds so quickly in transparent media (within several electronic cycles) that we may approximate it by the instantaneous form

$$\vec{\mathbf{P}}_{e}(\mathbf{\ddot{r}},t) = \frac{1}{2} \sigma \vec{\mathbf{E}}(\mathbf{\ddot{r}},t) \cdot \vec{\mathbf{E}}(\mathbf{\ddot{r}},t) \vec{\mathbf{E}}(\mathbf{\ddot{r}},t)$$
(1)

for any isotropic material. The electronic nonlinear susceptibility coefficient  $\sigma$  exhibits a small dispersion at the optical frequencies employed which we correct for when necessary. Evidently  $\sigma$  is independent of temperature at fixed density (although it varies with temperature at fixed pressure in a way not well understood).

The remaining part of  $\vec{P}^{\rm NL}$  is a nuclear part  $\vec{P}_n(\vec{r}, t)$  which is due to the linear response of the electronic currents about nuclear arrangements whose statistical probabilities are altered slightly in order to lower the average field-crystal interaction energy. When (as here) the medium is nonpolar, has no absorption near field frequencies and has little dispersion, the instantaneous fluctuation in this interaction energy density in a volume about  $\vec{r}$  which is small compared to a wavelength may be written  $-(1/8\pi)\vec{E}(\vec{r}, t) \cdot \vec{b} \in (\vec{r}, t)$ , where  $\vec{b} \in \vec{c}$  is the deviation from its average of the dielectric

permittivity tensor appropriate to the nuclear placements in the neighborhood of  $\vec{\mathbf{r}}$  at time *t*. Since  $\vec{\mathbf{P}}_n(\vec{\mathbf{r}},t)$  equals  $\delta \vec{\boldsymbol{\epsilon}} \cdot \vec{\mathbf{E}}(\vec{\mathbf{r}},t)/4\pi$  averaged with a weighting function (i.e., density matrix) expanded to first order in the above interaction energy, it must be proportional to the electric field at the same time (*t*) times a convolution of the square of the electric field at earlier times. In an isotropic medium, this means that the nuclear contribution to the nonlinear polarization must have the form

$$\vec{\mathbf{P}}_{n}(\vec{\mathbf{r}},t) = \vec{\mathbf{E}}(\vec{\mathbf{r}},t) \int a(t-s)E^{2}(\vec{\mathbf{r}},s)ds + \int \vec{\mathbf{E}}(\vec{\mathbf{r}},t) \cdot \vec{\mathbf{E}}(\vec{\mathbf{r}},s)b(t-s)\vec{\mathbf{E}}(\vec{\mathbf{r}},s)ds.$$
(2)

Here a(t) and b(t) are nuclear response functions for the "isotropic" and "anisotropic" parts of the nonlinear polarization, respectively, formed from the appropriate two-time correlations of components of  $\delta \vec{\epsilon}$ . The characteristic decay times in these response functions are several orders of magnitude longer than those for electronic nonlinearities. Also *a* and *b* are temperature-dependent, both at fixed density and fixed pressure, but in a way too poorly understood to yet be useful for the purpose of distinguishing nuclear and electronic contributions to nonlinear optical effects in liquids by observing their temperature variations.

Substituting into Eqs. (1) and (2) the specific forms of  $\vec{E}(\vec{r}, t)$  used in observing various nonlinear effects, we now proceed to solve Maxwell's equations to see what combinations of the infinitude of parameters contained in a(t) and b(t) describe the ellipse rotation and Kerr effects.

# Intensity-Induced Rotation of the Polarization Ellipse

To analyze this effect, we assume that there is propagating in the medium a z-directed monochromatic plane wave of frequency  $\omega$  composed of right and left circularly polarized components having complex vector amplitudes  $(\hat{x} \pm i\hat{v})E_{\pm}/\sqrt{2}$ . The two wavevectors  $\vec{k}_{\pm}$  corresponding to these components are found by substituting the field into Eqs. (1) and (2), transforming these expressions into the frequency domain, and substituting the resultant expression for  $\vec{P}^{\rm NL}(\omega)$  into Maxwell's equations, i.e., the wave equation in the frequency domain. Since the Fourier transforms of the nuclear response functions a(t) and b(t) are negligibly small at optical frequencies in  $E^2(t)$ , we find that

$$(ck_{\pm}/n\omega)^{2} = 1 + \pi [(\sigma + 2\alpha + \beta)E^{2} + (\sigma + 2\beta)|E_{\pm}|^{2}]/n^{2},$$
(3)

where  $E^2 \equiv |E_+|^2 + |E_-|^2$  is the time average of  $2\vec{E} \cdot \vec{E}$ ,  $\alpha \equiv \int a(s) \, ds$ ,  $\beta \equiv \int b(s) \, ds$ , and *n* is the (linear) refractive index at  $\omega$ . According to Eq. (3) the axes of the polarization ellipse rotate by  $\theta$  over a distance *z* so that a fraction

$$F(z) = (\sin 2\phi \, \sin \theta)^2 \tag{4a}$$

of the field at z becomes orthogonally polarized to the field at z=0;  $\tan \phi \equiv |E_*/E_-|$  and  $\theta = \frac{1}{2}(k_*-k_-)z$ . In our experiments  $|k_*-k_-| \ll n\omega/c$ , whence

$$\theta = \pi \omega_z E^2 (4nc)^{-1} (\sigma + 2\beta) \cos 2\phi.$$
(4b)

If the beam is weakly focused, and  $\theta << 1$  so that no further self-focusing due to nonlinear effects occurs, then we expect geometrical optics to be valid. In this case the ellipse-rotation angle may be computed for each ray by substituting  $\int_{z_1}^{z_2} E^2(z) dz$ for  $zE^2$  in Eq. (4b); here the integral is taken along the ray path. If this integral is calculated along an axial ray through the focus of an ideal Gaussian beam, we have<sup>12</sup>

$$\int_{-\infty}^{\infty} E^2(z) dz = 8\pi\omega P/c^2, \qquad (5)$$

where P is the total power in the beam. In our experiments we take advantage of this independence of  $\theta$  on beam dimensions when the liquid sample completely surrounds the focal region, measuring  $\theta$  by measuring the orthogonally polarized fraction F of the emerging beam.

In terms of the "B" coefficient defined by Maker et al.<sup>10</sup> in their original description of ellipse rotation, and in terms of the appropriate commonly used "c coefficients" defined by Maker and Terhune,<sup>13</sup> we see from Eq. (3) that

$$\sigma + 2\beta = 4 "B" = 24 c_{1221} (-\omega, \omega, \omega, -\omega) .$$
 (6)

# Kerr Effect

In an isotropic material a test beam  $\vec{\mathbf{E}}_{\omega}(\vec{\mathbf{r}}, t)$  of frequency  $\omega$  will exhibit birefringence in the presence of a strong beam  $\vec{\mathbf{E}}_{\nu}(\vec{\mathbf{r}}, t)$  of frequency  $\nu$ . This electric-field-induced birefringence is called the "ac Kerr effect" when  $\nu$  is an optical frequency, and called the "dc Kerr effect" when  $\nu$  is a radio frequency or lower. Both cases are usually described by a Kerr constant B (not the "B" above) defined by

$$B = \frac{\omega (\delta n_{\parallel} - \delta n_{\perp})}{2\pi c \langle E_{\nu}^{2} \rangle_{av}} , \qquad (7)$$

where  $\delta n_{\parallel} - \delta n_{\perp}$  is the difference between the induced changes in the refractive index parallel and perpendicular to the direction of  $\vec{E}_{\nu}$  whose mean square value in time is  $\langle E_{\nu}^2 \rangle_{av}$ .

Again  $\delta n_{\parallel}$  and  $\delta n_{\perp}$  may be calculated by substituting the two monochromatic plane waves into Eqs. (1) and (2) in the frequency domain and using  $\vec{P}^{\rm NL}(\omega)$ in Maxwell's equations. Substituting the resulting refractive index changes (second order in *E*) into Eq. (7) we find that

$$B = \omega(\sigma + \beta) / nc. \tag{8}$$

Here it is assumed that b(t) has no appreciable Fourier component at  $2\nu$  and  $|\omega - \nu|$ . In terms of the appropriate c coefficients, we have

$$\sigma + \beta = 12 [c_{1212}(-\omega, \omega, \nu, -\nu) + c_{1221}(-\omega, \omega, \nu, -\nu)].$$
(9)

Now one can see the important consequence that (small-angle) ellipse rotation measures  $\sigma + 2\beta$ , while the Kerr effect measures  $\sigma + \beta$  and together the effects yield the electronic parameter  $\sigma$  and the nuclear parameter  $\beta$  separately.

## **III. EXPERIMENT**

In the present ellipse-rotation experiments on CCl<sub>4</sub> we have attempted to avoid the difficulties encountered in earlier such measurements in several ways. First, we have employed a single-(transverse and longitudinal) mode beam, calibrated by measuring the ellipse rotation of  $CS_2$ , whose  $\sigma + 2\beta$  value we are able to determine to within 2% from other experiments. (Fortunately, the Kerr constant of CCl4 is also known most accurately in terms of that of  $CS_2$ .) We have also used stronger focusing into the sample so as to ensure that the ellipse rotation takes place entirely within the focal volume, thus taking advantage of the resulting independence of the ellipse-rotation angle of sample and focal dimensions. This arrangement also allows the optical intensity at the entrance and exit air-glass interfaces to be much lower for a given ellipse rotation angle, thus eliminating the danger of a nondamaging, absorbing plasma forming at the entrance face. As a result we observe a value of  $F/P^2$  reproducible to within ten percent, while using different focal length lenses, and samples, and also after changing the ruby laser-beam diameter and employing both active and passive Q-spoiling techniques.

The experimental configuration is shown in Fig. 1. The laser is a water-cooled room-temperature ruby laser Q-switched with a dye of cryptocyanine in acetone. Mode selection is performed by aperturing the  $\frac{9}{16}$ -in. diam×4-in. ruby to give a 3 mm output spot employing a sapphire etalon as the output reflector. The laser output is 0.05 J in a 20-nsec pulse under single-mode operation. Power monitoring of the laser output is performed via a beam splitter which directs a portion of the beam to an ITT FW 114A S-20 biplanar photodiode (D1). The rest of the beam is coupled through a Rochon prism (P-1) to define its plane of polarization prior to its introduction into the fresnel rhomb (R-1) which is oriented so as to produce an elliptically polarized input of desired eccentricity. The beam is then focused into the sample centrally by lens L-1 and then recollimated by lens L-2. A second Fresnel rhomb (R-2) is oriented parallel to R-1 so as to

2344



FIG. 1. Schematic diagram of the experimental arrangement used to observe ellipse rotation. BS: beam splitter, P-1: Rochon prism, P-2: Wollaston prism, F-1 and F-2: Schott neutral density stacks totaling N.D. = 4.0. F-3, F-4, and F-5: 6943-Å spike filters, D-1, D-2, and D-3: ITT FW 114A biplanar photodiodes, R-1 and R-2: fresnel rhombs, L-1 and L-2: lenses [(10-15)-cm focal length].

produce a linearly polarized output in the absence of ellipse rotation. This is followed by a Wollaston prism (P-2) oriented to direct a maximum "transmitted" signal into D-3 and a minimum "nulled" signal into D-2 in the absence of ellipse rotation.

The laser power delivered to the sample is adjusted by moving the Schott high-power neutraldensity filters from neutral-density stack F-1 to F-2, thus ensuring a constant reference power level into the diodes in the absence of a nonlinearity. Any rotation of the polarization ellipse during propagation through the sample thus reveals itself as a relative increase in the "nulled" signal. Monitoring of the transmitted beam in D-3 reveals any induced changes in the transmission path or changes in the spatial profile of the laser. A He-Ne laser operating at 6328 Å and adjusted collinearly with the ruby laser beam was used continuously to ensure proper alignment of the system.

Figure 2 shows the result of a typical run on the liquids  $CCl_4$  and  $CS_2$ . From this and similar other data we conclude that at 6943 Å and 23 °C.

$$\frac{(\sigma + 2\beta)/n \text{ of } CS_2}{(\sigma + 2\beta)/n \text{ of } CCl_4} = 56 \pm 6.$$
(10)

Using Eq. (8) we find that a previous direct measurement of the ratio of the Kerr constants of  $CS_2$  and  $CCl_4$  gives <sup>3</sup>

$$\frac{(\sigma+\beta)/n \text{ of } CS_2}{(\sigma+\beta)/n \text{ of } CCl_4} = 40.8 \pm 0.8$$
(11)

at 6943 Å and 23 °C. (This measurement, which was performed at 6328 Å, actually gave 41.6 for this ratio, and we have applied a small dispersion correction obtained from the wavelength variation data given in the Landolt-Bornstein tables.<sup>14</sup>)

Mayer<sup>5</sup> and Hauchecorne *et al.*<sup>6</sup> have found  $\sigma$  to be unobservable in CS<sub>2</sub> by a sensitive method (second-harmonic generation in the presence of an electric field) with which they are able to observe the electronic hyperpolarizability in 15 other molecules (including CCl<sub>4</sub>). From their data we conclude that  $\sigma < 0.01 \ \beta$  for CS<sub>2</sub>,<sup>15</sup> and so Eqs. (10) and (11) combine to give for the electronic fraction  $\sigma/(\sigma + \beta)$  of the Kerr constant of CCl<sub>4</sub> (at 6943 Å and 23 °C)

 $\sigma/(\sigma + \beta) = 0.54 \pm 0.16.$  (12)

We can derive from Eqs. (10) and (11) absolute values for  $\sigma$  and  $\beta$  of CCl<sub>4</sub> with the aid of the recent very accurate determination of Kerr constant of CS<sub>2</sub> by Volkova *et al.*,<sup>16</sup> who found it to be 3494  $\pm 4 \times 10^{-10}$  esu at 546 nm and 23 °C. Using the less accurate data of McComb to estimate the wavelength variation of this "constant," we deduce that it is  $253 \pm 5 \times 10^{-9}$  esu at 6943 Å and 23 °C.<sup>14</sup> Using refractive index values 1.62 and 1.46 for CS<sub>2</sub> and CCl<sub>4</sub>, respectively, the previously mentioned measurements imply that

$$\sigma + \beta = 100 \pm 4 \times 10^{-15} \text{ esu} \tag{13}$$

for CCl<sub>4</sub> at 6943 Å and 23  $^{\circ}$ C. (This value is well within the range of values deduced from the literature, and a little more accurate.)

#### IV. DISCUSSION

Maker *et al.*<sup>10</sup> and Wang<sup>9</sup> have also measured the ellipse-rotation constant for both liquids CS<sub>2</sub> and CCl<sub>4</sub> but used multimode beams. Their results give 32 and 34, respectively, for the ratio of Eq. (10) instead of 56 as we have found. (Wang's absolute value of  $\beta$  for CS<sub>2</sub> however agrees well with our value.) However, Wang believed his relative values for various liquids to be accurate only to roughly  $\pm$  25% and so the discrepancy is not surprising especially in view of the fact that CCl<sub>4</sub>, having a relatively weak ellipse rotation, was extremely susceptible to errors from self-focusing and other spurious effects.

One of us has derived a relation between the con-



FIG. 2. Typical graph of F vs optical power P. Unit abscissa corresponds approximately to an absolute power P=0.6 kW and the ordinate 10 corresponds to an angular ellipse rotation of  $\theta \simeq 2^{\circ}$ . Both of these are cross-sectional averages of uncertain precision. [From Eq. (4) it is to be noted that for each ray,  $F \propto \theta^2$  for small  $\theta$ .]

stant  $\beta$  and the total of the depolarized light scattering intensity for a classical liquid.<sup>4</sup> Although it was derived on the basis of a microscopic approximation (the linear dipole approximation), we have recently shown it to be a somewhat more general theorem than that derivation implies, and probably reliable to within a percent or so.<sup>11</sup> With the six (prelaser) measurements of the absolute depolarized light scattering intensity from liquid  $CCl_4$  and the existing Kerr data the theorem predicted that  $\beta/(\sigma + \beta)$  was between 0.36 and 0.60 for CCl<sub>4</sub>,<sup>4</sup> in almost perfect agreement with Eq. (12). However a laser measurement of the scattering intensity was about 50% greater than previous values.<sup>4</sup> There were also prelaser experimenters who disagreed with the scattering values for other liquids given by the sources of the  $CCl_4$  data.<sup>4</sup> Therefore, no definite conclusions could be drawn at the time. Our present results now lead us to believe that the standard prelaser literature values for both the depolarized scattering and Kerr constants of liquid CCl<sub>4</sub> are correct within their limits of error. Furthermore, the strengthening of the basis for the scattering-Kerr-effect relation now leads us to prefer the "high" values for the light scattering from CS<sub>2</sub> (and also from benzene) that are quoted many places in the literature in competition with a large number of experimentors who prefer "low" values (about 40% lower).<sup>11</sup>

Attempts to distinguish electronic and nuclear contributions to the Kerr constant of  $CCl_4$  (and other symmetric-molecule liquids) by studying its temperature dependence have proved inconclusive, mainly because one has no accurate way to calculate the dependence in such simple liquids.

One might hope that picosecond laser pulses could be produced short enough to "freeze out" nuclear motions and see only  $\sigma$ . From the frequency width of the depolarized scattering spectrum of liquid CCl<sub>4</sub>, one can see that the nuclear response time of the main central component is a good deal shorter than the shortest of present laser pulses (~1 psec). But even if this central component could be frozen out, the Raman lines which produce a nuclear contribution in around 10<sup>-13</sup> sec would still compete

<sup>†</sup>Research supported in part by the Air Force Office of Scientific Research and Clarendon Laboratory, Oxford, England.

<sup>‡</sup>Permanent address: Sandia Laboratories, Albuquerque, N. M. 87115. with the electronic mechanism, and thus picosecond pulse methods give little promise of distinguishing the electronic effect unambiguously.

Two methods have been used recently to measure the electronic hyperpolarizability of CCl<sub>4</sub> molecules in the vapor phase. Mayer<sup>5</sup> and Hauchecorne et al.<sup>6</sup> have measured the second harmonic produced by a ruby laser beam in the presence of a static electric field. Bogaard and Buckingham have measured the Kerr constant of CCl<sub>4</sub> vapor.<sup>7</sup> If we use these data in the standard theory of the electronic Kerr effect in liquids, with local-field corrections, <sup>1, 2</sup> we predict a value of  $\sigma$  for liquid  $CCl_4$  that is just below the lower limit of Eq. (12) from the second-harmonic measurements and a value just above the upper limit of Eq. (12) from the Kerr measurements. Considering that the standard theory of the Kerr effect for liquids errs commonly by a factor of 2 or more in predicting the nuclear contribution from vapor data, it is difficult to say more than that, though inconsistent with each other, neither of the above hyperpolarizabilities may be said presently to be inconsistent with our results.

Aside from third-harmonic generation, which has proven to be extremely difficult to calibrate in dense media, we know of no methods other than those which we have just discussed for distinguishing the electronic from nuclear mechanisms in the third-order nonlinear polarization of liquids. Now that the techniques for producing a single-Gaussianmode laser pulse have been developed, it would seem fairly evident from the foregoing discussion of these other methods that the comparison of Kerr and ellipse-rotation constants is the simplest and most accurate method presently available for comparing the electronic and nuclear contributions to these effects in isotropic media.

## ACKNOWLEDGMENTS

One of us (R.W.H.) would like to thank the Clarendon Laboratory, Oxford, for hospitality extended, and the National Science Foundation for support, during part of this work.

<sup>4</sup>R. W. Hellwarth, J. Chem. Phys. <u>52</u>, 2128 (1970).

<sup>5</sup>G. Mayer, Compt. Rend. <u>B267</u>, 54 (1968).

<sup>6</sup>G. Hauchecorne, F. Kerheve, and G. Mayer, J. Phys. (Paris) <u>32</u>, 47 (1968).

<sup>7</sup>M. P. Bogaard and A. D. Buckingham (private communication) have measured the Kerr constant of CCl<sub>4</sub> vapor, and deduced a value for the molecular hyperpolarizability  $\gamma$  of 9.96 ± 0.32 × 10<sup>-36</sup> esu.

2346

<sup>\*</sup>Present address: Clarendon Laboratory, Oxford, England. Permanent address: University of Southern California, Los Angeles, Calif. 90007.

<sup>&</sup>lt;sup>1</sup>W. Voight, Ann. Physik 4, 197 (1901).

<sup>&</sup>lt;sup>2</sup>T. H. Havelock, Proc. Roy. Soc. (London) <u>A80</u>, 28 (1907); <u>A84</u>, 492 (1911); P. Langevin, Radium <u>7</u>, 249 (1910); P. Debye, *Marx's Handbuch der Radiologie VI* Akademische Verlagsgesellschaft, Leipzig, 1925), Chap.

V; A. D. Buckingham and R. E. Raah, J. Chem. Soc. <u>2</u>, 2341 (1957); S. Kielich, Acta Phys. Polon. <u>30</u>, 683 (1966); and others.

<sup>&</sup>lt;sup>3</sup>N. George, R. W. Hellwarth, and C. R. Cooke, Electron Technol. (Poland) <u>2</u>, 229 (1969).

<sup>8</sup>A. Owyoung, R. W. Hellwarth, and N. George, Phys. Rev. (to be published).

<sup>10</sup>P. D. Maker, R. W. Terhune, and C. M. Savage,

Phys. Rev. Letters <u>12</u>, 507 (1964).

<sup>11</sup>R. W. Hellwarth (unpublished).

<sup>12</sup>H. Kogelnik, Appl. Opt. <u>4</u>, 1562 (1965).

<sup>13</sup>P. D. Maker and R. W. Terhune, Phys. Rev. <u>137A</u>, 801 (1965).

<sup>14</sup>Landult-Bornstein, Zahlenwerte and Functionen II/8, edited by K. H. Hellwege and A. M. Hellwege (Springer, Berlin, 1962), pp. 840-860.

<sup>15</sup>M. P. Bogaard, A. D. Buckingham, and G. L. D.

PHYSICAL REVIEW A

Kerr measurements on  $CS_2$  vapor to infinite temperature. Their value, with standard but unreliable theory, predicts that about a tenth the Kerr constant of liquid  $CS_2$  is electronic. However, the temperature extrapolation from data whose origin is primarily not electric would seem to be much less direct. If this extrapolation were correct however, we would have to alter the values in Eq. (12) to  $0.61 \pm 0.16$ . <sup>16</sup>Y. A. Volkova, V. A. Zamkoff, and L. V. Nalbandoff,

Ritchie, Mol. Phys. 18, 575 (1970), have deduced a value

for the molecular hyperpolarizability by extrapolating

<sup>10</sup>Y. A. Volkova, V. A. Zamkoff, and L. V. Nalbandoff, Opt. i Spectroskopiya <u>30</u>, 556 (1971).

VOLUME 4, NUMBER 6

DECEMBER 1971

Magnetic and Gravitational Energy Release by Resistive Instabilities<sup>\*</sup>

Mark A. Cross<sup>†</sup> and Gerard Van Hoven

Department of Physics, University of California, Irvine, California 92664 (Received 2 June 1971)

The resistive magnetohydrodynamic "tearing" and "gravitational-interchange" instabilities are investigated in the linear incompressible limit in the absence of heating. A periodic model is used for the initial magnetic field. The requirement that the spatial Fourier series of the perturbation must converge uniquely determines the growth rate of the instability. At the most unstable wavelength the tearing mode has a growth rate  $\omega \tau_r = 3.4S^{0.57}$ , where S is the magnetic Reynolds number at the Alfvén velocity and  $\tau_r$  is the resistive diffusion time. The interchange mode exhibits behavior, at small wavelengths and large gravitational fields, similar to that of the magnetohydrodynamic Rayleigh-Taylor instability. The resistive mode grows for gravitational fields smaller than the equivalent Suydam threshold for infinite-conductivity instability in a sheared magnetic field. Application of these results to the solar-flare problem is briefly discussed, in the context of the temporal behavior exhibited.

## I. INTRODUCTION

When one attempts to understand the instability responsible for solar flares one is led to the consideration of resistive mechanisms for the decoupling of magnetic fields and particles.<sup>1,2</sup> These processes allow a transformation into kinetic energy of the abundant magnetic and/or gravitational energy stored in the sunspot-connected fields of "active regions." The resistivity can be that due to collisions in the chromosphere or the higher turbulence-induced value resulting from the "flashphase" instability.<sup>3</sup>

The basic paper on resistive magnetohydrodynamic instabilities of this kind was written by Furth, Killeen, and Rosenbluth.<sup>4</sup> This massive work treated, in various analytically solvable limits, all of the possible linear resistive instabilities of a basic sheared magnetic field configuration. In addition, the relevance of the theory to laboratory experiments was discussed. The formulation and the solutions were expressed in a unique system of normalized variables which requires careful attention in interpretation. The sheer magnitude of the work and the specialized notation have generated errors, of partial quotation or misquotation, in later citations of the results obtained.

Furth *et al.*<sup>4</sup> were most successful in explaining the driving forces of the instability. However, in view of the manifold possible equilibrium configurations and of the many approximations needed to make the problem tractable analytically, they were not able to describe the general parameter dependence of the instability growth rates.

Wesson<sup>5</sup> treated these instabilities by solving their differential equations on a computer for several different equilibrium configurations of magnetic field and resistivity. He found that the growth rate of the tearing mode depends on wavelength in a different way from that described by Furth *et al.*, but showed that their central approximation is valid. He also investigated the gravitational interchange mode under conditions in which the tearing mode is stable.

Barston<sup>6</sup> put analytic limits on the growth rate in the case of uniform resistivity and uniform shear. Van Hoven and Cross<sup>7</sup> were able to remove the uniform shear condition in the case of the tearing mode. In this paper we investigate the tearing and gravitational modes by a new method which involves a computer search for the unique value of the growth rate which brings about the convergence of the Fourier series describing the per-

<sup>&</sup>lt;sup>9</sup>C. C. Wang, Phys. Rev. <u>152</u>, 149 (1966).