Measurement of nonlinear polarizability of air

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The results are reported of the measurement of the component χ_{1221} of the nonlinear susceptibility tensor of air by a method based on the effect of nonlinear precession of the polarization ellipse. A new procedure for accumulating the nonlinear signal is used and makes it possible to increase substantially the accuracy of the measurements. It is shown experimentally that the polarization parameters of Gaussian beams are preserved in weak self-focusing in a nonlinear medium with cubic nonlinear polarization. Relative measurements, accurate to 20%, are made of the χ_{1221} component for air under normal conditions. It is shown that the contribution of the electronic mechanism to the nonlinear polarization of air is negligibly small. An estimate is presented for the critical self-focusing power in air.

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The measurement of the components of nonlinear polarization of air and of gases is a timely problem, inasmuch as many experiments with powerful and superpowerful lasers are carried out in the atmosphere, and in the design and planning of the experiment it is apparently advantageous to take into account the nonlinearity of the gaseous medium or of atmospheric air. In addition, it is of interest to compare the results of laser experiments with nonlinear polarization of a gas medium calculated accurately using either data from independent experiments or with rigorous quantum-mechanical calculation.[1,2] Finally, results of direct measurements of the Kerr constant in gases at various pressures in a low-frequency electric field are presently known.[3-5] On the other hand, to our knowledge, the direct measurements of the linear susceptibility tensor in the optical band (e.g., with the aid of third-harmonic generation[6]) have yielded only estimates, and no measurements of the component χ_{1221} have been made at all to this day.

We analyze first the principal relations that make it possible to compare the results of various measurements. We start from the known phenomenological coefficients of the nonlinear susceptibility, introduced by Hellwarth[7] and making it possible to separate the various physical mechanisms of the nonlinearity in the analysis. Namely, in the Born-Oppenheimer approximation it is quite simple to separate the purely electronic part of the nonlinear polarizability from the electron-nuclear part. The electronic part, i.e., the one that follows instantaneously all the high-frequency field changes, is given by

$$\mathbf{P}^{e}={}^{1}/_{2}\sigma\mathbf{E}(\mathbf{E}\mathbf{E}), \qquad (1)$$

while the electron-nuclear symmetrical part takes the form

$$\mathbf{P}^{s}=\mathbf{E}(t)\int a(t-\tau)(\mathbf{E}(\tau)\mathbf{E}(\tau))d\tau, \qquad (2)$$

where $a(t-\tau)$ is the response function of the nuclei to the perturbing electro-magnetic fields.

Similarly, the electron-nuclear antisymmetrical part is

$$\mathbf{P}^{as}=\int\mathbf{E}(\tau)b(t-\tau)(\mathbf{E}(t)\mathbf{E}(\tau))d\tau, \qquad (3)$$

where $b(t-\tau)$ is the corresponding response function.

The spectral representation of the response functions can be expressed in the form[8]

$$B(\nu)=\int b(t-\tau)e^{i\nu\tau}d\tau=\sum_j\left(\frac{B_j}{\omega_j-\nu+i\Gamma_j}+\text{c.c.}\right) \qquad (4)$$

$$A(\nu)=\int a(t-\tau)e^{i\nu\tau}d\tau=\sum_j\left(\frac{A_j}{\omega_j-\nu+i\Gamma_j}+\text{c.c.}\right) \qquad (5)$$

where in the case $\omega_j\neq 0$ each term of the sum corresponds, generally speaking, to some line in the Raman-scattering spectrum, and the ratio of the complex quantities A_j and B_j can be obtained by measuring the depolarization of the corresponding Raman lines. At $\omega_j=0$ the expressions (4) and (5) describe the orientational contribution to the nonlinearity, and the corresponding parameters can be obtained from the spectra of the depolarized scattering of the Rayleigh line wing. Using the relations introduced above, we can obtain an expression for the polarizabilities measured in different experiments. For a linearly polarized electromagnetic field, the nonlinear increment to the refractive index, expressed in terms of the coefficients introduced above, takes the form

$$\Delta n=\langle E^2\rangle n_2=\pi n^{-1}[{}^{3}/_{2}\sigma+2B(0)+2A(0)]\langle E^2\rangle. \qquad (6)$$

Performing simple calculations for the case of two fields, namely a strong field

$$E_L=\vec{\mathscr{E}}_{xL}\cos(\omega_Lt+\varphi_L)$$

and a weak signal field

$$\mathbf{E}_s=\mathbf{e}_x\mathscr{E}_{xs}\cos(\omega_st+\varphi_{xs})+\mathbf{e}_y\mathscr{E}_{ys}\cos(\omega_st+\varphi_{ys}),$$

we obtain expressions for the nonlinear polarization:

$$P_x(\omega_s)=[{}^{3}/_{2}\sigma+A(0)+B(0)+A(\omega_s-\omega_L)+B(\omega_s-\omega_L)]E_{sx}\langle E_{xL}^2(t)\rangle, \qquad (7)$$

$$P_y(\omega_s)=[{}^{1}/_{2}\sigma+A(0)+{}^{1}/_{2}B(\omega_s-\omega_L)]E_{sy}\langle E_{xL}^2(t)\rangle. \qquad (8)$$

From the foregoing definitions it follows that for a monochromatic wave $E_x(t)=E_x\cos(\omega t-kz)$ the nonlinear increment to the refractive index is

$$\Delta n=n_2\frac{E_x^2}{2},\quad n_2=\frac{\pi}{n}\left(\frac{3}{2}\sigma+2B(0)+2A(0)\right).$$

Using (6) and (7), we easily obtain expressions for the high-frequency Kerr constant:

$$\mathscr{K}=\frac{n_\parallel-n_\perp}{\lambda_0\langle E_L^2\rangle}=\frac{2\pi}{\lambda_0 n}\left[\sigma+B(0)+A(\omega_s-\omega_L)+\frac{1}{2}B(\omega_s-\omega_L)\right]. \qquad (9)$$

For a molecular medium with nonpolar molecules the expression for the Kerr constant in a stationary field

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is

$$\mathcal{H}=\frac{2\pi}{\lambda_0 n}(\sigma+B(0)). \qquad (10)$$

Thus, investigations of the spontaneous-scattering spectra together with measurements of the Kerr constant in a constant field make it possible to calculate the high-frequency Kerr constant. Without dwelling in detail on the possible variants of the measurement of the phenomenological coefficients defined above, we note that experiments on third-harmonic generation make it possible to obtain the electronic part of the nonlinear polarizability σ, inasmuch as it follows from (1) that

$$P^e(3\omega)={}^1/_8\sigma\mathscr{E}^2\mathscr{E}\cos(3\omega t-3kz), \qquad (11)$$

where $\mathscr{E}$ is the amplitude of the initial wave at the frequency ω.

We note in addition that in those cases when the main contribution to the nonlinearity is made by orientation of the optically anisotropic molecules, the constants $A(0)$ and $B(0)$ are connected by the relation

$$-3A(0)=B(0)=\frac{N_0}{15}\frac{(\alpha_\parallel-\alpha_\perp)^2}{kT}, \qquad (12)$$

where $\alpha_\parallel$ and $\alpha_\perp$ are the principal polarizabilities of the axisymmetric molecule, and N_0 is their concentration.

The relations given above make it possible to determine the rate of nonlinear precession of an elliptically polarized monochromatic wave in a medium with cubic nonlinearity:

$$\frac{d\theta}{dz}=\frac{\pi\omega}{4nc}(\sigma+2B(0))\mathscr{E}_x\mathscr{E}_y=\eta_2\mathscr{E}_0^2\sin 2\Omega, \qquad (13)$$

where θ is the angle of rotation of the principal axes of the polarization ellipse; $\mathscr{E}_x$ and $\mathscr{E}_y$ are respectively the major and minor semiaxes of the polarization ellipse. The quantity $\sigma+2B(0)$ and the component χ_{1221} of the nonlinear polarization tensor are connected by the simple relation

$$\sigma+2B(0)=24\chi_{1221}.$$

Thus, the rotation constant is equal to

$$\eta_2=\frac{\pi\omega}{4nc}(\sigma+2B(0))=6\frac{\pi\omega}{nc}\chi_{1221}.$$

From the experimental point of view, measurements of the rotation of the polarization ellipse are quite simple, so that the measurements of the nonlinearity by this method yield the highest measurement accuracy, see, e.g., Ref. 9. On the other hand, in experiments one uses not a plane wave (although this is feasible in principle), but as a rule a smooth beam of a single-mode laser with an near-Gaussian intensity distribution. The laser beam is focused into the sample. The structure of the focused Gaussian beam has been investigated in detail. In particular, the intensity distribution is given by

$$\mathscr{E}^2(z,r_\perp)=\frac{\mathscr{E}_0^2}{1+z^2/z_0^2}\exp\left(-\frac{r_\perp^2}{w^2(z)}\right), \qquad (14)$$

where $\mathscr{E}_0$ is the amplitude of the field on the axis in the constriction of the beam, $z=kw_0^2/2$ is the radius of the diffraction divergence of the beam, and $w^2(z)=w_0^2(1+z^2/z_0^2)$. It is obvious that the precession rate of the polarization ellipse varies in the sample both with the coordinate z and with $r_\perp$. It is not quite obvious here whether the parameters of the polarization ellipse of an inhomogeneous beam are preserved when the nonlinearity and the diffraction act simultaneously.

We have considered within the framework of perturbation theory ($P_\perp \ll P_{cr}$) the nonlinear precession of a Gaussian elliptically polarized beam focused into the sample with simultaneous account taken of the nonlinearity and the diffraction.[13] In particular, in the asymptotic region, i.e., at $z_0 \ll l_2-l_1$, the polarization characteristics of the beam are the same at the entry and at the exit, i.e., the polarization ellipse is preserved, notwithstanding the inhomogeneous spatial distribution of the intensity, and consequently the inhomogeneous distribution of the rate of rotation of the polarization ellipse. For the nonlinear rotation angle of the polarization ellipse we obtain the expression

$$\theta={}^1/_2\pi\eta_2 z_0\mathscr{E}_0^2\sin 2\Omega. \qquad (15)$$

It is assumed in a number of experimental papers[9,7] that if one records the polarization ellipse parameters only in the region near the beam axis, then it is possible to calculate the phase advance or the corresponding equivalent rotation of the polarization ellipse within the framework of geometric optics, i.e., the corresponding axial rotation

$$\theta=\sin 2\Omega\int_{l_1}^{l_2}\eta_2\mathscr{E}^2(z,r_\perp)|_{r_\perp=0}dz$$

$$=\eta_2\mathscr{E}_0^2 z_0\left(\operatorname{arctg}\frac{l_1}{nz_0}-\operatorname{arctg}\frac{l_2}{nz_0}\right)\sin 2\Omega,$$

where l_2-l_1 is the sample length, and n is the refractive index in the sample. For the case of deep focusing of the radiation in the sample, $l_2-l_1 \gg z_0$, we get

$$\theta=\eta_2\mathscr{E}_0^2 z_0\pi\sin 2\Omega. \qquad (16)$$

Expression (16) does not take into account the diffraction mixing of the phase or of the polarization of the beam on passing through the constrictions. It is easily noted that this result differs from the correct result (15) by a factor of two.

The preservation of the ellipse parameters and the homogeneity of its rotation over the entire transverse distribution make it possible to realize an experimental procedure for measuring lenticular-waveguide type nonlinearities in which a nonlinear signal (in our case, the rotation angle of the polarization ellipse) is produced. The advantage of using such a scheme for measuring the nonlinearity in gases is due to the fact that the nonlinear phase advance or the corresponding polarization-ellipse rotation angle after passing through a single constriction is small in absolute magnitude, and is limited above by breakdown (laser spark). Increasing the constriction dimension, which in general makes it possible to increase the nonlinear signal, is not effective in our case, because, first, it necessitates a substantial increase of the dimensions of the installation, and second calls for increasing the signal power to a level greatly increasing the critical self-focusing power in the laser glasses of the amplifier; this in turn can distort substantially the geometric parameter of the

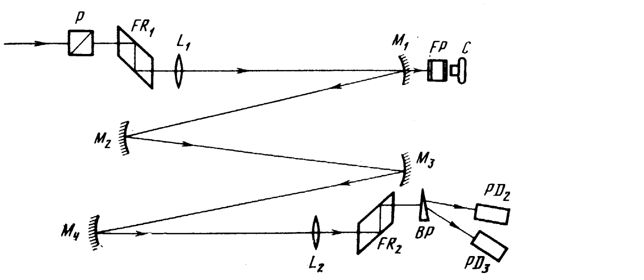

FIG. 1. Measurement setup: P—polarizer, FR_1 and FR_2—Fresnel rhombs, L_1 and L_2—lenses, M_1–M_4—spherical mirrors, FR—Fabry-Perot interferometer, C—photographic camera, BP—birefringent prism; PD_2 and PD_3—calorimeters.

initial quasi-Gaussian beam.

In our experiments we used as the radiation source a neodymium-glass laser with passive Q switching, operating on one axial and one angular (TM_{00}) modes, and two two-pass amplifiers. For better stability of the laser installation we used an automatic control system that made it possible a sequence of laser pulses with adjustable repetition frequency at periods from 2 to 10 min. The spatial distribution of the beam at the entry to the sample was smooth, and the profile of the intensity distribution was close to Gaussian.

The radiation from the laser setup was applied to the measurement system illustrated in Fig. 1. The laser radiation was focused by lens L_1 and by four spherical mirrors in tandem (M_1, M_2, M_3, M_4) so that the laser radiation passed in succession through five necks and was only then fed to the polarization analyser and the calorimeters PD_2 and Pd_3. In the experiments we used the standard[9] scheme for obtaining and analyzing the elliptic polarization. The scheme consisted of an input polarizer, a Fresnel rhomb FR_1 oriented at an angle 22.5° to the transmission plane of the input polarizer, and a similar unit (Fresnel rhomb FR_2 and output polarizer BP in the form of a spar wedge) at the exit from the system. The only difference between our polarization system and that described in Ref. 9 was that the radiation polarization analyzer was turned through a small angle $\theta_0 \lesssim 1°$ to the position of the total ($\sim 5\times 10^{-4}$) extinction in a direction opposing the nonlinear precession of the polarization ellipse. This organization of the experiment made it possible to separate unambiguously the nonlinear precession of the polarization ellipse, which increases the signal of the PD_3 calorimeter, from the nonlinear polarization of the radiation by the particles suspended in air and from other parasitic effects.

To check on the premise that the nonlinear signal is cumulative and that the necks are equivalent, an LGS-247 glass sample was placed in the first and fifth constrictions in tandem, and the corresponding nonlinear constant was measured in the glass by the usual method. The measurement results are shown in Fig. 2. These results show that the first and fifth constriction used in our experiment are equivalent with ~4%, thus fully confirming the correctness of the organization of the experiment.

To measure the nonlinear polarization of the air we

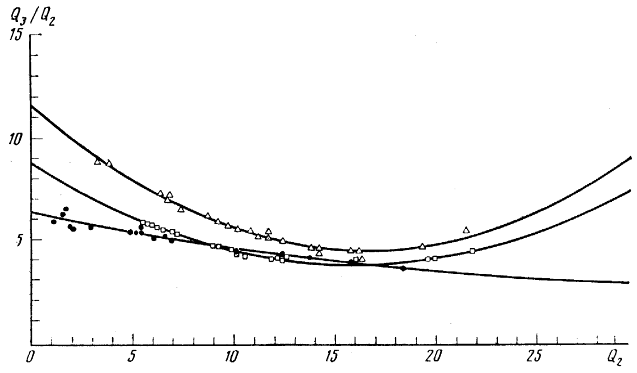

FIG. 2. Nonlinear signal registered by calorimeter PD_3 and normalized to the total radiation energy, as a function of the total energy passing through the radiation system (arbitary units): □—LGS-27 sample in the first constriction, △—the same sample in the fifth constriction, with different filters in front of the calorimeter PD_3; ●—air.

used a laser pulse of 30 nsec duration with maximum energy ~1 J. The distance between the waveguide mirrors was ~4 m at a mirror radius 2 m. The geometric parameters of the radiation intensity distribution near the constrictions was monitored by the method described in Refs. 10 and 11, and the deviations from the Gaussian distribution of the intensity within the half-width W of the Gaussian intensity distribution were negligible. The relative measurements of the component χ_{1221} for air yielded $\chi_{1221}^{air}/\chi_{1221}^{glass} = 1.25\cdot 10^{-3}$. The accuracy of the relative measurements, determined by averaging over several measurement runs, was 15%. The characteristic results of one measurement run on air are shown in Fig. 2 (dark circles). The curves were obtained by reducing the experimental points by least squares with a computer. Using the previously obtained value of χ_{1221}^{glass} for glass, namely 3.7×10^{-15} cgs esu, we obtain

$$\chi_{1221}^{air} = 4.6\cdot 10^{-18} \text{ cgs esu.}$$

On the basis of measurements performed in this study we can calculate, within the framework of certain assumptions, the critical self-focusing power for air. Using (6) and neglecting σ in accord with the results of Ref. 12, where it was shown that $\sigma = 5\times 10^{-15}$ cgs esu, i.e., it is negligibly small compared with $\sigma + 2B(0)$, we obtain

$$n_2^{air} = \pi[2B(0)+2A(0)]/n.$$

For further estimates we must measure $A(0)$, but if we assume that the main contribution to the nonlinear polarizability is due to the orientation of the optically anisotropic molecules, then $A(0) = -B(0)/3$ and

$$n_2^{air} \approx \frac{\pi}{n}\frac{4}{3}B(0) = 2.5\cdot 10^{-16} \text{ cgs esu.}$$

The critical self-focusing power is accordingly

$$P_{cr} \approx c\sqrt{\varepsilon_0}/2n_2k^2 \approx 1.6 \text{ GW.}$$

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High pressure plasma pinch under conditions of ion "free flight" conditions

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A plasma pinch in dynamic equilibrium with a surrounding dense gas is considered under conditions when the ion mean free path in the hot region of the pinch is much longer than the characteristic dimensions of the pinch. An ion flux that hinders the penetration of the neutral particles into this region is produced on the boundary of the hot region of the pinch so that in the ion "free flight" regime the energy losses of the electrons turn out to be lower than under the conditions of the diffusion regime. The structure of the plasma pinch is investigated.

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1. High-power high-frequency electromagnetic radiation can produce in a dense gas a plasma pinch in which the electron temperature is several orders of magnitude higher than the temperature of the gas surrounding the pinch. This interesting phenomenon was observed by Kapitza.[1] He obtained and investigated a pinch in gas at atmospheric pressure at a power input ≈20 kW. The pinch thickness was several millimeters, the length several centimeters, and the electron temperature $T_e \approx 3 \cdot 10^5 - 10^6$ K.

The electrons are heated in the pinch under conditions of the anomalous skin effect, and the containment of the high-temperature electrons is due to the presence in the boundary region of the pinch of a constant electric field that is the result of the separation of the charges. In this boundary region, called in Ref. 1 a double layer, the densities of both the hot electrons and of the neutral particles are high. The structure of the double layer influences strongly the energy lost by the hot electrons. If it is assumed[1] that the thickness of the double layer is of the order of the pinch diameter, then the power of the RF field is by far not enough to compensate for the hot-electron energy loss due to elastic collisions with the neutral particles and to the collisions with the cold electrons, which result from impact ionization. The structure of the pinch was investigated in Refs. 2 and 3 on the basis of the balance equations for the number of particles under conditions of the diffusion regime, when the mean free path of the ions between the collisions with the neutral particles is small compared with the characteristic dimensions of the pinch. In this case the thickness of the double layer turns out to be smaller by two or three orders of magnitude than the dimension of the pinch. The inelastic energy lost by the hot electron makes in this case a negligible contribution to the energy balance of the system. However, the energy lost by the hot electrons in collisions with the cold ones is nevertheless large: the power necessary for the pinch to exist in the diffusion regime is 10–30 times larger than the experimental value of the RF field power fed to the pinch.

The present paper deals with a plasma pinch under conditions when the ion mean free path in the hot region of the pinch exceeds the characteristic dimensions of the pinch. It appears that this is precisely the case realized in Kapitza's experiment,[1] since the regime of the ion "free flight" is energywise favored over the diffusion regime. The point is that under ion free flight conditions the neutral particle density in the entire pinch region where hot electrons are present is much less than the density of the gas far from the pinch. The reason is that the ions accelerated by the constant electric field produce on the boundary of the pinch a convection pressure that balances the action of the neutral gas surrounding the pinch. Beyond the limits of the hot region of the pinch the ions lose velocity as they collide with the neutral particles, the convection pressure of the ions decreases, the partial pressure of the neutral particles increases. Thus, the partial pressure of the hot electrons on the pinch boundary is transformed into convection pressure of the ions, which prevents the neutral particles from penetrating

