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Self-diffraction of CO_2-laser radiation in SF_6

L. T. BOLOTSKIKH, V. G. POPKOV, A. K. POPOV, V. M. SHALAEV
L. V. Kirensky Institute of Physics, USSR Academy of Sciences, Siberian Branch, SU-660036 and Krasnoyarsk University, Krasnoyarsk, USSR

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Self-diffraction of CO_2-laser radiation ($\lambda = 10.51\,\mu m$) has been observed in gaseous SF_6. Different orders of self-diffraction have been studied, up to the third. The amplification due to degenerate resonant multiphoton parametric processes has been measured to be as large as eight. The experimental estimates have been derived for the effective nonlinear susceptibilities (coefficients of nonlinear interaction) of the third, fifth, and seventh orders. The self-diffraction may essentially affect nonlinear processes including optical phase conjugation.

1. Introduction

In a nonlinear medium irradiated by two light beams of equal frequencies and at an angle θ with respect to each other energy transfer may occur from one wave to another (see, e.g. [1, 2]). This can also give rise to new light beams propagating at angles $\pm\theta$, $\pm 2\theta$, . . . to the incident ones [3–6]. The appearance of the new beams may be accounted for by both a self-diffraction from the laser-induced amplitude-phase grating and a multiphoton parametric interaction between different waves. Physically, both approaches are equivalent and within a certain limit yield similar results [7].

The interest in the interaction between two waves propagating at a small angle with respect to each other is associated with the problem of obtaining dynamic holograms [8] used in optical phase conjugation (OPC) [8, 9]. Investigation of OPC of CO_2-laser radiation using vibrational–rotational molecular transitions is of particular importance [10–15]. In a conventional OPC experiment one has an induced signal wave and one of the pump waves travelling at a small angle θ. Therefore, generation of a reflected PC wave can be accompanied by self-diffraction of the signal beam and by creation of the wave travelling at the angle $-\theta$ with respect to the pump beam. As a result of the six-wave interaction, the OPC efficiency can essentially be improved [16]. Thus, to optimize OPC of infrared radiation from a CO_2 laser the self-diffraction effects have to be taken into account.

2. Experimental

A single-mode pulsed (TEM_{00q}) CO_2-laser radiation tuned with a diffraction grating was used. The output pulse consisted of a narrow peak on a wide background with a duration of $\tau_{0.5} = 100$ ns and $\tau_{0.1} = 1.5\,\mu s$.

A NaCl plate split the CO_2-laser radiation into two beams. One of these formed the A_0 wave and the other, of lower intensity, formed the A_θ wave. The beams were directed into a cell ($L = 2.5$ cm) with a molecular gas SF_6. The angle θ between the beams could be varied from 5×10^{-2} to 8.6×10^{-3} rad. The input intensities were $I_0 \simeq 8\,MW\,cm^{-2}$ and $I_\theta \simeq 230\,kW\,cm^{-2}$. The power was monitored by photon-drag detectors ($\tau_{const} = 1$ ns) combined with broadband amplifiers ($\Delta\nu \simeq 100$ MHz). The stimulated parametric forward scattering (SPS) generated new light beams in the resonant nonlinear medium (see Fig. 1). The SPS efficiency studied as a function of the angle θ exhibited an inverse dependence on it. At $\theta = 8.6 \times 10^{-3}$ rad the weak beam A_θ was amplified and the new beams $A_{-\theta}$, $A_{\pm 2\theta}$, $A_{\pm 3\theta}$ could be observed to arise over a wide pressure range of SF_6. The pressure dependences of the gain (η_θ) and the conversion efficiency ($\eta_{-\theta}$, $\eta_{\pm 2\theta}$, $\eta_{\pm 3\theta}$) of the A_θ beam to the $A_{-\theta}$, $A_{\pm 2\theta}$, $A_{\pm 3\theta}$ beams are presented in Fig. 2 for the $P(12)$ ($\lambda = 10.51\,\mu m$) CO_2-laser

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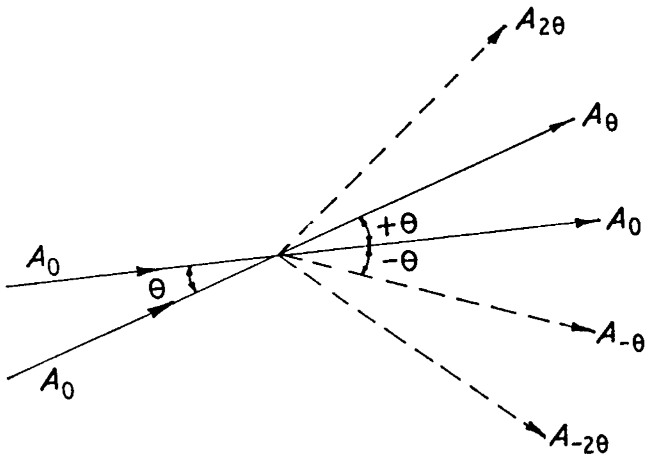

Figure 1 Experimental geometry.

line. The gain and the conversion efficiency were measured as ratios of the output beam powers to the input power of the A_0 beam. Even at a SF_6 pressure of 15 torr the weak beam A_θ was amplified and an additional beam could be observed at $-\theta$ angle. A further increase of the gas pressure stimulated the appearance of the beams at $\pm 2\theta$, $\pm 3\theta$ angles. As seen from Fig. 2, the output power exhibits a strong dependence on the SF_6 pressure with a maximum at about 30 torr. At low pressures ($P < 30$ torr) the output power grows because of the increased number of the molecules involved in SPS process. At high pressure ($P > 30$ torr) the major limitation on the conversion efficiency η is imposed by the pump absorption. The joint contribution of these two processes results in the pressure dependence of η obtained.

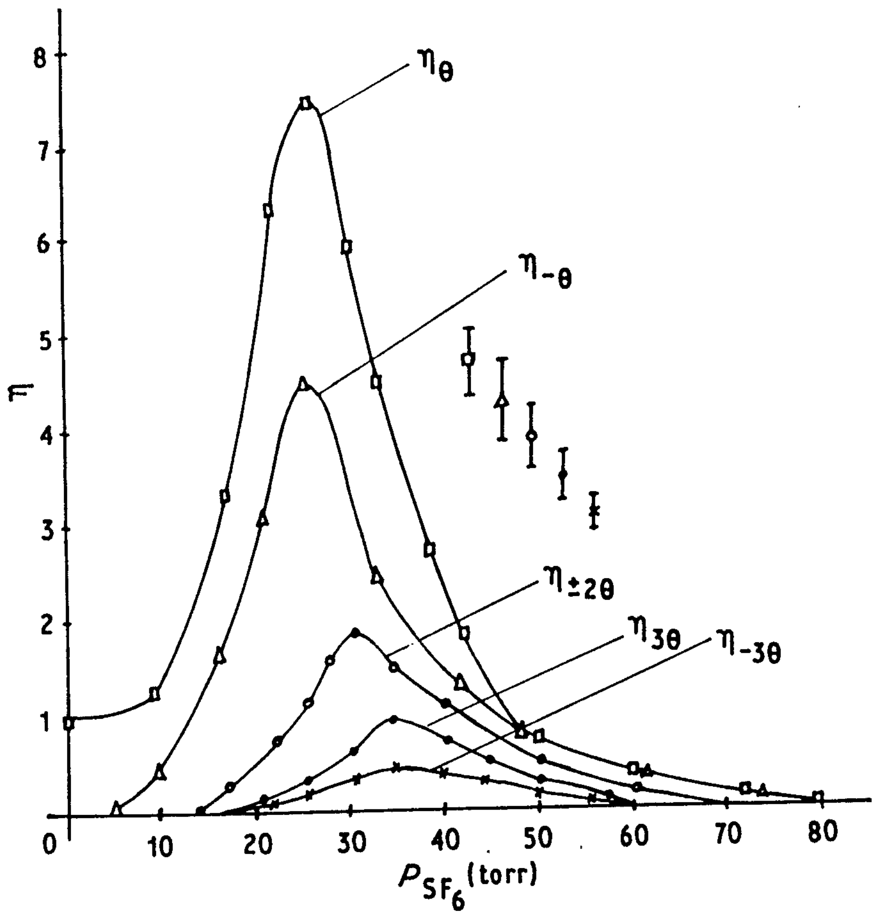

Figure 2 SF_6 pressure dependence of the gain η_θ and conversion efficiencies $\eta_{-\theta}$, $\eta_{\pm 2\theta}$, $\eta_{\pm 3\theta}$ of the A_θ beam to the $A_{-\theta}$, $A_{\pm 2\theta}$, $A_{\pm 3\theta}$ beams for the $P(12)$ ($\lambda = 10.51\ \mu m$) CO_2 laser line. $\theta \simeq 8.6 \times 10^{-3}$ rad; $I_0 = 8\ MW\,cm^{-2}$.

Figure 3 A picture of the spatial distribution of the $A_{-3\theta}$, $A_{-2\theta}$, $A_{-\theta}$, A_0, A_θ, $A_{2\theta}$, $A_{3\theta}$ beams originating from SPS of CO_2 laser radiation in SF_6 gas.

In Fig. 3 a picture of the spatial radiation distribution in the distant zone is given. The seven detected beams, $A_{-3\theta}$, $A_{-2\theta}$, $A_{-\theta}$, A_0, A_θ, $A_{2\theta}$, $A_{3\theta}$ (from left to right) were produced by SPS of CO_2-laser radiation in molecular SF_6 at a pressure of 34 torr. The picture was obtained by a thermal sensitization of the emulsion [17]. The effect is based on the absorption of the infrared radiation in the emulsion and a subsequent exposure of the film to visible light [18]. Infrared irradiated spots on the negative show a more intense blackening compared with those exposed only to the visible light and correspond to infrared images of the beams. We used a camera "Zenit TTL" with the lens replaced by a device similar to that reported in [17].

The pictures of the beams were taken in the focal plane of the lens ($F = 125$ mm) and the lens placed 80 mm from the cell. Photometering of the images obtained revealed a smaller diameter of the central pump beam spot compared with that of the A_θ beam. This was probably owing to self-focusing of the pump beam at $P_{SF_6} = 34$ torr. Spatial distribution measurements of the pump beam which passed through the medium showed that its diameter at the $1/e$ level changes from 3.7 mm for $P_{SF_6} = 0$, to 0.8 mm for $P_{SF_6} = 34$ torr. No self-focusing of the beam A_θ was observed in the experiment.

At high pressures the fundamental A_0 is not only absorbed, but also substantially depleted because of the A_0 wave converted to output radiation. Thus, the ratio $I_0/I_\theta \sim 30$ measured at the cell entrance at lower pressures, $P \lesssim 10$ torr, remained almost the same at the exit, whereas at $P \simeq 34$ torr the output pump power was measured to be 1.4 times lower than that of the A_θ wave.

When studying the SPS dependence on the pump beam wavelength the maximum scattering efficiency was observed for a $P(12)$ CO_2-laser line. An optimum value of the gas pressure was found to exist for each generation line and intensity value. A similar behaviour of PC reflectivity was reported in [13, 14].

Besides SPS, the process of spontaneous parametric scattering was observed in the experiment when only one strong pump beam was supplied to the cell with SF_6 gas. Unlike SPS processes which developed in a pressure range of 5–80 torr, the maximum spontaneous parametric scattering occurred in a pressure range of 40 to 75 torr. In the pressure range of the maximum SPS signals (about 30 torr) the signal from the spontaneous parametric scattering appeared to be negligible.

We proceed to analyse SPS processes at low pressures where the pump absorption, self-focusing and depletion due to conversion may be neglected.

3. Theory

Consider a simultaneous interaction of infrared radiation

$$E = \tfrac{1}{2}e^{i\omega t}\{A_0 e^{-ik_0\cdot r} + A_\theta e^{-ik_\theta\cdot r} + A_{-\theta}e^{-ik_{-\theta}\cdot r} + A_{2\theta}e^{-ik_{2\theta}\cdot r} + A_{-2\theta}e^{-ik_{-2\theta}\cdot r} + \ldots\} + \text{c.c.} \tag{1}$$

with a nonlinear gaseous medium.

The expression for the electromagnetic field E consists of the waves with non-zero amplitudes, A_0 and A_θ, at the cell entrance as well as the waves $A_{-\theta}, A_{\pm 2\theta}, \ldots$ arising inside the gas-phase medium owing to the nonlinear parametric interaction (see Fig. 1).

We assume $A_0 \gg A_\theta$. For the pulse duration τ much longer than the inverse transition halfwidth $\Gamma^{-1}(\tau\Gamma \gg 1)$ the expression for the polarization causing the waves $A_{-\theta}, A_{-2\theta}, A_{-3\theta}, \ldots$ can generally be written as

$$P = \tfrac{1}{2}e^{i\omega t}\{\bar{\chi}^{(3)}A_0^2A_\theta^* e^{-i(2k_0-k_\theta)\cdot r} + \bar{\chi}^{(5)}A_0^3A_\theta^{*2} e^{-i(3k_0-2k_\theta)\cdot r} + \bar{\chi}^{(7)}A_0^4A_\theta^{*3} e^{-i(4k_0-3k_\theta)\cdot r} + \ldots\} + \text{c.c.} \tag{2}$$

We neglect cascade generation of the waves $A_{-2\theta}, A_{-3\theta}, \ldots$ (generation due to a sequence of low-order nonlinear processes), which is confirmed by the estimations.

Some remarks should be made concerning the coefficients $\bar{\chi}^{(j)}$. A generalized susceptibility is introduced by Fourier transformation of the response function [19], and is by definition time independent. If coefficient $\bar{\chi}^{(j)}$ in Equation 2 depends on t it cannot strictly be referred to as the susceptibility of the corresponding order j. Therefore, after [20], we shall denote these values as $\bar{\chi}^{(j)}$ (with a bar above). Generally $\bar{\chi}^{(j)}$ are thought of as proportionality coefficients for the corresponding interaction. For example, $\bar{\chi}^{(3)}$ is the four-photon parametric interaction coefficient.

The first, second and third terms in Equation 2 are responsible for generation of the $A_{-\theta}$, $A_{-2\theta}$, $A_{-3\theta}$ waves, respectively. As an example, consider the physical sense of the third term in Equation 2. In the given form it determines the appearance of the $A_{-3\theta}$ wave due to the scattering of four photons of the strong pump beam A_0 into three photons of the A_θ wave and one photon of the $A_{-3\theta}$ wave, i.e. it corresponds to an eight-photon parametric interaction. In terms of the grating description this corresponds to the diffraction of the pump beam A_0 from the population grating at the -3θ angle. The spatial modulation of the grating is determined by the factor $A_0^3A_\theta^{*3}\exp[-3i(\boldsymbol{k}_0 - \boldsymbol{k}_\theta)\cdot \boldsymbol{r}]$, i.e. its period Λ is equal to $2\pi/3|\boldsymbol{k}_0 - \boldsymbol{k}_\theta|$. The period of the grating scattering the pump beam at -3θ is 3 and 1.5 times smaller than the grating scattering at $-\theta$ and -2θ angles, respectively.

The new waves $A_{-\theta}$, $A_{-2\theta}$, $A_{-3\theta}$ generated in the medium interact with the A_0 and A_θ waves to give rise to the waves $A_{2\theta}, A_{3\theta}, \ldots$.

At a sufficiently low gas pressure when the change of A_0 and A_θ along the medium may be neglected, substituting the expression for P into Maxwell's equation we obtain the expression for the conversion efficiency of the wave at θ to the wave at $-\theta$

$$\eta_{-\theta} = \frac{I_{-\theta}(L)}{I_\theta(0)} = 4\pi^2k_0^2\left(\frac{8\pi}{c}\right)^2 I_0^2|\bar{\chi}^{(3)}|^2L^2\,\text{sinc}^2\left(\frac{\Delta k_\theta L}{2}\right) \tag{3}$$

where $I_{\pm\theta} = c/8\pi|A_{\pm\theta}|^2$; $\Delta k_\theta = \boldsymbol{k}_\theta + \boldsymbol{k}_{-\theta} - 2\boldsymbol{k}_0$; $\boldsymbol{k}_0, \boldsymbol{k}_{\pm\theta}$ are the wave vectors; L is the resonant medium length. Note that the phase of the complex amplitude $A_{-\theta}$ is conjugate with respect to A_θ. By a backward reflection from ordinary mirrors one can obtain a phase-conjugate wave.

If

$$\kappa L \gtrsim 1 \tag{4}$$

where

$$\kappa = \left(|\gamma|^2 - \left(\frac{\Delta k_\theta}{2}\right)^2\right)^{1/2}, \qquad \gamma = 2\pi k_0\bar{\chi}^{(3)}A_0^2 \tag{5}$$

the wave A_θ is amplified along the medium and depends on z as

$$A_\theta(z) = \tfrac{1}{2}A_\theta(0)\exp\left[i\frac{\Delta k_\theta}{2}z\right]\exp[\kappa z]\left(1 - i\frac{\Delta k_\theta}{2\kappa}\right) \tag{6}$$

Using Equations 6 and 2 in Maxwell's equation one has

$$\eta_{-2\theta} = \frac{I_{-2\theta}(L)}{I_\theta(0)} = \frac{\tfrac{1}{4}\pi^2k_0^2\left(\frac{8\pi}{c}\right)^4 I_0^3I_\theta(0)}{4\kappa^2 + (\Delta k_{-2\theta} - \Delta k_\theta)^2}\frac{|\gamma|^4}{\kappa^4}\exp[4\kappa L]|\bar{\chi}^{(5)}|^2 \tag{7}$$

$$\eta_{-3\theta} = \frac{I_{-3\theta}(L)}{I_\theta(0)} = \frac{\frac{1}{16}\pi^2 k_0^2 \left(\frac{8\pi}{c}\right)^6 I_0^4 I_\theta^2(0)}{9\kappa^2 + (\Delta k_{-3\theta} - \frac{3}{2}\Delta k_\theta)^2} \frac{|\gamma|^6}{\kappa^6} \exp\,[6\kappa L] |\bar{\chi}^{(7)}|^2 \quad (8)$$

where $\Delta k_{-2\theta} = k_{-2\theta} + 2k_\theta - 3k_0$, $\Delta k_{-3\theta} = k_{-3\theta} + 3k_\theta - 4k_0$; $I_{-2\theta} = (c/8\pi)|A_{-2\theta}|^2$, $I_{-3\theta} = (c/8\pi)|A_{-3\theta}|^2$ are the intensities of the waves at -2θ and -3θ.

Theoretical formulae for conversion efficiencies, Equations 3, 7 and 8, have been derived by substituting Equation 2 for polarization into Maxwell's equation. Equation 2 represents a general form (if $\bar{\chi}^{(3)}$, $\bar{\chi}^{(5)}$, $\bar{\chi}^{(7)}$ are not specifically defined) and is independent of the microscopic model of radiation interaction with molecules. This only indicates the type of parametric wave scattering. The coefficient $\bar{\chi}^{(j)}$ indicates how intense the process is. Thus, the above formulae (Equations 3, 7 and 8) are independent of the microscopic model and have a general character. These formulae when compared with the experimental results enable us to estimate the values $\bar{\chi}^{(j)}$ for the given frequency and intensity of laser radiation.

Solving the set of kinetic equations for the density matrix within a strong collisonal model, $\bar{\chi}^{(j)}$ can be found in an explicit form. In general, the interaction of infrared radiation with multi-atomic molecules depends on the number of relaxation collisional processes, such as: collisions characterized by the frequency ν_R and resulting in rotational relaxation and a Boltzmann distribution $W_B(J)$ over the rotational levels J; intramode energy VV-exchange with frequency ν_{VV}; intermode vibrational energy VV'-exchange; and energy VT-exchange between vibrational and translational degrees of freedom with frequency ν_{VT}.The intramode energy exchange proceeds with a substantially higher probability than the intermode vibrational energy exchange [21]. Assuming

$$\nu_{VT}\tau \ll 1, \qquad \nu_R\tau \gg 1 \quad (9)$$

(τ is the pulse duration) and solving the set of equations with regard to the above relaxation processes for the relation of different-order j coefficients, $\bar{\chi}^{(j)}$, we obtain

$$\frac{\bar{\chi}^{(3)}}{\bar{\chi}^{(5)}} = \frac{\bar{\chi}^{(5)}}{\bar{\chi}^{(7)}} = -\frac{1 + 2R_0T}{c\sigma T/4\pi\hbar\omega} \quad (10)$$

where $R_0 = (c/8\pi)|A_0(t)|^2\sigma/\hbar\omega$, and σ is the transition cross-section. Depending on the value $\nu_{VV}\tau$ the expression for T has the form

$$T = \nu_R^{-1}, \qquad \text{if } \nu_{VV}\tau \ll 1 \quad (11)$$

$$T = \frac{1 + \frac{\nu_R}{\nu_{VV}} \sum_{i=1}^{k} W_B(J_i')}{\nu_{VV} + \nu_R}, \qquad \text{if } \nu_{VV}\tau \gg 1 \quad (12)$$

In deriving Equations 10 to 12 we assumed the infrared radiation to interact simultaneously with k vibrational–rotational transitions (of approximately equal cross-sections σ) $0J_i' \leftrightarrow 1J_i$ ($i = 1, 2, \ldots k$) between different rotational sublevels of a vibrational molecular transition 0–1.

4. Discussion

Parametric wave interaction discussed in Section 3 is effective only provided the phase-matching conditions are satisfied: $\Delta k_{\beta\theta} L < 2\pi$ ($\beta = 1, 2, 3$). Considering that $\Delta k_{\beta\theta} \simeq k(\beta\theta)^2$ this requirement may be expressed

$$L\lambda/\Lambda_\beta^2 < 1 \quad (13)$$

where $\Lambda_\beta \simeq \lambda/\beta\theta$ is the period of the grating diffracting the A_0 beam. Inequality 13 corresponding to our experimental conditions is typical of 'thin holograms' [22]. Thus, phase-matched parametric wave scattering corresponds to light diffraction from 'thin holograms'.

According to the experimental dependence (Fig. 2), at a SF_6 pressure of $P = 10$ torr the change of the A_0 wave in the medium may be neglected. At $P = 10$ torr the absorption of the A_0 wave was also small (only about 10%) which is obviously due to saturation of the absorption coefficient at $I_0 = 8\,\mathrm{MW\,cm^{-2}}$. Thus, the value $\bar{\chi}^{(3)}$ can be derived from Equation 3. The value of $\eta_{-\theta}$ for $P = 10$ torr is measured to be as large as 0.42. Substituting this value into Equation 3 and taking $k_0 \simeq 6 \times 10^3\,\mathrm{cm^{-1}}$, $I_0 = 8\,\mathrm{MW\,cm^{-2}}$, $L = 2.5\,\mathrm{cm}$, $\Delta k_\theta L = 1.11$, we obtain for the value of $|\bar{\chi}^{(3)}|$ corresponding to the maximum conversion efficiency in time at $P = 10$ torr

$$|\bar{\chi}^{(3)}| \sim 10^{-11}\,\mathrm{esu} \tag{14}$$

This value is commensurable with that of the susceptibility $\chi^{(3)}$ in semiconductors determining the reflectivity at optical phase conjugation [23] and is greater by many orders of magnitude than for the THF of CO_2-laser radiation in SF_6 at an appropriate pressure [24].

At a SF_6 pressure of $P = 15$ torr, as seen from Fig. 3, $\eta_\theta \simeq 2.5$, i.e. the change of the A_θ wave in the medium has to be taken into account. The experimental data analysed at $P = 15$ torr yielded $2\kappa L = 1.6$ and the $\bar{\chi}^{(3)}$ value of the order of that in Equation 14.

We now estimate the value of $\bar{\chi}^{(5)}$. As follows from measurements and calculated results, at $P = 15$ torr a major contribution to the wave $A_{-2\theta}$ is expected from the six-photon parametric scattering. Since for $P = 15$ torr one has $2\kappa L = 1.6$, the requirement of Equation 4 is sufficiently satisfied and the conversion efficiency $\eta_{-2\theta}$ is determined by Equation 7. Substituting all the required values into Equation 7 at $P = 15$ torr, we have for $\bar{\chi}^{(5)}$ ($\eta_{-2\theta} \simeq 0.12$)

$$|\bar{\chi}^{(5)}| \sim 10^{-15} \text{ to } 10^{-16}\,\mathrm{esu} \tag{15}$$

The minimum SF_6 pressure necessary to detect the signal $I_{-3\theta}$ is as high as 22 torr. At this pressure absorption and depletion of the pump radiation and its self-focusing have been observed which are not included in Equation 8. Therefore, to calculate $\bar{\chi}^{(7)}$ we use Equation 10.

In SF_6 the typical relaxation times of the vibrational states for different relaxation processes are as follows: $v_R^{-1}P \simeq 35$ ns torr [25], $v_{VV}^{-1}P \simeq 31$ ns torr [26], $v_{VT}^{-1}P \simeq 122\,\mu$s torr [27]. Since the experimental pressure was about 10 to 60 torr and the pulse duration of the order of 100 ns, the Conditions 9 may be considered satisfied. At $P \sim 10$ torr one has $T \sim 1$ ns (see Equations 11 and 12) which corresponds to $R_0 T \sim 10$ ($\sigma = 8 \times 10^{-18}\,\mathrm{cm^{-2}}$). Finally, for the relaxation between different-order coefficients we find

$$\frac{\bar{\chi}^{(3)}}{\bar{\chi}^{(5)}} \sim \frac{\bar{\chi}^{(5)}}{\bar{\chi}^{(7)}} \sim 10^5\,\mathrm{esu} \tag{16}$$

Note that the experimental values $\bar{\chi}^{(3)}$ and $\bar{\chi}^{(5)}$ (see Equations 14 and 15) agree with the theoretical estimate in Equation 16.

Using Equation 15 and 16, the magnitude $\bar{\chi}^{(7)}$ can be evaluated

$$|\bar{\chi}^{(7)}| \sim 10^{-20} \text{ to } 10^{-21}\,\mathrm{esu} \tag{17}$$

The high values of $\bar{\chi}^{(j)}$ obtained for different multiphoton processes are favoured by the resonant character of the radiation interaction with gas. Thus, resonant multiphoton parametric processes involved in forward scattering arrangement may affect other nonlinear processes. In particular, they should be taken into account in resonant optical phase-conjugation studies.

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