

# Resonant four-photon parametric processes in Ne in the field of a single-frequency He-Ne laser, and their use in nonlinear spectroscopy

Im Thek-de, O. P. Podavalova, A. K. Popov, and G. Kh. Tartakovskii

*L. V. Kirenskiĭ Physics Institute, Siberian Division, USSR Academy of Sciences*

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A heterodyning method was used to register parametric radiation at the frequency  $\omega_\nu = 2\omega_0 - \omega_\mu$ . All the frequencies lie within the natural linewidth. The feasibility of using this process for nonlinear spectroscopy of ultrahigh resolution is demonstrated.

1. We investigated four-photon parametric processes within the natural and collision line widths of an electronic transition of an atom. The effect is observed in the field of a weak highly-monochromatic laser. Resonances made it possible to realize nonlinear susceptibilities  $\chi^3 \sim 10^{-7}$  cgs esu units, exceeding by nine orders of magnitude the nonresonant susceptibilities in a solid. The resonance of the interaction has resulted in large growth rates and absorption rates of the waves. We investigated simultaneously the dependence of the absorption of a weak field in the presence of a strong one. A strong dependence of the components of the nonlinear susceptibility tensors on the wave polarization, on the collision effect, and on the axial magnetic field was registered. The experimental results are compared with the theory. A new method of ultra high-resolution nonlinear spectroscopy, based on resonant four-photon parametric processes is proposed.

2. Quasiresonant four-photon parametric processes are customarily investigated in fields of high-power pulse lasers having complicated spectral-temporal structures.<sup>[1,2]</sup> The recently developed cw dye lasers with relatively narrow spectra, have made it possible to observe a number of interesting strongly nonlinear effects in the wings of the Doppler resonances.<sup>[3]</sup> Low-power gas lasers can produce radiation of hitherto unattained monochromaticity. The present study has demonstrated the feasibility of studying strongly nonlinear resonant interactions in the field of such a laser and of realizing nonlinear ultrahigh-resolution spectroscopy on the basis of nonlinear processes that have not been used for this purpose before.

3. The four-photon parametric processes were ex-

cited by mixing two waves with frequency difference  $\Delta = \omega_0 - \omega_\mu$ . The beats of the population differences at the frequency  $\Delta$  lead both to a change in the polarization of the medium (absorption) at the frequency  $\omega_0 - \Delta = \omega_\mu$ , and to the appearance of polarization (parametric radiation) at a new frequency  $\omega_\nu = \omega_0 + \Delta$ . This radiation was registered by heterodyning with a stronger field at the frequency  $\omega_\mu$ . A heat signal  $I_{\nu\mu}$  is produced at the frequency  $|\omega_\mu - \omega_\nu| = 2\Delta$ . This method has made it possible to register parametric-radiation powers on the order of  $10^{-9}$  W. The experimental setup is shown in Fig. 1. Linearly-polarized radiation of a single-frequency He-Ne laser ( $\lambda = 0.63 \mu$ ) with mode selection by pressure and with output power 1.5 mW was broken up with the aid of a system of mirrors 2, 3, and 5 into two waves of different intensity. The strong field  $E_0$  was reflected from the mirrors 4 that were set in motion with the aid of a sawtooth voltage, and acquired a Doppler frequency shift  $\Delta = 32$  Hz relative to the weak field. Thus, two strongly collinear waves were produced at the entrance of cell 7 with the aid of mixing plate 6. The frequencies and phases of these waves were rigidly interconnected. The cell had a length  $\lambda = 60$  cm, an inside diameter 3.5 mm, and was filled with Ne, which was excited with a direct-current discharge. The wave polarizations were

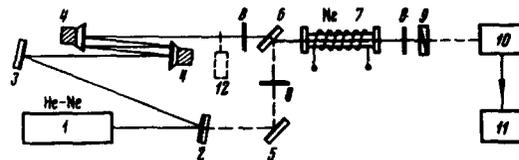


FIG. 1.

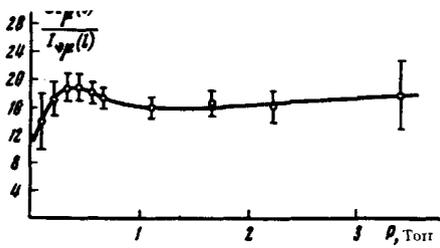


FIG. 2.

regulated with the aid of phase plate 8. A polaroid 9 has made it possible to block the strong field  $E_0$  and to pass the weak fields  $E_\mu$  and  $E_\nu$ , which were registered with a square-law photodetector 10 and a selective amplifier 11 tuned to frequency  $2\Delta = 64$  Hz. While the nonlinear increment to the field absorption  $E_\mu$  was registered, the mirrors 4 remained stationary, and the strong field was modulated with the aid of a chopper at a frequency 64 Hz. The signal of amplifier 11 was proportional in this case to the change  $\delta I_\mu$  in the intensity of the transmitted field  $E_\mu$ . By varying the Ne pressure in the cell 7 and the magnetic field  $H$  applied to it, it is possible to investigate the influence of the collisions and of the screening of the degenerate levels on the nonlinear effect.

4. The equations for the slow complex amplitudes of the interacting waves  $E_j^\alpha$  ( $j=0, \mu, \nu$ ) can be represented in the form

$$2 \frac{dE_j^\alpha}{dz} = -\Lambda E_j^\alpha + \theta_j^\alpha \delta_\gamma \beta E_0^\delta E_\nu^{\delta\gamma} E_\mu^{\beta\gamma} + \delta_{j\nu} \delta_{\alpha+\beta} \delta + \gamma \sigma_\nu^\alpha \delta_\gamma \beta E_0^\delta E_\nu^{\delta\gamma} E_\mu^{\beta\gamma} \quad (1)$$

Here  $\alpha, \beta, \gamma, \delta = \pm 1$  are the indices of the circular polarizations,  $\Lambda$  is the linear-absorption index,  $\hat{\theta}$  is the tensor of the nonlinear susceptibility and is responsible for the change in the absorption of the weak field  $E_\mu$  in the presence of a strong field  $E_0$ , and  $\hat{\sigma}$  is responsible for the parametric generation. Summation over repeated upper and lower indices is implied. The second Kronecker symbol reflects the law of conservation of the total angular momentum of the photons in the absence of coherent collisions of the atoms with momentum transfer. For perpendicular polarizations of  $E_0$  and  $E_\mu$ , the solution of (1) takes the form

$$I_{\nu\mu} = \frac{c}{8\pi} |E_\mu(0)|^2 \frac{F_2}{2(\Lambda - F_0)} \left[ 1 - \exp\{-(\Lambda - F_0)l\} \right] \times \exp\{-(\Lambda - F_1)l\};$$

$$\delta I_\mu = \frac{c}{8\pi} |E_\mu(0)|^2 \exp\{-\Lambda l\} [\exp\{F_1 l\} - 1];$$

$$F_0 = (\theta_{0111}^1 + \theta_{0-1-1+1}^1) \frac{|E_0(0)|^2}{2};$$

$$F_1 = (\theta_{\mu 111}^1 + \theta_{\mu-1-1+1}^1 - \theta_{\mu+1-1-1}^1) \frac{|E_0(0)|^2}{2};$$

$$F_2 = (\sigma_{1111}^1 - \sigma_{1+1-1-1}^1) \frac{|E_0(0)|^2}{2}.$$

The tensors  $\theta$  and  $\sigma$  can be calculated in the "strong" collision model<sup>[4]</sup> with the aid of the density matrix. For a degenerate transition with total angular momenta  $J_m = 1 \rightarrow J_n = 2$ , corresponding to the investigated Ne transition, the calculation yields

$$\frac{F_1}{F_2} = \frac{23}{6} + \frac{25}{3} \frac{\sum_{j=m,n} \left\{ \tau_{2j}'' + \tau_{2j}'' \frac{2\sqrt{\pi}\Gamma}{k_0 \bar{v}} \exp\left[-\left(\frac{\Omega_0}{k_0 \bar{v}}\right)^2\right] \right\} / (2J_j + 1)}{\sum_{j=m,n} \left\{ \tau_{1j} + \tau_{2j}' \frac{2\sqrt{\pi}\Gamma}{k_0 \bar{v}} \exp\left[-\left(\frac{\Omega_0}{k_0 \bar{v}}\right)^2\right] \right\}} \quad (3)$$

Here  $\Gamma$  and  $k_0 \bar{v}$  are the homogeneous and Doppler-line half-widths, respectively, and  $\Omega_0 = \omega_0 - \omega_{mn}$ . The state of the atom is characterized by energy values  $E_m > E_n$ , a velocity  $v$ , and a projection of the total angular momentum  $M$ .  $\tau_{ij}$ ,  $\tau_{2j}'$ ,  $\tau_{2j}''$ , and  $\tau_{2j}'''$  are the lifetimes in the states  $jMv$ ,  $jMv_p$ ,  $jM_p v$ , and  $jM_p v_p$ , respectively. The first is the time prior to collisions of any type, and the remaining ones after collisions that cause only one or both parameters to assume equilibrium values.<sup>[4]</sup>  $F_2$  does not contain contributions of the depolarizing collisions that upset the coherence of the sublevels and cause the four-photon parametric processes to vanish. To the contrary, the incoherent part of  $F_1$  depends on the times  $\tau_{2j}''$  and  $\tau_{2j}'''$ .

5. a) The polarization measurements have shown that if  $E_0$  and  $E_\mu$  have opposite circular polarizations there is no beat signal  $I_{\nu\mu}$ , apart from the noise; this corresponds to absence of coherent collisions. When the polarizations of  $E_0$  and  $E_\mu$  were perpendicular, the signal to noise ratio was 10.

b) The dependence of the experimental values of  $\delta I_\mu / I_{\nu\mu} \approx 2F_1 / F_2$  on the Ne pressure, which were calculated from measurements of  $I_{\nu\mu}$  and  $\delta I_\mu$  and with the aid of formulas (2), is shown in Fig. 2. This ratio should not depend on the population difference  $N_n - N_m$ . Extrapolation of the curve to zero pressure yields a value close to critical. The increase of the ratio  $F_1 / F_2$  with increasing pressure can be attributed to the increasing role of the depolarizing collisions that are

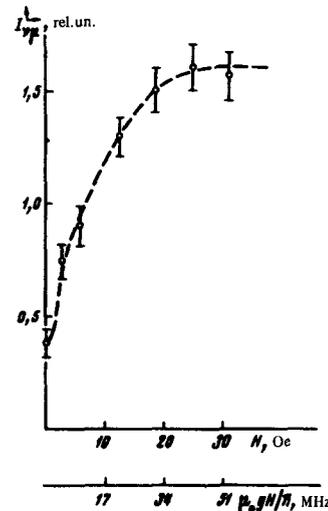


FIG. 3.

characterized by the times  $\tau''_{2j}$  and  $\tau'''_{2j}$ . In the pressure region up to 1 Torr, the main mechanism of collisions of this type is the dragging of the radiation from the level  $m$ . At higher pressures, the dragging can be regarded as complete. This seems to explain the flattening of the curve. In this region, the experimental value of the ratio of the sums in formula (2) is close to 1/2. To determine the concrete times and cross sections it is necessary to have additional data on the ratios  $\tau'''_{2j}/\tau''_{2j}$  and  $\tau_{1j}/\tau'_{2j}$ .

c) The experimental dependence of  $I_{\nu\mu}$  on the magnetic field in the region of weak magnetic fields is shown in Fig. 3. Owing to the Zeeman splitting of the levels, the real part of the nonlinear susceptibility appears and increases with increasing field. The solution of Eqs. (1) becomes more complicated, in view of the appearance of effects such as phase locking etc. An analysis of the components of the tensor  $\hat{\sigma}$  shows that  $\sigma^1_{\nu 111}$  does not depend on the field  $H$ , while  $\sigma^1_{\nu-11-1}$  decreases with in-

creasing  $H$  like a sum of two Lorentzians. This explains the course of the curve of Fig. 3. A reduction of these curves yields information on the line width and on the levels. At large values of  $H$ , the method makes it possible to investigate the electrodynamics of strongly nonlinear interactions. The proposed method does not call for stabilization of the laser frequency.

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