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FOUR-PHOTON RESONANT PARAMETRIC INTERACTION IN LASERS USING DYE SOLUTIONS

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Submitted 31 March 1969
ZhETF Pis. Red. 9, No. 10, 564 - 567 (20 May 1969)

We report in this paper observation of four-photon parametric interaction in dye solutions; this interaction leads to the appearance of a narrow intense line with controllable frequency in the spectral region of the dye emission.

The frequency-variation effect was obtained by using the experimental setup shown in Fig. 1. The dye solution was contained in a cylindrical cell, one face of which was coated with a dielectric coating having a reflectance 85% in the region of $\lambda \sim 1\ \mu$. The cell was placed in the field ofa neodymium laser with pulsed Q-switching, having a radiation power $\sim$50 MW/cm^2. The optical axis of the resonator with the dye was inclined at an angle α to the direction of the neodymium-laser radiation. The dye radiation was registered with an STE-1 spectrograph. The working media were solutions of an analog of pentacarbocyanine, the lasing of which was reported earlier [1]. The solvents were nitrobenzene and ethyl alcohol. The molecule concentration in the solution was of th the order of 10^{18} mol/cm^3.

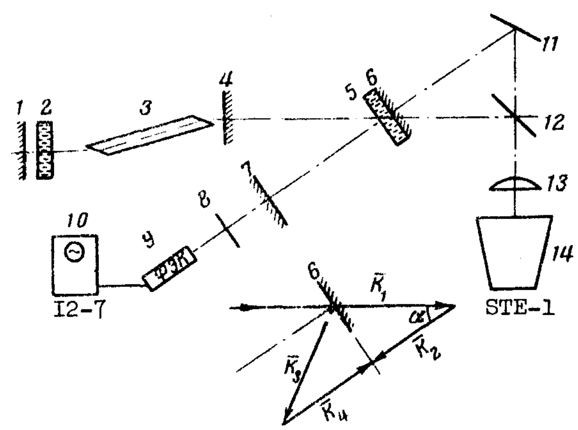

Fig. 1. Experimental setup: 1, 4, 6, 7 - mirrors; 2 - cell with passive shutter, 3 - neodymium-glass rod, 5 - cell with investigated dye solution, 8 - light filter, 9 - coaxial photocell, 10 - oscilloscope, 11, 12 - tilting plates, 13 - cylindrical lens, 14 - spectrograph.

We investigated the generation spectra of the dye at different angles α. The results are shown in Fig. 2. At $\alpha < 9°$, the spectrum represented the broad band (10850 - 11050 Å) customarily observed in the generation of the dye (Fig 2a). At $\alpha = 9°10'$ (Fig. 2b) we observed, besides the aforementioned band, also an intense spectral line shifted towards the shorter wavelengths relative to the generation spectrum of the dye (λ = 10790 Å). An increase of the angle α led to a shift of this line towards the long-wave side (Figs. 2c - g). In many cases, when the line fell in the spectral region of the

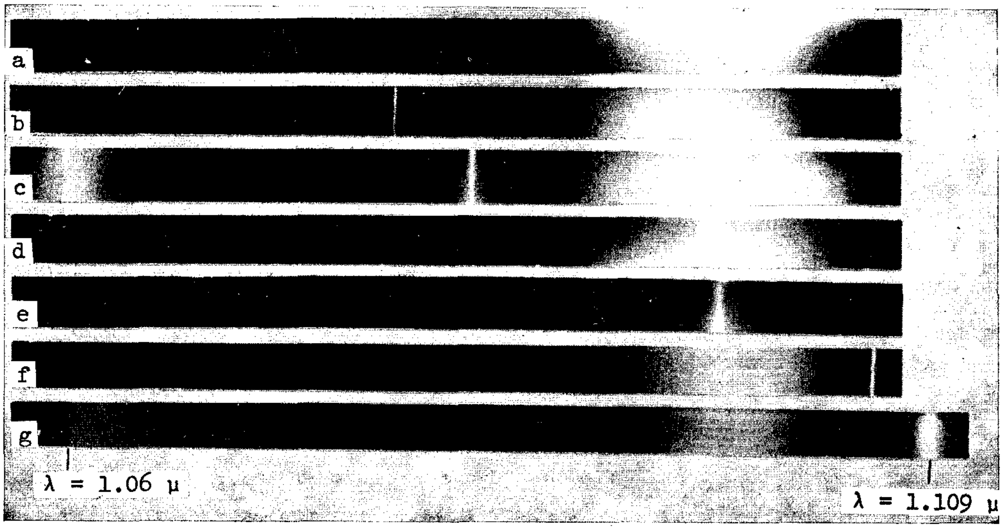

Fig. 2. Emission spectra of dye at different angles α between the direction of the neodymium-laser radiation and the dye-laser radiation: a - $\alpha < 9^\circ$, b - $\alpha = 9^\circ 10'$, c - $\alpha = 10^\circ 40'$, d, e - $\alpha = 14^\circ$, f - $\alpha = 15^\circ 30'$, g - $\alpha = 16^\circ 10'$.

dye generation, the generation band of the dye either weakened greatly or disappeared completely (Figs. 2d, e). Further increase of the angle α led to a shift of the line behind the "red" edge of the generation region of the dye (Figs. 2f, g). At $\alpha > 16^\circ 30'$, only the typical generation spectrum of the dye was again observed.

A reduction of the spectra has shown that the wavelength of the single line λ is conncected with the cosine of the angle α by the relation $\lambda = \lambda_n/\cos\alpha$, where λ_n is the wavelength of the neodymium-laser emission.

Variation of the dye concentration led to a shift of the generation spectrum, whereas the position of the line was determined only by the angle α.

The appearance of intense radiation in the form of a single line whose frequency depends on the angle between the directions of the neodymium-laser and dye-laser emissions is interpreted by us as a manifestation of parametric four-photon interaction.

In our experiment the dye, whose electronic excited level is inversely populated relative to the system of electron-vibrational levels of the ground state, is acted upon by the field of the neodymium laser with frequency ω_1 and wave vector $\vec{k}_1$, the field of the reflected radiation of the neodymium laser (ω_3, $\vec{k}_3$), and the field of the dye emission (ω_2, $\vec{k}_2$; ω_4, $\vec{k}_4$). The proposed vector diagram of the interaction is shown in Fig. 1. The dye molecules can virtually absorb two quanta $\hbar\omega_1$ and $\hbar\omega_2$ and then emit the quanta $\hbar\omega_3$ and $\hbar\omega_4$. Of importance for such a parametric transformation are the conditions of the space-time synchronism ($\omega_1 + \omega_2 = \omega_3 + \omega_4$, $\vec{k}_1 + \vec{k}_2 = \vec{k}_3 + \vec{k}_4$). In our case $\omega_1 = \omega_3$ and $\omega_2 = \omega_4$, and the conditions on the wave vectors are $|\vec{k}_2| = |\vec{k}_4| = |\vec{k}_1|\cos\alpha$.

As indicated above, the last condition is satisfied for the observed line. Since the frequency $\omega_{2,4}$ is the natural frequency of the dy emission, the interaction has a resonant character.

A theoretical model of four-photon parametric resonant interaction was considered in

[2]. The expression obtained there for the time variation of the inverted population ΔN is in our case of the form

$$\frac{d\Delta N}{dt} = -\frac{2\Delta N}{\hbar}\chi^{(1)''}(\omega_2)A^2(\omega_2) - \frac{\Delta N}{\hbar}\chi^{(3)''}(\omega_2)\sin^2\phi A^2(\omega_2)A^2(\omega_1) - \frac{\Delta N - \Delta N_0}{T} .$$

Here $A(\omega)$ are the real amplitudes of the field of frequency ω, $\chi^{(1)''}$ is the imaginary part of the component of the second-rank nonlinear-suseptibility tensor, $\chi^{(3)''}$ is the imaginary part of the component of the fourth-rank nonlinear-susceptibility tensor, $\sin^2\phi$ is a factor connected with the phase of the interacting fields, and T is the lifetime of the excited state.

It follows therefore that the ratio of the terms determining the parametric interaction and the ordinary generation is proportional to the intensity of the field of frequency ω_1. An increase of this field in the experiment led to an increase of the contrast of the parametric line relative to the spectrum of the ordinary generation.

The proposed scheme of parametric interaction in which resonant radiation takes part can be used to develop radiation generators with smoothly variable frequency in practically any region of the spectrum. This is realizable by using an extensive class of generating dyes and other compounds having broad radiation bands and various exciting lasers[1)]. The range of frequency tuning can be extended by increasing the densities of the exciting flux and by changing the solvent and the concentrations of the solution.

The authors are deeply grateful to A. S. Markin for a useful discussion of the results.

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NARROW RESONANCES IN THE SATURATION OF ABSORPTION OF SF_6 BY CO_2-LASER EMISSION

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Submitted 3 April 1969
ZhETF Pis. Red. 9, No. 10, 568 - 571 (20 May 1969)

In [1 - 3[it was proposed to use the effect of saturation of the absorption of rotational-vibrational transitions of a low-pressure molecular gas in a coherent light field to obtain narrow resonances within the Doppler line and to stabilize the frequency of a laser by means of these resonances. Ultrahigh stabilization schemes were proposed using nonlinear absorbing cells either inside [1 - 3] or outside the resonator [4, 5], by means of the power peak [2, 4, 5] or by means of the auto-stabilization effect [1, 3, 5]. The efficacy of the proposed method is evidenced by the attainment of stability and reproducibility of the freqeuncy within 10^{-11} in an He-Ne laser operating at $\lambda = 3.39\ \mu$ by saturating the ab-

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[1)] Suppression of one spectral line and its shift as a function of the angle was observed by us also in cyan dyes excited by ruby-laser emission.

The footnote of the article by L. D. Derkacheva and A. I. Krymova (Vol. 9, No. 10), on p. 345, should read "Appearance of one spectral line," not"Suppression of one spectral line."