Four-photon parametric luminescence during noncollinear two-frequency excitation

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The spatial spectra of the four-photon parametric luminescence in sodium vapor have been measured during noncollinear two-frequency excitation of the $3S$-$3D$ transition. These spectra have a clearly defined ring structure. This structure is interpreted on the basis of an analysis of the conditions for frequency and spatial phase matching in the four-photon parametric luminescence.

The characteristics of four-photon parametric luminescence are of interest in connection with a variety of applications. An important aspect of this research is to determine the spatial structure of the luminescence. As a rule, such studies have been carried out during the two-photon excitation of a metal vapor by a frequency-degenerate collinear pumping.[1–3]

We have now studied the frequency and angular structure of the four-photon parametric luminescence in sodium vapor during two-frequency (ω_{H1}, ω_{H2}) excitation of the $3S$-$3D$ transition and its subsequent decay, accompanied by emission at frequencies ω_1 and ω_2 near the frequencies of the $3S$-$3P$ and $3P$-$3D$ transitions.

The experimental arrangement is shown in Fig. 1. The two pump components are put on different sides of the frequency of the $3S$-$3P$ transition for this switch from frequency-degenerate pumping to two-frequency pumping. One of the pump compo-

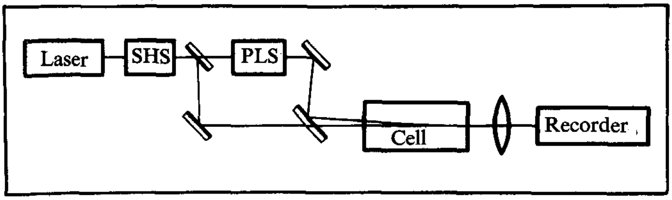

FIG. 1. The experimental arrangement.

nents is the second harmonic (SHS) from a single-pulse picosecond Nd:YAG laser ($\lambda_{H1} = 0.5332\,\mu$m, $P_{H1} \simeq 10^9$ W, $\tau_{H1} \simeq 30$ ps); the other pump component is the emission from a parametric light source (PLS) using KDP crystals ($\lambda_{H2} = 0.964\ \mu$m, $P_{H2} \simeq 10^7$ W). The density of sodium vapor in the cell is $N \simeq 10^{17}$ cm^{-3}. The spatial spectrum of the four-photon parametric luminescence is recorded on film through a Gelios-44-2 objective. The frequency spectrum of the luminescence is measured with a DFS-452 spectrograph.

Frame 1 in Fig. 2 shows the experimental spatial spectrum of the four-photon parametric luminescence in the vicinity of the 3*S*-3*P* transition in the case of collinear pump components. This spectrum has a clearly defined ring shape, which corresponds to the emission of the luminescence in a cone with an inner angle ~2.8° and an outer angle ~4.8°. The luminescence frequency is in the low-frequency region with respect to the frequency of the 3*S*-3*P* transition. The spectral width of the luminescence is ~30 Å. When the pump components meet at an angle of 1.5°, the angular dimensions of the ring in the spatial spectrum of the luminescence increase to ~3° and ~5°, respectively (frame 2 in Fig. 2).

As the angle between the pump components is increased further, to 3.5°, the angular dimensions of the ring in the spatial spectrum of the luminescence increase to ~4° and ~5.6°, and at the center of the ring we see the appearance of emission with an angular divergence ~2.2° (frame 3 in Fig. 2). The frequency of this new emission is on the high-frequency side of the 3*S*-3*P* transition, and its spectral width is 40 Å.

These results can be interpreted by working from the conditions for frequency and spatial phase matching in the four-photon parametric luminescence:

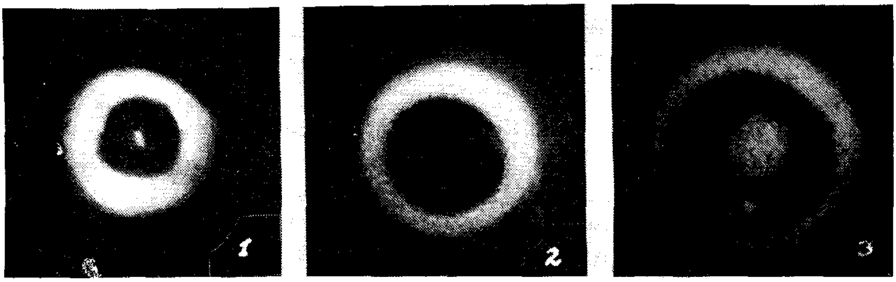

FIG. 2. Spatial spectra of the four-photon parametric luminescence near the frequency of the 3*S*-3*D* transition. The angle between the pump components is as follows: 1—$\alpha_1 = 0$; 2—$\alpha_2 = 1.5°$; 3—$\alpha_3 = 3.5°$.

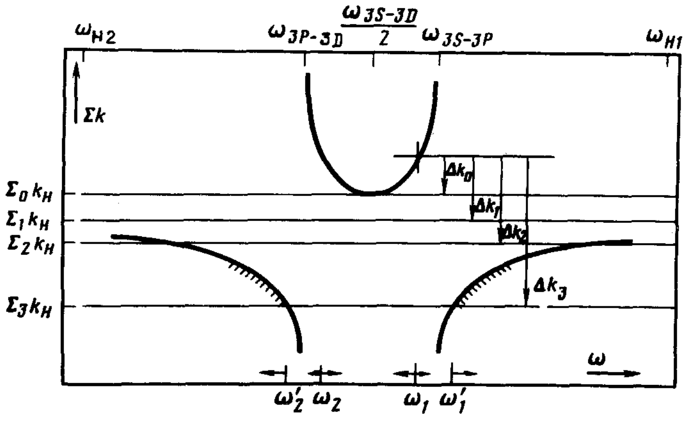

FIG. 3. Spatial-frequency diagram of the four-photon parametric luminescence.

$$\omega_{H1} + \omega_{H2} = \omega_{3S-3D} = \omega_1 + \omega_2,$$

$$k_{H1} + k_{H2} = \Sigma k = k_1 + k_2 .$$

Figure 3 shows the spatial-frequency diagram of the luminescence process in sodium vapor for two-frequency excitation of the 3*S*-3*D* transition, constructed in accordance with these conditions. On this diagram, the frequency regions with $\Sigma k \geqslant \Sigma k_H$ correspond to the four-photon parametric luminescence which is propagating along the generatrices of a cone whose angle is determined by the difference $\Delta k = \Sigma k - \Sigma k_H$. The value $\Delta k = 0$ corresponds to the collinear interaction.

Analysis of this diagram yields a good explanation for the transformation of the spatial and frequency spectrum of the four-photon parametric luminescence which is observed experimentally. Specifically, in the case of collinear and frequency-degenerate pumping ($\Sigma_0 k_H$ in the diagram), the luminescence near the frequency of the 3S-3*P* transition should consist of a "halo" around the pump-propagation direction. The spatial spectrum of this emission can become ring-shaped only by virtue of the frequency dependence of the efficiency of the luminescence, which is determined by the nonlinear susceptibility $\chi^{(3)}(\omega)$. When we switch to two-frequency pumping, with one component having $\omega_{H1} > \omega_{3S\text{-}3P}$ ($\Sigma_1 k_H$ in the diagram), a ring structure definitely must appear in the spatial spectrum of the luminescence.

With noncollinear pumping or with pumping at different frequencies, we find a decrease in the resultant wave vector ($\Sigma_2 k_H$ in the diagram) and thus an increase in the angular dimensions of the cone in which there is no luminescence. Finally, when $|\Sigma k_H|$ crosses the lower branch of the spatial-frequency diagram ($\Sigma_3 k_H$ in the diagram), four-photon parametric luminescence with a frequency $\omega_1' > \omega_{3S\text{-}3P}$ appears along the direction of the resultant pump vector at sufficiently large values of $\chi^{(3)}$ (see the hatched region on the diagram). The structure of the spatial spectrum of the luminescence for frequencies ω_2 near the frequency of the 3*P*-3*D* transition should be analogous to that shown here. (In the present experiments, we detected this emission, but we did not measure its spatial spectrum.)

These results are of fundamental importance for implementing specific schemes based on nonlinear interactions in gaseous media. For example, in schemes involving frequency conversion in metal vapor, the four-photon parametric luminescence is one of the basic factors limiting the sensitivity.[4] As follows from the results above, however, when the geometry is chosen correctly, and when the relationship between the pump frequencies is chosen correctly, it is possible to achieve, in a certain solid angle, the noise-free conversion regime which has been achieved previously in frequency-conversion schemes in condensed media.[5]

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