Splitting of superradiance emission lines of pulsed lasers

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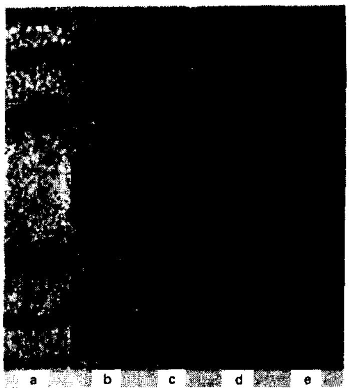
We observed experimentally a stochastically periodic structure in the superradiance lines of a nitrogen laser. The analog of this phenomenon in other pulsed radiation sources is discussed.

1. A fine structure was observed in[1–5] in the superradiance lines of pulsed lasers, namely, the lines $\lambda = 6143$ Å, $\lambda = 5944$ Å, $\lambda = 5401$ Å of Ne and $\lambda = 5350$ Å of Tl consisted of several components situated within the limits of the Doppler width. The main feature of the phenomenon was its stochastic character, namely, the number and location of the components varied over the beam cross section and were not duplicated from flash to flash. The interpretation proposed for the phenomenon was "... that the components of the structure appear to belong to the superradiant modes of the pulsed laser".[1] The investigations of[1–5] were the only ones in their field and it remains unclear whether the effect is common to some degree or whether it is peculiar to the conditions of[1–5] (discharge low-pressure atomic gas, Doppler broadening).

The experiments described below and the analysis of the published data lead to the conclusion that this phenomenon is usually common and the line splitting is observed in all pulsed sources with large gain, regard-



Interference patterns of the N_2^+ line $\lambda = 4278$ Å: a- with polyethylene film in front of the Fabry-Perot etalon, b, c, d, e- without a film.

less of the type of the radiating system of the method of excitation. In addition, a new property was observed, namely quasiperiodicity of the line splitting.

2. We investigated the structures of the lines $\lambda = 3371$ Å of N_2 and $\lambda = 3914$ Å and $\lambda = 4278$ Å of N_2^+ in the pulsed laser described in[6]. A transverse discharge excited an He-N_2 mixture in a volume $0.5 \times 3 \times 300$ mm^3 at a pressure 8—10 atm (the He : N_2 pressure ratio was approximately 100 : 1). The duration of the superradiance pulse was 2.5 nsec (at half-height). We used a Fabry-Perot etalon with a dispersion region 0.313 cm^{-1} and an approximate resolution 0.015 cm^{-1}. The exit face of the laser was projected in the plane of the rings.

3. The intensity distribution in the beam cross section had a characteristic grainy structure. The superradiance line consists of 1—6 components, the number and position of which varies from grain to grain and from laser flash to flash. If a scattering element (ground glass, polyethylene film) is placed ahead of the etalon, then the line contour becomes smooth (see Fig. 8) with a width $\gamma_{gen} = 0.15$ cm^{-1}. This means that the scatterer averages the spectrum over many grains.

For a more detailed study of the spectrum of a single grain, the laser end face was projected in the plane of the interference rings with a magnification (~100) such that the dimension of the grain image was larger than the diameter of several rings. In these experiments, performed for the $\lambda = 4278$ Å line, we observed a new and unexpected property, namely quasiperiodicity of the spectral structure of the line: on 30 photographs (out of the 51 scanned), the line components were approximately equidistant, although the periods of the structure were different in different photographs (Figs. b, c, d, e). A statistical reduction of the measurement results has led to the following values of the width $\delta\Omega$ of the component, the average period $\bar{\Omega}$, and the scatter[1)] in the periods over the flashes and $\Delta_2\Omega$ within a single flash:

$$\bar{\Omega} = 0.058\ \mathrm{cm}^{-1};\quad \Delta_1\Omega = \pm 0.021\ \mathrm{cm}^{-1};\quad \Delta_2\Omega = \pm 0.005\ \mathrm{cm}^{-1};$$

$$\delta\Omega \cong 0.020\ \mathrm{cm}^{-1};\quad \gamma_{gen} \cong 0.15\ \mathrm{cm}^{-1}.$$

Thus, the average period $\bar{\Omega}$ is comparable with its variation $2\Delta_1\Omega$ in different flashes, but is much larger than the deviations $\Delta_2\Omega$ from the equidistant distribution within the limits of one photograph; in addition, $\Delta_2\Omega$ is one fourth as large as the width $\delta\Omega$ of the individual component. From the listed aggregate of relations we can conclude that the superradiance line has a quasiperiodic discrete structure.

We measured also also the frequency of the most intense component and its scatter $\Delta\omega_m = \pm 0.44$ cm^{-1} turned out to be close to $\bar{\Omega}$. Finally, the distance 0.143 ± 0.048 cm^{-1} between the outermost components of the structure almost coincided with the width γ_{gen} of the smooth spectrum.

Outwardly, the line structure observed by us and the structures in[1-5] are practically the same, and it can be assumed that in both cases we are dealing with the same phenomenon. The difference lies only in the scale of the picture, which in our case is larger by one order of magnitude. We emphasize that there are great differences in a number of traditionally important characteristics, namely: molecular and molecular-ion lines in our case and atomic lines in[1-5]; interaction broadening in our case as against Doppler broadening; the spontaneous-emission line widths differ by tens of times, and the oscillator strengths differ by three orders of magnitude. Such large changes in the indicated parameters allow us to think that the line splitting is a common property of high-power superradiance pulses. However, it is observed not only in gas-discharge systems. The recently observed fine structure of the SRS lines in calcite[7,8] and in liquid nitrogen[8] is surprisingly similar to ours: approximately the same number of components, stochastic behavior, quasiperiodicity, etc. Thus, the splitting effect does not depend either on the excitation method or on the number of photons participating in the radiative process.

5. The universality of the phenomenon of superradiance line splitting (the first Stokes SRS falls in this group) induces us to search for sufficiently common causes. The stochasticity of the spatial and spectral characteristics can be easily explained. The emission of a pulse-inverted medium is amplified spontaneous emission and possesses its inherent randomness features. The known considerations concerning the narrowing of the radiation spectrum in an amplifying medium[9] pertain to average quantities and correspond to our "smooth spectrum" of the line. The discrete line structure and the spatial separations should be regarded as fluctuation deviations. In this respect, there is full analogy with the fluctuations of transients in stationary lasers (see, e.g., [10]) and with the fluctuations in the emission of "ordinary" pulsed resonator lasers. [11] The stochasticity is therefore in itself not of very great interest. What is nontrivial is the quasiperiodicity (i.e., the ordering) of the fluctuation spectral structure. This fact points clearly to an obviously nonlinear transformation of the spectrum. In addition to the general considerations, the nonlinearity is favored also by two facts: 1) in systems without resonators based on superradiance, the product of the gain α by the length L of the active medium is of the order of 20 and more, whereas in resonator lasers $\alpha L \lesssim 1$ and even $\alpha L \ll 1$; 2) the energy dE of the interaction of the atom with the

field is given, according to estimates, by $dE/(2\pi\hbar c)$ $\sim 0.04\ \mathrm{cm}^{-1}$, which is comparable with Ω.

Several nonlinear mechanisms of periodization of the fluctuation structure of the spectrum can be indicated (splitting of the gain contour in the presence of a strong field, 4-photon resonant scattering of light, light echo). Each of them can lead to a periodicity with a random period. It is difficult, however, to choose from among them on the basis of the presently available data.

We assume therefore that the stochastically-periodic line structure should be inherent in the radiation from any pulse-inverted system with sufficiently large value of αL. In addition to the cases indicated above (gas-discharge pulsed lasers, SRS), these systems include SMBS and a photon avalanche following pulsed optical excitation of an adjacent transition (experiments of this type were carried out, for example, in potassium vapor[12] and in hydrogenfluoride vapor[13]).

[1)] By "scatter" we mean here and below an interval with a fudicial probability 0.84.

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