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NONSTATIONARY GENERATION OF SOLUTIONS OF ORGANIC DYES IN PICOSECOND OPTICAL PUMPING

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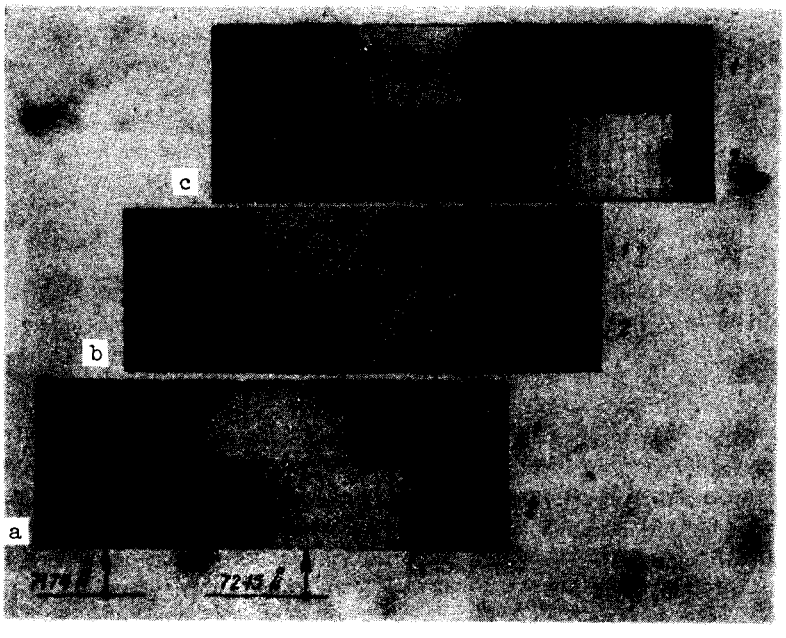
The establishment of stationary laser generation is preceded by the following transient processes: a) deviation of the inverted population of the working level from the stationary value - kinetic nonstationarity; b) nonstationarity due to the finite time of formation of the half-width of the resonator mode - spectrum nonstationarity; c) nonstationarity due to the finite rate of relaxation of the active centers over the states participating in the transition.

In the case of optical pumping by picosecond pulses, we can expect all the aforementioned nonstationarities to be manifest to the greatest extent.

We report here observation of nonstationary processes of type (a) and (b) in the generation of lasers based on solutions of organic dyes.

The work was based on comparison of the generation spectra obtained by varying the pump pulse duration from 30 nsec to several times ten picoseconds. To vary the pulse duration we placed alternately in the resonator of a ruby laser with an RL2B 10 × 120/180 element an inertial modulator based on Vaphthalocyanine in dimethylsulfoxide (nanosecond regime) and a modulator based on a solution of tricyanine dye in ethanol (picosecond regime).

We investigated several types of dye solutions generating in the spectral region 7100 - 9100 Å [1].



Generation spectra of solutions of cyanine dye-1 [1]: 1 - picosecond optical pumping, 2 - nanosecond optical pumping; the optical density of the solution increases from a to c, base of the resonator is ℓ = 15 mm.

At T = 300°K, equilibrium with air, and a pulse repetition period 10 nsec in the train, no accumulation effects were observed at all in the investigated dye solutions.

The generation spectra shown in the figure (spectrograph with diffraction grating, dispersion 7 Å/mm) were obtained with almost longitudinal pumping of the dye solution-1 [1] in a plane-parallel cell without reflecting coatings, of 15 mm length. They are characterized by the following features: a) the average frequency of the generation band of the entire solution is shifted towards the short-wave region in the case of pumping with picosecond pulses (1), compared with pumping with a nanosecond pulse (2) - Fig. a, b, c; b) the shift increases in proportion to the optical density of the solution and does not depend on the individual spectroscopic properties of the dye solutions; c) the generation spectra in the case of picosecond pumping (1) differ from the generation spectra in nanosecond pumping (2) in the partial or complete absence of an interference (mode) structure.

The results can be explained qualitatively within the framework of the notions concerning generation occurring under nonstationary conditions.

When the dye solution is excited by a picosecond pulse, the population of the working level occurs with a characteristic time $\tau = \tau_{21}[1 + \alpha(\nu)I(\nu)]^{-1} < \tau_{21}$, where τ_{21} is the total relaxation time of the level without allowance for the induced transition, $\alpha(\nu) = \sigma_{12}(\nu)\tau_{21}$ is the nonlinearity parameter for the two-level system, and $\sigma_{12}(\nu)$ and $I(\nu)$ are the absorption cross section and the pump intensity. For the value $\alpha(\nu) = 10^{-24}$ $cm^{-2}sec^{-1}$, which is typical of cyanine dyes and $I(\nu) = 1$ GW/cm^2, we have $\tau \sim 10^{-4}\tau_{21}$. This means that an appreciable population of the excited state is reached within one picosecond pulse and can greatly exceed the population determined by the stationary-generation condition. It is known that the average frequency of the stationary-generation band of the organic dyes is determined by the value of the inverted population of the working states [2]. In pumping by nanosecond pulses, the average frequency of the generation band remains practically unchanged with changing pump intensity, i.e., a case close to stationary generation is realized. The use of picosecond pumping, at equal Q of the resonator, causes the generation to develop when the population greatly exceeds the stationary value. By virtue of the frequency dependence of the maximum gain of the dye solutions from the inverted population, this leads to a shift of the average frequency of the generation band to the short-wave region. For solutions of low optical density, when there is no possibility of a large difference in the gains on going from nanosecond to picosecond pumping, the shift decreases and tends to zero (see the figure).

Upon excitation by picosecond pulses under the conditions of the given experiment, no generation is observed in the spectral region of wavelengths shorter than the maximum of the fluorescence band. This indicates that the nonstationarity of type (c), which is connected with the violation of the equilibrium in the excited state, does not arise.

A second interesting feature of generation of dye solutions with picosecond pumping is the partial or total vanishing of the interference (mode) structure, and can be attributed to spectral nonstationarity. It should be noted that what is realized in the performed experiments is the generation regime and not superluminescence, which is also characterized by a diffuse spectrum [3].

According to [4], the stationary mode is formed during a finite time. For times $\tau = (tc/2\ell)|1 - R\exp(k\ell')| \ll 1$ (t - running time, ℓ' and ℓ - lengths of the active medium and of the resonator base, R and k - reflection and gain coefficients) the process of formation of the stationary mode is described by the expression

$$I(\phi) = S^2 \frac{\theta}{\delta\Phi} r^2 \left[\frac{\sin(\phi r/2)}{\phi r/2}\right]^2, \qquad (1)$$

where S^2 is the intensity of the spontaneous radiation, $\theta = (2\ell/c)|1 - R\exp(k\ell')|$, $\delta\Phi \simeq |1 - R\exp(k\ell')|$, and $\phi = 4\pi\ell\nu/c$ is the phase shift per pass. According to [1], the half-width of the mode decreases in proportion to the time (to the number of passes), and the peak power increases like the square of the time. Accordingly, the decrease of the generation time in picosecond pumping should become manifest in a broadening of the generated modes and in the occurrence of a diffuse generation band. Comparing the generation spectra obtained for different durations of the picosecond pulse, it can be concluded that at a constant resonator length the diffuseness of the spectrum increases with decreasing pulse duration. Analogously, the diffuseness of the generation spectrum increases also with increasing resonator base. These results lead to a preliminary conclusion that the duration of the picosecond pulse can be estimated from the form of the generation spectrum of an organic dye in the resonator with a specified base.

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PULSED NARROWING OF p-n JUNCTION IN TWO-PHOTON EXCITATION

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Two-photon absorption in homogeneous crystals has been investigated in sufficient detail. There are practically no published data, however, on two-photon excitation of p-n junctions and heterojunctions. The reason for this can be understood if it is recognized that such crystals contain impurities that lead to the single-photon excitation mechanism. One should expect, however, the impurity centers to become depleted at sufficiently high excitation levels and generation of electron-hole pairs in the volume to begin. If this process is active enough, then considerable changes in the properties of the p-n junction should occur. To reveal such changes, the following experiment was organized.

A p-ZnTe - n-CdSe heterojunction was excited with a neodymium laser (pulse energy 0.4 J, pulse duration at half-width 35 nsec, $\hbar\omega = 1.17$ eV), as a result of which one could expect two-photon generation of electron-hole pairs both in the zinc telluride ($E_g = 2.26$ eV) and in the cadmium selenide ($E_g = 1.7$ eV). We measured the no-load voltage V_{nl}, the short-circuit current I_{sc}, and the lux-ampere characteristic of the heterojunction in the photodiode regime. A standard registration procedure with a long-persistence oscilloscope was used. The time resolution of the circuit was 10^{-8} sec.

The results of the measurement of no-load voltage V_{nl} (measured at the peak value) are shown in Fig. 1. Figure 1b shows, in a logarithmic scale, a section of the curve a corresponding to two-photon absorption. Figure 2 shows the kinetics of the relaxation of V_{nl} at different excitation intensities.