Interference character of the "gap" produced by laser radiation in continuous dye-solution absorption spectra

B. S. Neporent and A. G. Makogonenko

S. I. Vavilov State Optical Institute
(Submitted October 12, 1975)
Pis'ma Zh. Eksp. Teor. Fiz. **22**, No. 11, 542–544 (5 December, 1975)

A simple experiment is used to prove the previously advanced hypothesis that the selective transmission ("spectral gap") in spectra of dye solutions bleached by laser radiation is brought about to a considerable degree by the interference between the bleaching and measuring laser beams in the solution zone.

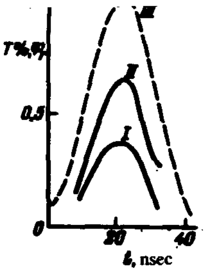
PACS numbers: 42.60.Nj, 82.50.–m

It was established in[1,2] by indirect experiments that a gap of width $1.0<\delta\nu<40$ cm^{-1} is produced in the continuous spectra of cryptocyanine solutions bleached by the action of a nanosecond ruby-laser pulse. The "gap," of width $\delta\nu\approx 8$ cm^{-1} and lifetime $\delta t\approx 10$ psec, was observed directly in[3] by applying picosecond pulses and by using these pulses for the measurements. It was concluded in these studies that the laser radiation "burns out" the component of the inhomogeneously-broadened absorption band, and that the freed vibrational levels are then populated as a result of molecular relaxations. At the bleaching-pulse powers employed in[1,2], however, a "gap" with the parameters given in[3] can have only a very small average depth. Indeed, attempts[4–6] at directly observing the "gap" in the absorption spectra of cryptocyanine solutions bleached by nanosecond pulses, using probing radiation of relatively broad spectral composition, have led to negative results, and to the conclusion[4] that the vibronic bands are homogeneously broadened. In[7], likewise, no "gap" has been observed in the spectra of bleached phthalocyanine solutions, in which the rates of the vibrational relaxation are large.[8] In[5,6], on the other hand, using the same bleaching radiation of frequency ν_b for the measurements of the absorption, it was possible to observe in the spectra of the bleached solutions of both substances—phthalocyanine and cryptocyanine—similar singularities of the "gap" type near the frequency ν_b.

It was indicated in[6] that the aforementioned and other contradictions disappear if it is recognized that a system of standing optical waves is produced in the investigated solution, owing to the reflection of the bleaching radiation with narrow spectral composition from the rear wall of the cell or from other elements of the optical system. This decreases the amount of the bleaching light in the high-intensity zones as a result of the enhanced bleaching of the dye, and in the low-intensity zones because of the small amount of the active light. Thus, for a coherent bleaching radiation of frequency ν_b, the bleached system can be more transparent than for the narrow section near ν_b, which is separated from the probing nonmonochromatic radiation after passage through the cell with the solution.

The foregoing mechanism of formation of a "spectral gap" in absorbing matter, which is analogous to the "burning of a spatial gap" by a system of standing waves in a laser as proposed in[9], can be called, not quite accurately, an "interference" mechanism.

We describe in this communication the results of simple direct experiments that demonstrate the appreciable part played by the interference mechanism in the



formation of the "spectral gap." To this end, we measured the transmission of the bleaching radiation through a solution of cryptocyanine in ethanol placed in a cell with windows at the Brewster angle. A wedge-shaped plate was placed behind the cell, by rotating the plate it was possible to reflect from one of its faces a part of the discoloring radiation back into the cell with the solution. The measurements were made in two regimes: without a beam returned to the cell and with the beam returned. The intensities of the incident light (I_0) and the light passing through the cell and the plate (I) were registered with an oscilloscope and coaxial photocells. The concentration of the cryptocyanine solution was 0.8×10^{16} molecules/cm^3, the path length in the cell was 8 mm, the optical thickness of the initial solution for the bleaching ruby-laser radiation was $D = 4.5$, the duration of the radiation single pulse was $\Delta t \approx 20$ nsec, and its average intensity was $\Phi_m = 2\times 10^{26}$ quanta/cm^{2}sec.

The results of a typical experiment are shown in the figure in the form of plots of the transmission $T = I/I_0$ of the bleaching radiation through the investigated solution against the time t for both regimes—without the returned beam in the cell (curve I) and after introducing the returned beam (II). The dashed curve III is the waveform of the bleaching pulse. It is obvious from the figure that the measured transparency of the solution is greatly enhanced when only 4% of the transmitted light is reflected to the cell, an amount that could not cause noticeable additional discoloring in the absence of interference. Under the same conditions, however, the modulation of the spatial intensity of the interference light field in the cell reaches almost 40%, which agrees with the observed decrease of the absorption of the discoloring coherent radiation. We indicate here that the labile amplitude or phase diffraction grating produced in the interference field weakens the considered effects somewhat, reflecting back some of the incident discoloring radiation.

The described results prove that the interference mechanism takes a part in the formation of the "spectral gap" by the action of the laser radiation. The features of this mechanism make it possible to explain without contradictions all the details of the researches in[1–7], and the participation of the interference processes in the experiments of[3] is directly confirmed by certain results of[10].

1C. R. Giuliano and L. P. Hess, Appl. Phys. Lett. 9, 196 (1966).
2M. L. Spaeth and W. R. Sooy, J. Chem. Phys. 48, 2315 (1968).
3G. Mourou, B. Drouin, and M. M. Denariez-Roberge, Opt. Communs. 8, 56 (1973).
4A. M. Bonch-Bruevich, T. K. Razumova, and I. O. Starobogatov, Opt. Spektr. 35, 640 (1973).
5V. A. Kryukov, G. V. Lukomskiǐ, A. G. Makogonenko, and V. B. Shilov, Abstracts of Internat. Conf. on Luminscence, Leningrad, 17–22 August, 1972.
6N. S. Neporent and A. G. Makogonenko, Spectroscopy Lett. 8, No. 10 (1975).
7F. Gires, L'onde Electrique 47, 976 (1967).
8B. S. Neporent and V. B. Shilov, Opt. Spektr. 30, 1074 (1971).
9A. Schmackpfeffer and H. Weber, Z. Angew. Phys. 23, 413 (1967).
10C. V. Shank and E. P. Ippen, Appl. Phys. Lett. 26, 62 (1975).