

OBTAINING THE SECOND HARMONIC OF A NEODYMIUM GLASS LASER IN DYE POWDERS

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 Submitted 18 March 1970
 ZhETF Pis. Red. 11, No. 10, 469 - 472 (20 May 1970)

The nonlinear effects observed in dyes until recently (absorption saturation, two-photon absorption [1], four-photon parametric interaction [2]) result from the presence of a polarization proportional to the cube of the electric-field amplitudes. Second harmonic generation, which is connected with a polarization that is quadratic in the field and is due to the appreciable value of a lower-order nonlinear susceptibility, was observed only in several dyes of the coumaran and triphenylmethane class [3]. We report here observation of frequency doubling in powders of dyes of the aminophthalimide, diarylmethane, and benzanthrone-derivative classes.

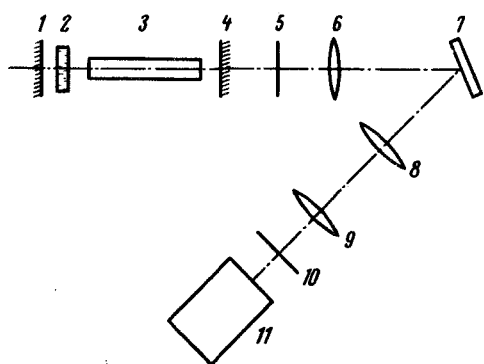


Fig. 1. Experimental setup: 1, 4 - mirrors, 2 - passive shutter, 3 - neodymium-glass rod, 5, 10 - light filters, 6, 8, 9 - lenses, 7 - investigated sample, 11 - spectrograph.

We used the experimental setup shown in Fig. 1. The laser radiation was focused by lens 6 on the sample 7, comprising a dye powder placed between two glass plates. The powders were not specially purified, nor were the crystallites sorted out by sizes. The employed neodymium-glass laser was Q-switched with a passive shutter. The laser radiation-power density on the sample surface was $\sim 200 \text{ MW/cm}^2$. The dye emission was focused with a two-lens condenser on the slit of a Fuess spectrograph.

We investigated about 30 dyes and observed the occurrence of a strong second harmonic in three substances: 3-aminophthalimide (from among the phthalimides), auramine (from along the diarylmethanes), and 3-metoxigenzathrone (from among the benzathrone derivatives). The structure of these compounds are quite different.

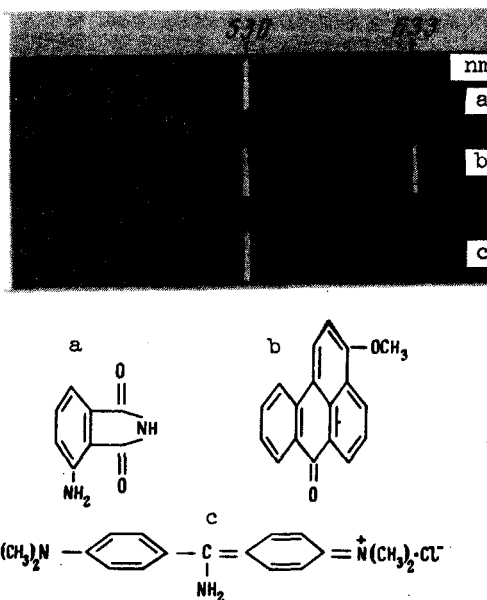


Fig. 2. Emission spectra of dye powders and structure formulas: a - auramine, b - metoxybenzathrone, c - 3-aminophthalimide. Right - reference spectral line of Ne, $\lambda = 633 \text{ nm}$.

The emission spectra of these dye powders, obtained by irradiation with a giant laser pulse, are shown in Fig. 2. They constitute a single line whose wavelength ($\lambda = 530$ nm) corresponds to the second harmonic of the neodymium-glass laser, and whose width is determined by the apparatus function of the instrument.

We compared the intensities of the second harmonic generated by the investigated dyes, by the better dyes described in [3], and by powders of the substances widely used for frequency doubling. To this end, the radiation incident on the spectrograph slit was attenuated by filters selected such that the radiation produced normal density in the photographic film. The measurement results are listed in the table in relative units.

It should be noted that the dyes are much more stable against intense illumination than, say, lithium niobate, which burned out in our experiments after the very first flash, whereas the dyes withstood repeated irradiation.

Substance	Relative second-harmonic intensity	Maximum phosphorescence spectrum, cm^{-1}
Lithium niobate	10	—
7-diethylamino-4-methylcoumaran	10	19 200
3-aminophthalimide	3	19 400
3-metoxybenzathrone	1	18 500
Auramine	1	18 850
7-hydroxy-4-methylcoumaran	1	18 850
Potassium dihydrophosphate	<1	—

The dyes investigated by us were powders of green and yellow-green color. The absorption spectra of the dye solutions, obtained with a Hitachi spectrophotometer, are shown in Fig. 3. We see that the absorption of the dyes in the region of the neodymium-laser second harmonic is small compared with their absorption at the maximum. However, it can be quite appreciable in powders. The possibility of absorbing the second harmonic on the edge of the spectrum is apparently larger in the case of 3-metoxybenzathrone and coumaran, the spectra of which lie in the longer-wavelength region. This can cause the intensity of the harmonic of these substances to be lower than, say, 7-diethylamino-4-methylcoumaran and 3-aminophthalimide.

An interesting common feature of these dyes is the presence of triplet-singlet emission in the green region of their spectra. We measured the phosphorescence spectra of alcohol solutions of the investigated substances at $T = 77^\circ\text{K}$. The measurements were made with a UM-2 monochromator with an FEU-51 photoelectric receiver using a single-disc phosphoroscope to cut off emission with duration $<10^{-3}$ sec. The frequencies of the maxima of the measured phosphorescence spectra are listed in the table. It is seen from the table that the neodymium-laser second-harmonic frequency ($18\,870\text{ cm}^{-1}$) falls in a region close to the frequencies of the triplet-singlet transition of the dyes.

It is known [4] that the dispersion dependence of the nonlinear susceptibility of lower order has a resonant character. Thus, in the classical approximation of an anharmonic oscillator we have

$$\chi_{111}(2\omega, \omega, \omega) = \frac{-N_0 \frac{e^3}{m^2} V}{(\omega_0^2 - \omega^2 - i\Gamma\omega)^2 (\omega_0^2 - 4\omega^2 - i\Gamma 2\omega)}$$

The approach of the fundamental frequency or of a harmonic to the natural frequency of the substance leads to an increase of the nonlinear susceptibility. Such an effect is observed, in particular, in semiconductors [5]. It is possible that the large nonlinear susceptibilities of the dyes are due to the proximity of the neodymium-laser second harmonic to the frequencies of the triplet-singlet transitions of the investigated compounds.

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STRUCTURE OF DEPOLARIZED COMPONENT OF SCATTERED LIGHT IN α -CHLORONAPHTHALENE

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 Submitted 18 March 1970
 ZhETF Pis. Red. 11, No. 10, 473 - 474 (20 May 1970)

The temperature dependence of the frequency of the transverse doublet observed in the depolarized component of scattered light [1 - 3] cannot be explained within the framework of the Leontovich phenomenological theory [4]. It is therefore of interest to measure the anisotropy relaxation time and the viscosity as functions of the temperature.

The measurements were made on α -chloronaphthalene, in which a depolarized doublet was distinctly observed and its behavior with changing temperature was investigated [3].

According to the Leontovich theory, the anisotropy relaxation time should be calculated from the half-width of the depolarized $I_{xy}(\omega)$ component of the scattered light. For many liquids, this component can be represented by a dispersion curve. The measurement setup was described in [3].

We have noted in the study of the $I_{xy}(\omega)$ component of scattered light in α -chloronaphthalene that a strong deviation from the dispersion curve with a single time τ is observed already in the region where the intensity decreases by a factor 10 - 15. The contour of the $I_{xy}(\omega)$ component can be described by a sum of two dispersion contours, one narrow with a sharp maximum and the other broad with a flat maximum; the half-widths of the contours are determined respectively by relaxation times τ_{cg} and τ_1 . The relaxation time τ_{cg} was determined from the half-width of the narrow contour at height $I/2$. τ_1 was determined from the slope of a plot of $1/I = f(\Delta\omega^2)$. It should be noted that in this case I is the intensity in that frequency region $\Delta\omega$ where the contribution of the intensity from the narrow contour does not exceed 5%. In measurements of this type it is necessary to be certain that correct allowance has been made for the intensities due to the different orders of the interference. All the measurements were therefore made in the dispersion regions of 5 and 16.6 cm^{-1} .