

Standing Wave Effects in Interactions between Intense Light Beams and Liquid Dielectrics

Operation condition of a Nd^{3+} : glass laser containing absorbing liquids within its resonator was investigated in detail. Quasi Q-switching of the laser emission caused by the liquids as dependent on the kind of liquid, its thickness, pulse energy, and on the standing wave ratio was determined.

Changes in the refractive index induced by electrostriction, molecular redistribution, and by thermal effects were calculated. A proposed explanation of the observed effects takes into account reflection of the laser light on isobaric periodic fluctuations of the refractive index: the latter being induced by the standing wave structure.

1. Introduction

Various interactions of light beams and liquid dielectrics are now at the centre of interest in numerous branches of physical research, like optics, quantum electronics and nonlinear optics [1-4] and others in particular. In the field of an (at least partially) standing light wave, the nature of these interactions undergoes some modifications. Although the elementary mechanism of the phenomenon remains the same, nevertheless there appear certain interesting effects, unobserved in the case of travelling waves.

In order the light beam be, at least partially, a standing one, it has to be introduced into an optical resonator. In the present investigation, we had recourse to the resonator of a neodymium laser. Provided that the liquid sample in the laser resonator absorbs radiation at the laser wavelength, a change in its transmission or reflexion coefficient (the result of its interaction with the light beam) will lead to Q-switching of the resonator. Hence, measurements of Q-switching effects can serve as a convenient and highly accurate method for the study of interaction effects between the light beam and liquid in the resonator, provided that the interaction involves changes in the liquid transmittivity or reflectivity (which is almost always the case). The elucidation of the Q-switching mechanism is then equivalent to that of the interaction effect.

Effects of Q-modulation of a plane-parallel laser resonator, containing an absorbant liquid sample, have been observed by PLANNER [5],

KATZENSTEIN [6, 7], BŁASZCZAK [8], GAPONOV [9], and their co-workers. Some explanations of the effects observed have been put forward, but none appears to account completely for the experimental facts.

In certain conditions, interaction of the laser beam and liquid in the resonator leads to a very strong Q-factor modulation (especially when having recourse to backwards stimulated Mandelshtam-Brillouin scattering). To this aim, lasers with resonator conformations ensuring extreme power densities of the beam acting on the liquid are constructed [10-14]. Such set-ups are of essential practical importance; however, the interaction mechanism are so well-known that it is hardly worth while to consider the matter in too much detail here.

Let us consider the block-diagram of a laser with liquid cell (Fig. 1.). If the liquid absorbs the laser wavelength, the Q-factor of the re-

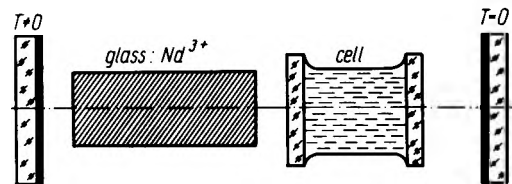


Fig. 1. Scheme of the laser resonator containing liquid absorbing cell

sonator decreases, whereas a positive variation in Q-factor during laser operation can arise due to all those effects which lower the losses, as the beam traverses the cell twice. A decrease in loss can occur by transmission enhancement (e.g. by way of a saturable absorption process) or by directional reflection of part of the energy of the beam inside the cell. Partial reflection of the beam can be due, on the one hand, to effects occurring as well in the field of a travelling wave (of the type of stimulated Mandelstam-

* Quantum Electronics Laboratory, Institute of Physics, A. Mickiewicz University, 60-780 Poznań, Poland.

-Brillouin scattering — SMBS or stimulated Rayleigh scattering — SRS) and, on the other, to effects apparent only in the field of a beam with non-zero standing wave coefficient, such as: e.g. changes in refractive index of the liquid caused by electrostriction, molecular reorientation and redistribution, as well as thermal changes in density induced by the laser beam.

In a liquid dielectric medium placed in an optical resonator these effects can produce a regular layer structure, with a refractive index varying periodically in space in conformity with the spatial distribution of nodes and anti-nodes of the laser light standing wave.

The reflection coefficient R at perpendicular transition of the beam through the boundary between layers with relative refractive index μ , is given [15] by the expression:

$$R = \left(\frac{\mu - 1}{\mu + 1} \right)^2. \quad (1)$$

If the beam has to traverse N double layers of quarter-wave thickness and relative refractive index μ , the reflectivity of the pile in the direction perpendicular to the layer surface is:

$$R_{2N} = \left(\frac{\mu^{2N} - 1}{\mu^{2N} + 1} \right)^2. \quad (2)$$

Eq. (2) can be approximated by the formula [6]:

$$R_{2N} \simeq \operatorname{tgh}^2 \left(\frac{1}{2} \frac{\Delta n}{n_0} N \right), \quad (3)$$

where n_0 and Δn are the mean refractive index and its variation, respectively. Eq. (3) is a good approximation of (2) as long as $\Delta n \ll n_0$.

2. Experimental set-up, and method of measurement

Measurements were performed in nine liquids, namely: benzene, toluene, ethylene dichloride, nitrobenzene, chlorobenzene, ethylene tetrachloride, acetone, carbon disulphide, and methanol, placed in the plane-parallel resonator of a Nd: glass laser. The construction of the laser, which consisted of separate elements mounted on an optical bench, provided for wide universality of the measuring system and ready adaptability of the resonator set-up to various measurements. The Nd-activated glass rod, of a length of 13 cm and an effective diameter

of 9,5 mm, was situated in a water-cooled head, and optically pumped by means of two linear VQX 15 15 type xenon flashtubes. The optical resonator was composed of two plane dielectric mirrors, of 0 and 40 % transmission, respectively (in some experiments, both mirrors had zero transmission).

The time-characteristics of laser operation were studied with photoelectric circuits feeding the signal to the input of an OSA-601 or I 2-7 type oscilloscopes. For measurements of the parameters of the single pulse we used a probe with fast germanium photodiode of the FDWCz type, made at the Institute of Electron Technology in Warsaw. When an accurate mapping of the amplitude of pulses grouped in longer trains was necessary another photoelectric probe with an FEU 22 photomultiplier was used. Weak photoelectric signals were amplified with an IPE 306 pulse distributed amplifier (pulse rise time = 3 ns) constructed in the Institute of the Fundamentals of Electronics of the Warsaw Technical University. The total pulse rise time of the device amounted, to 3, 5 and to about 10 ns for the fastest and slowest circuits, respectively.

The laser energy was measured with an accuracy of 0.03 J by means of a bolometric sphere meter with thermocouple. For current, automatic monitoring of the laser pulse energy, use was made of a self-recording photoelectric meter. Many measurements consisted in a comparison of the intensities or energies of laser pulse light beams, originating from the same source. To this aim a pulse photometer [19], consisting of photoelectric probes, electric signal integrators and formers, as well as reading recorders, was constructed. This set-up permitted to perform direct measurements of transmittivity and reflectivity coefficients of samples throughout and arbitrarily wide range of radiation power density, and the measurements of relative mean intensities and energies of pulsed light beams, the error committed in a single measurement amounted to less than 2 %.

Spectra of the laser emission signals were studied with a grating spectograph of medium resolution rate, and an appropriately modified IT51-30 interferometer; the modification consisted in a change of the mirror coatings from metallic to dielectric ones and in addition of elastic spacing rings which extended the free spectral range to 112 Å (for the 1.06 μm line).

All optical systems were adjusted with an autocollimator and low-power He-Ne laser. The measurements were carried out at room tem-

Table 1

perature applying pro analysi grade liquids (filtered in order to eliminate all foreign impurities and dusts).

On introducing the liquid cell into the laser resonator, variations in laser operation were observed to occur similar to those reported by KATZESTEIN [6], BŁASZCZAK [8], and GAPONOV [9]. The total energy of the laser pulse decreased; however the number of spikes during operation decreased too. The energy conveyed in a single spike changed but slightly, the mean duration time of the spike simultaneously decreased. This resulted in an increase in power of the pulse by a factor of several. On the cross-section of the beam a filamentary emission pattern appeared. Its diameter of the order of fractions of a millimeter suggested a filament structure of the radiation emitted by the laser. The power density of the radiation in a filament during action was very high compared with the mean power density in the beam emitted by the laser in the absence of the liquid cell in the resonator.

The number of well apparent spots of the pattern on the cross-section of the beam was observed to be equal to the number of strong spikes in the laser action oscillogram. Hence, with an appropriate liquid sample in the resonator, laser action consists of numerous elementary events weakly correlated in space and time. The relationships are characterized quantitatively in Table 1. In certain measurements, it was necessary to place one or two beam splitters in the resonator in order to direct part of the radiation perpendicularly to the axis of the laser one, or two beam splitters had to be placed in the resonator. The influence of the beam splitters on the conditions of generation was checked by appropriate measurements carried out for the resonator with beam splitters. Table 1 gives, for comparison, some results for laser action parameters, measured in such resonators. The results of measurements given in Table 1 prove that partial Q-switching takes place in the laser resonator, when the latter contains a cell with an appropriate liquid. The Q-switching effects are the strongest in methanol, rather strong for toluene, acetone and ethylene dichloride, weaker for benzene, barely perceptible for nitrobenzene and chlorobenzene, and are absent in the case of carbon disulphide and ethylene tetrachloride. For a given liquid, the effects are the stronger the thicker is the liquid layer.

As already mentioned, the Q-switching in these conditions can be due to variations in

Mean values of spike energy \bar{E}_s , duration half-time $\bar{\tau}_{1/2}$, power \bar{P}_s , and power density $\bar{I}_s \cdot \bar{d}_s$ is the mean diameter of the light filaments W —the thickness of the liquid layer

Liquid	W [mm]	\bar{E}_s [J]	$\bar{\tau}_{1/2}$ [ns]	\bar{P}_s [MW]	\bar{d}_s [mm]	\bar{I}_s [MW/cm ²]
resonator without beam splitters						
Methanol	2	0.020	183	0.111	1 *	7.0
	5	0.014	262	0.053	1 *	3.4
	10	0.012	156	0.080	0.5	40
	20	0.010	131	0.080	0.4	64
	30	0.011	133	0.086	0.4	69
Benzene	2	0.033	330	0.098	9	0.1
	5	0.030	359	0.083	9	0.1
	10	0.030	257	0.117	9	0.2
	30	0.040	171	0.234	1 *	15
	50	0.037	170	0.220	1.1	23
	100	0.038	138	0.273	0.8	55
	200	0.021	107	0.201	0.8	72
Toluene	2	0.035	406	0.086	9	0.1
	5	0.026	310	0.085	9	0.1
	10	0.032	275	0.116	1*	4.9
	30	0.049	116	0.423	0.9*	33
	50	0.027	128	0.211	0.8	42
	100	0.028	107	0.263	0.6	93
	200	0.023	102	0.228	0.4	180
Carbon disulphide	30	0.031	368	0.088	9	0.1
	200	0.031	332	0.094	9	0.1
Ethylene tetrachloride	30	0.027	285	0.096	9	0.1
Nitrobenzene	30	0.018	259	0.069	9	0.1
	200	0.012	207	0.056	9	0.1
Chlorobenzene	30	0.022	263	0.085	9	0.1
	200	0.026	230	0.115	9	0.2
Acetone	30	0.023	110	0.212	0.8	42
	200	0.023	108	0.218	0.4	170
Ethylene dichloride	30	0.026	187	0.139	0.7	36
	200	0.015	136	0.111	0.4	88
no cell		0.030	386	0.077	9	0.1
resonator with beam splitters						
Methanol	30	0.019	173	0.109	0.6	38
Benzene	200	0.043	146	0.295	0.8	59
Toluene	200	0.031	174	0.177	0.6	63
Carbon disulphide	200	0.023	403	0.057	9	0.1
Ethylene tetrachloride	200	0.039	388	0.010	9	0.2
Nitrobenzene	200	0.028	234	0.120	9	0.2
Chlorobenzene	200	0.020	315	0.085	9	0.1
	200	0.029	129	0.227	0.7	60
Acetone	200	0.024	183	0.131	0.6	46
Ethylene	200	0.024	183	0.131	0.6	46
no cell		0.029	372	0.079	9	0.1

The asterisk* at the value of \bar{d}_s signifies that the filament emission pattern (spots) appeared against a background of homogeneous radiation. When calculating the mean power density, the distribution of energy emitted in the filaments and homogeneous beam was evaluated from the picture of traces on the cross-section of the beam and from the structure of the oscillograms.

transmittivity or reflectivity coefficient of the liquid-variations which are the result of interaction between the latter and the light beam. In order to verify whether transmission depends on the incident power density, we performed transmission measurements in the laser beam behind the resonator using a pulse photometer. The beam was narrowed by means of a telescopic system of lenses, thus achieving power densities up to 640 MW/cm². In the case of methanol and toluene, transmission was found to increase with increasing power density; these variations however, were but slight and could thus hardly be of significance in the effect of Q-switching. The lack of a change in transmission of these liquids even at very high incident power densities can be interpreted as being due to a low value of the effective cross-section and to a short lifetime of excited states. In the liquids studied by us, the absorption band at 1.06 μm was caused by absorption on harmonics of the vibration frequencies. In these cases the lifetimes of the higher vibration levels are rather short, amounting to about 10⁻¹¹s [20-23].

The feasibility of the occurrence of absorption saturation can be assessed on the basis of the formulae (4) and (5), proposed by the author [24]. When a liquid sample of thickness W , transmission T , density ρ and molar mass M_m acted on during the time t by a light beam of power density I , then the probability that a quantum of radiation is absorbed per molecule is equal to:

$$W = \frac{I\tau_{1/2}M_m(1-T)}{h\nu W \rho 6.02 \cdot 10^{23}} \quad (4)$$

The change in transmission ΔT (for weak absorption saturation effects), determined at kinetic equilibrium of transmissions, is given by the formula

$$\Delta T = \frac{P(1-T)\tau}{h\nu \cdot \left(\frac{M}{M_m} \cdot 6.02 \cdot 10^{23} - \frac{P\tau_{1/2}(1-t)}{h\nu} \right)} \quad (5)$$

where P is the power of the radiation incident on the absorbing sample. With the following typical parameters

$$\begin{aligned} I &= 300 \text{ MW/cm}^2, & T &= 0.7, & \tau_{1/2} &= 100 \text{ ns}, \\ M_m &= 30 \text{ g}, & h\nu &= 1.874 \cdot 10^{-12} \text{ erg}, \\ W &= 100 \text{ mm}, & \rho &= 0.9 \text{ g/cm}^3, \\ \tau &= 10^{-11} \text{ s}, & d &= 0.1 \text{ mm}, \end{aligned}$$

one obtains from Eqs (4) and (5): $W \simeq 2.7 \cdot 10^{-4}$ and $\Delta T \simeq 2.6 \cdot 10^{-6} \%$.

Hence, of ten thousand only as few as three molecules have a chance to absorb a photon (during one flash), and the change in transmission thus induced will be as small as $3 \cdot 10^{-6} \%$. The preceding results explain why transmission, in the liquids in question, is independent of the power density.

In order to describe the ability of Q-factor modulation of the respective liquids, we have measured the coefficient of Q-factor modulation A , defined as

$$A = \frac{\left(\frac{S_2}{S_1}\right)_{\text{meas}} - \left(\frac{S_2}{S_1}\right)_{\text{calc}}}{\left(\frac{S_2}{S_1}\right)_{\text{calc}}}, \quad (6)$$

versus the liquid and the layer thickness. In Eq. (6) $(S_1/S_2)_{\text{meas}}$ denotes the experimentally measured ratio of probe signals S_2 and S_1 of

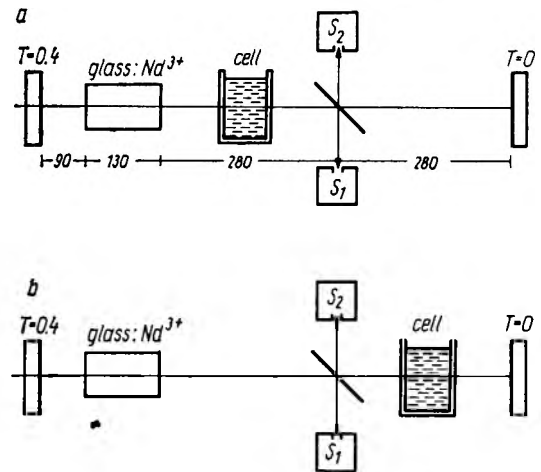


Fig. 2. Scheme of laser resonator enabling measurements of the Q-switching factor as depending on the kind of liquid and its length. The liquid cell was dismatch from the optical axis by an angle of 5°
a) Calibration setup, b) Measuring device

the pulse photometer referred to calibration signals, whereas S_1/S_2 is the signal ratio, calculated on the assumption of the Lambert-Beer law. The measurements were carried out in the circuit shown schematically in Fig. 2. The experimental results are to be found in Table 2.

On inspection of Table 2, one notes that the largest changes in Q-factor of the laser resonator occur for methanol, toluene, acetone and ethylene dichloride. The changes are the larger the thicker is the layer of the liquid.

The next step consisted in measurements of the coefficient A for the various liquids versus the laser light standing wave coefficient. The measuring circuit differed from that of

Table 2

Experimental results for A , the coefficient of Q-factor modulation, for various liquids and cell thickness

Liquid	W [mm]	$\left(\frac{S_2}{S_1}\right)_z$	$\left(\frac{S_2}{S_1}\right)_0$	A
Methanol	2	0.895	0.788	0.136
	5	0.838	0.679	0.234
	10	0.755	0.575	0.313
	20	0.643	0.461	0.395
	30	0.602	0.401	0.501
Benzene	2	0.990	0.830	0.193
	5	0.976	0.808	0.208
	10	0.957	0.788	0.214
	30	0.922	0.796	0.158
	50	0.889	0.764	0.164
	100	0.958	0.667	0.436
Toluene	2	0.828	0.819	0.011
	5	0.918	0.785	0.169
	10	0.903	0.776	0.164
	30	0.890	0.745	0.195
	50	0.819	0.707	0.158
	100	0.745	0.559	0.333
Carbon disulphide	30	0.892	0.885	0.008
	200	0.812	0.801	0.014
Ethylene tetrachloride	30	0.894	0.882	0.014
	200	0.827	0.812	0.018
Nitrobenzene	30	0.883	0.843	0.047
	200	0.708	0.663	0.068
Chlorobenzene	30	0.875	0.857	0.021
	200	0.735	0.709	0.034
Acetone	30	0.948	0.817	0.160
	200	0.718	0.507	0.416
Ethylene dichloride	30	0.957	0.814	0.176
	200	0.702	0.503	0.396

Fig. 2 since the resonator contained two mirrors of zero transmission one of them being shielded with a filter. By varying the transmission T of the filter F , it was possible to determine the standing wave ratio. The results are given in Fig. 3.

These measurements show that, in the experimental conditions stated, effective interaction between the light beam and the liquid is possible only at high values of the standing wave coefficient. The latter conclusion was confirmed by transmission and reflection measurements of the liquid, performed beyond the resonator i.e. in conditions of zero standing wave coefficient.

It is thus plausible that Q-factor modulation of the resonator is caused by an increase in reflectivity of the liquid during the evolu-

tion of laser action. Our measurements indicate that the reflecting structure arises as result of the interaction between the standing wave and the liquid: within the latter, the refractive index varies periodically, in spatial correlation with the nodes and anti-nodes of the standing light wave.

In order to gain insight into the magnitude of the effects causing Q-factor modulation, we carried out measurements for the determination of absolute refractive index variations in a liquid subjected to the action of a standing light wave. The procedure used to determine the energy distribution in the resonator is shown in Fig. 4. With the known energy ratios of the beams extracted from the resonator through the beam splitters, the absolute reflectivity coefficients of the liquids can be calculated by solving the following set of equations:

$$\frac{E_2}{E_1} = [(T + \Delta T) - (R_1 + \Delta R_1)] \times \sum_{i=0}^{N_0} R_p^{2i} (R_2 + \Delta R_2)^i,$$

$$\frac{E_4}{E_1} = \frac{1}{R_p^2} (R_1 + \Delta R_1) + [(T + \Delta T) - (R_1 + \Delta R_1)] \times [(T + \Delta T) - (R_2 + \Delta R_2)] \sum_{i=0}^{N_0} R_p^{2i} (R_2 + \Delta R_2)^i,$$

$$\frac{E_4}{E^3} = [(T + \Delta T) - (R_2 + \Delta R_2)] + \frac{R_1 + \Delta R_1}{R_p^2} \frac{E_2}{E_1}, \quad (7)$$

$$i = 1, 2, 3, \dots, N_0.$$

where E_1, E_2, E_3, E_4 are the corresponding relative energy values of the beams marked in Fig. 4; T — the transmission of the liquid layer for a light beam of low power density; ΔT — the change in transmission as a function of the power density; R_1, R_2 — the reflection coefficients of the cell windows; $\Delta R_1, \Delta R_2$ — the reflection coefficients of the liquid in the cell, in the direction of the laser rod and back (towards the mirror), respectively; R_p — the reflection coefficient of the beam splitter set at 45° to the axis of the beam; N_0 — the number of times the beam traverses the resonator there and back during one pulse.

The set of Equations (7) holds on the assumption of a rectangular pulse shape, and constant $\Delta T, \Delta R_1$ and ΔR_2 throughout the pulse duration. Thus, by solving (7), we obtain time-averaged values of ΔR_1 and ΔR_2 . If the transmission of the liquid does not depend on the power density of the beam, the number of

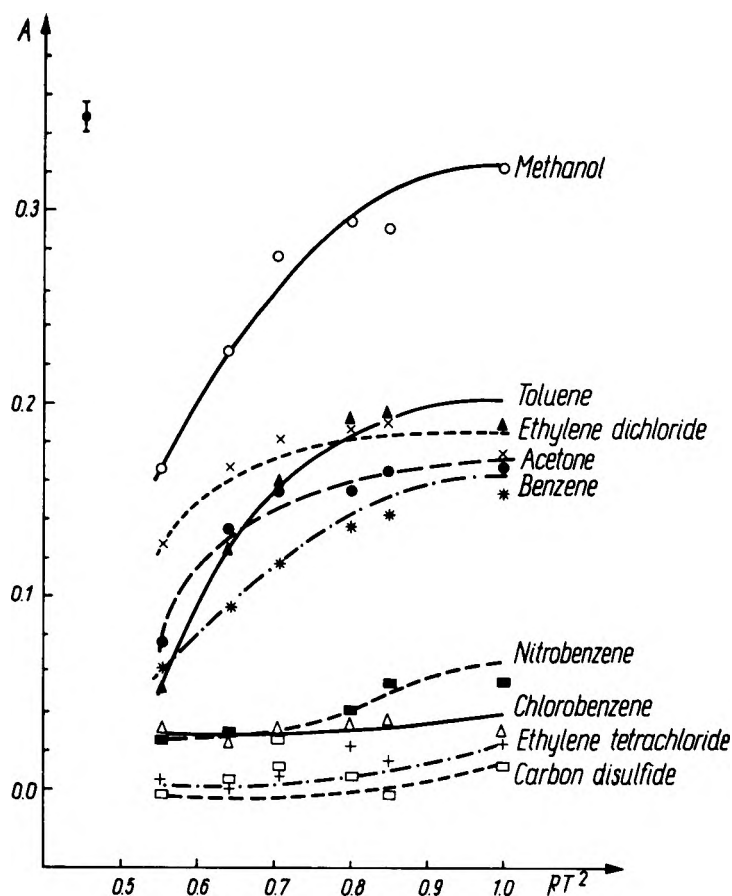


Fig. 3. Q-switching factor A as function of reflectivity of the arrangement: mirror-filter, for various liquids

equations is reduced to 2. The set of Equation (7) was solved by numerical methods. The solutions obtained, for various liquids and layer thickness values are given in Table 3.

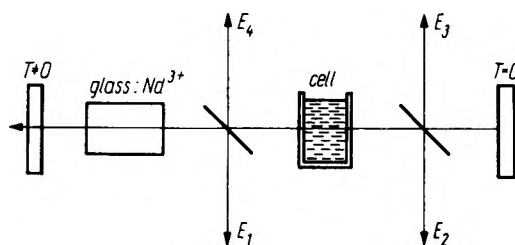


Fig. 4. Scheme of the laser containing active medium, liquid cell and two beamsplitters

It is noteworthy that, in conformity with our other experiments the largest values of ΔR were obtained for methanol, toluene, acetone and ethylene dichloride; smaller values for benzene; very small values for chlorobenzene and nitrobenzene; and practically zero for carbon disulphide and ethylene tetrachloride. As a rule, $\Delta R_2 > R_1$ is in agreement with the measurements described above, since the standing wave coefficient in the cell, is much closer to the mirror than to the rod.

3. Tentative mechanism of the effect

The preceding experiments give a direct evidence that changes in transmission play no significant role in the interactions between a light beam of high standing wave coefficient and a liquid dielectric. The results of our measurements, and the analysis of the spectral structure of the radiation emitted by lasers with organic solvents in the resonator lead to the conclusion that, owing to reflectivity variations in the liquids, the effects travelling wave, thus back-stimulated Mandelstam-Brillouin and back-stimulated Rayleigh scattering, are not apparent. This is obvious, considering that the assessed radiation power densities in the resonator (even at the anti-nodes of the standing wave) lie well below the threshold power density for stimulated Mandelstam-Brillouin scattering. Stimulated temperature Rayleigh scattering presents also a high power density threshold. HERMAN and GRAY [25], and RANK et al. [26] have found that, for a given liquid, there exists a critical value of the absorption coefficient α_{cr} :

$$\alpha_{cr} = \left(\frac{n^2 - 1}{2} \right) \left(\frac{n^2 + 2}{3} \right) \frac{\gamma c_v \omega_B}{\beta_n v^2 c} \frac{\Gamma + \Gamma_R}{\Gamma + \Gamma_B} \quad (8)$$

at which the threshold power density values for stimulated Mandelstam-Brillouin and Rayleigh scattering are equal. In Eq. (8) n denotes the refractive index of the liquid; β — its

Table 3

Values of ΔR_1 and ΔR_2 , calculated

Liquid	W [mm]	ΔR_1	ΔR_2
Methanol	2	0.098	0.099
	5	0.113	0.176
	10	0.216	0.239
	20	0.248	0.301
	30	0.321	0.364
Benzene	2	0.034	0.042
	5	0.030	0.042
	10	0.035	0.051
	30	0.162	0.206
	50	0.116	0.157
	100	0.120	0.181
Toluene	2	0.101	0.101
	5	0.079	0.101
	10	0.162	0.213
	30	0.185	0.239
	50	0.158	0.208
	100	0.175	0.264
Carbon disulphide	30	0.031	0.032
	200	0.024	0.026
Ethylene tetrachloride	30	0.032	0.033
	200	0.021	0.023
Nitrobenzene	30	0.055	0.085
	200	0.046	0.056
Chlorobenzene	30	0.042	0.047
	200	0.027	0.047
Acetone	30	0.236	0.266
	200	0.196	0.316
Ethylene dichloride	30	0.244	0.273
	200	0.211	0.279

compressibility coefficient; c_v — its specific heat at constant volume; v — hypersonic velocity; and Γ , Γ_R , Γ_B — the spectral linewidths of the incident light, spontaneous Rayleigh line, and spontaneous Mandelstam-Brillouin scattering component, respectively; $\gamma = C_p/C_v$.

The value of α_{cr} amounts to: 0.34 in acetone, 0.31 in methanol, 0.62 in carbon disulphide, and to 0.65 in benzene. Hence, for the liquids under investigation, stimulated temperature Rayleigh scattering (which could raise considerably the reflectivity of the liquid), at the

wavelength 1.06 μm , is established at a power density much in excess of that required for stimulating Mandelstam-Brillouin scattering.

The experimental results show clearly that the observed increase in reflection of the liquid is the resultant effect of elementary reflections of the beam on the successive layers (with different refractive indices) of the periodic laminar structure formed within the liquid. The layer structure is the result of the action of the

Table 4

Pulse parameters adopted in calculation of the variations in refractive index. I_0 is the radiation power density in the resonator at an anti-node of the standing wave

Liquid	W [mm]	I_0 [MW/cm ²]	$\tau_{1/2}$ [ns]	$E \cdot 10^{-4}$ [V/cm]	E [i.e.s.]
Methanol	2	20.2	237	12.3	410
	5	27.0	340	14.3	476
	10	252	201	43.6	1451
	20	253	170	43.7	1454
	30	280	173	46.0	1530
Benzene	2	0.8	330	2.4	81
	5	0.6	360	2.2	73
	10	1.1	250	2.9	96
	30	83	170	25.1	837
	50	110	170	28.8	961
	100	294	140	47.1	1570
Toluene	2	0.9	400	2.6	87
	5	0.8	310	2.4	82
	10	1.0	270	2.7	90
	30	69	120	22.8	762
	50	140	130	32.5	1084
	100	327	110	49.6	1654
Carbon disulphide	200	509	100	62.0	2065
	30	0.8	370	2.5	82
Acetone	200	0.6	370	2.1	68
	30	107	110	28.4	941
	200	474	110	59.8	1991

standing wave on the latter. In these conditions, significantly large changes in the refractive index of the liquid can arise due to electrostriction, molecular reorientation and redistribution, as well as to thermal interactions. When calculating the changes in refractive index due to these various effects, we have employed the experimental pulse parameters given in Table 4.

When a liquid dielectric (of permittivity ϵ and density ρ) is subjected to the action of an electric field E (in c.g.s.e.s.u.), the electrostric-

tive increase in pressure ΔP and change in refractive index Δn_{es} are, after KACZMAREK [18]:

$$\Delta p = \frac{1}{8\pi} E^2 \left(\rho \frac{\partial \epsilon}{\partial \rho} \right), \quad (9)$$

$$\Delta n_{es} = \frac{\left(\rho \frac{\partial \epsilon}{\partial \rho} \right)^2 \beta_s E^2}{n_0}, \quad (10)$$

where β_s is the adiabatic compressibility coefficient. The values of ΔP and Δn_{es} calculated from Eqs. (9) and (10) are given in Table 5.

Table 5

Electrostrictive changes in pressure and refractive index

Liquid	W [mm]	$\rho \frac{\partial \epsilon}{\partial \rho}$	$\beta_s \cdot 10^{12}$	ΔP [atm]	$\Delta n_{es} \cdot 10^8$
Methanol	2	0.82	100	0.54	16.9
	5			0.72	22.7
	10			6.8	212
	20			6.8	212
	30			7.5	235
Benzene	2	1.56	52.6	0.04	1.1
	5			0.03	0.9
	10			0.06	1.6
	30			4.3	117
	50			5.7	155
	100			15.1	413
Toluene	2	1.60	70.0	0.05	2.0
	5			0.04	1.7
	10			0.05	2.1
	30			3.6	151
	50			7.4	305
	100			17.2	713
Carbon disulphide	30	2.39	49.5	0.06	2.2
	200			0.04	1.6
Acetone	30	1.34	89.0	4.	189
	200			20.97	899

Orientalional-redistributional effects can cause considerable variations of the refractive index (KIELICH [27, 28]; HELLWARTH [29]), particularly, if the light beam incident on the medium is completely linearly polarized. An approximate, but at the same time, simple formula for the calculation of the above variations has been proposed by HELLWARTH [29]:

$$\Delta n = n_2 E^2, \quad (11)$$

where n_2 is the value of the nonlinear term of the index. Values of n_2 for the liquids studied here calculated by HELLWARTH [29] as well as

values of the variations in index Δn for the condition of our experiments are given in Table 6.

Table 6

Changes in refractive index due to molecular redistribution effects, calculated

Liquid	W [mm]	$n_2 \cdot 10^{12}$	$\Delta n_r \cdot 10^7$
Methanol	2	50	84
	5		112
	10		1053
	20		1057
	30		1170
Benzene	2	37	2.4
	5		2.0
	10		3.4
	30		259
	50		341
	100		912
Toluene	2	45	3.4
	5		3.0
	10		3.7
	30		261
	50		528
	100		1234
Carbon disulphide	30	96	6.4
	200		4.5
Acetone	30	6.7	60
	200		266

Temperature changes in index Δn_t were calculated by the method of SCARLET and led finally to the following expression for thermally induced changes in index [30]:

$$\Delta n_t = \frac{\partial n}{\partial T} \cdot \frac{2I_0 \tau_{1/2} (1-T)}{W \cdot c_p}. \quad (12)$$

The amplitudes of the above variations, calculated from Eq. (12), are to be found in Table 7.

It is apparent from the data of Tables 6, 7 and 8 that, in nine of the liquids studied, the largest changes in refractive index are caused by thermal effects. This is additionally corroborated by the well marked correlation between the absorption coefficient value of each of the liquids and the magnitude of the Q-factor modulation effects caused by the liquid when in the laser resonator (see Table 8).

The above facts point to thermal interactions as the principal factor giving rise to a reflecting structure. Absorption of the laser beam

Table 7

Changes in temperature Δt and refractive index Δn due to absorption of radiation

Liquid	W [mm]	c_p [J/cm ³ K]	$\frac{\partial n}{\partial t} \cdot 10^5$ [K ⁻¹]	Δt [K]	$\Delta n \cdot 10^5$
Methanol	2	2.18	40.5	0.88	36
	5			1.87	75
	10			4.04	164
	20			5.21	211
	30			4.71	191
Benzene	2	1.57	64	0.03	2
	5			0.02	1
	10			0.01	1
	30			0.32	20
	50			0.29	18
	100			0.50	32
	200			0.46	29
Toluene	2	1.48	61	0.06	4
	5			0.03	2
	10			0.04	2
	30			0.31	19
	50			0.47	28
	100			0.81	50
	200			0.93	57
Carbon disulphide	30	1.30	81.5	0.00	0
	200			0.00	0
Acetone	30	1.74	50	0.18	9
	200			0.60	30

by the liquid plays a double role: on the one hand, light absorption at the laser wavelength permits the evolution of thermal effects (we neglect weak electrocaloric phenomena) whereas, on the other, the presence in the resonator of an absorbant sample, initially lowering the Q-factor is a necessary condition for the occurrence of Q-factor modulation. The absence of Q-factor modulation when a liquid not absorbing the laser wavelength (carbon disulphide, ethylene tetrachloride) is placed in the resonator is by no means evidence that a reflecting structure is not formed in the liquid. Nonetheless, if spatially periodic changes in index were to appear in such a liquid (e.g. due electrostriction or redistribution), symptoms of Q-factor modulation should become apparent in the system on lowering the Q-factor of the resonator by introducing an absorbing filter. Thus, the facts show that the periodic structure of refractive index variations in the liquid is essentially due to thermal interaction of the beam and medium. A lesser role is played by molecular redistribution effects though, if taken into account, they permit to explain certain irregularities in Table 8. Thermal chan-

Table 8

Calculated values of the absorption coefficient α and experimentally measured values of the power density and duration time of the pulse, the Q-switching factor, and coefficients ΔR_1 and ΔR_2 , for the different liquids and a layer thickness of 30 mm

Liquid	α [1/cm]	Q-switching coefficient	Mean duration time of the pulse [ns]
Methanol	0.127 ± 0.005	0.501	133
Toluene	0.0157 ± 0.0007	0.195	116
Ethylene dichloride	0.0113 ± 0.0006	0.176	188
Acetone	0.0105 ± 0.0006	0.160	110
Benzene	0.0087 ± 0.0006	0.158	171
Nitrobenzene	0.0049 ± 0.0005	0.047	259
Chlorobenzene	0.0036 ± 0.0005	0.021	263
Ethylene tetrachloride	0.0000	0.014	285
Carbo disulphide	0.0000	0.008	368

	Mean power density of the pulse [MW/cm ²]	ΔR_1	ΔR_2
Methanol	68	0.321	0.364
Toluene	33	0.185	0.239
Ethylene dichloride	36	0.244	0.273
Acetone	42	0.236	0.266
Benzene	15	0.162	0.206
Nitrobenzene	0.1	0.055	0.085
Chlorobenzene	0.1	0.042	0.047
Ethylene tetrachloride	0.1	0.032	0.032
Carbo disulphide	0.1	0.031	0.032

ges in index are characterized by the longest rise time (of the order of several ns [30]) but are still fully able to follow the changes in radiation power density, considering the relatively slow time-evolution of the pulses (half-times of the order of 100 ns). Although interferometric and spectroscopic measurements of the laser emission signal spectra failed to provide decisive data concerning the interaction mechanism, they nevertheless permitted to describe an interesting dependence of the spectral line-width on the magnitude of Q-factor modulation [14]. The conclusions of the above cited authors regarding the mechanism of interaction between light waves of nonzero standing wave coefficient and liquid dielectrics

differ essentially from those of the present paper. However, the mechanism proposed here is in agreement with all the experimental facts available.

* *
*

The author is indebted to Prof. Dr F. Kaczmarek for his helpful comments and discussions, and to K. Flatau, M. Sci., for the English version.

**Влияние стоячих волн на взаимодействие
между пучками интенсивного света и жидкими
диэлектриками**

Подробно обсуждены условия действия лазера из неоднородного стекла, в резонаторе которого содержится поглощающая жидкость. Определено квазипереключение Q лазерной эмиссии, вызванной жидкостями, в зависимости от вида жидкости, ее плотности, энергии пульсирования, а также от коэффициента стоячей волны.

Вычислены изменения в коэффициенте преломления, вызываемые электрострикцией, молекулярным перераспределением и термическими явлениями. Предложено истолкование обнаруженных явлений. В нем учитывается воздействие лазерного света на периодические изобарные флуктуации коэффициента отражения. Последний индуцировался структурой стоячей волны.

References

[1] KACZMAREK F., *Wstęp do fizyki laserów*, PWN, Warszawa (in press).
 [2] KIELICH S., *Podstawy optyki nieliniowej*, A. Mickiewicz University Press, Poznań, Part I – 1972, Part II – 1973.
 [3] STANKOWSKI J., GRAJA A., *Wstęp do elektroniki kwantowej*, PWN, Warszawa 1972.
 [4] FABIELINSKI L. L., *Molekularnoye rasseyanye sveta*, Izd. "Nauka", Moscow 1965.
 [5] PLANNER A., Doctor's Thesis, Institute of Physics of A. Mickiewicz University, 1971.
 [6] KATZENSTEIN J., MAGYAR G., SELDEN A. C., *J. Opto-Electronics* **1**, 13 (1969).

[7] HARRISON R. G., KEY P., LITTLE V. I., MAGYAR G., KATZENSTEIN J., *Appl. Phys. Letters* **13**, 253 (1968).
 [8] BŁASZCZAK Z., DOBEK A., PATKOWSKI A., *Acta Phys. Polon.* **A42**, 349 (1972).
 [9] GAPONOV S. V., GONCHAROV A. G., KRAFTMAKHER G. A., KHANIN J. I., *ZhETF, Pisma v redaktsiyu* **11**, 370 (1970).
 [10] POHL D., *Phys. Letters* **24A**, 239 (1967).
 [11] PLANNER A., SZYMAŃSKI M., *Acta Phys. Polon.* **A41**, 241 (1972); *Postępy Fizyki* **23**, 313 (1972).
 [12] GRASYUK A. Z., RAGULSKII V. V., FAYZULLOV F. C., *ZhETF, Pisma v redaktsiyu* **9**, 11 (1969).
 [13] POPOVICHEV V. I., RAGULSKII V. V., FAYZULLOV F. C., *Kvantovaya Elektronika* **5**, 126 (1972).
 [14] SZYMAŃSKI M., *Acta Phys. Polon.* (in preparation).
 [15] BORN M., WOLF E., *Principles of Optics*, Pergamon Press 1964.
 [16] KASPROWICZ B., KIELICH S., *Acta Phys. Polon.* **31**, 787 (1967).
 [17] SHEN R. Y., *Phys. Letters* **20**, 378 (1966).
 [18] KACZMAREK F., *Acta Phys. Polon.* **32**, 1003 (1967).
 [19] SZYMAŃSKI M., VI-th Conference *Quantum Electronics and Nonlinear Optics*, **A20**, 109, Poznań 1974.
 [20] ANDRAE J. H., LAMB J., *Proc. Roy. Soc.* **A226**, 51 (1954).
 [21] ANDRAE J. H., HEASELL E. L., LAMB J., *Proc. Phys. Soc.* **L XIX**, 6-B, 625 (1956).
 [22] LAMBEREAU A., von der LINDE D., KAISER W., *Opt. Commun.* **7**, 173 (1973).
 [23] LAMBEREAU A., KIRSHNER K., KAISER W., *Opt. Commun.* **9**, 182, (1973).
 [24] SZYMAŃSKI M., Doctor's Thesis, Institute of Physics of A. Mickiewicz University, Poznań 1975.
 [25] HERMAN R. M., GRAY M. A. *Phys. Letters* **19**, 824 (1967).
 [26] RANK D. H., CHO C. W., FOLTZ N. D., WIGGINGS T. A., *Phys. Rev. Letters* **19**, 828 (1967).
 [27] KIELICH S., *Acta Phys. Polon.* **23**, 321 (1964); **A37**, 719 (1970); **27**, 305 (1965); **28**, 95 (1965); *J. Phys.* **28**, 519 (1967); *Mol. Phys.* **6**, 49 (1963).
 [28] PIEKARA A., KIELICH S., *J. Chem. Phys.* **29**, 1297 (1958).
 [29] HELLWARTH R. W., *Phys. Rev.* **152**, 156 (1967).
 [30] SCARLET R. I., *Phys. Rev. A*, **6**, 2281 (1972).

Received February 9, 1976