SPECTRAL-TIME METHOD FOR INVESTIGATING PARTIAL MODE LOCKING IN RUBY AND NEODYMIUM GLASS LASERS

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A fluctuation representation of the output radiation is used to explain an irregular structure of the free-oscillation spectra of solid-state lasers. A spectral-time method is suggested for investigating partial mode locking in lasers with passive Q-switches. It was found experimentally that the selection of a single pulse (full mode locking) in ruby and neodymium glass lasers became less efficient when the width of the emission spectrum was increased. This investigation was carried out on traveling- and standing-wave lasers using passive Q-switches made of various materials.

IN an earlier paper,[1] we reported the discovery of a structure in the emission spectra of solid-state lasers operating under free-oscillation conditions. Further investigations showed that the suppression of the mode selection and of the spatial inhomogeneity in the inversion depletion did not destroy the structure in the emission spectra even in the case of lasers with uniformly broadened luminescence lines.[2] An irregular structure was also observed by us in the spectra of ruby and neodymium glass lasers with passive Q-switches.

The present paper gives an explanation of the observed structure on the basis of fluctuation representations of the laser radiation, and suggests a spectral-time method for investigating partial mode locking, which can be used to determine the effectiveness of a passive Q-switch as a mode-locking device.

The emission spectra of lasers operating under free-oscillation conditions and of lasers with passive Q-switches are formed, during the linear stage of the development of laser emission, from a background of noise along the time axis. We shall assume that the radiation field of a laser during one axial period consists of short identical pulses f(t); the moments of appearance of these pulses θ_j and their relative amplitudes are random quantities:

$$E(t)=\sum_j a_j f(t-\theta_j)\exp\{2\pi i c\nu_0 t+i\varphi_j\}.$$

The complex amplitude of the field spectrum is

$$\tilde{E}(\nu)=\tilde{f}(\nu)\sum_j a_j\exp\{-2\pi i c(\nu-\nu_0)\theta_j+i\varphi_j\}.$$

Since the number of terms in this sum is large (it is governed by the width of the spectrum and is $\sim 10^2$ for a ruby laser and $\sim 10^3$ for a neodymium glass laser), the value of the sum has a normal distribution in the complex plane. The intensity measured as a percentage of $[\tilde{f}(\nu)]^2$ is distributed in accordance with the law $I_0^{-1}\times\exp(-I/I_0)$. If the distribution of pulses in one axial period is more or less uniform, the frequency interval of correlation of the random function $\tilde{E}(\nu)\tilde{E}^*(\nu)/[\tilde{f}(\nu)]^2$ is of the order of the mode spacing $\delta\nu_m$. When an emission spectrum is recorded with a spectroscopic device whose resolution is $\delta\nu$, the average relative deviation of the intensity from the mean is $\approx\sqrt{\delta\nu_m/\delta\nu}$ and it usually amounts to a few tens of a percent.

In the nonlinear region, the interaction of a passive Q-switch with laser radiation results in the selection of the strongest peaks along the time axis and in the narrowing of these peaks.[3,4] In spectroscopic language, this represents the flattening of the structure and the broadening of the spectrum. The case of full mode locking corresponds to the selection of a single ultrashort pulse in one axial period, i.e., in this case, the spectrum becomes "smooth" to within one mode spacing. In the case of partial mode locking, only a few ultrashort pulses remain in one axial period and the spectrum has a structure which carries information on the time dependence of the emitted radiation. We shall illustrate this point by considering the emission spectrum of a laser with several pulses in one axial period. Using the notation established earlier, the energy spectrum of the radiation field is now

$$I(\nu)=\tilde{E}(\nu)\tilde{E}^*(\nu)=$$
$$=[\tilde{f}(\nu)]^2\left\{\sum_j a_j^2+2\sum_{j>k}a_j a_k\cos[2\pi(\nu-\nu_0)c(\theta_j-\theta_k)-(\varphi_j-\varphi_k)]\right\}.$$

We can see that the spectral pattern is formed by the superposition of $[\tilde{f}(\nu)]^2$ "beats" on the spectrum of a single pulse. The period of such beats is governed by the time intervals between spikes and the amplitude is determined by the relative energies of the spikes.

Generally speaking, the spectra obtained with a spectroscopic instrument cannot be used to reconstruct the emission pattern in relation to time. However, in tackling specific problems, it is found that the information contained in the structure of a spectrum is sufficient to determine the time dependence of the emitted radiation. Thus, for example, in our case, the structure of a spectrum whose period is $\Delta\nu_s$ should be interpreted as being due to the presence, along the time axis, of pulses separated by an interval $1/c\Delta\nu_s$. The important point is that the interval between pulses decreases with increasing value of the structure period. In the case of two ultrashort pulses in one axial period, the emission spectrum has a sinusoidal structure with a period $1/c\delta\Theta$, where $\delta\Theta$ is the time interval between pulses. If $\delta\Theta \lesssim T$ (where T is the axial period), the structure can be

discovered with a spectroscopic instrument which can resolve spectral intervals down to the mode spacing. If such an instrument cannot resolve a mode spacing, time intervals of the order of T must be investigated additionally by using simultaneously a spectroscopic instrument and a device such as a photoelectric detector with an oscillograph having a relatively "coarse" time resolution (~ 1–2 nsec). If only one ultrashort pulse is generated in one axial period, the spectrum has only the mode structure, i.e., by our definition such a spectrum is "smooth."

In the foregoing discussion, we have assumed that there is no phase modulation of the output radiation pulses. In the field description such modulation is represented by factors of the exp [$i\varphi(t)$] type. The presence of a nonoscillating phase modulation (of the $\varphi(t) = \beta t^2$ type) does not cause any difficulty in the interpretation of the "smooth" spectra because this modulation does not produce any additional structure. A structure can be introduced into a spectrum only by an oscillating phase modulation, which is unlikely to occur in a laser.

Our discussion shows that the "smoothness" of a spectrum is a sufficient condition for the selection of a single ultrashort pulse in one axial period, i.e., for the full mode locking. This criterion was used in our investigation of the mode locking in ruby and neodymium lasers with passive Q-switches.

The apparatus used to investigate partial mode locking in ruby and neodymium glass lasers is shown schematically in Fig. 1. We used a ring resonator configuration because it prevented the appearance of an additional spectral structure associated with the actual position of a passive switch (PS) cuvette in the resonator. The use of mirrors M_1, M_2, and M_3, supported by wedge-shaped bases, and the alignment of the cuvette and an active rod (AR) at the Brewster angle with respect to the resonator axis, prevented accidental mode selection. The nonaxial modes were suppressed by introducing stops (S_1, S_2) with apertures of 3 mm diameter into the resonator. The spectroscopic instrument (SP) was either a Fabry–Perot interferometer with a frequency resolution down to $\delta\nu = 0.008$ cm^{-1}, or a diffraction-grating spectrograph with $\delta\nu = 0.1$ cm^{-1}. The time dependence was recorded with a photodiode (PD) of the FÉK-0.9 type and an oscillograph (O) of the I2-7 type with a combined time resolution of not less than 1.5 nsec. The passive Q-switch for the neodymium glass laser was a solution of a polymethenyl dye in nitrobenzene, giving a transmission $T_0 = 35\%$. A solution of kryptocyanine in ethyl alcohol, with $T_0 = 25\%$, was used for the ruby laser. The resonator length was ≈ 170 cm. The cuvette was 1 mm thick.

Our investigation was carried out on spectra of different widths. The width of a spectrum was altered by inserting various plane-parallel plates (P) in the resonator, which acted as selectors. For each width of the spectrum we obtained a series of spectrograms and oscillographs keeping the laser parameters fixed. The number of spectrograms and oscillograms in each series was not less than 20 for the neodymium laser and not less than 30 for the ruby laser. This investigation demonstrated that in each series spectra of different types were obtained: both structured and "smooth" spectra were observed.[1)] The relative numbers of the structured and "smooth" spectra depended on the spectral width. Typical microphotograms and oscillograms are presented in Fig. 2.

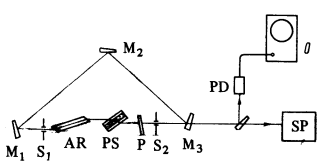

FIG. 1. Schematic representation of the apparatus.

When the spectral width had its maximum value ($\Delta\nu = 20$ cm^{-1} for the neodymium laser and $\Delta\nu = 0.8$ cm^{-1} for the ruby laser), the "smooth" spectra were practically never observed and the pulse duration in the oscillograms was equal to the width of the apparatus function of the recording system (Figs. 2a and 2e). The irregular structure observed in the spectra indicated the presence of a large number of pulses in a time interval, which was not resolved oscillographically.

We estimated the relative deviation from the mean in a spectrum whose width was $\Delta\nu = 20$ cm^{-1} and which was

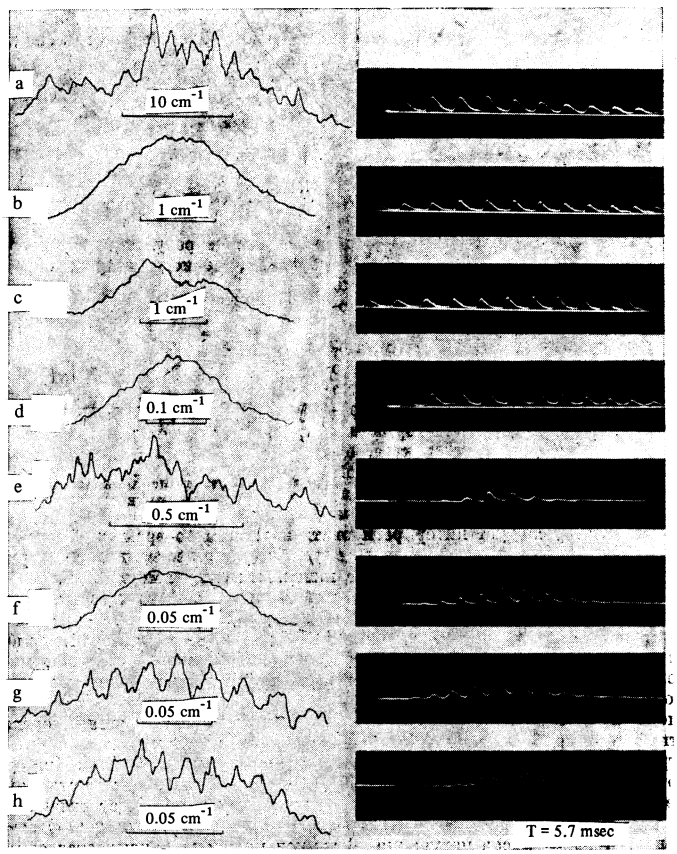

FIG. 2. Microphotograms of the spectra and the corresponding oscillograms of the radiation emitted by a neodymium glass laser (a-d) and a ruby laser (e-h). Microphotograms a-c were obtained using a diffraction-grating spectrograph with a $\delta\nu = 0.1$ cm^{-1} resolution; the other five microphotograms were obtained using a Fabry-Perot interferometer and the following resolutions: d) $\delta\nu = 0.015$ cm^{-1}; e) $\delta\nu = 0.05$ cm^{-1}; f), g), h) $\delta\nu = 0.008$ cm^{-1}.

[1)] A spectrum was regarded as "smooth" when the scatter of the values of the blackening in microphotograms did not exceed the mean square value of the photographic emulsion noise.

recorded with a frequency resolution of 0.1 cm^{-1}. Radiation pulses were located, as indicated by the oscillograms in Fig. 2, in a time interval shorter than one-quarter of the axial period. Consequently, the frequency interval of correlation in the spectrum was not less than $4\delta\nu_m$. The relative deviations from the mean in the spectrum were not less than $\sqrt{4\,\delta\nu_m/\delta\nu} = 0.5$ and therefore they should be observed easily with the apparatus employed in our study.

When the spectral width was reduced to $\Delta\nu \leq 2$ cm^{-1} for the neodymium laser and to $\Delta\nu = 0.12$ cm^{-1} for the ruby laser, more "smooth" spectra were observed (Figs. 2b, 2d, 2f). The structure of these spectra became simpler and a periodic pattern predominated in the time dependence, indicating that there were fewer ultrashort pulses in the emitted radiation (Figs. 2c and 2g).

Difficulties were encountered in the interpretation of the "smooth" spectra which were $\Delta\nu = 2$ cm^{-1} wide. Several ultrashort pulses, separated by time intervals of 0.3–1.5 nsec, could appear in one axial period in an oscillogram but a structure in the corresponding frequency spectrum (0.1–0.02 cm^{-1}) could not be resolved by the spectroscopic instruments used in our investigation. A similar situation was encountered when the frequency resolution of the spectroscopic instrument, expressed in time units ($1/c\delta\nu$), was less than the time resolution of the photodiode-oscillograph system, i.e., when the frequency and time resolutions did not overlap (Fig. 3). The use of an oscillograph made it possible to resolve time intervals longer than 1.5 nsec, which could not be resolved by our spectroscopic apparatus.

The frequency and time resolutions did not overlap either in the case $\Delta\nu = 20$ cm^{-1} but this did not impede our interpretation because the "smooth" spectra were not observed for the number of modes ($\sim 10^3$) excited in this case.

However, it should be stressed that these difficulties were not of a basic nature because they could be avoided by using spectroscopic instruments of higher resolution.

When narrow spectra ($\Delta\nu = 0.2$ cm^{-1} and $\Delta\nu = 0.12$ cm^{-1}) were recorded with the Fabry–Perot interferometer using resolutions of $\delta\nu = 0.015$ cm^{-1} and $\delta\nu = 0.008$ cm^{-1}, respectively, the regions of frequency and time resolution were found to overlap. This made it possible to observe simultaneously the structure in the time dependence of the emission spectrum and the corresponding structure in the frequency spectrum, as shown in Fig. 2h. The time dependence had its two strongest peaks separated by $\delta\Theta \approx T/2$, where $T = 5.7$ nsec. The structure corresponding to these two peaks ($\Delta\nu_s \approx 2\,\delta\nu_m = 0.011$ cm^{-1}) was observed clearly in the frequency spectrum.

Thus, the suggested spectral-time method can be used to estimate qualitatively the efficiency of a passive Q-switch in the selection of a single ultrashort pulse and to determine whether full mode locking occurs under practical conditions.

Table I gives the results of a statistical analysis of the spectra obtained. For each spectral width, we give the corresponding number of cases, expressed in percent, when a single ultrashort pulse was isolated in accordance with the aforementioned criterion.

It is evident from Table I that an increase in the width of a spectrum makes the conditions for full mode locking less favorable and such locking is practically unattainable for spectra whose width is 0.8 cm^{-1} in the case of the ruby laser and 20 cm^{-1} in the case of the neodymium glass laser (these values apply to the passive Q-switches and the resonators used in our investigation). This conclusion is in agreement with the results reported in [4], where it is shown that an increase in the number of excited modes makes it more difficult to select a single ultrashort pulse in one axial period.

The suggested spectral-time method was also used by us to investigate the influence of the laser resonator parameters on the mode locking phenomenon. For example, in addition to the ring resonator, we investigated mode locking in a straight resonator. This investigation showed that the frequencies of the appearance of the "smooth" spectra differed slightly from those given in Table I. When the mode locking occurred in the resonator of a traveling-wave laser (the traveling-wave conditions were achieved by the use of a back-reflecting mirror[2]), the frequencies of appearance of the "smooth" spectra were considerably lower than those given in Table I. These observations indicated that the use of standing-wave conditions in a laser was advantageous from the point of view of the selection of a single ultrashort pulse in one axial period.

In addition to a solution of kryptocyanine in ethyl alcohol, we used a Q-switch for the ruby laser which consisted of a solution of vanadium phthalocyanine in chlorobenzene. However, when this dye was used and the width of the spectrum was set at $\Delta\nu = 0.12$ cm^{-1}, the "smooth" spectra were practically never observed, which indicated that this dye was less efficient in mode locking.

The spectral-time method described in the present paper can be used not only in investigations of the mode locking phenomenon in solid-state lasers but also in investigations of fast light-pulse processes with a resolution down to time intervals of the order of the pulse width.

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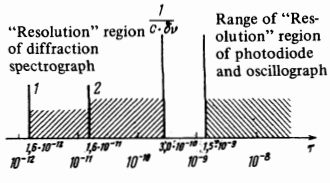

FIG. 3. Regions of time resolutions in the spectral-time method: 1) lower limit of the resolution for a spectrum 20 cm^{-1} wide; 2) lower limit of the resolution for a spectrum 2 cm^{-1} wide.

Table I.

	Neodymium glass laser			Ruby laser	
Width of spectrum, cm^{-1}	20	2	0,2	0.8	0.12
Frequency of occurrence of "smooth" spectra, %	< 5	40	70	< 3	50

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