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# Examination of thermal stability and laser characteristics of LiF: [$F_2^-$] modulators*

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In this paper, the results of examination of both thermal and photochemical stability of colour centres in the LiF monocrystals used to passive modulation of the YAG: $Nd^{3+}$ laser resonator Q-factor (Q-switch) are reported. The interaction kinetics of the ionized colour centres of M- and R-types has been examined. The activation energy for particular structural transitions, the limiting temperatures of the $F_2^-$ centres stability as well as the basic spectroscopic and laser parameters for LiF: [$F_2^-$] modulators have been determined. The experimental results of the measurement of the thermal, strength and generation characteristics of the YAG: $Nd^{3+}$ single pulse laser with passive LiF: [$F_2^-$] modulator have been compared to the results of the theoretical calculations.

## 1. Introduction

The works on different ways of Q-factor modulation in laser resonators have been stimulated by the need of generation of the high power laser pulses of short duration.

The effect of the so-called nonlinear absorption, i.e., the effect of transmission enhancement in some materials interacting resonantly with the high intensity radiation is exploited in the passive method. The passive modulators require neither external control nor power supply and are characterized by a much simpler design compared to the active modulators.

The parameters of radiation emitted by the laser with a passive Q-factor modulator depend essentially on the spectroscopic characteristics and also on the optical and thermal stability of active centres [1]–[3].

The organic dye solutions applied commonly in the form of translucent filters suffer from many shortcomings of which the low thermal and photochemical resistivities are the most important [4], [5]. For example, the foil modulators, produced on the base of thiolene BDN dye suspended in the matrix of methyl polimethacrylate and employed to passive Q-factor modulation (Q-switch) in YAG: $Nd^{3+}$ lasers, are characterized by the following strength parameters [6], [7]:

– working temperature $< 55°C$,

– generation repetition rate $< 0.3$ Hz,

– energy resistivity of the modulator to the single pulses of laser radiation $< 100$ MW/cm$^2$.

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The single crystals of lithium fluoride with the colour centres $F_2^-$ indicating the effect of nonlinear dependence of absorption on the radiation power density (of wavelength $\lambda = 1064$ nm) are applied to both the passive Q-switching in YAG: $Nd^{3+}$ laser resonators [8]–[10] and to passive mode synchronization [11] since the year 1980.

The examinations of those crystal have shown that the basic parameter determining the durability of a single pulse laser is the thermal stability of the $F_2^-$ colour centres which in turn depends on the monocrystallization technology, the way of colour centres generation, the LiF crystal structure and the dopings [12]–[14].

The purpose of these investigations was to recognize the influence of the colour centres stability in the LiF single crystal on the basic generation and strength characteristics of the single pulse YAG: $Nd^{3+}$ laser.

## 2. Experimental part

The $6 \times 35$ mm samples of LiF cut out from the single crystal along the [100] direction were subject to investigation. The irradiation by radiation in the field of isotopics Cf–252, Cs–137 and Co–60 was performed in 293 K for the exposure doses of $4 \times 10^5 = 10^8$ R. The power of the exposure dose, for the particular series of crystals, was measured directly with the help of a A-143 diode equipped with a indicator of STUDSVIK 3809 A type. Transmission spectra of the samples were examined on the Beckman Acta MVII spectrophotometer equipped with a temperature attachment. The temperature stability of the dye centres $F_2^-$, $F_3^-$ and $F_2$ was examined within the 213–473 K temperature range by using isothermal heating [2], [12] and differential thermal analysis (DTA) methods.

The final losses and the basic spectroscopic parameters of an absorber have been determined by the method reported in [15], [16]. The source of diagnostic radiation was a single pulse YAG: $Nd^{3+}$ laser of the following parameters:

– YAG: $Nd^{3+}$ rod, $\varnothing = 4 \times 55$ mm, $\varrho = 0.01$ cm$^{-1}$,
– passive dye modulator (BDN), $T_0 = 20\%$,
– exit mirror $R = 15\%$,
– output energy $E_{out} = 35 \pm 1$ mJ,
– single pulse duration 5 ns,
– angular divergence of radiation 5 mrad.

The influence of initial transmission of the modulator LiF: $[F_2^-]$ and of initial amplification in the resonator on the basic generation characteristics of the single pulse YAG: $Nd^{3+}$ laser was examined. The results of the examinations have been compared with those obtained from numerical calculations of the energy parameters of the investigated laser, which were carried out for the model of immediate Q-switching [17], the final losses of the multiple reflections of the radiation in the resonator being taken into account [18].

## 3. Discussion of the results of examinations

As it follows from the examinations of the effectiveness of the colour centre generation in the single LIF crystal samples the most advantageous irradiation parameters and a simultaneous repeatability of the exposure conditions are offered by Co-60.

The results of examinations of the temperature dependence of the transmission in the maximum of the absorption band of the examined centres, for a constant sample heating time (5 h) presented in Fig. 1, indicate that the $F_2$ centres ($\lambda_{max} = 450$ nm) are

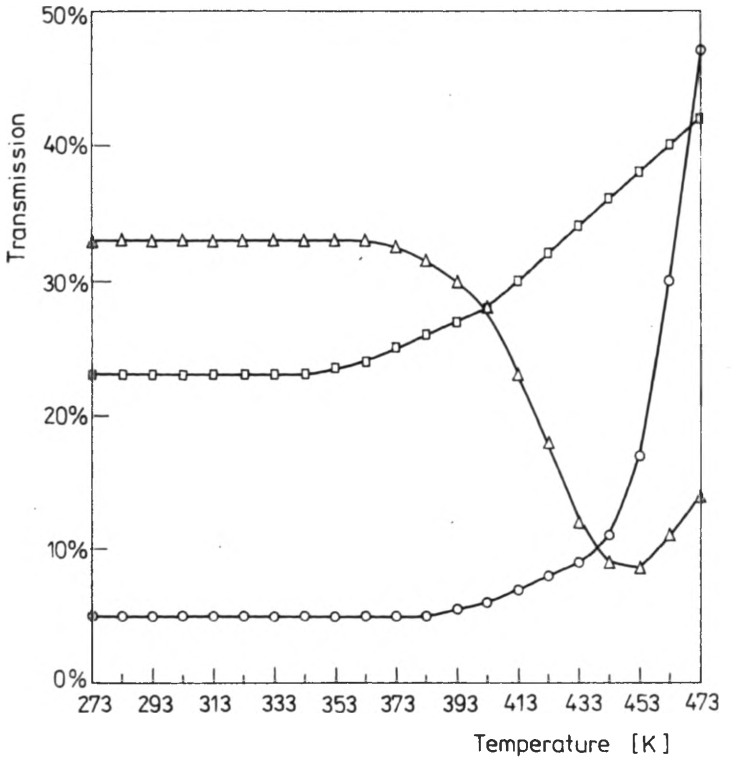


Fig. 1. Thermal stability examination for the colour centres in LiF: △ – $F_3^-$ centre ($\lambda_{max} = 800$ nm), □ – $F_2^-$ centre ($\lambda_{max} = 960$ nm), ○ – $F_2$ centre ($\lambda_{max} = 450$ nm)

stable up to 403 K; above this temperature there occurs their recombination. On the other hand, the centres $F_2^-$ ($\lambda_{max} = 960$ nm) and $F_3^-$ ($\lambda_{max} = 800$ nm) are stable up to the temperature of 353 K above which one observes the concentration decay of the $F_2^-$ centres as well as an increase of the $F_3^-$ centres (which recombine above 443 K).

These results have been confirmed by other examinations carried out by the method of the differential thermal analysis, which indicate that during heating of LiF with colour centres there occurs an exothermic process the maximum of which occurs at the temperature of about 403 K.

The time-dependent changes of transmission spectra of the LiF samples with the colour centres at the temperature of 423 K are presented in Fig. 2. The observed points of constant absorption coefficient for the wavelength $\lambda = 880$ nm and 1040 nm prove the existence of the dynamic balance connected with the structural

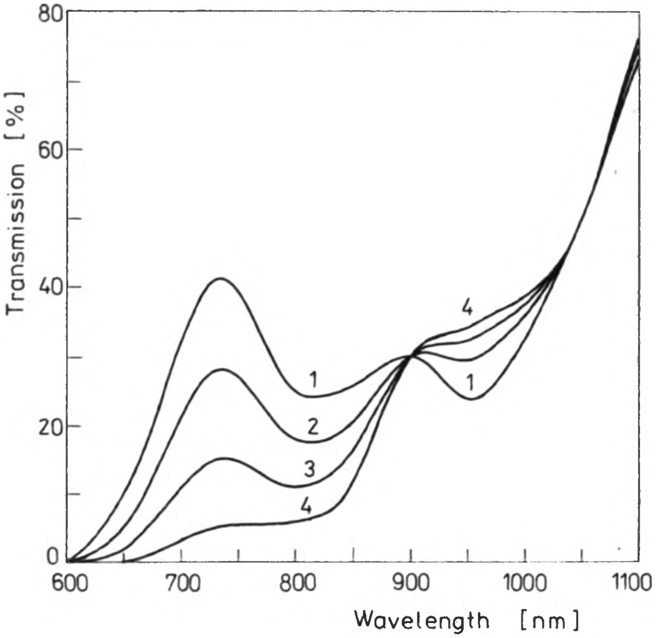


Fig. 2. Transmission spectrum of LiF (Ø6 × 35 mm) with the colour centres vs the time at 423 K: 1 – $t = 0$ h, 2 – $t = 10$ h, 3 – $t = 10$ h, 4 – $t = 30$ h

transitions: $F_2^- \rightarrow F_3^-$ ($\lambda = 880$ nm) and $F_2^- \rightarrow F_3^- \rightarrow F_4^-$ ($\lambda = 1040$ nm). Similar transitions are characteristic of the nonionized colour centres occuring in other alkali metal halogens [19]–[21].

It has been stated that the reactions of the $F_2^-$ centre decay is described by the kinetic equation of the first order. Constant reaction rates of $F_2^-$ centres decay have been calculated for the following temperatures: 373, 423 and 473 K. Taking advantage of the Arrhenius equation

$$k(T) = A_0 \exp(-E_a/RT),$$

the activation energy $E_a = 76.7$ kJ/mol as well as the factor $A_0 = 5.84 \times 10^6 \mathrm{h}^{-1}$ have been determined for $F_2^-$ centres. For the examined LiF samples the half-decay time the 293 K estimated from these results amounts to ca 600 years.

In Figure 3, the dependence between the final transmission $T_k$ of the absorber

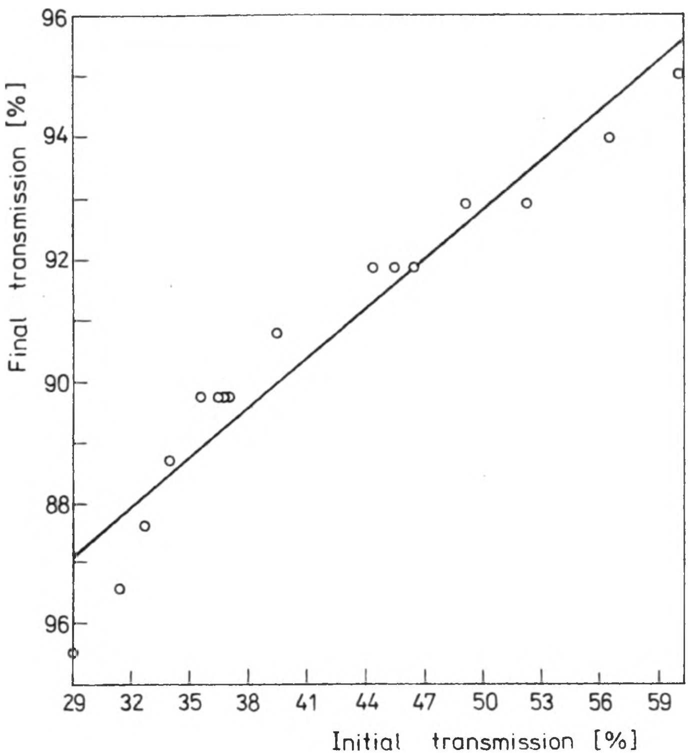


Fig. 3. Final transmission vs initial transmission of the LiF: $[F_2^-]$ modulator

and the initial transmission $T_0$ is shown for the wavelength $\lambda = 1064$ nm. The experimental results have been compared with the theoretical relation $T_k = AT_0+B$ ($A = 0.27002$, $B = 0.79266$) using rms approximation method. The results obtained allowed us to analyse the thermal field distribution in a passive crystal Q-factor modulator of the YAG: $Nd^{3+}$ laser resonator after single pulse generation.

The results of calculations of the temperature increment of the LiF: $F_2^-$ modulator due to the emittance of heat in the absorber after single pulse generation are shown in Fig. 4. In the assumed model of laser with immediate switch of losses, the output energy of single pulse is described by the dependence

$$E_{in} = E_{gen} - E_{\varrho} - E_A$$

where:

$E_{gen}$ – energy generated in the active material,

$E_{\varrho}$ – energy absorbed inside the active material,

$$E_{\varrho} = E_{gen}\frac{\varrho}{k_s^k},$$

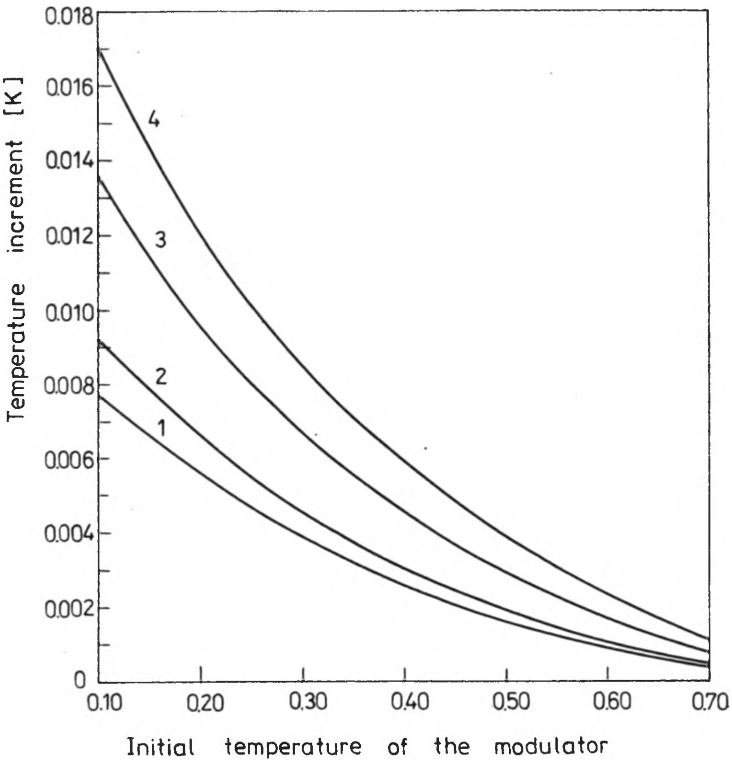


Fig. 4. Temperature increment in the LiF: $[F_2^-]$ modulator after single pulse generation by the laser (YAG: $[Nd^{3+}]$ rod of ∅4 × 55 mm, $\varrho = 0.01\ \mathrm{cm^{-1}}$): 1 – $R_2 = 0.1$, 2 – $R_2 = 0.2$, 3 – $R_2 = 0.5$, 4 – $R_2 = 0.7$

$\varrho$ – loss coefficient of the active material,
$k_s^k$ – loss coefficient of the final resonator,
$E_A$ – energy absorbed in nonlinear absorber,

$$E_A = E_{gen}\frac{k_r^*(T_k)}{k_s^k}\frac{1-R_2^*-T^*}{1-R_2^*},$$

$k_r^*(T_i)$ – effective loss coefficient in which the interference effects, multiple reflection of the radiation and the absorber transmission [9] are taken into account,

$$k_r^*(T_i) = \frac{1}{2l}\ln\frac{1}{R_1^* R_2^*(T_i)},$$

$$k_s^k = \varrho + k_r^*(T_k),$$

$R_2^*$, $T^*$ – effective reflection coefficient of the output mirror and the effective transmission of the absorber [9], respectively,
$l$ – absorber length.

Taking into account the system geometry (Fig. 5) and the thermal parameters of

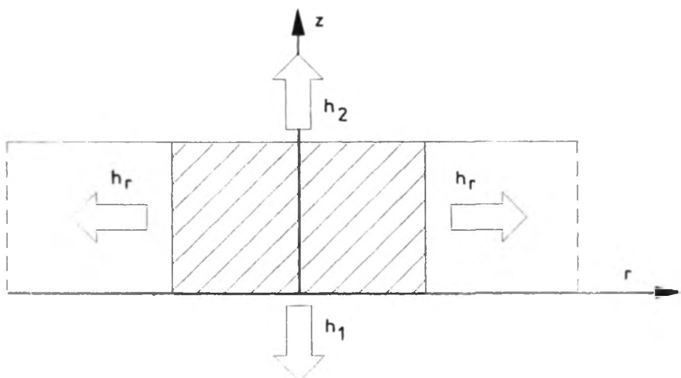


Fig. 5. Coordinate system used to analyse of the temperature field in the modulator (for explanation, see text)

the modulator we have accepted the two-dimensional model of the temperature field with the boundary conditions of the third kind. The temperature in the region of heat emission, after each pulse generation being repeated every $\vartheta$, is described with the recurrence formula

$$T_i(r, z, t) = T_0 + \sum_{n=1} \sum_{m=1} A_{mn}\left(\cos\frac{\delta_m z}{r_0} + \frac{h_1 r_0}{\delta_m}\sin\frac{\delta_m z}{r_0}\right) J_0\left(\sigma_n \frac{r}{r_0}\right) \exp\left(-\frac{a^2}{r_0^2}\gamma_{nm}^2 t\right)$$

where: $\sigma_n$, $\delta_m$, $\gamma_{nm}$ and $A_{nm}$ are determined from the dependences:

$$\frac{\sigma_n}{r_0} J_1(\sigma_n) = h_r J_0(\sigma_n),$$

$$\mathrm{ctan} = \frac{\delta_m z_0}{r_0} = \frac{1}{h_1 + h_2}\left(\frac{\delta_m}{r_0} - \frac{h_1 h_2 r_0}{\delta_m}\right),$$

$$\gamma_{nm}^2 = \delta_m^2 + \sigma_n^2,$$

$$A_{nm} = \frac{4 T_p^* r_0^2 \left[\frac{\delta_m}{r_0}\sin\frac{\delta_m z_0}{r_0} + h_1\left(1 - \cos\frac{\delta_m z_0}{r_0}\right)\right]}{\sigma_n \left[J_1(\sigma_n)\left(1 + \frac{\sigma_n^2}{r_0^2 h_r^2}\right)\right](r_0^2 h_1^2 + \delta_m^2)} \left[z_0 + \frac{\left(\frac{\delta_m^2}{r_0^2} + h_1 h_2\right)(h_1 + h_2)}{\left(\frac{\delta_m^2}{r_0^2} + h_1^2\right)\left(\frac{\delta_m^2}{r_0^2} + h_2^2\right)}\right]^{-1}$$

$T_0$ – temperature of the environment, 293 K,
$T_p$ – temperature increment after a single pulse generation,

$$T_p = \frac{E_A}{\Pi r_0^2 z_0 c \varrho},$$

$$T_p^* = T_p + T_{i-1}(\vartheta),$$

$z_0$, $2r_0$ – length and diameter of the heat emission region,
$h_1$, $h_2$, $h_r$ – coefficients of heat acceptance in the direction of $-z$, $z$ and $r$ axes, respectively,

$J_0(x)$, $J_1(x)$ – Bessel functions of the first kind and of zero and first order, respectively,

$$a = \frac{c\varrho_c}{\lambda},$$

$c$ – specific heat of the modulator (1.56 J/gK),
$\varrho_c$ – specific weight of the modulator (2.6 g/cm$^3$),
$\lambda$ – heat conductivity of the modulator (9.142 W/cmK).

The temperature distribution in the heat emission region was studied depending on the resonator parameters (initial transmission of the absorber, reflection coefficient of the exit mirror), the generation repetition time and the conditions of heat removal. From the calculations carried out and presented in Fig. 6, as well as

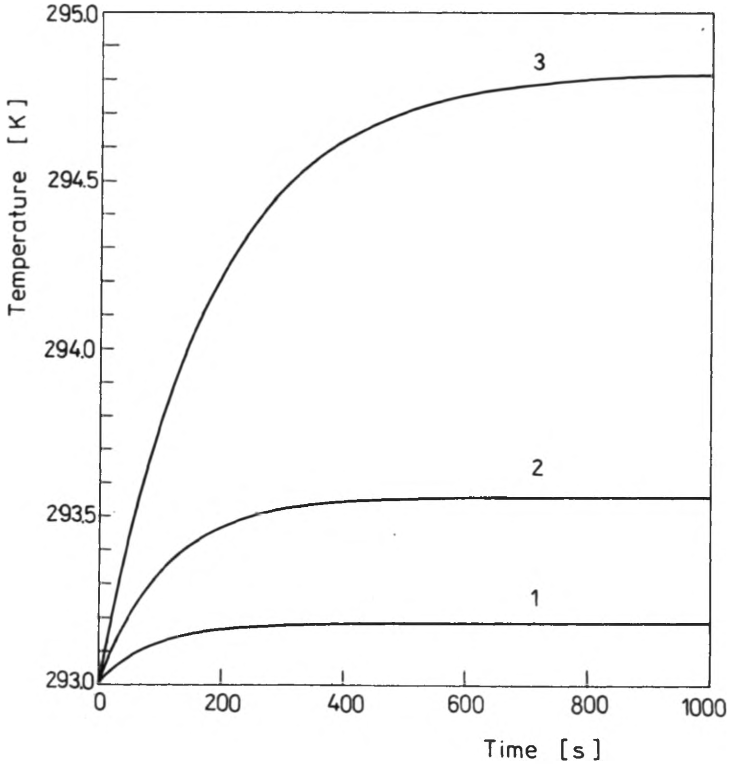


Fig. 6. Maximal temperature dependence of the LiF: $[F_2^-]$ modulator during the single pulse generation of different repetition frequency (YAG: $Nd^{3+}$ rod of $\varnothing 4 \times 55$ mm, $\varrho = 0.01$ cm$^{-1}$): 1 – 0.2 Hz, 2 – 0.5 Hz, 3 – 1 Hz

from the thermal stability examinations it is possible to estimate the limiting working conditions of the YAG: $Nd^{3+}$ laser with a passive LiF: $[F_2^-]$ modulator. If we assume that the maximal working temperature of the LiF: $[F_2^-]$ modulators amounts to 343 K, the typical values of the heat acceptance coefficient being

$h_1 = h_2 = 0.017$ cm$^{-1}$, $h_r = 0.034$ cm$^{-1}$, then the limiting repetition frequency for the surrounding temperature up to 343 K is equal to 2 Hz. On the other hand, the LiF: [$F_2^-$] modulator is resistive to a series of single pulses as high as 7000 for the 100 Hz repetition frequency at 293 K surrounding temperature.

The spectroscopic thermal and laser examinations allowed us to explain the kinetics of the colour centres interaction in the LiF single crystals. They also showed a high stability of the optical parameters of LiF with the colour centres for the radiation of the wavelength $\lambda = 1064$ nm.

In comparison to the passive foil dye modulators used nowadays the crystal modulators assure more stable work of the single pulse laser, higher thermal photochemical resistivities and also offer a higher frequency of pulse generation.

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## Исследование стабильности и лазерных характеристик модуляторов LiF: [$F_2^-$]

В работе представлены результаты исследования термической и фотохимической стабильностей центров окраски в монокристаллах фтористого лития, использованных в качестве пассивных модуляторов добротности резонатора лазера YAG:$Nd^{3+}$. Измерено изменение пропускания пассивного лазерного затвора на LiF: [$F_2^-$] в зависимости от температуры. Методом изотермического отжига радиационно окрашенных кристаллов в течение различного времени исследована кинетика реакции между центрами окраски типа M и R и ее влияние на основные генерационные характеристики моноимпульсного лазера YAG: $Nd^{3+}$. Экспериментальные результаты сравнены с теоретическими расчетными оценками изменения температуры модулятора во время генерации моноимпульсов.