

Bleaching of the F_2^- color centers in an LiF crystal due to two-photon absorption from the excited state.

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The bleaching of F_2^- color centers in an LiF crystal exposed to intense IR radiation with wavelength $1.06 \mu\text{m}$ was studied. It is shown that the bleaching of the color centers is due to two-photon absorption from the excited state, and is accompanied by formation of a pronounced dichroic absorption band. It was observed that the direction of minimum absorption coefficient did not coincide with the direction of polarization of the IR radiation. The two-photon cross section for absorption from the excited state of the color centers was calculated from independent experimental data on the absorption dichroism and on the dynamics of the decay process.

INTRODUCTION

Lithium fluoride crystals with F_2^- color centers ($\text{LiF}(F_2^-)$) are currently used as passive Q switches (PQS) of neodymium lasers¹⁻³ and as the active medium in tunable lasers.^{4,5} In both cases, these crystals are exposed to intense infrared radiation. It is assumed that the stability of the F_2^- color centers to such a radiation is rather high.⁶ An early (1967) publication, however, indicates that the M' color centers (which are currently named F_2^- centers) in LiF are bleached by infrared radiation. The recent publication of Asayenok *et al.*⁸ confirms the results of that paper.

In our studies of the long term stability of $\text{LiF}(F_2^-)$ -based PQS neodymium lasers we have observed that the initial PQS transmittance increases irreversibly in time in the crystal region exposed to the laser radiation. This phenomenon was attributed by us to the bleaching of the color centers. In terms of the practical applications of the LiF-based PQS, this instability of the color centers restricts the radiation intensity in the cavity. On the other hand, studies of color-center bleaching by exposure to intense infrared radiation are of independent interest, since they permit a better understanding of the nature of the color centers. This is the motivation for our studies of the features and bleaching mechanism of the F_2^- color centers in LiF.

The objectives of the present study of the photo-endurance of $\text{LiF}(F_2^-)$ in the infrared are:

—to elucidate the dependence of the variation of the weak-signal absorption coefficient (the initial PQS transmittance) on the energy density of a strong signal that bleaches the color centers;

—to elucidate of the dependence of this variation on the number of laser pulses incident on the PQS (the irradiation dose);

—to study the effect of the intense radiation on the transmittance of $\text{LiF}(F_2^-)$ PQS in the open state;

—to study the polarization properties due to irradiation of the region with lower concentration of color centers;

—to study the stability of $\text{LiF}(F_2^-)$ crystals after bleaching of the color centers.

Obviously, these objectives have both scientific and practical importance. Interesting is also the effect of the preparation method on the photostability of the crystals containing color centers. In our studies, we used $\text{LiF}(F_2^-)$

crystals prepared by two different groups (at the Irkutsk State University and at the Institute of General Physics of the USSR Academy of Sciences).

The main results of our work are the following. The bleaching of the F_2^- color centers in LiF has a two-photon character, and the two photons are absorbed from the excited state. In the decay process in PQS, a region of lower concentration of the color centers of given type is formed and has a strong absorption anisotropy (dichroism) (The occurrence of dichroism at the moment of action of a strong laser pulse on PQS is known⁹). The polarization properties of the induced absorption dichroism depend on the direction of polarization of the intense radiation with respect to the crystallographic axes of LiF. The low color center concentration zone formed has a prolonged stability. Differences, if any, in the preparation methods of various groups practically do not affect the photostability of the $\text{LiF}(F_2^-)$ crystals.

EXPERIMENTAL SETUP

It is obvious that, in order to obtain reliable data on the photostability of $\text{LiF}(F_2^-)$ crystals, good experimental conditions should be assured. The data obtained in the measurements inside the resonators have shown that the properties of the laser radiation are not sufficiently reliable means of determining the change of properties of PQS. It is also difficult to assure a wide range of variation of the conditions of color center bleaching in PQS, as well as the probing of the irradiated region. We, therefore, decided to perform out-of-cavity experiments.

In order to study the photostability of $\text{LiF}(F_2^-)$ crystals, we used the radiation of a pulsed-periodic neodymium phosphate glass laser (repetition frequency 2 kHz). The output energy density distribution was close to the TEM_{00} mode. In the LiF crystal exposed to this radiation, a zone with increased transmittance was created. The energy density variation of the decay of the color centers was determined from the measured distribution of the absorption coefficient in this zone, compared to the energy density distribution of the intense radiation, led to unambiguous conclusions concerning the dependence of the color-center bleaching on the energy density.

The experimental setup is shown in Fig. 1. The laser was operated in two different regimes: with Q -switching by using

a DKDP electrooptic shutter, and in the probing mode, without Q -switching (with the shutter disconnected). This assured that the intense and the measuring beams had the same direction, which was important since the crystals were rather long (3–4 cm). In the probing regime, absorbing filters were placed in front of the LiF crystal, so that the intensity density ($\sim 1 \text{ kW/cm}^2$) was not sufficient to bleach the color centers.

The investigated LiF crystal and the diaphragm D were placed on micrometric stages moving perpendicular to the direction of the laser beam. The lens L, with magnification $2 \times (\pm 0.01)$, formed the image of the exit end surface of the crystal on the diaphragm (the magnification was measured behind an IKS-6 filter, using an electrooptic converter). The diaphragm measured $0.15 \times 0.35 \text{ mm}$ (the smaller side in the direction of orientation of the stages).

The measurements were performed by two photodiodes, PD1 and PD2, the first of which was used as the reference. The photodiodes used were of PD-24K type, with an integrating RC circuit (with time constant 6 ms) as the load. The output signal was fed into a digital voltmeter of ATsP-14-type in the CAMAC reference. The experiment was run, and the data were collected and processed, by an "Agat" PEVM computer connected on-line with the CAMAC through a monitor.¹⁰

The measurements were performed as follows:

1. Determination of the energy distribution in the intense beam. The crystal was removed from the laser beam, the diaphragm was displaced (in steps of 0.1 mm), and the signal ratio was measured with the photodiodes. The measurements were averaged over 10 pulses. The distribution was measured before and after exposure of the crystal.

2. Exposure of the crystal to the intense beam. This operation was typically performed in 10 series of 10^4 pulses each. The polarization of the laser radiation was practically linear (the intensity ratio of the polarization-ellipse axes was 1:40).

3. Measurement of the distribution of the absorption coefficient of the crystal after each series of exposures was performed with the crystal displaced in steps of 0.05 mm perpendicular to the axis of the laser beam (and the diaphragm fixed). The laser was operated in the free-running regime. The polarization was linear (the same as in the case of Q -switching). The ratio of the signals from the photodiodes was measured as in Item 1 above.

4. Measurement of the dichroism. A $\lambda/4$ plate was placed at the output of the laser (which was operated in the free-running regime), as well as a Glan prism whose rotation set the direction of polarization of the probing radiation. The measurements were performed as shown above (Item 3), for

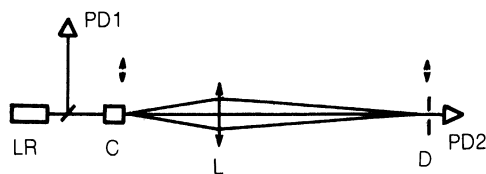


FIG. 1. Schematic diagram of the experimental setup: LR—laser; C—crystal; L—lens; D—diaphragm; PD1 and PD2—photodiodes. The arrows indicate the direction of displacement of the LiF crystal and the diaphragm.

each direction of polarization (in steps of 10° on the axis of the exposed region of the crystal).

5. The experimental setup also allowed the measurement of the effect of crystal exposure to the intense radiation on the transmittance of $\text{LiF}(\text{F}_2^-)$ in the open state (i.e., on the magnitude of the residual losses). In this case, the laser was operated in the Q -switching regime.

EXPERIMENTAL RESULTS

The $\text{LiF}(\text{F}_2^-)$ crystals used had a length 30 mm (our Institute) and 40 mm (Irkutsk) and an initial transmittance of 16% and 50%, respectively. Their open state transmittance before exposure was 77.7% and 95%. For the first sample, it was found that the intense radiation (10^5 pulses, energy 28 mJ, length 18 ns, maximum intensity density 150 MW/cm^2) had no effect on the residual losses.

The experimental data shown below refer to the first of the two types of samples. Here, k is only the absorption-coefficient bleaching component obtained by subtracting the residual losses from the measured absorption coefficient.

Figure 2 shows the energy-density distribution (in relative units) of the intense laser beam in the plane of the output end face of the LiF crystal as a function of the transverse coordinate. This energy distribution approximated by a function of the form:

$$I = I_0 \exp(-2x^2/w^2) \quad (1)$$

by using the least squares method, with $x < 0.3 \text{ mm}$, yields $2w = 0.99 \text{ mm}$ and $\sigma_w = 0.01 \text{ mm}$, where σ_w is the variance of the quantity $2w$.

Figure 3 shows an example of the results of the action of intense radiation having the distribution shown in Fig. 2 and the characteristic parameters given above (number of pulses $n = 2 \times 10^4$) on the F_2^- color centers in LiF. Since the effect on the LiF crystal is manifested in the reduced density of the color centers, Fig. 3 shows the measured variation of the absorption coefficient, $\Delta k(x)$. It was approximated by the function given in Eq. (1), with $2w_{\Delta k} = 0.76 \text{ mm}$, and $\sigma_w = 0.03 \text{ mm}$ (0.69 and 0.06 mm for $n = 1 \times 10^4$).

Figure 4 shows the variation of the absorption coefficient

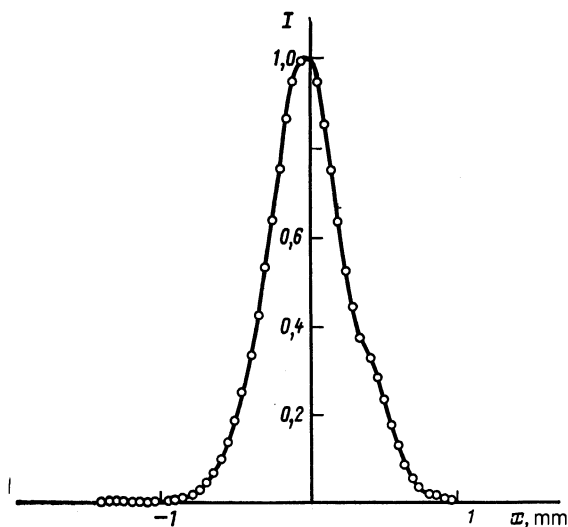


FIG. 2. Energy density distribution of the intense radiation as a function of the transverse coordinate.

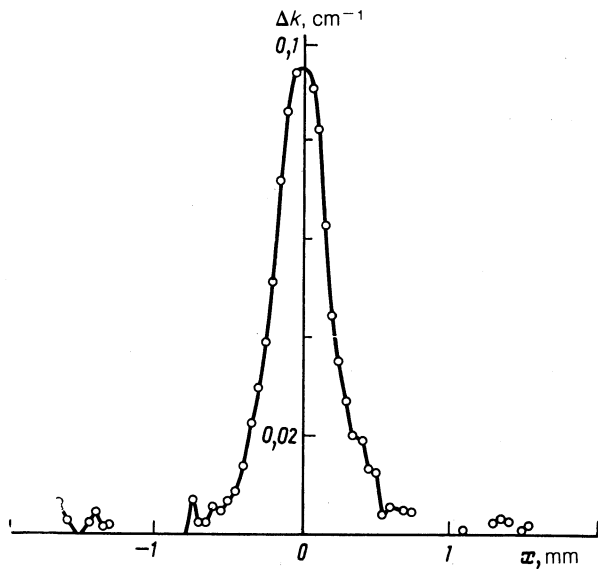


FIG. 3. Absorption coefficient of the $\text{LiF}(\text{F}_2^-)$ crystal, as a function of the transverse coordinate, after interacting with the intense radiation pulses.

cient with the irradiation dose for the axial zone of the intense radiation, where the energy density is maximum. Assuming that the bleaching of the color centers is purely exponential, the experimental data can be approximated by a curve of the form:

$$k(n) = A[1 - \exp(Cn)], \quad (2)$$

with $A = 0.4 \text{ cm}^{-1}$ and $C = -1.3 \times 10^{-5}$. It should be mentioned that, in this series of measurements, the transmitted energy decreased after irradiation from 28 to 23 mJ.

Figure 5 shows the absorption coefficient of $\text{LiF}(\text{F}_2^-)$ versus the transverse coordinate, for two orientations of the linearly polarized probing radiation: parallel to the direction of polarization of the intense beam (curve 1), and perpendicular to it (curve 2). The data were obtained for $n = 10^5$ laser pulses (28 mJ, 18 ns, 250 MW/cm^2). Figure 5 shows that, for an initial absorption coefficient $k_0 = 0.51 \text{ cm}^{-1}$, the parallel and perpendicular components of k have the values $k_{\parallel} = 0.15$ and $k_{\perp} = 0.28 \text{ cm}^{-1}$, which indicates a rather significant absorption dichroism.

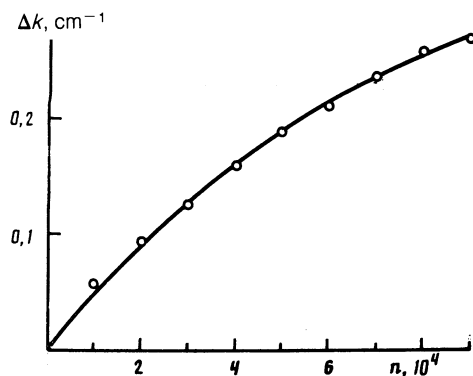


FIG. 4. The absorption coefficient as a function of the number of pulses of intense radiation.

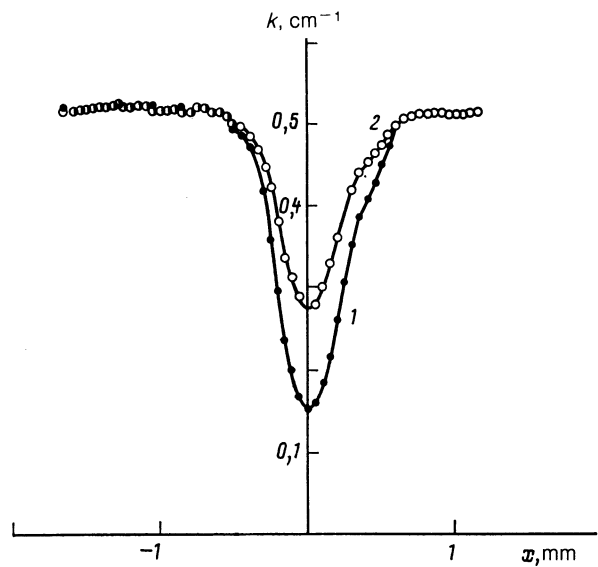


FIG. 5. Variation k_{\parallel} (1) and k_{\perp} (2) for the $\text{LiF}(\text{F}_2^-)$ crystal as a function of the x coordinate, after interaction with 10^5 pulses of intense radiation.

Analysis of these data shows that the direction of polarization of the probing radiation, for which the k value in the LiF crystal is minimum, may not coincide with the direction of polarization of the intense radiation. The transmission coefficient of the PQS was, therefore, measured as a function of the direction of polarization of the probing radiation (Fig. 6, curve 1), and the exact direction of polarization of the intense beam was determined using a Glan prism (Fig. 6, curve 2). The angle of deflection from the direction of polarization of the intense beam, calculated from the data in Fig. 6, give at an absorption coefficient k_{\min} the value $\Delta\theta = -7^\circ$ (the intrinsic birefringence of the crystal gives a contribution that does not exceed 0.1°).

Measurements of the dichroism induced by the intense radiation were repeated three months after the experiment. No differences in the curves shown in Fig. 5 were observed (within the experimental errors). This indicates that no deformation of the zone with lower concentration of color centers, or variation of its characteristics, were observed in

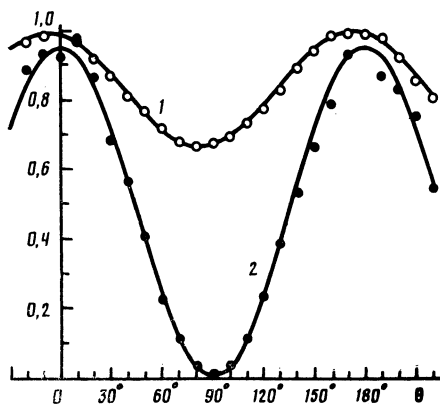


FIG. 6. Relative variation of the transmission coefficient of an $\text{LiF}(\text{F}_2^-)$ crystal ($n = 10^5$ pulses) as a function of the direction of polarization of the probing radiation (curve 1), and intensity variation of the intense radiation as a function of the rotation angle of the Glan prism (curve 2).

an $\text{LiF}(\text{F}_2^-)$ crystal kept for a prolonged time at room temperature.

DISCUSSION

It is known that the F_2^- color centers in LiF are bleached by either the third (0.35μ) or the second (0.53μ) harmonic of the neodymium laser radiation. In the first case, this process is due to the direct photoionization of the color centers. In the second case, the bleaching occurs in steps consisting of formation of the F_2^+ color centers from the neutral F_2 color centers, excitation of these F_2^+ centers, and nonradiative transfer of energy to the F_2^- centers, after which a second photon (0.53μ) bleaches the color center with transfer of an electron into the conduction band.

Under simultaneous exposure to the IR radiation (0.9μ) and to the second harmonic of the neodymium laser, the bleaching of the F_2^- color center proceeds in two steps: excitation of the color center by the IR radiation, followed by absorption from this state of a second-harmonic photon and bleaching of the color center.¹²

If only the IR radiation acts upon the F_2^- color center, it is correct to assume that the decay process proceeds in a similar way, i.e., the color center absorbs one photon of IR radiation, is excited, and is then bleached by absorbing two more photons from the excited state. In other words, in the second stage we have a two-photon process (In this case, the effect is expected to vary as the square of the intensity of the IR radiation, and to have a special dependence on the polarization¹³). In the following description of the bleaching of the F_2^- color centers by the intense radiation, it will be assumed that all the color centers are in an excited state, and that the two-photon absorption proceeds from this excited state. This assumption can be supported by the following facts: 1) the intense laser pulse (15–20 ns) is much shorter than the lifetime of the excited state of the F_2^- color centers ($\sim 100 \text{ ns}$ ^{11,14}), i.e., its decay during the action of the intense pulse can be neglected; 2) the energy density at the maximum of the laser pulse, $E_0 = 4 \text{ J/cm}^2$, is significantly larger than the saturation energy of the color center, $E_s = h\nu\sigma = 10^{-2} \text{ J/cm}^2$, and the transition cross section for $\lambda = 1.06 \mu$ is equal to $\sigma = 2 \times 10^{-17} \text{ cm}^2$ (Ref. 14). Therefore, for such a large E_0/E_s ratio (~ 400), neglect of the polarization dependence of the excitation of the color centers seems to be justified.

The F_2^- color centers can occupy six equivalent positions in the LiF crystal lattice. Following Ref. 9, the coordinate axes were chosen along the directions [100], [010], and [001], and the unit vectors along these directions were \mathbf{e}_i ($i = 1, 2, 3$). We assume that the intense and the probing waves propagate along the \mathbf{e}_1 direction, and that their polarizations (the directions of the electrical field) is given by the vectors \mathbf{e} and \mathbf{e}_0 , respectively, such that $(\mathbf{e}_3 \cdot \mathbf{e}) = \cos \theta$, and $(\mathbf{e}_3 \cdot \mathbf{e}_0) = \cos \varphi$.

Let us introduce the unit vectors \mathbf{a}_i ($i = 1, 2, \dots, 6$) in the direction of the fluorine vacancies in the F_2^- color center:

$$\begin{aligned} \mathbf{a}_1 &= (\mathbf{e}_1 + \mathbf{e}_2)/2^{1/2}, & \mathbf{a}_2 &= (\mathbf{e}_1 - \mathbf{e}_2)/2^{1/2}, \\ \mathbf{a}_3 &= (\mathbf{e}_2 + \mathbf{e}_3)/2^{1/2}, & \mathbf{a}_4 &= (\mathbf{e}_2 - \mathbf{e}_3)/2^{1/2}, \\ \mathbf{a}_5 &= (\mathbf{e}_3 + \mathbf{e}_1)/2^{1/2}, & \mathbf{a}_6 &= (\mathbf{e}_3 - \mathbf{e}_1)/2^{1/2}. \end{aligned} \quad (3)$$

Let us assume that the cross section for absorption from

the ground state of the color center, for a wavelength of 1.06μ , depends on the relative orientation of \mathbf{e}_0 and \mathbf{a}_i , as $\sigma_i = \sigma_0 (\mathbf{a}_i \cdot \mathbf{e}_0)$, where σ_0 is the maximum value of the absorption cross section. The absorption coefficient of the probing wave is then obtained as follows:

$$k = \sigma_0 \sum_i N_i (\mathbf{a}_i \cdot \mathbf{e}_0)^2, \quad (4)$$

where N_i is the concentration of each of the six "types" of F_2^- color center. If it is assumed that, prior to exposure of the $\text{LiF}(\text{F}_2^-)$ to the intense radiation, the concentration of the color centers is N_0 , then $N_i = N_0/6$, which gives for the initial absorption coefficient:

$$k_0 = \sigma_0 N_0/3. \quad (5)$$

It is assumed that exposure to one pulse of intense radiation changes insignificantly the density of the color centers. In this case, the relationship between the initial (N_{i0}) and final ($N_i(n)$, after interaction with n pulses) densities is given by the formula:

$$N_i(n) = N_{i0} \exp \left[-n \gamma_i \int_{-\infty}^{\infty} I^2(t) dt \right], \quad (6)$$

where γ_i is a coefficient connected with the two-photon absorption, and $I(t)$ is the intensity of the intense laser pulse. From Eqs. (4) and (6), the absorption coefficient of the $\text{LiF}(\text{F}_2^-)$ crystal, after transmission of n laser pulses, is:

$$k(n) = \sigma_0 \sum_i N_{i0} (\mathbf{a}_i \cdot \mathbf{e}_0)^2 \exp \left[-n \gamma_i \int_{-\infty}^{\infty} I^2(t) dt \right]. \quad (7)$$

The coefficient γ_i in Eq. 6 is determined by the polarization dependence of the two-photon absorption. In the simplest case of a three-level scheme of the F_2^- color center, where the ground level is level 1, the first excited level is level 2 ($\lambda_{12} = 0.96 \mu$), and the highest level is level 3 ($\lambda_{13} = 0.35 \mu$), the two-photon absorption cross section for the 2–3 transition in the second order of the perturbation theory (see, e.g., Refs. 13 and 15) is given by $\sigma_{2ph} \sim |d_{12} e|^2 |d_{13} e|^2$, where d_{ij} are the matrix elements of the i – j transitions. For the 1–2 transition, it was assumed that $|d_{12} e_0|^2 \sim (\mathbf{a}_i \cdot \mathbf{e}_0)^2$. For the 1–3 transition we assume two different versions: $d_{13} = 1$ (the isotropic case), and $|d_{13}|^2 = |d_{12}|^2$. Then:

$$\gamma_i = \sigma_{20} (\mathbf{a}_i \cdot \mathbf{e})^2 S^2/2, \quad (8)$$

where the parameter S is equal to unity or $(\mathbf{a}_i \cdot \mathbf{e})$, and the coefficient $1/2$ accounts for bleaching of one color center upon absorption of two photons.

Calculating (7) with γ_i given by Eq. (8), the following expression is obtained:

$$k(n) = (k_0/2) [b_1 \sin^2 \varphi + b_2 \cos^2 \varphi + b_3 \sin^2(\varphi + \pi/4) + b_4 \cos^2(\varphi + \pi/4)], \quad (9)$$

where

$$\begin{aligned} b_1 &= \exp [(-Y/\kappa) \sin^* \theta], \\ b_2 &= \exp [(-Y/\kappa) \cos^* \theta], \\ b_3 &= \exp [-Y \sin^* (\theta + \pi/4)], \\ b_4 &= \exp [-Y \cos^* (\theta + \pi/4)], \end{aligned} \quad (10)$$

$$Y = (n\sigma_{20}/2) \int_{-\infty}^{\infty} I^2(t) dt,$$

$\kappa = 2$ for $S^2 = 1$, and $\kappa = 4$ for $S^2 = (\mathbf{a}_i \cdot \mathbf{e})^2$.

Equations (9) and (10) indicate a rather complex dependence of the attenuation of the probing radiation on its polarization and on the polarization of the intense radiation that bleaches the F_2^- color centers. Let us find the values of the angle φ corresponding to the extremal values of k ($dk/d\varphi = 0$):

$$\operatorname{tg} 2\varphi_m = (b_4 - b_3)/(b_1 - b_2). \quad (11)$$

It is found that the direction of minimum k does not generally coincide with the direction of polarization of the intense radiation (i.e., $\varphi_m \neq \theta$). This is clearly seen in Fig. 6 (which, in general, demonstrates that θ is different from zero). Since b_i depends on Y (10), it follows that φ_m depends both on the energy density of the intense radiation and on the number of pulses. An interesting fact should be emphasized: in a crystal with residual losses, the quantity Y , and thus the directions of k_{\min} and k_{\max} , should vary along the direction of propagation of the radiation.

Using Eqs. (11) and (9), the following expressions are found:

$$k_{\min} + k_{\max} = k_0(b_1 + b_2 + b_3 + b_4)/2, \quad (12)$$

$$k_{\max} - k_{\min} = k_0(b_1 - b_2)/2 \cos 2\varphi_m. \quad (13)$$

From Eq. (9), it is seen that $k(n)$ is not described by a single exponential. Such a simple relationship would be valid only under some specific certain, e.g., for $\theta \ll 1$:

$$k(n) = k_0 \exp(-Y/\kappa). \quad (14)$$

If in this case we have also $Y \ll 1$, then k varies linearly with the number of pulses of the intense radiation

$$k(n) = k_0 \left[1 - \left(n\sigma_{20}/2\kappa \right) \int_{-\infty}^{\infty} I^2(t) dt \right]. \quad (15)$$

If $I(t)$ is a Gaussian distribution in time and space

$$I(x, t) = \frac{1}{\tau\pi^{1/2}} \frac{E_0}{\hbar\nu} \exp\left(-\frac{2x^2}{w^2}\right) \exp\left(-\frac{t^2}{\tau^2}\right), \quad (16)$$

then Eq. (15) becomes

$$k(n) = k_0 \left[1 - \frac{n\sigma_{20}}{2\kappa\tau(2\pi)^{1/2}} \left(\frac{E_0}{\hbar\nu}\right)^2 \exp\left(-\frac{4x^2}{w^2}\right) \right]. \quad (17)$$

Let us return to the experimental results. The first two points in Fig. 4 correspond to a linear variation of $\Delta k(n)$ ($n = 10^4$ and 2×10^4). For these points, the transverse distribution $\Delta k(x)$ should thus, according to Eq. (17), be described by the square of the function that describes the initial distribution of the intense radiation vs x (the TEM_{00} mode), which corresponds to $2w_{\Delta k} = 2w\sqrt{2}$. For the distribution shown in Fig. 2, $2w = 0.99$ mm and, therefore, $2w/\sqrt{2} = 0.71$ mm. Comparison of this value with the measured $2w_{\Delta k}$ values, and allowance for the variance of the experimental data indicates that bleaching of the F_2^- color centers in LiF proceeds, indeed, via a two-photon process. One-photon and the three-photon processes (with contributions equal to $2w_{\Delta k} = 2w$ and $2w_{\Delta k} = 2w/\sqrt{3}$, respectively) are excluded. The same conclusion is also supported by the

measurements of $\Delta k(x)$ for n values larger than 2×10^4 (obtaining $w_{\Delta k}$ from these data involves a more complex procedure).

A root mean square approximation of the experimental data [Fig. 4, Eq. (2)] yield the values $A = 0.4 \text{ cm}^{-1}$ and $C = -1.3 \times 10^{-5}$. The initial absorption coefficient of the crystal is $k_0 = 0.51 \text{ cm}^{-1}$. According to Eq. (2), this means that $A \neq k_0$, which should be valid for $n \rightarrow \infty$. This discrepancy can be explained by recalling earlier remark that, in general, $k(n)$ [Eq. (9)] does not vary exponentially. A simpler explanation is also plausible: the energy of the intense radiation pulses fell during the experiment (10^5 pulses) from 28 mJ to 23 mJ.

By combining (2), (15), and (17), it is found that:

$$\sigma_{20} = -2\tau\kappa C (2\pi)^{1/2} (\hbar\nu/E_0)^2. \quad (18)$$

Taking into account that $\tau = \tau_p/[2(\ln 2)^{1/2}]$, where τ_p is the total pulse length at half maximum, substitution of the experimental values ($E_0 = 4 \text{ J/cm}^2$, $\tau_p = 18 \text{ ns}$, and $C = -1.3 \times 10^{-5}$) yields $\sigma_{20} = \kappa \cdot 1.56 \cdot 10^{-51} \text{ s} \cdot \text{cm}^4$.

Let us consider now the data on the induced dichroism of the absorption. Equation (12), with $\theta \ll 1$, gives:

$$2(k_{\max} + k_{\min})/k_0 - 1 = 3 \exp(-Y/\kappa). \quad (19)$$

From Eqs. (10), (16), and (19), a relationship similar to (18) is obtained, in which C is replaced by:

$$C' = -\frac{1}{n} \ln \left[\frac{2(k_{\max} + k_{\min})}{3k_0} - \frac{1}{3} \right]. \quad (20)$$

Substituting the numerical values of the experimental data from Fig. 6: $k_{\min} = 0.15 \text{ cm}^{-1}$, $k_{\max} = 0.28 \text{ cm}^{-1}$ (due to the small value of the angle $\Delta\theta$, these values coincide, within the experimental errors, with k_{\parallel} and k_{\perp}), $k_0 = 0.51 \text{ cm}^{-1}$, the total number of pulses $n = 10^5$, the duration of intense radiation, and the maximum energy density, we obtain: $C' = 1.47 \times 10^{-5}$, so that, $\sigma_{20} = \kappa \cdot 1.77 \cdot 10^{-51} \text{ s} \cdot \text{cm}^4$.

Thus, both the dynamics of variation of the absorption coefficient with the number of interacting intense pulses, and the data on the induced dichroism of the absorption yield independently close values for the cross section of the two-photon absorption process of the F_2^- color centers in the LiF crystal.

One more observation is warranted, related to the magnitude of the calculated cross section of the two-photon process. In the treatment developed above, it was assumed that each excited color center is bleached after absorption of two photons. This assumption is valid if the probability of ionization of the color centers from this level (state 3) is much greater than the probability of the electron returning to the ground state. This assumption is fully plausible. No related data are, however, available in the literature.

Finally, the bleaching of the color centers in the laser cavity is usually considered to proceed under the simultaneous action of the intense IR radiation and the scattered light of the pumping lamps. In this case, at these large energy densities, when the bleaching of the PQS is almost total, the number of bleachings of the color centers should not depend on the power density of the IR radiation.

CONCLUSIONS

This work demonstrates the existence of a two-photon bleaching mechanism of the F_2^- color centers in LiF. The

cross section of this two-photon absorption process was determined with accuracy up to the factor κ ($\kappa = 2$ or 4 , in the numerical model presented in this paper, depending on the form of the dipole moment of the two-photon transition). Accurate determination of the value of the cross section was possible only by studying in detail the transitions with wavelength near 0.35μ , which play an essential role in the stability of the F_2^- color centers under intense IR radiation. Based on the data of this study, important practical conclusions can be drawn regarding the possible regimes of operation (peak power, pulse length, and total number of pulses) of LiF (F_2^-) crystals as passive quality modulators and as active elements in lasers.

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