# The Effect of the Frequency Modulation Doubling of the $F_3^+$- Color Centers Luminescence in LiF Crystals with Induced Anisotropy

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**Abstract –In LiF crystals with induced anisotropy spatial-periodic picture of $F_2$– and $F_3^+$–centers luminescence intensity distribution is observed. Period of spatial-periodic modulation of luminescence intensity usually coincide with the period of exciting light polarization change. For $F_2$-centers the experimental axial spatial-periodic dependence of luminescence is similar with calculated when absorbing and radiating transitions are described by linear oscillators (oriented along axes $C_2$). However, at the same experiment for $F_3^+$ – centers period of modulation of luminescence is doubling.**
**It was shown that the doubling of the modulation frequency in spatial-periodic luminescence picture arises as a consequence of nonlinear dependence of luminescence intensity versus exciting light intensity. When $F_3^+$ – centers are excited in absorption band ($\lambda_{max}$= 452 nm) the doubling of the luminescence intensity modulation frequency proceeds from the saturation of color center metastable triplet state.**

## 1. Introduction

In the numbers of works [1–3] axial-periodic distribution of luminescence intensity of color centers is studded as in the nature anisotropic crystals as in cubic crystals with induced anisotropy. Modulation period of luminescence usually is closed to period of light polarization state changing then one propagates in the one-axis crystal. Such spatial-periodic pictures are observed [1–3] with luminescence of different color centers in number of crystals ($\alpha$-$Al_2O_3$, $MgF_2$, LiF). However in the experiment for F3+ – center in LiF crystals with induced anisotropy the spatial-periodic picture of luminescence is observed with doubly shorter intensity modulation period. This specific effect is investigated in this work

## 2. Experimental result

As is known cubic crystals can be converted artificially into lower symmetry crystals by using axial compression or imposition of electric field.

Also it is known that the elementary oscillators which describe transitions in the color centers in the cubic crystals are oriented along the pivot symmetry axis of the second, third and fourth order [4].

In Fig. 1 the scheme of experimental observation of spatial-periodic luminescence picture of the color centers in cubic crystals with induced anisotropy is shown.

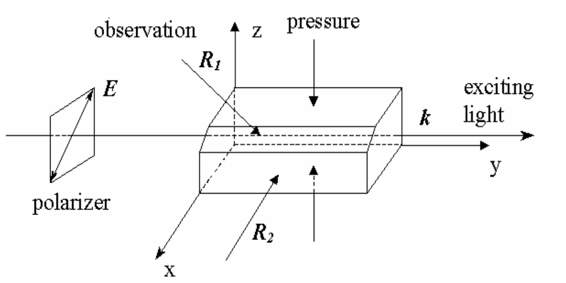


Fig. 1. Experimental configuration

In the development sample the optical axis c in the direction of compression was induced consequently linearly polarized exciting light is changed to right-circular then linear, left-circular and to initial polarized depending on phase change $2\pi y/\Lambda$, where $\Lambda = \lambda/\Delta n$; λ – the wavelength of the exciting light, $\Delta n = |n_o - n_e|$ – the value of the birefringence of crystal.

In Fig. 2 the photos of axial-periodic distribution luminescence of $F_2$ and $F_3^+$ color centers under argon laser excitation in pressured crystal LiF are shown. Exciting light of the argon laser ($\lambda$ = 488 nm) falls on a normal to crystallographic plane (100) of the sample, pressure is directed on axis $C_4$. Vector ***k*** is normal to stressing direction. The luminescence is observed in direction of axis $C_2$ (***$R_1$*** in Fig. 1).

In Fig. 2 photos of axial-periodic distribution luminescence of $F_2$ and $F_3^+$ color centers under argon

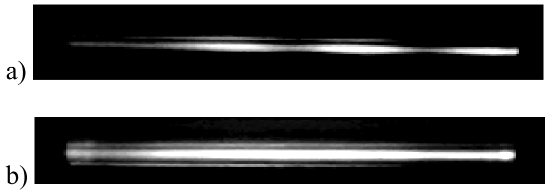

Fig. 2. Spatial distribution of luminescence intensity $F_2$ (a) and $F_3^+$ (b) – centers in LiF crystals with induced anisotropy. Exciting light was directed into normal to plane (100)

laser excitation in pressured crystal LiF are shown. Exciting light of the argon laser ($\lambda$ = 488 nm) falls on a normal to crystallographic plane (100) of the sample, pressure is directed on axis $C_4$. Vector ***k*** is normal to stressing direction. The luminescence is observed in direction of axis $C_2$ (***R₁*** in Fig. 1).

The axially- periodic dependence of the intensity (APD) of red luminescence ($F_2$ – centers) has a period which coincides with the period of polarization state change of the exciting light. The depth of modulation corresponds to calculated when the transition in the center is simulated by the linear oscillators oriented along six axes of second order. Modulation of the intensity of luminescence $F_3^+$-center is not observed in this experiment, as we see.

In Fig. 3 the spatial – periodic picture of color centers luminescence is shown when exciting light falls on normal to crystallographic plane (110) of sample, and stress is directed on axis $C_2$ perpendicularly to a vector ***k***, luminescence is observed in direction [111]. Photos have been made in the same experiment, optical filters through which observation was spent varied only.

a) b)

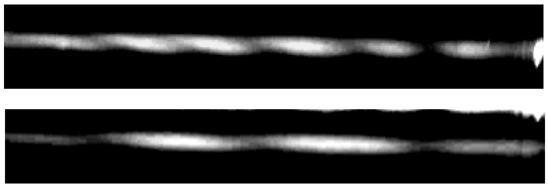

Fig. 3. Effect of frequency doubling spatial modulation of luminescence intensity of $F_3^+$ (a) – centers, in comparison with luminescence intensity of $F_2$; (b) – centers

As it is shown in Fig. 3 the spatial-periodic picture of green luminescence ($F_3^+$ – centers) has twice shorter period than red luminescence ($F_2$ the centers). It is impossible to explain such character of a picture, at the description of transition in the center elementary absorbing and radiating oscillators (linear dipoles or rotators) at any possible orientations of the oscillators.

**3. Discussion**

At calculation of axial-periodic dependence of luminescence intensity the absorbed power of *i*-oscillators group orientation is calculated:

$$W^{(i)} \sim n^{(i)}\left(\sigma^{(i)} I\right), \qquad (1)$$

where $n^{(i)}$ - concentration of the absorbing centers (in the ground state) *i* – number of orientation group, $w_{abs.}^{(i)} = \left(\sigma^{(i)} I\right)$ – probability of transition $1 \to 2$ for a unit of time, $\sigma^{(i)}$ - section of absorption of the color center of *i*- orientation group, *I*- intensity of exciting light. In dipole approximation section of absorption for $i$ – orientation group:

$$\sigma^{(i)} = C\left|\left(\langle 2|\vec{d}^{(i)}|1\rangle \cdot \vec{e}\right)\right|^2, \qquad (2)$$

where C – a constant, $\langle 2|\vec{d}^{(i)}|1\rangle$ – the electrodipole moment of transition, $\vec{e}$ - vector of polarization (Johnse's vector) of exciting light (generally both these vectors are complex and normalized on one).

Axial dependence of luminescence intensity, irradiated in direction of observation by all orientation groups of the centers:

$$J(y) \sim \sum_{i=1}^{p} W^{(i)}(y) q^{(i)}, \qquad (3)$$

where *q*(*i*) – the weight multiplier which proportional to radiation probability of oscillator *i*- orientation in observation direction. Summation is spent on number orientation groups of the oscillators *p* (at orientations of the oscillators on axes $C_2$, *p=6*; on axes $C_3$, *p=4*).

Usually consider that next condition are satisfied: Concentration of the absorbing centers of *i*-orientation groups in the ground state $n^{(i)} = n_0/p$ (in expression (1)) is constant does not depended from polarization and exciting light intensity, i.e. there are no effects of saturation.

It is possible to show that with satisfaction of this condition, i.e. when the intensity of luminescence linearly depends on the intensity of exciting light, the period of APD luminescence (3) is coincides with the period of polarization state change of the exciting light: $\Lambda = \lambda/\Delta n$. But with the disturbance, the intensity of luminescence nonlinearly depends on the intensity of exciting light and occurs the effect of the frequency doubling of modulation in APZ luminescence.

For $F_3^+$- center (the absorption bend with maximum on $\lambda_{abc} = 452$ nm) this condition is disrupted and concentration center of i- orientational group in ground (singlet) and in the metastable (triplet) states depends on polarization and intensity of exciting light. It is proves by the number of experimental data in the work [5].

Calculations for the three-level scheme showed that the expression for axial dependence (3) must be changed to:

$$J(y) \sim \sum_{i=1}^{4} \left( \frac{n_0\left(\sigma^{(i)} \cdot I\right)}{1 + \left(\sigma^{(i)} \cdot I\right)a} \right) \cdot q^{(i)} \qquad (4)$$

where $a = \dfrac{w_{s-t}}{w_{rel}\left(w_{em} + w_{s-t}\right)}$, $w_{em}$, $w_{s-t}$, $w_{rel}$ – the probability (for un. of time) respectively of emission, singlet- triplet transition and relaxation from the triplet level to the ground.

Fig. 4 shows calculated APD for the orientational groups of rotators by which they are described the absorbing and radiating transition in $F_3^+$- center. Experimental data (given in the work [5]) are used for determining the parameters in (4).

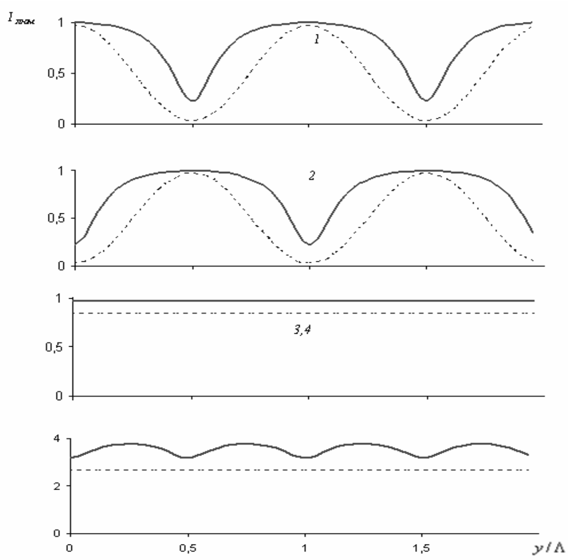


Fig. 4. Axial-periodic dependence intensity luminescence of rotators orientation group 1–4. In the bottom figure is shown summary APD. Continuous lines show dependences at saturation triplet level of the centre. Dotted line – dependences in absence saturation. The depth of modulation is 14 %

As can be seen from Fig. 4 the summary dependence of luminescence has doubly shorter period ($\frac{\Lambda}{2}$) than partial dependences 1,2. The contribution to the modulation of intensity give rotators 1 and 2 (rotators 3 and 4 give the unmodulated emission). The depth of modulation $\frac{J_{max} - J_{min}}{J_{max} + J_{min}}$ composes 14%.

For the experimental dates which shown in Fig.2 (exciting light falls along the normal to the plane (100) of crystal) the calculation of the axially-periodic dependence of luminescence $F_3^+$ -center shows the depth of modulation without saturation equal to 12,5% and with saturation ~2%. Latter fact is explained the absence of modulation of the luminescence intensity in our conditions for experiment.

## 4. Conclusion

Thus the effect of the frequency modulation doubling in APD luminescence is appears as the consequence of the nonlinear dependence of luminescence the intensity from the exciting light intensity. For $F_3^+$ – center with the excitation of luminescence into absorption band the frequency modulation doubling of luminescence occurs as result of metastable triplet level of color center saturation.

Calculations APD luminescence $F_3^+$ – center with saturation of the triplet level are in accordance with experimental dates and explain the observation of effect in Fig. 3,4 and the absence of modulation of the luminescence intensity in Fig.2.

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