



Color center based scintillation in LiF crystals



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Introduction

Color centers (CC) formation during crystal irradiation reflects nonradiative channel of energy losses. Therefore, potential ability of CC luminescence use for scintillation application could be important as an additional way for scintillators development and study. LiF crystals were chosen as the subject for investigation. It is known that F⁻ type aggregate color centers in irradiated LiF crystals are the basis for the laser generation of ultra fast pulses in the range of 820 – 1210 nm. Detectors are extremely suitable for laser plasmas and for X-ray lasers sources.

The intense fast luminescence, generated by optical pumping in the absorption bands of color centers, is widely used for LiF tunable lasers (Table 1).

The aim of the study is to investigate the radioluminescence of color centers in as-grown and irradiated LiF and LiF:Mg, OH crystals.

Of particular interest is the creation of F₂⁺, F₂⁻ and F₃⁻ centers emitting in near IR range under a selective irradiation by photons of different energy and excitation density.

Absorption and emission bands of CC in LiF crystal [G. Baldacchini, 2002]

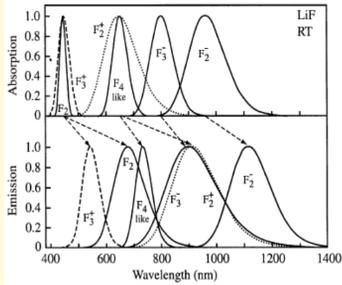


Table 1. Laser' active color centers in LiF crystals

[Basiev, 1997; Ter-Mikirtychev, 1997; Dergachev, 1998; Khulugurov, 2002; Baldacchini, 2002; 2003, 2007]

Color center	Absorption, nm	Emission, nm	Decay, ns	Stability	Laser efficiency %
F ₂ ⁻	443	678	17	<460°C	100
F ₃ ⁻	448	~528	8-11	<200°C	100
F ₂ ⁺	625	910	18	unstable	
F ₂ ⁺ (OH, Mg)	640 (770)	910	19.5	stable	62
F ₂ ⁺ like	600-780	1085 (796 -1210)		stable	28-53
F ₃ ⁻	820	900	10	unstable	10
F ₂ ⁻ (OH, Mg)	960	1120 (1080 - 1220)	55	stable	30

Experiment

The absorption and luminescence spectra of pure and OH, Mg containing LiF crystals were investigated at 10 and 297 K. The emission was excited by synchrotron pulses with energies of 4 – 22 eV and 130 eV (Superlumi and BW3, DESY, Hamburg) as well as by X ray (35 keV) irradiations. Cathodoluminescence of crystals was excited by electron beam (10 keV; 0.04, 0.4 and 20 μA).

Results

X-ray irradiation. Infrared absorption. LiF:Mg, OH → LiF:Mg, O

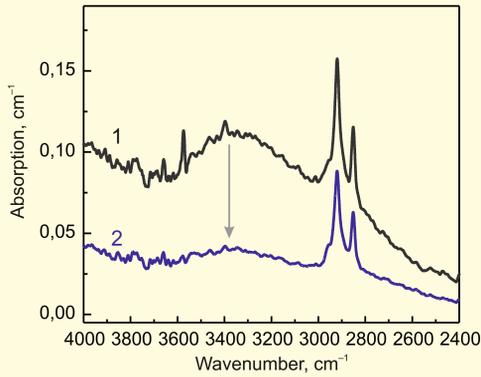


Fig. 1 Infrared absorption spectra of as-grown (1) and X-ray irradiated LiF:Mg,OH crystal (2) at 297 K. Dose 8.4·10³ Gy.

As-grown high-purity LiF crystal does not reveal emission and absorption in VUV- IR range. IR absorption spectrum of LiF: Mg,OH crystal consists of bands at ~3730 cm⁻¹ due to (OH⁻) and 3560 / 3610 cm⁻¹ (OH⁻ Mg²⁺).

X - irradiation leads to IR bands suppression. Process is attributed to the highly interacting hydroxide ions and products of their destruction.

X-ray and e-beam irradiation of LiF:Mg,O. Induced UV-VIS absorption

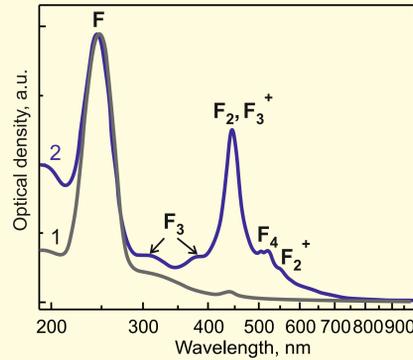


Fig. 2 Absorption induced by X-ray (1) and e-beam (2) irradiation in LiF: Mg,O crystal (spectra are normalized to the F⁻ band optical density). RT

X-ray exposure (penetration depth ~12 mm) induces much stronger volume coloration of LiF: Mg,O than pure LiF.

Electron irradiation (penetration depth ≤1μm) leads to intense surface coloration. Distinct peaks of F, F₂ and F₃⁻ centers are revealed.

Emission of LiF crystals excited by SR: hv ≥ E_g

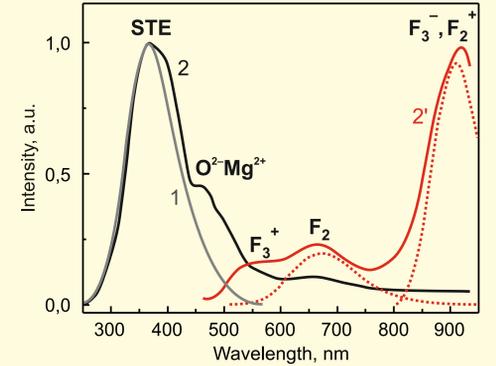


Fig. 3 Luminescence of LiF (1) and irradiated LiF:Mg,O (2) excited by energy of 14.0 eV at 10 K. Emission of irradiated LiF:Mg,O excited by 21 eV at 297 K (2'). Dotted - Gaussian of 2'. Spectra are normalized to STE emission intensity.

E_{exc} ≈ E_g (14.0 eV)

STE emission (~350 nm) dominates for LiF and LiF:Mg,O at 10K O²⁻ / Mg²⁺O²⁻ emission (400 / 440 nm) reveals in LiF:Mg,O

E_{exc} > E_g (hv = 21.0 eV)

Intense NIR emission of F₂⁺ and F₃⁻ and weak bands of F₃⁺/F₂⁻ are found for LiF:Mg,O at RT

Cathodoluminescence of LiF and LiF: Mg,O

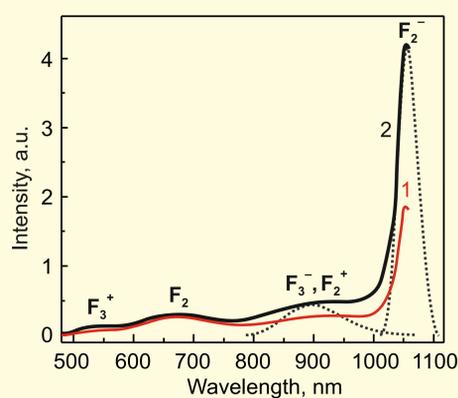


Fig. 4 Cathodoluminescence spectra of as-grown LiF (1) and LiF: Mg,O (2) crystals. 297 K

Electronic excitation leads to the intense color centers' luminescence at RT.

Spectra are the similar, and fitting allows separating the main peak at 1050 nm (F₂⁻) and weak ones corresponding to F₂, F₂⁺ and F₃⁻ centers.

Cathodoluminescence intensity of LiF:Mg,O is far above than LiF.

Density dependence of cathodoluminescence spectra

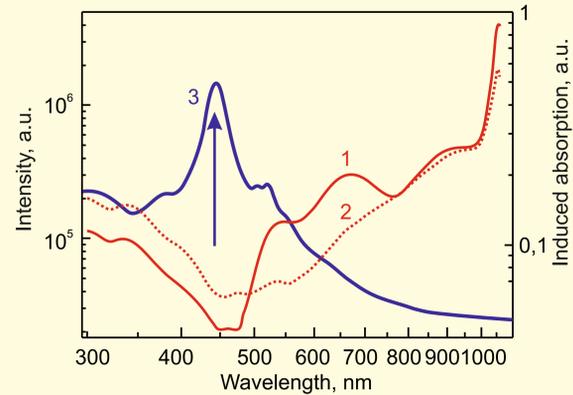


Fig. 5 Luminescence of LiF:Mg,O excited by e-beam with low (1) and high (2) density. Absorption spectrum induced by high density e-beam irradiation (3). 297 K. Cathod-beam with density of 8·10¹⁷ and 4·10¹⁹ (e-h)/s cm³ was used.

- High density excitation leads to intense coloration.
- Induces absorption overlaps the range of F₃⁺/F₂⁻ centers emitting.
- Spectra of cathodoluminescence depend on e-beam density.
- Emission in visible (F₃⁺, F₂⁻ centers) is suppressed whereas NIR bands remains stable with density rises.

Conclusions

- Near-infrared broadband luminescence of color centers (F₂⁺, F₃⁻ and F₂⁻) was revealed in LiF crystals under high energy excitation.
- Efficiency of NIR cathodoluminescence is higher in the presence of O²⁻ and Mg²⁺ ions.
- NIR emission is weakly dependent on the e - beam density, while visible luminescence significantly decreases due to induced absorption.
- Color centers' radioluminescence phenomenon indicates the possibility of LiF crystals using for "in-situ" detection of high-density ionizing radiation.

Generally, it has to be noted that near-infrared luminescence of LiF allows to use crystal for the high-energy registration. Taking into account the high-efficiency and fast decay of color centers emission, we can assume that phenomenon is useful for scintillation application. Moreover, LiF CC radioluminescence indicates a possibility of other dielectrics feasible to the in-situ detection of ionizing radiation.

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