Radioluminescence of color centers in LiF crystals

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Introduction

A new imaging detector for VUV or soft X-ray radiation based on optically stimulated luminescence of LiF crystals recently was presented [1, 2]. Detectors are extremely suitable for laser plasmas and X-ray lasers sources.

The intense fast luminescence of color centers in a range from visible to IR, generated by optical pumping in the absorption bands, is widely used for LiF based tunable lasers (Table 1). Contrary to photoemission, the radioluminescence of color centers has not been studied in detail. The investigation in this direction is perspective for detection of high density ionizing radiation.

The aim of the present study is to investigate the radioluminescence of color centers in as-grown and irradiated pure and activated LiF crystals. Of particular interest is the creation of F_2^+, F_2^- and F_3^- centers emitting in near IR range under a selective irradiation by photons of different energy and excitation density.

Table 1. Parameters of laser' active color centers in LiF

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	<260°C	100
F_2^+	640	910	18	unstable	
F_2^+ (Mg,OH)	770	910	19.5	stable	62
F_2^- (Mg,OH)	960	1080	55	stable	30

Experiment

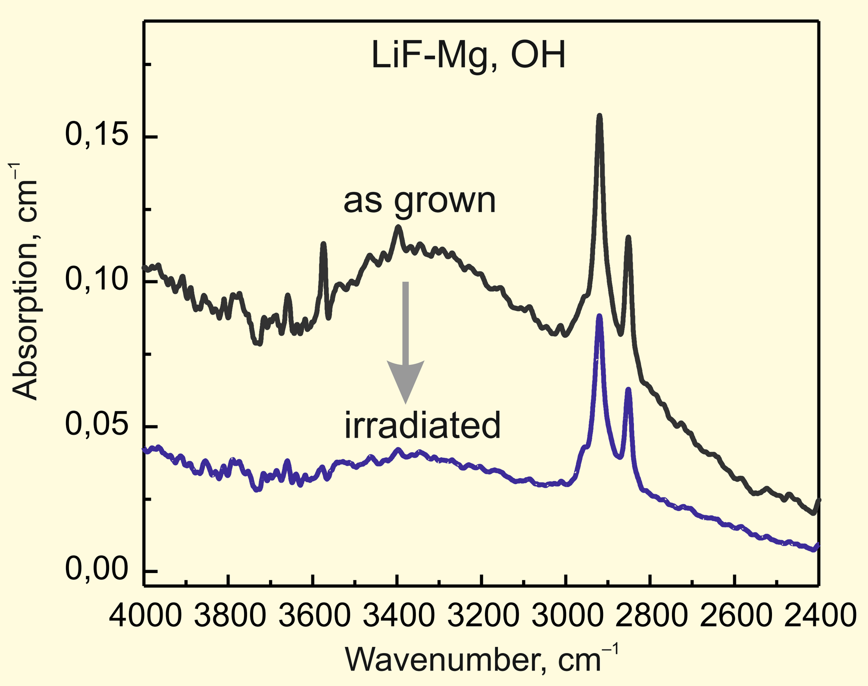
The absorption and luminescence spectra of ultra pure and containing the traces of hydroxyl and magnesium LiF crystals were investigated at 10 and 300 K.

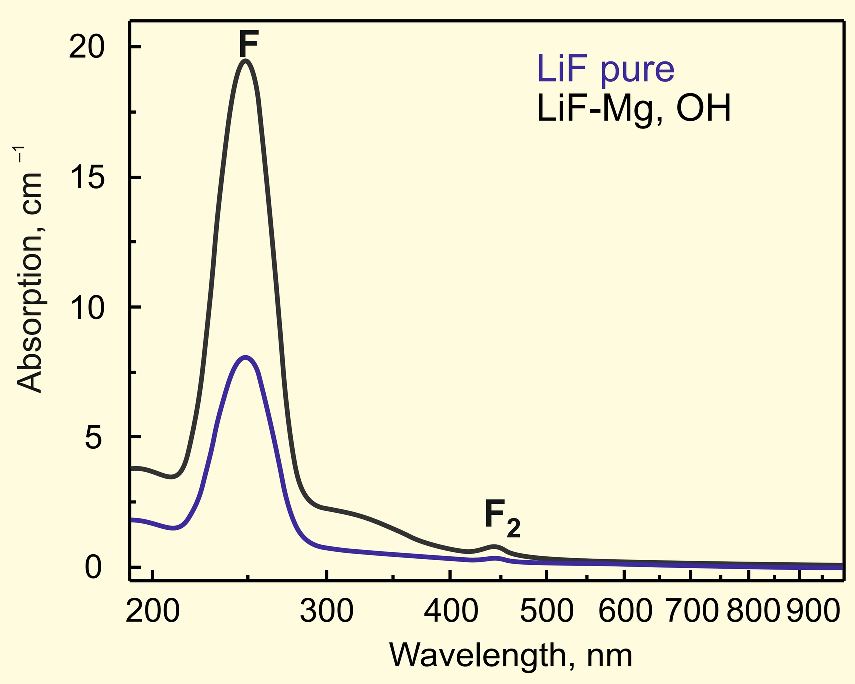
The emission was excited by synchrotron pulses with energies of 4 – 22 eV and 130 eV (Superlumi and BW3, DESY, Hamburg) as well as by X-ray (35 keV) irradiation.

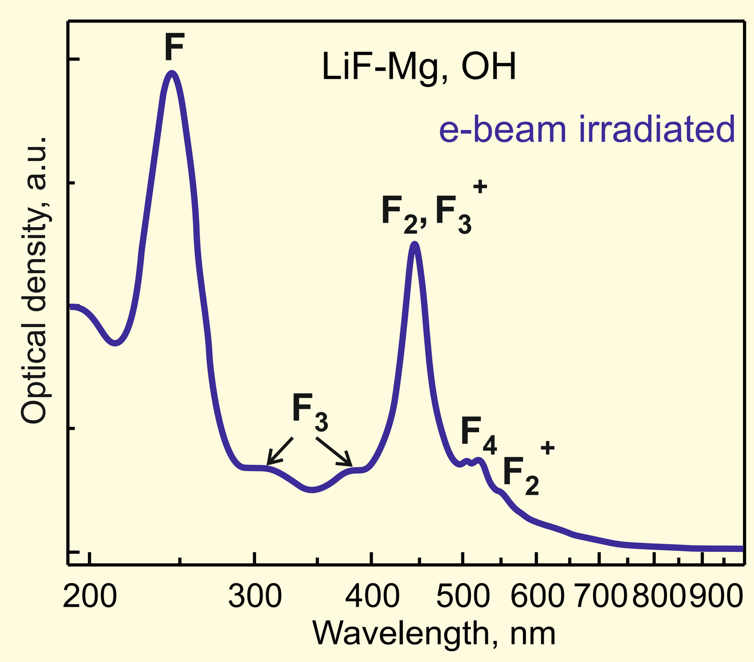
Cathodoluminescence of crystals was studied using excitation by a 2 mm diameter electron beam of energy 10 keV with beam currents of 0.04, 0.4 and 20 μA.

Results

1. Absorption spectra of irradiated crystals

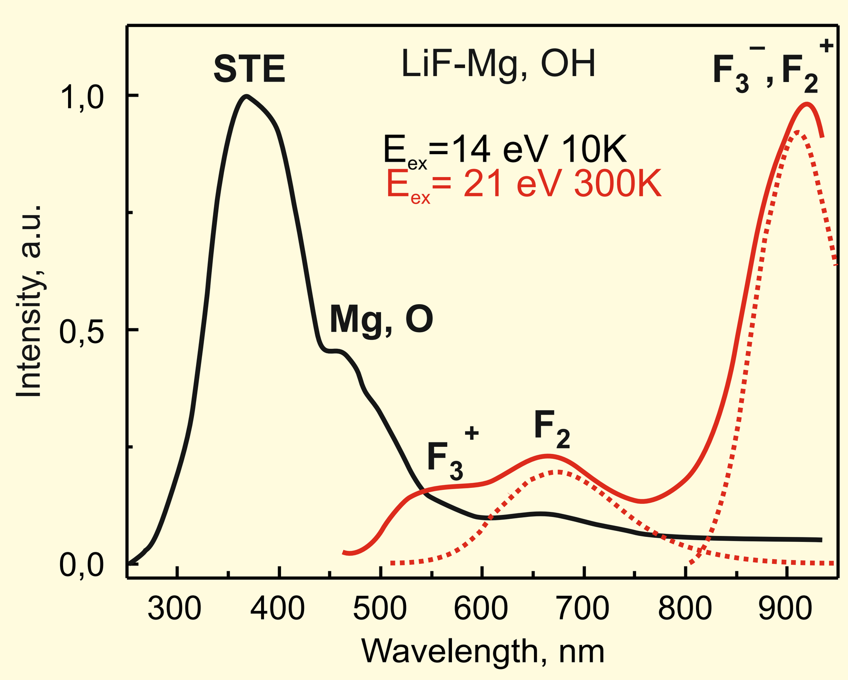


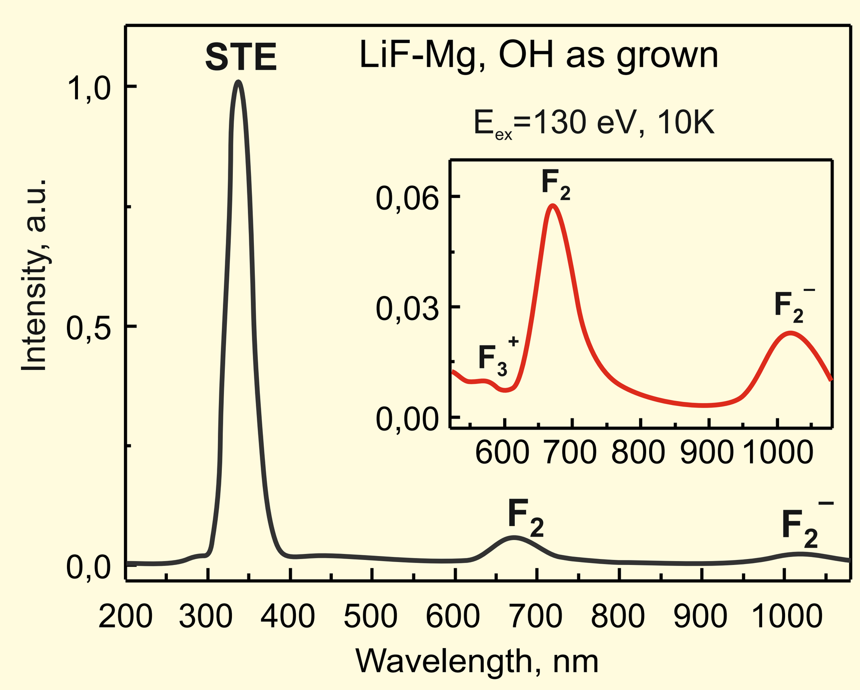




- IR - absorption spectrum of LiF:Mg, OH crystal reveals bands of OH^- and OH^--Mg^{2+}. Irradiation leads to their suppression. As a result the formation of O_2^- and $Mg^{2+}O_2^-$ centers takes place. This process is commonly used to stabilization of F_2^+ and F_2^- color centers in LiF based laser [3, 4].
- X-ray induced absorption spectrum demonstrates the intense peak of F centers and weak band of F_2 and F_3^+ centers.
- Electron beam irradiation leads to intense coloration of the thin subsurface. Colorability of LiF:OH, Mg is much higher than that of LiF crystal. Several peaks of F type aggregate centers arise beside F and F_2 bands.

2. VUV-excited luminescence

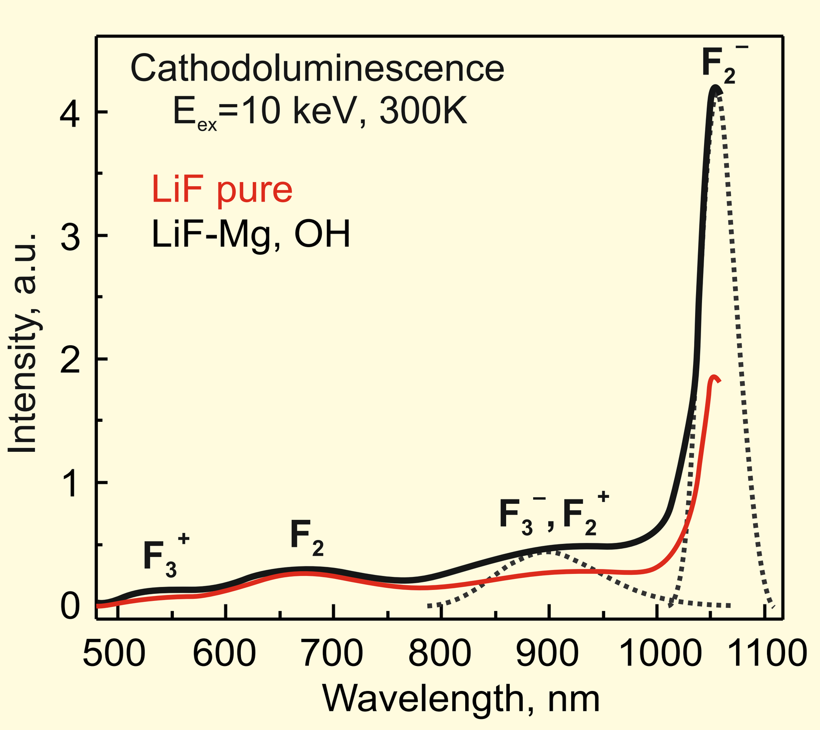


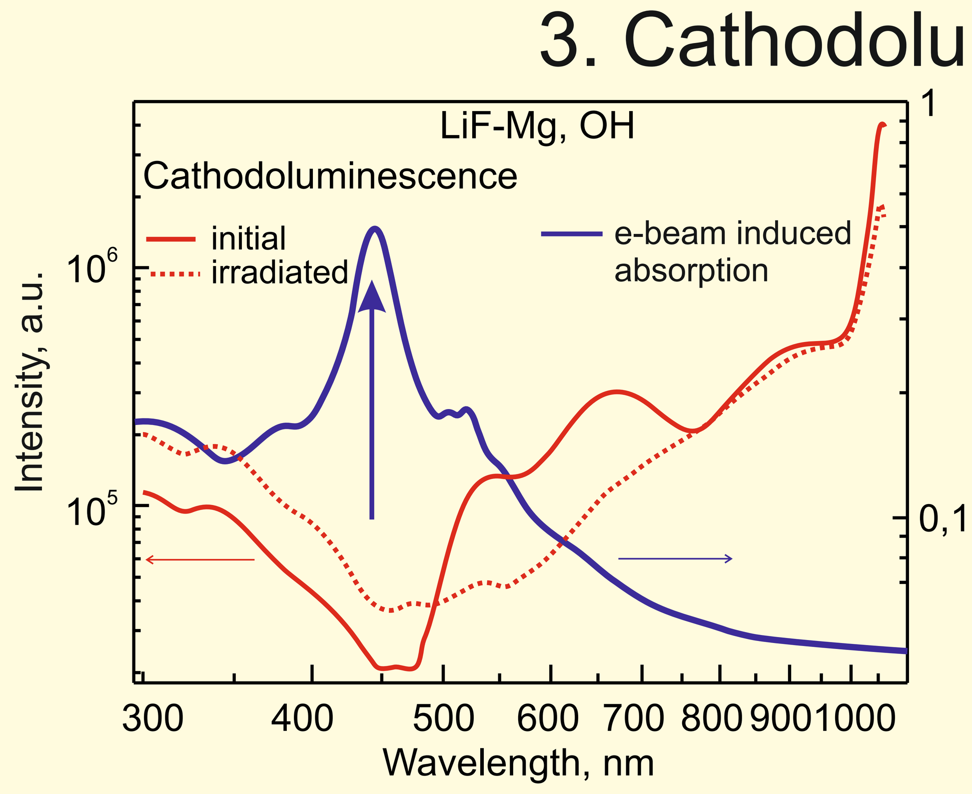


Of interest is the excitation region of 14 eV ($E_{exc} \approx E_g$) as well as the region of $h\nu_{ex} > E_g$ connected with the creation of separated electrons and holes.

- Excitation of colored LiF:OH, Mg crystal by energy of 14 eV leads to the appearance of STE emission (350nm) and weak bands in visible range at 10K.
- If the excitation energy is 21 eV (density ~10^{13} ph/cm^2 sec), strong luminescence bands are revealed in near IR, specified for F_3^+, F_2, F_2^+ and F_3^- centers, at RT.
- In case of $E_{exc} \approx 130$ eV (10^{15} ph/cm^2 sec), as-grown LiF:OH, Mg crystal demonstrates the color centers' emission at 680 and ~1010 nm, besides the intense STE band.
- The inhomogeneous distribution of emission bands was found by fiber-optic scanning, which is evidence of color center accumulation. The yield of color centers' luminescence is higher than STE emission at certain points.

3. Cathodoluminescence





The excitation density of e-beam (10 keV) was varied from $8 \cdot 10^{17}$ to $4 \cdot 10^{19}$ (e-h)/sec cm^3. Penetration depth into LiF crystal is near 1.2 μm.

In this case the color centers emission reveals in the range of 500-1100 nm at RT.

- The band with maximum at ~1050 nm (F_2^- -centers) dominates. Addition weak bands at 910 nm (F_2^+, F_3^-), 520 and 680 nm (F_3^+ and F_2) occur as well.
- The spectra of the observed emission do not depend on the presence of impurities and preliminary irradiation of crystals, but cathodoluminescence yield of LiF:Mg, OH crystals is far above that of pure LiF.
- With the increasing of density and exposition the intensity of IR-bands almost unchanged, whereas the visible emission decreases. Induced absorption overlaps the optical range, in which F_3^+ and F_2 centers are emitting.

Conclusions

Cathode beam application allows obtaining high excitation density, comparable with density in the track of ionizing particles which, can not be realized by photoexcitation.

- ✓ Excitation of color centers' luminescence in near IR region was firstly revealed in LiF crystals directly in the process of ionizing excitation with high density. The decisive role plays the small penetration depth of radiation (d ≤ 1.2 μm). Note that this effect was not detected for X-ray excitation with d~12 mm.
- ✓ It is known that the efficiency of lasers based on colored LiF crystals can be improved by doping with small and strictly controlled admixtures of anion and cation impurities [3, 4]. The similar phenomenon was determined for investigated samples. Namely, the intensity of F_2^+ and F_3^- color centers luminescence is higher in samples, containing O_2^- and Mg^{2+} - ions, than that in ultra-pure crystals. This fact opens the possibilities of searching for the optimal impurity composition of the material.
- ✓ If the density of the electron beam and the accumulation dose increase, the luminescence yield in near IR range remains practically unchanged, while the visible emission of F_3^+ and F_2 centers decreases due to induced absorption. So, the crystals are suitable for micro-radiography images of VUV and soft X-ray sources.
- ✓ The observed nonhomogeneous luminescence distribution of F_2^+, F_3^- and F_2^- centers is evidence of color centers clustering. This is consistent with the suggestion [5] that the irradiation providing high density of electronic excitations leads not only to the creation of stable Frenkel' defects but also to the excitation of a whole group of crystal ions, thus, causing the creation of bi-vacancies, lithium and fluorine interstitials as well as their associations.

In general, the mechanism of energy transfer to the color centers requires a detail study, but its presence indicates the possibility of using LiF crystals for "in-situ" detection of ionizing radiation.

References

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