Radioluminescence of pure and doped LiF crystals

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- Results. Possibility of radiation detection by LiF crystals

  I. LiF as detector for dark matter search

  II. Photo - and radioluminescence of color centers
- Conclusions

MOTIVATION
Possibility of applying LiF to dark matter search

A new field of application for scintillators emerged recently through strong interest in ***dark matter searches***.

Detecting *Weakly Interacting Massive Particles* (WIMP) requires **large detectors capable of discriminating the weak signal over the background caused by natural radioactivity. Light yield have to be ≥15 000 Ph/MeV at LNT.**

Why LiF may be suitable for the detection ?

- both Li^{+} and F^{-} ions have high interaction rates with such particles
- material behave the high radiation purity
- experience in growing large crystals in ISMA (up to ~200kg).

LY for LiF is not known.

Question: How great the light yield of pure and doped LiF? Could LiF crystal be a detector for dark matter search?

Scintillation efficiency of LiF crystals has been studied by us in cooperation with Institute for nuclear research of the Russian Academy of Sciences

MOTIVATION. Scintillation efficiency of LiF

Fundamental limits to the scintillation yield

Traditional approach: efficiency is determined by the energy gap

$$LY \approx \eta\, 10^6/E_g \approx 5 \cdot 10^5/\, E_g \quad \text{(LY – light yield, } E_g \text{ – gap)}$$

.

Scintillation efficiency of LiF?

Since $E_g \simeq 14.2$ eV, LY should be near 35.000 Ph/MeV.

In fact, the output of X-luminescence is negligible (0.02% of CsI at 5 K) [Pooley, 1970].

So, the first assumption does not work properly in this case.

T*he second assumption has to be based on the track structure analysis?*

Main requirements depending on the usage and LiF properties

Application	Requirements	LiF crystal
Scintillator	$E_g \leq 7eV$ LY high fast emission	$E_g \approx 14.2$ eV LY is low - *Why?*
Dosimeter	TSL / OSL $T_m \approx 200$ C	LiF (Mg,O) Effective dosimeter
Laser	Intense color centers' photoemission	LiF (Mg,OH) Effective color center based lasers
"In-situ" detector	Fast intense radioluminescence	*Is it possible?*

Motivation.

- LY of pure and doped LiF crystal is unknown.
- Intense and fast luminescence of CC may be promising for detectors.
- Color Centers emission has not been studied under high-energy excitation.

Research purposes

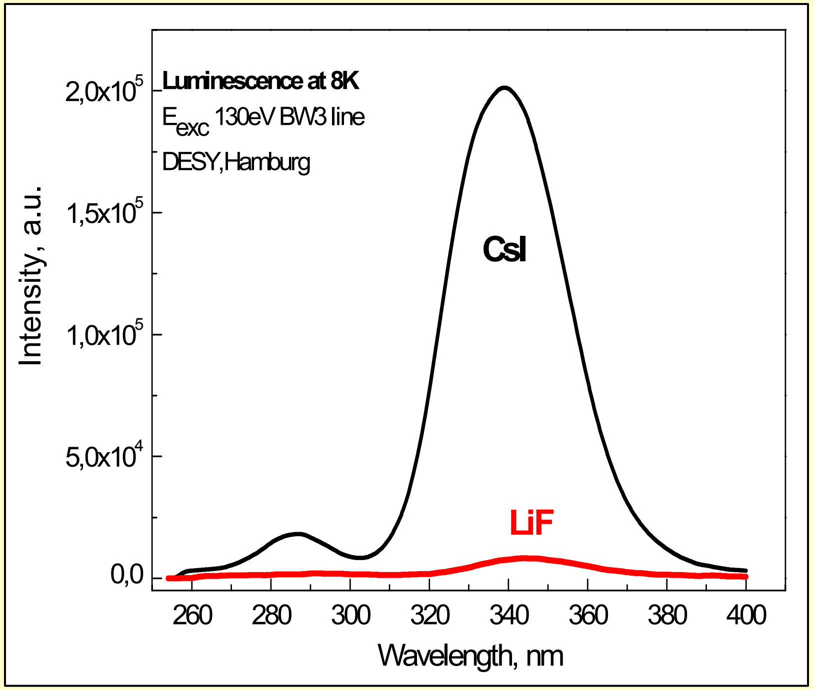
Questions

- How great the light yield of pure and doped LiF? Could LiF crystal be the detector for dark matter search?
- Could the phenomenon of color centers' luminescence be promising for "in-situ" detection of high density radiation?

Tasks

- Estimation the light output value of LiF at room and low temperatures
- Color centers' radioluminescence detection

I. Results. LiF pure as detector for dark matter



Emission spectra pure LiF and CsI crystals recorded using 130 eV SR excitation at 8K. Full absorption of excitation energy is the similar (~$1.4 \cdot 10^5$ см$^{-1}$).
http://11bm.xor.aps.anl.gov/absorb/absorb.php

Light output was evaluated as squares under curves. Based data for CsI ~100.000 ph/MeV at LNT *[Moszynski, et al. NIM A 537 (2005) 357]*

Our result:
STE emission was revealed at 8K
LY is low (≤ 5% of CsI)

LiF reveals too weak intrinsic luminescence at low temperature

Question: Why the output of LiF is so low?

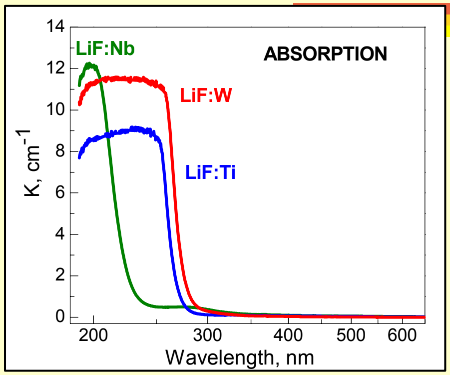
Light yield of pure LiF crystal at low temperature

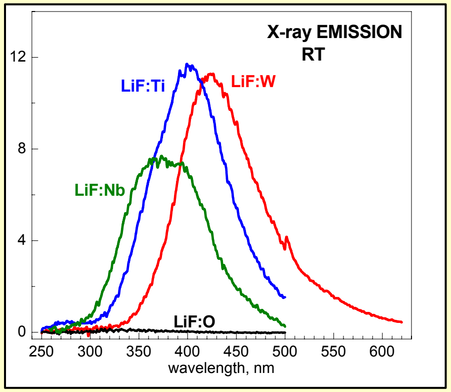
Temp.	Light yield Ph/MeV	Source	Reference
1.7 K	≤ 1 000	α ^{241}Am	C.Sailer et al., 2011
5 K	≤ 12 000	X-ray compared to CaF_2:Eu	E.Radzhabov et al., 2011
8 K	< 5 000	SR 130 eV compared to CsI pure	our data

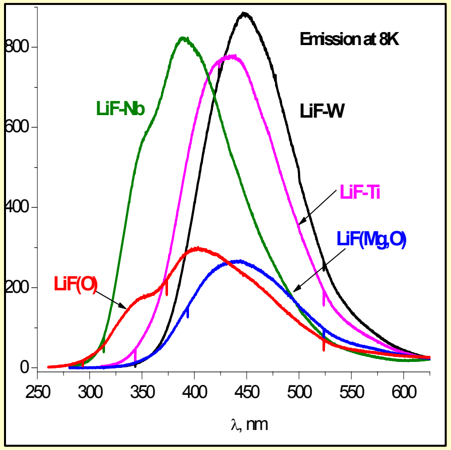
Value of LY obtained by various authors differs significantly. The best result seems overestimated. But even it does not reach the value required for the detection of dark matter particles.

Preliminary conclusion:
too weak STE luminescence does not allow to use pure LiF crystal to search dark matter particles.

Light Yield of doped LiF crystals







Can we improve efficiency LiF by doping?

Crystals LiF(Nb_2O_5), LiF(TiO_2) , LiF(WO_3)

At room temperature

Scintillation response of LiF crystal has been investigated both pure and doped with W, Ti and Nb with ^{60}Co and ^{239}Pu sources.

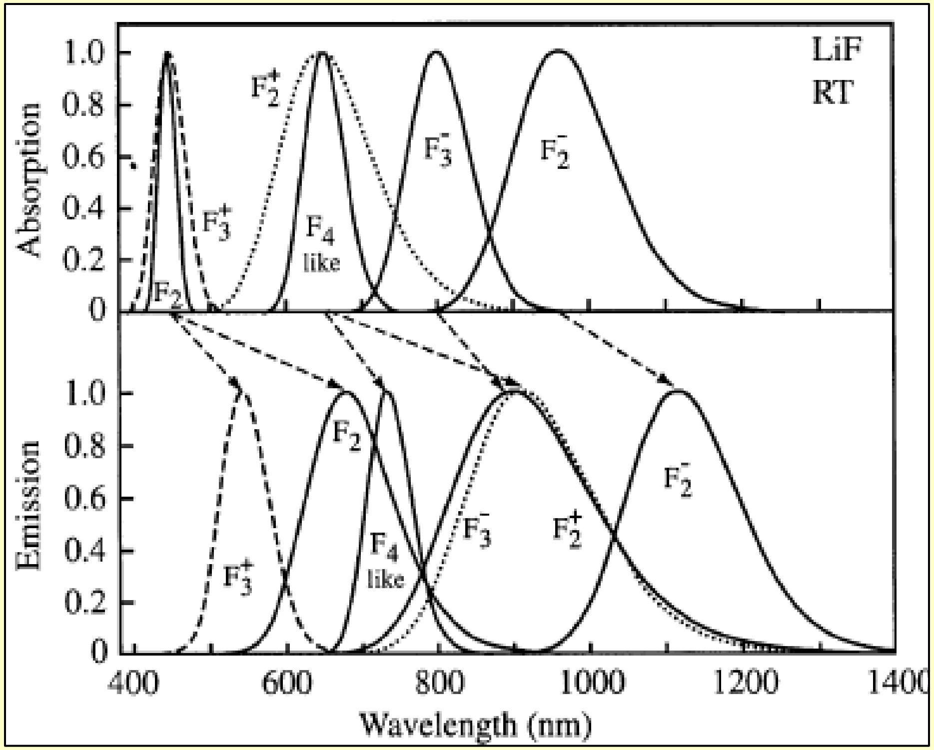
- LY <<0.2% of NaI:Tl for pure LiF.
- LY ~1.8% of NaI:Tl for LiF:W and LiF:Ti (decay closed to 35 and 115 μs, respectively).

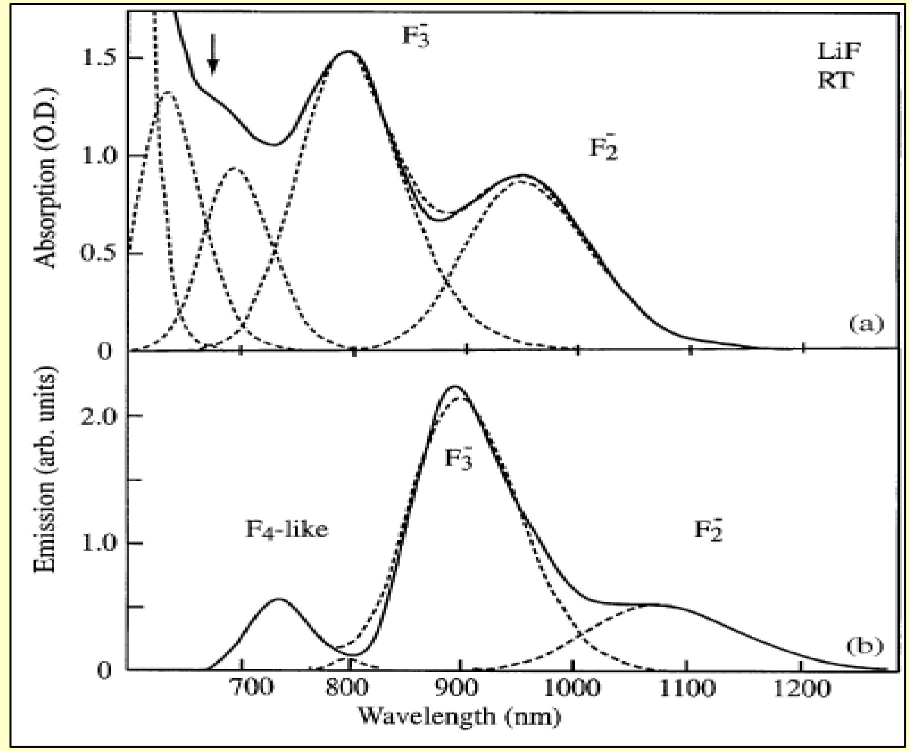
At low temperatures

Intensity of blue (oxygen) emission is higher while STE band is suppressed. How great the light yield - requires a special study.

Doping does not considerably improve the LY of LiF

II. Photo - and radioluminescence of LiF. Motivation. Color centers absorption and emission





Schematic absorption and emission bands in a colored LiF crystal

[G. Baldacchini, J. Lumin. 100 (2002) 333–343]

Absorption at RT above 600 nm crystal colored with a high-energy e-beam, and photoluminescence excited by a diode laser at 672 nm.

Laser-active color centers in LiF crystals

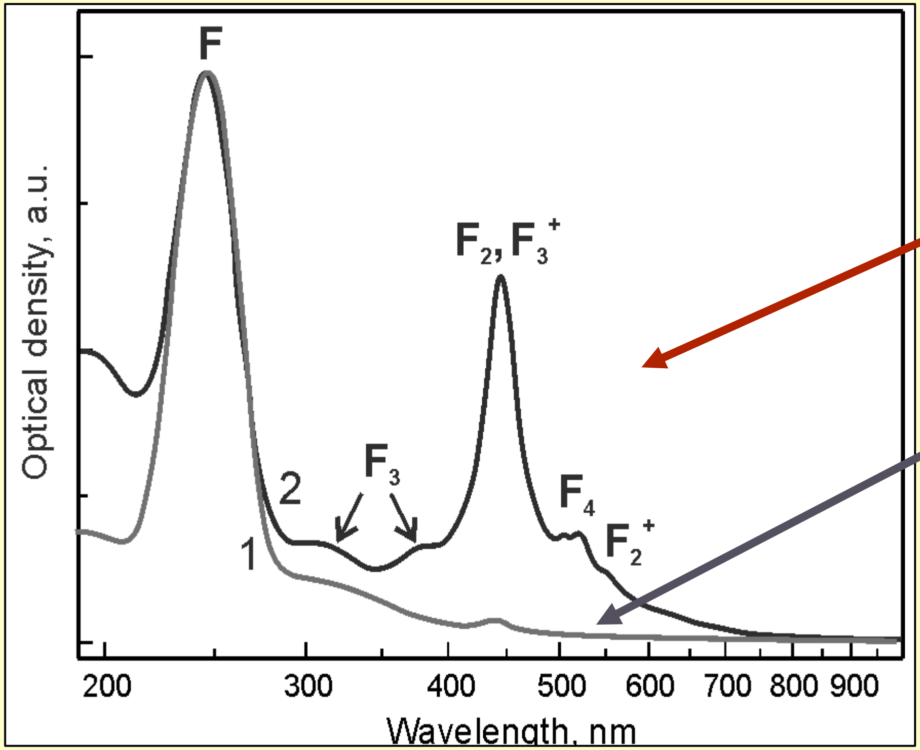
Spectral and kinetic parameters

Color center	*Absorpion, nm*	*Emission, nm*	*Decay, ns*	*Thermal 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^+ (Mg,O)	640 (770)	910	19.5	stable	62
F_2^{+*} (Mg,O)	600-780	1085 (796 - 1210)		stable	28-53
F_3^-	820	900	10	unstable	10
F_2^- (Mg,O)	960	1120 (1080 - 1220)	55	stable	30

[Basiev et al., 1997; Ter-Mikirtychev, 1997; Dergachev and Mirov, 1998; Khulugurov et al., 2002; Baldacchini, 2002; Flora et al., 2003; Baldacchini et al. 2007; Lushchik et al., 2012]

- **Color Centers emission under high-energy excitation has not been studied up to now.**
- **Taking into account the high efficiency and fast kinetics of these centers, research in this direction can be promising for the creation of new radiation detectors.**

II Results. Absorption induced by X-ray and e-beam irradiation



Absorption induced by X-ray (1) and
e-beam (2) irradiation of LiF: Mg,O at RT.
Normalized to the optical density of F- band.

Penetration depth

http://henke.lbl.gov/optical_constants/atten2.html

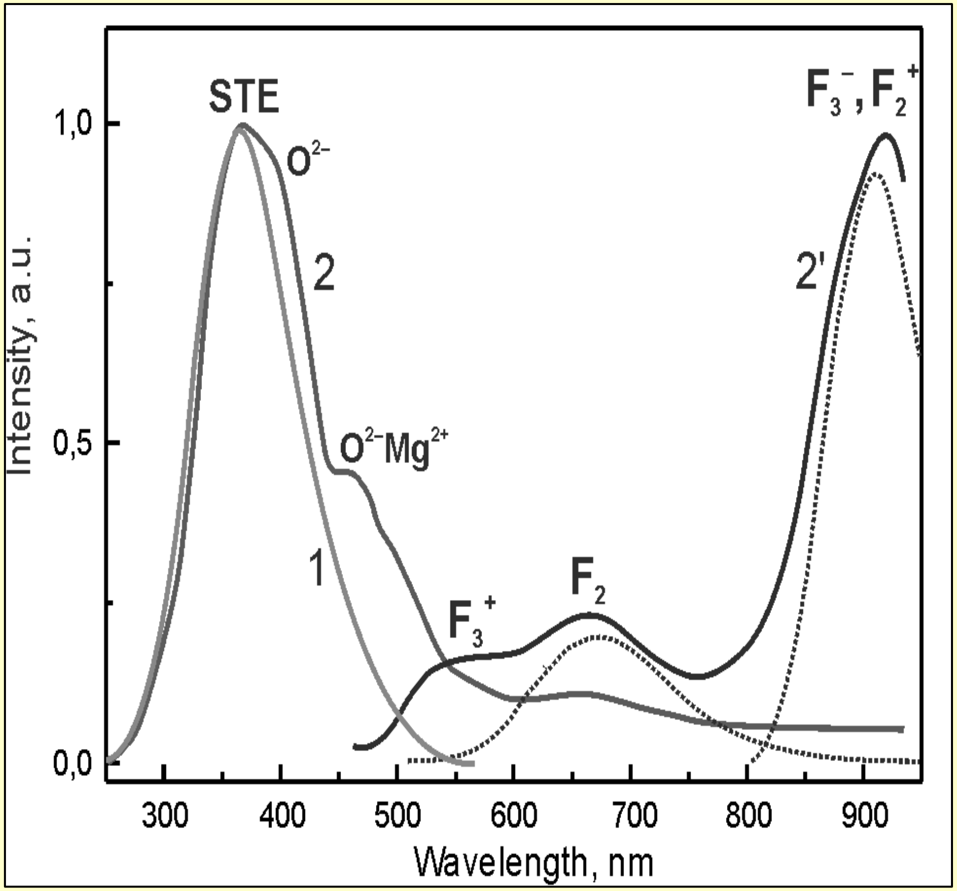
Induced absorption in UV - NIR of LiF:Mg,O crystal

Cathode-beam induces coloration with the intense F, F_2 and F_3^+ peaks

X-ray exposure induces volume coloration with the intense F, but weak F_n- peaks (penetration depth ~12 mm)

Electron-beam give rise intense coloration caused by complex centers in thin subsurface layer (penetration depth ≤ 1μm)

Excitation by hν~E_g and hν > E_g



Luminescence of as grown LiF (1) and irradiated LiF:Mg,O (2) crystals excited by 14.0 eV at 10 K. Emission of irradiated LiF: Mg,O excited by 21 eV at 297 K (2’). Normalized to the STE emission.

Luminescence of
as-grown LiF and colored LiF:Mg,O

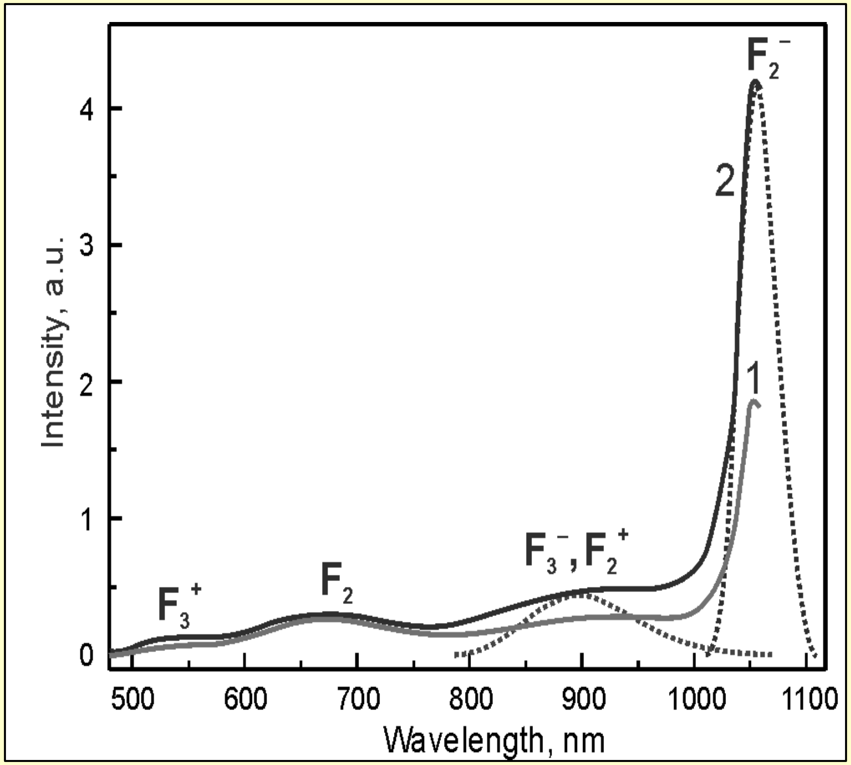
Excitation by hν ~E_g (14.0 eV) at 10 K

- *as-grown LiF* → STE emission.
- *x-irradiated LiF:Mg,O* → STE and blue (O^{2-} / $Mg^{2+}O^{2-}$) bands

Excitation by hν > E_g (21.0 eV) at RT

of colored LiF:Mg,O give rise to strong emission in NIR of F_2^+ and F_3^- centers.

Cathodoluminescence of as-grown LiF and LiF:Mg,O crystals

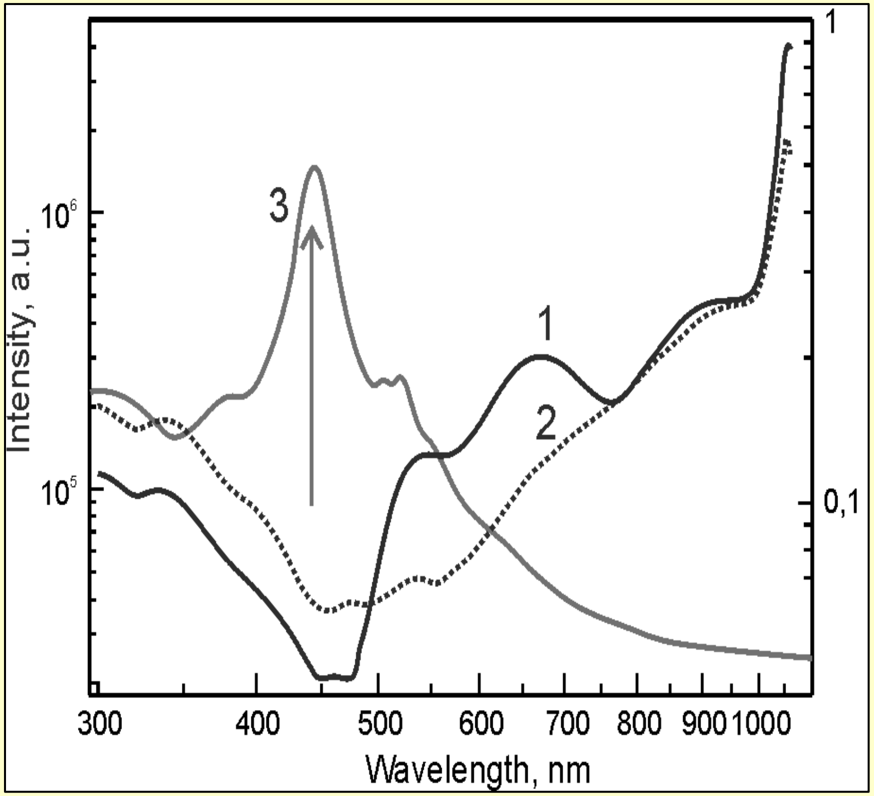


Cathodoluminescence spectra of as-grown LiF (1) and LiF:Mg,O (2). 297 K.

- Electronic excitation leads to the intense NIR emission of color centers for as-grown LiF and LiF:Mg,O at RT.
- Spectra of CL are the similar, but intensity of LiF:Mg,O is far above than LiF.
- Gaussian decomposition separates the main peak at 1050 nm (F_2^-) and several weak bands at 520, 680 and 910 nm of F_2, F_2^+ and F_3^- centers.

The phenomenon indicates the color centers luminescence directly in the process of electron excitation.

Cathodoluminescence dependence on density of secondary electron-hole pairs



CL spectra of LiF:Mg,O excited by e-beam with low (1) and high (2) density. Absorption induced by high density irradiation (3). 297 K.

- Cathodoluminescence spectra are changed with the density increase from $8 \cdot 10^{17}$ to $4 \cdot 10^{19}$ (e-h)/s cm^3.

- Since high density excitation leads to the intense coloration, VIS emission (F_3^+, F_2) is suppressed due to induced absorption, while NIR bands remain persistent.

The increased density of secondary electron-hole pairs leads to point radiation defects association and complex centers formation.

Conclusions

- Scintillation efficiency of LiF is low, so its prospect as a dark matter detector is questionable.
- NIR emission of F_2^+, F_3^- and F_2^- color centers under high-energy excitation is firstly revealed in LiF crystal.
- The phenomenon of color centers NIR luminescence point to possibility of LiF for "in-situ" detection.

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