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Article

Pulsed cathodoluminescence of BaFBr crystals irradiated by swift heavy ions

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Abstract. This study investigates the influence of swift heavy ion irradiation on the pulsed cathodoluminescence properties of BaFBr crystals. The objective is to analyze how ion species and fluence affect exciton luminescence behavior under pulsed electron beam excitation. BaFBr crystals were irradiated with 147 MeV krypton and 227 MeV xenon ions at varying fluences, and their PCL spectra were recorded at room temperature. Time-resolved measurements revealed the presence of luminescence bands centered at 4.2 eV, attributed to self-trapped excitons. A comparative analysis of irradiated and unirradiated samples showed that ion irradiation led to notable modifications in the intensity and decay dynamics of the nanosecond and microsecond components of the emission. The fast component dominated at lower fluences, while higher fluences induced a shift toward slower decay processes, likely due to the accumulation of radiation-induced defects. The experimental results suggest that controlled ion irradiation can be used to tailor the luminescence characteristics of BaFBr crystals, with implications for improving materials used in digital imaging and radiation detection technologies.

Keywords: BaFBr, luminescence, pulsed cathodoluminescence, pulsed electron flow, fast heavy ions.

1. Introduction

Barium fluorobromide (BaFBr) is a mixed halide crystal known for its application in image plate technology due to its ability to store and release optical signals upon stimulation. Crystallizing in a tetragonal PbFCl-type structure, BaFBr is characterized by its layered nature and capacity to form self-trapped excitonic states upon irradiation or stimulation. Its exceptional performance as an X-ray storage phosphor, particularly when doped with Eu^{2+}, makes it highly relevant for use in digital radiography and dosimetry. Understanding the luminescence behavior of this material under various excitation conditions is crucial for improving image quality and detection sensitivity in pulsed radiographic systems.

Despite the technological importance of BaFBr-based phosphors, a comprehensive understanding of the luminescence mechanisms—particularly those involving self-trapped excitons (STEs)—remains elusive. The processes of energy storage and release involve complex electronic transitions and defect dynamics, especially under high-energy excitation or irradiation. Recent advances in radiation detection demand materials that can reliably function under extreme conditions, such as exposure to swift heavy ions (SHIs), yet the influence of such irradiation on the time-resolved luminescence characteristics of BaFBr is not fully understood.

Several studies have attempted to elucidate the mechanisms underlying luminescence in BaFBr. For instance, [1] investigated phonon and electronic transport in related systems, indirectly contributing to the understanding of energy transfer in irradiated crystals. [2] discussed strategies for controlling thermal conduction in crystalline materials, a key factor in luminescent efficiency. More directly relevant, [3] examined electron-phonon interactions in semiconductors, offering insights into scattering mechanisms that may also govern exciton behavior in BaFBr. However, these studies either focus on different material systems or do not explicitly address the influence of ion irradiation on the luminescence decay dynamics of BaFBr.

The lack of targeted experimental data on the pulsed cathodoluminescence (PCL) behavior of BaFBr under high-energy ion bombardment represents a significant research gap . Previous work has largely overlooked how ion species, energy levels, and fluences modulate the temporal characteristics of excitonic emission, especially in the nanosecond to microsecond range. Moreover, the interplay between induced defects and exciton localization, which ultimately affects the emission decay kinetics, remains insufficiently characterized [4], [5], [6].

Based on these gaps, we hypothesize that irradiation with fast heavy ions (specifically Kr and Xe at energies of 147 and 227 MeV, respectively) significantly alters the PCL behavior of BaFBr crystals by modifying defect landscapes and affecting the balance between fast and slow excitonic decay components. This alteration can be quantitatively analyzed through time-resolved luminescence spectroscopy under pulsed electron beam excitation at room temperature.

The goal of this study is to investigate how swift heavy ion irradiation influences the spectral and temporal properties of pulsed cathodoluminescence in BaFBr crystals. By comparing irradiated and unirradiated samples across different ion species and fluences, we aim to identify key trends in exciton dynamics and define the role of induced defect states. The novelty of this work lies in its detailed analysis of PCL decay kinetics, using nanosecond-resolution time-resolved spectroscopy to reveal radiation-induced changes in the excitonic landscape of BaFBr crystals—a topic with direct implications for the development of advanced imaging phosphors and radiation detection materials.

In this work we present results on the exciton luminescence attenuation in barium fluorobromide crystals unirradiated and irradiated with 147 meV K and 227 MeV Xe ions up to fluences (10^{10}-10^{14}), (10^{10}-10^{12}) ion/cm^2, respectively, under excitation by pulsed electron flux at room temperature.

2. Methods

BaFBr crystals were grown by the Shteber method (Vinogradov Institute of Geochemistry SB RAS, Irkutsk, RF), in a graphite crucible in helium-fluoride atmosphere using stoichiometric mixtures of $BaBr_2$ and BaF_2.

Elemental analyses were performed by energy-dispersive X-ray spectroscopy (EDX or EDS) using an SEM Hitachi TM3030 (Hitachi High - Technologies Corporation, Tokyo, Japan) with a Bruker attachment and software quantax 70 (Bruker Nano GmbH, Berlin - Germany) EDX analysis. EDX is a method used to identify the composition of solid materials. This technique relies on exciting electrons close to the nucleus, which then causes distant electrons to lower their energy levels to fill the resulting holes. Each element releases a different set of X-ray frequencies when filling these holes, which can provide both qualitative and quantitative information about the near-surface composition of a sample.

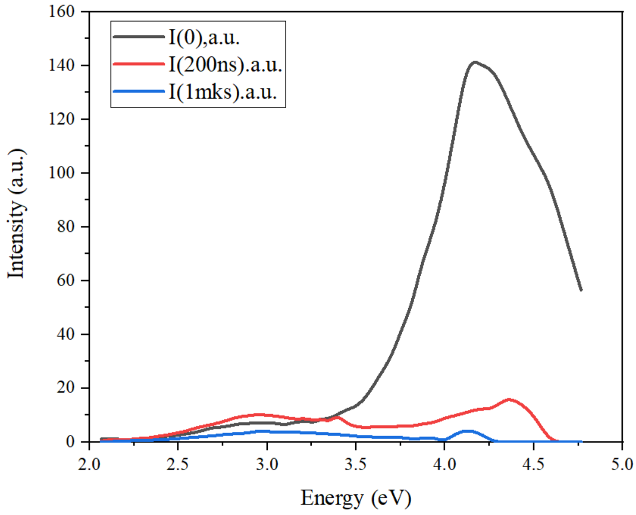
The plate-shaped samples prepared for irradiation were approximately 1 mm thick. Irradiation was carried out at the heavy ion gas pedal DC-60 (Astana, Kazakhstan) by high-energy krypton ions with energy 147 MeV up to fluences: 10^{10}-10^{14} ions/cm^2 and xenon ions with 227 MeV energy up to fluences of 10^{10}-10^{12} ions/cm^2 at room temperature.

Pulsed cathodoluminescence (PCL) was excited by a pulsed electron beam by the accelerator GIN-600 with parameters: $E = 0.25$ MeV, $t_{1/2} = 15$ ns, $P_p = 40$ mJ/cm^2. The spectra were measured on an optical spectrometer consisting of MDR-3 monochromator, PEM-97 photomultiplier tube, and a four-channel 350 MHz oscilloscope LeCroy WR 6030A. Luminescence oscillograms were recorded for photons with a certain energy between 1.0 and 5.0 eV at room temperature. The oscillograms were then converted into luminescence kinetic curves to determine the luminescence attenuation parameters and record the luminescence spectrum at a given time delay with a time resolution of 7ns statistical processing of data was carried out in the OriginPro program.

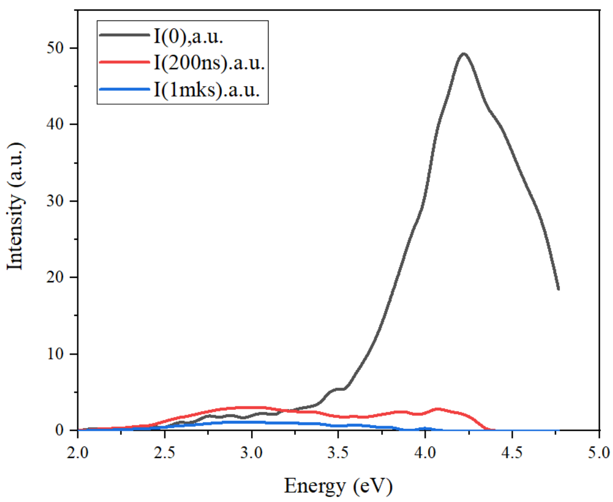
3. Results and Discussion

The spectrum of pulsed cathodoluminescence of unirradiated and irradiated crystals consists of a band with a maximum in the region of 4.2 eV, which, according to [7], corresponds to the luminescence of an self -trapped exciton (STE) of bromine. In [8], the authors hypothesized that the STE in BaFBr crystals has a two-center type structure consisting of an electron and a self-trapped hole $V_k(Br_2^-)$.

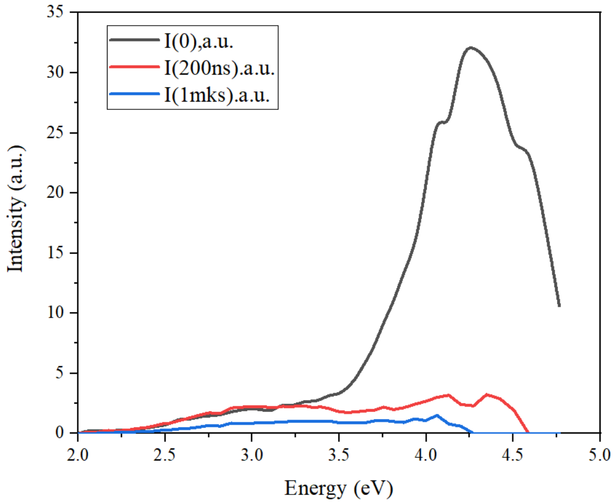
In each of the studied samples irradiated with krypton ions, a gradual decrease in intensity and a change in the shape of the luminescence curve in the ultraviolet region of the spectrum can be traced. According to the graphs shown in Figures 1 and 2, the dependence of the band intensity (PCL) 4.2 eV, on the fluence at irradiation with fast heavy ions ^{84}Kr and ^{130}Xe can be traced. The nanosecond component of the pulsed cathodoluminescence spectra flares up to fluence 1x10^{12} ions/cm^2 followed by slow decay up to fluence 1x10^{14} ions/cm^2, the microsecond component is not subject to noticeable intensity changes depending on the absorbed irradiation dose.



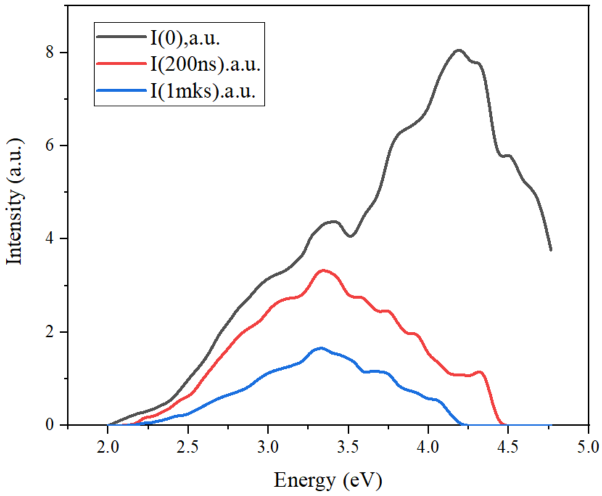
a) Unirradiated sample



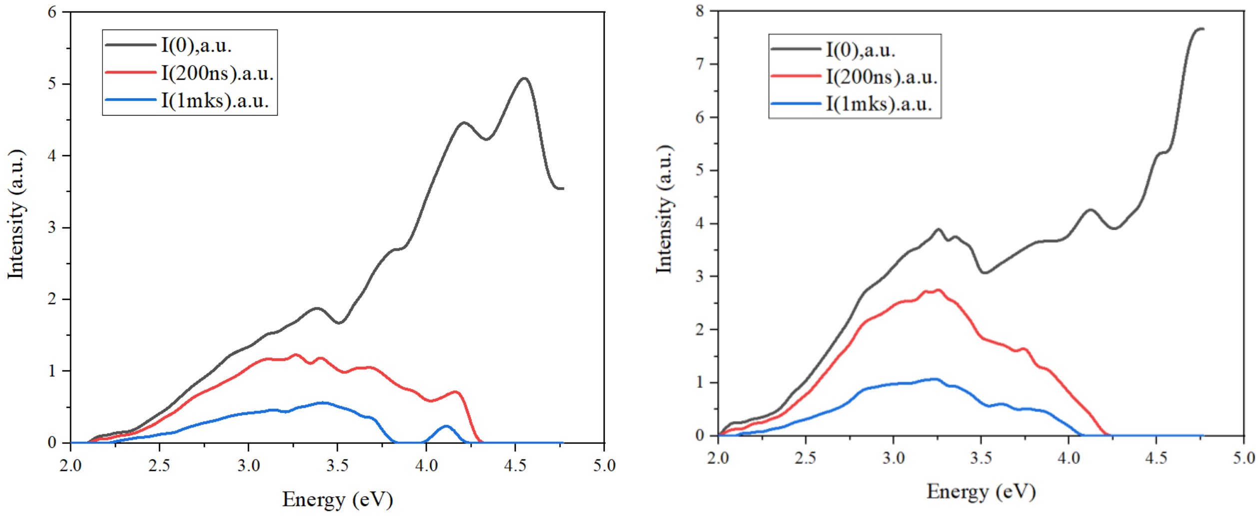
b) fluence 1x10^{10} ions/cm^2



c) fluence 1x10^{11} ions/cm^2



d) fluence 1x10^{12} ions/cm^2



e) fluence 1x10^{13} ions/cm^2 f) fluence 1x10^{14} ions/cm^2

Figure 1 – Spectra of microsecond (blue curve) and nanosecond (red curve) attenuation components of pulsed cathodoluminescence of BaFBr crystals irradiated with ^{84}Kr ions, E=147 MeV to fluences of 1x10^{10} - 1x10^{14} ions/cm^2 at room temperature

A distinctive characteristic of the 4.2 eV luminescence band is its good intensity at temperatures much higher than 77K. The data obtained by measuring pulsed cathodoluminescence at room temperature clearly demonstrate this (Figure 1a). According to [9], this band may originate from perturbed STE near point defects or impurity ions and arise according to the scheme “F+$V_k(Br_2^-)$” with the neighboring impurity ion O^{2-}.

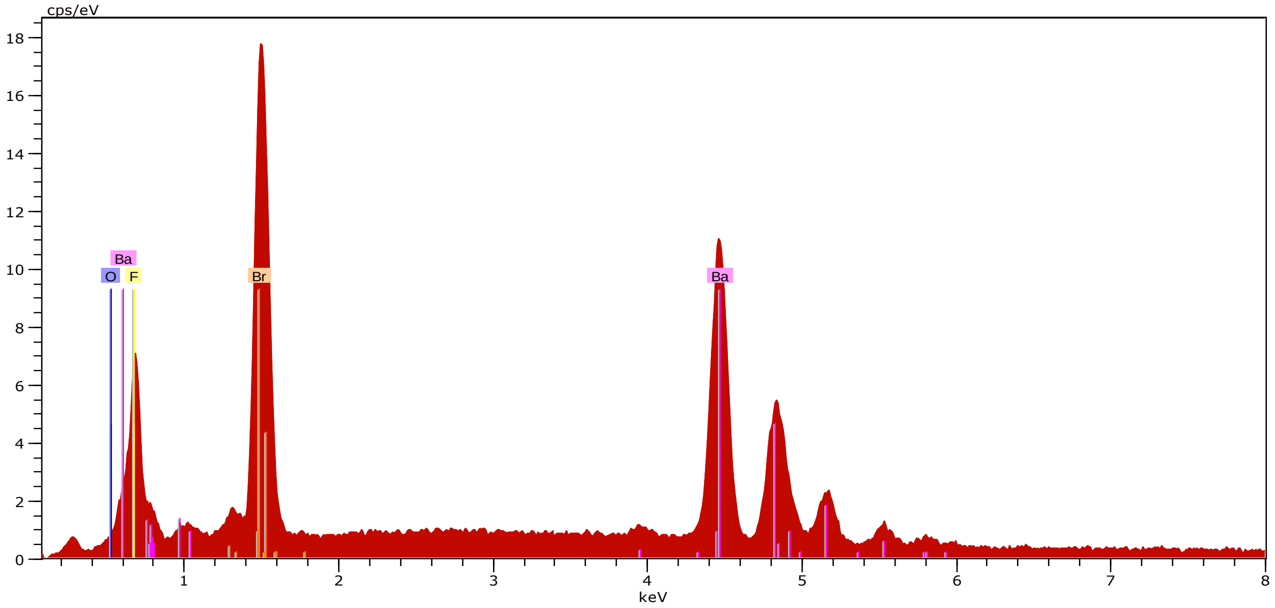
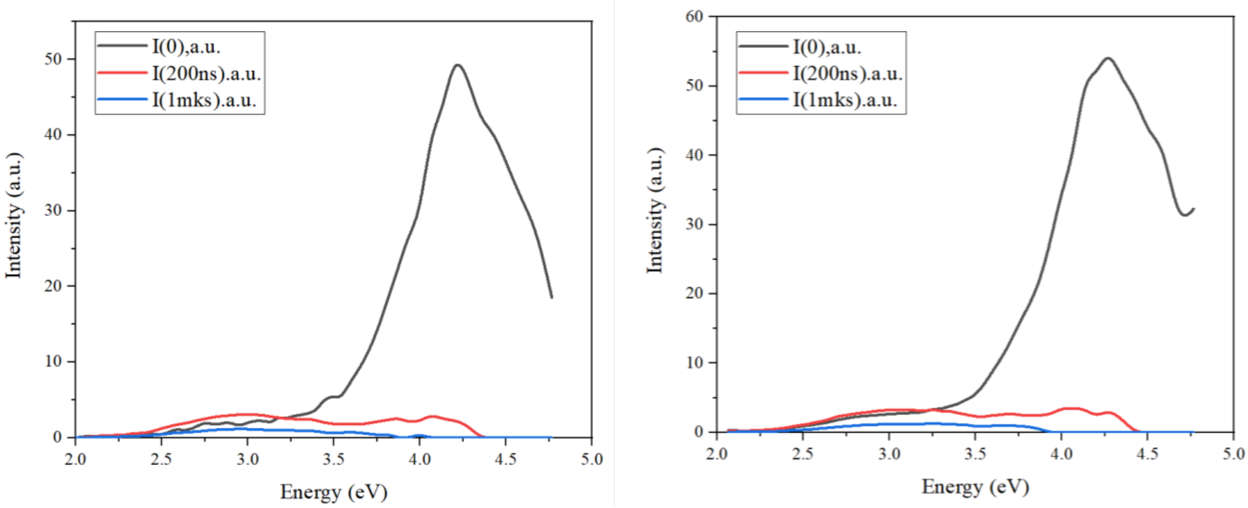
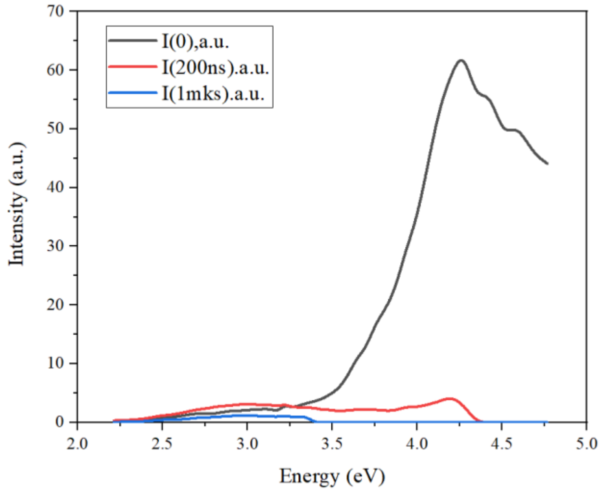


Figure 2 – EDX analyses for BaFBr

It should be noted that the BaFBr crystal contains some amount of oxygen O^{2-} on the crystal surface, this is evident from the data carried out by elemental dispersion analysis on the composition (Figure 2).



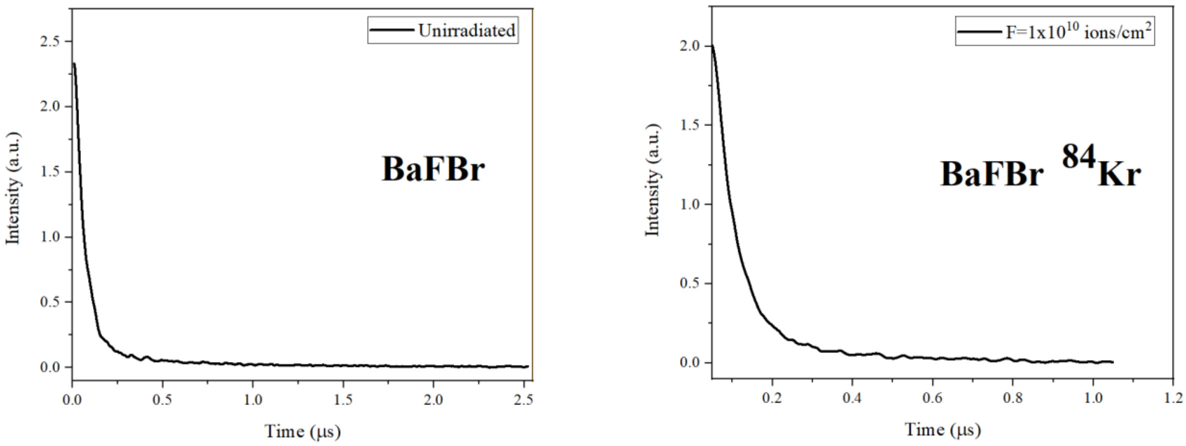
a) fluence 1x10^{10} ions/cm^2 b) fluence 1x10^{11} ions/cm^2



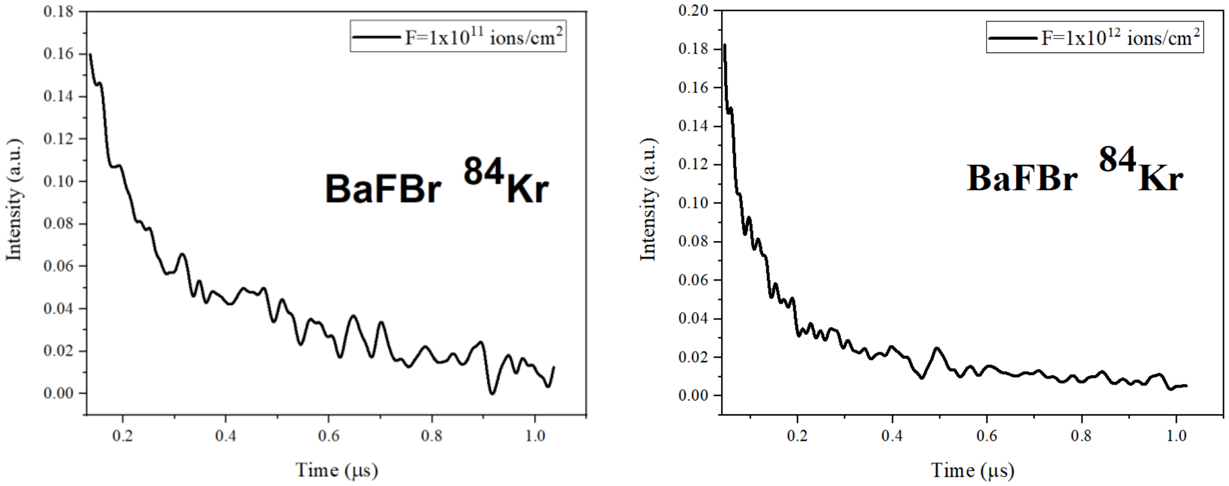
c) fluence 1x10^{12} ions/cm^2

Figure 3 – Spectra of microsecond (blue curve) and nanosecond (red curve) decay components of pulse cathodoluminescence attenuation of BaFBr crystals irradiated with ^{130}Xe ions, E=227 MeV to fluences of 1x10^{10} - 1x10^{12} ions/cm^2 of unalloyed BaFBr crystal, at room temperature

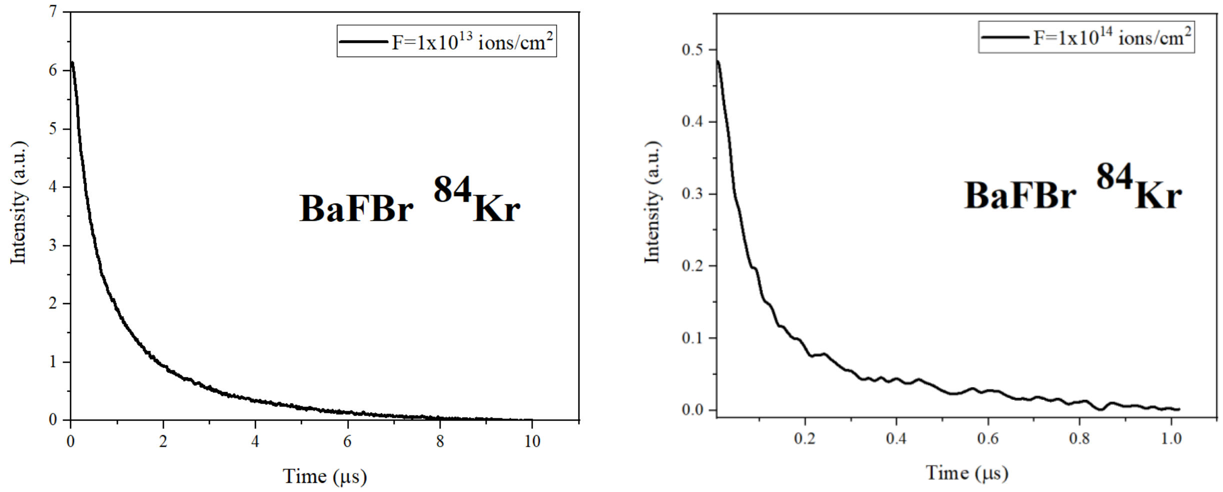
In BaFBr crystals irradiated with xenon ions, the glow of autolocalized bromine excitons and the flaring of the nanosecond component are also observed. It is noteworthy that at high fluences of 1x10^{13}-2x10^{13} ion/cm^2 the crystal began to peel and collapse, most likely this is due to track overlap [10]. The attenuation curves of the studied samples are shown in Figure 4a – i.



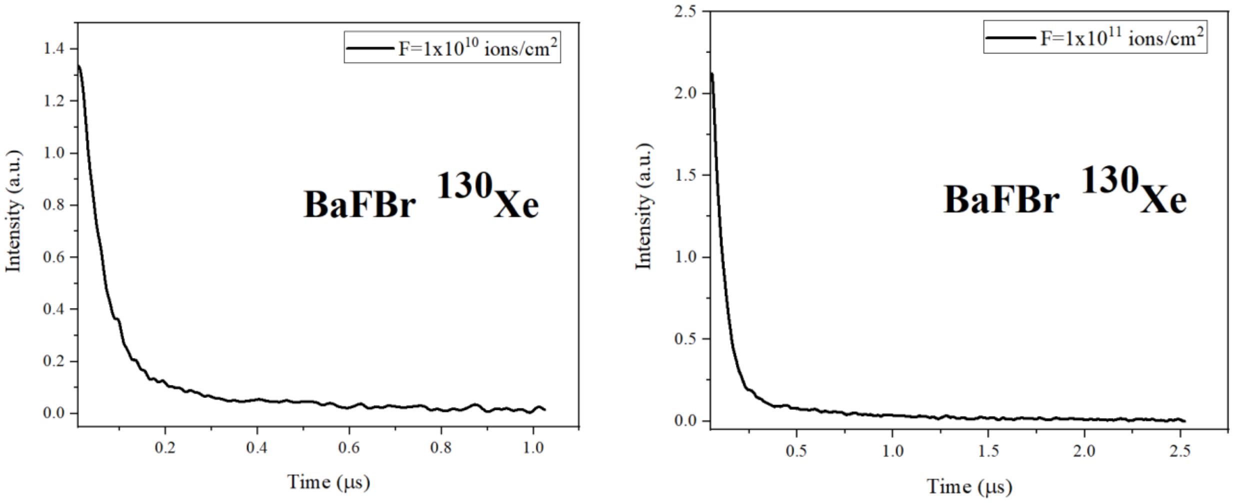
a) unirradiated sample b) fluence 1x10^{10} ions/cm^2 irradiated with ^{84}Kr



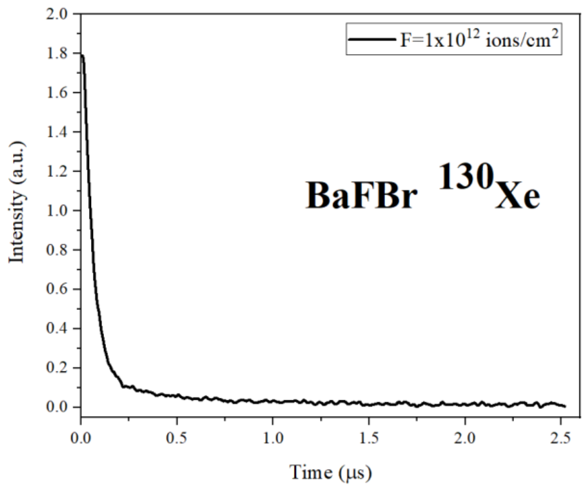
c) fluence 1x10^{11} ions/cm^2 irradiated with ^{84}Kr d) fluence 1x10^{12} ions/cm^2 irradiated with ^{84}Kr



e) fluence 1x10^{13} ions/cm^2 irradiated with ^{84}Kr f) fluence 1x10^{14} ions/cm^2 irradiated with ^{84}Kr



g) fluence 1x10^{10} ions/cm^2 irradiated with ^{130}Xe h) fluence 1x10^{11} ions/cm^2 irradiated with ^{130}Xe



i) fluence $1x10^{12}$ ions/cm^2 irradiated with ^{130}Xe

Figure 4 – Kinetics of luminescence attenuation in 4.2 eV and 3.93 eV bands during pulsed electronic beam excitation of BaFBr crystal samples irradiated with ^{84}Kr ions with energy E=147 MeV, ^{130}Xe with energy E=227 MeV to fluences $1x10^{10}$ - $1x10^{12}$ ion/cm^2 of unalloyed BaFBr crystal, at room temperature

The band attenuation kinetics at 4.2 eV for BaFBr crystals unirradiated and irradiated by fast heavy ions with different energies is described by a set of two exponentials at T=296 K with different values of the characteristic attenuation time, recorded in the table below.

Table 1 – Parameters of PCL on ion type and fluence

Ion	(Φ), ion/cm^2	(λ), nm	(τ), max		(A)		S= τ x A		S_1/S_2 (%)
			τ_1	τ_2	A_1	A_2	S_1	S_2	
-	0	295	0.057	0.541	2.841	0.116	161.937	0.062756	99.96/0.04
^{84}Kr	$1x10^{10}$	295	0.057	0.571	4,853	0,146	276.621	0.083366	99.96/0.04
	$1x10^{11}$	295	0.062	0.721	0.817	0.108	0.05065	0.077868	39.42/60.58
	$1x10^{12}$	295	0.0478	0.266	0.274	0.065	0.01309	0.01729	43.11/56.89
	$1x10^{13}$	290	0.392	1.985	4.093	2.551	1.60446	5063.735	0.04/ 99.96
	$1x10^{14}$	310	0.059	0.467	0.454	0.125	0.02678	0.058375	31.45/68.55
^{130}Xe	$1x10^{10}$	295	0.051	0.498	1.656	0.114	0.08446	0.056772	59.81/40.19
	$1x10^{11}$	315	0.062	0.650	5.172	0.144	0.32066	0.0936	77.41/22.59
	$1x10^{12}$	295	0.054	0.555	2.135	0.103	0.11529	0.057165	66.85/33.15

The data presented in Table 1 show that the initial amplitude of the PCL attenuation kinetics is significantly larger for the fast component in the nanosecond range, regardless of the fluence in BaFBr crystals irradiated with krypton ions. In samples irradiated with 130Xe up to fluences 1011-1012 ions/cm2 there is an increase in the amplitude of the slow component. The fast component of the unirradiated sample gives a significant contribution to the percentage of the light-sum of the luminescence kinetics, starting from fluence1011 ion/cm2 at irradiation with 84Kr ions there is a shift towards the slow component. When irradiated with xenon ions, the fast component contributes to the light sum percentage.

4. Conclusions

PCL measurements of BaFBr crystals at room temperature revealed a dominant luminescence band at 4.2 eV, attributed to self-trapped excitons of bromine.

Irradiation with swift heavy ions (147 MeV Kr and 227 MeV Xe) at fluences ranging from 1×10^{10} to 1×10^{14} ions/cm² led to significant changes in luminescence intensity and decay kinetics.

At low fluences ($\leq1\times10^{11}$ ions/cm²), the fast nanosecond component dominated the emission with a light-sum contribution of up to 99.96%, while at higher fluences ($\geq1\times10^{13}$ ions/cm²), the slow microsecond component became more prominent, contributing up to 99.96% of the emission in some samples.

The shift in decay dynamics reflects increased defect formation and track overlap effects at higher ion fluences, which alter the excitonic recombination processes.

The study successfully addressed the research problem by demonstrating how ion irradiation affects both spectral and temporal PCL characteristics in BaFBr, revealing patterns of transition between fast and slow components based on irradiation dose.

These findings can be applied to optimize the performance of image plate phosphors and radiation detectors by tailoring the excitonic response through controlled ion modification.

The study was limited to room temperature measurements and selected ion species; future research could explore temperature-dependent behavior, other dopants, and extended dose ranges to further refine material performance.

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Alexei Shalayev – modeling, interpretation.
Marina Konuhova – resources, editing, funding acquisition.

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