ФИЗИКА

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Effect of Xenon Ion Irradiation on the Raman Spectra of BaFBr Crystals

Abstract: Luminescent properties of barium fluorobromide crystals irradiated with xenon ions with energy E=229 MeV up to fluences (1×10^{10} - 1×10^{12}) ion/cm^2 are investigated in this work. The photoluminescence spectra show the presence of an uncontrolled impurity, which is involved in the mechanisms of defect formation. Raman spectra from the surface of BaFBr single crystals were used to determine the lattice vibration frequencies as a function of the fluence. A decrease in Raman spectra intensity is traced as the fluence increases at frequencies belonging to fluorine ions.

Keywords: BaFBr crystals, heavy ions, luminescence, photoluminescence, Raman spectra, defects.

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Introduction. BaFBr crystals doped with Eu^{2+} as an activator are widely used as scintillators and X-ray-preserving phosphors [1-7]. Due to the presence of two types of anion sites in these materials, two types of F centers were found, in which electrons captured on fluorine and bromine vacancies participate: F(F$^-$) and F(Br$^-$) centers, respectively [8-9]. In detectors based on BaFBr - the image created by ionizing and nuclear radiation remains stable in the dark at room temperature, which is successfully used to create storage screens (imaging plate - IP). Memory screens were developed initially for X-rays, but their application is currently extended to other types of ionizing radiation, such as neutrons, electrons, gamma rays, or ion beams [10]. BaFBr-based detectors have several advantages: high sensitivity, wide dynamic range, and high spatial resolution [11]. Thus, it is challenging to find optically isotropic information storage materials with the same characteristics as Europium-doped BaFBr.

The studied BaFBr crystal has a matlockite structure, which is similar in physical properties to the PbFCl-type structure, which has a tetragonal type of structure with a space symmetry group P4/nmm and a point symmetry group C_{4h}, having a density of ρ=4.95 g/cm^3, this type of structure also includes crystals: BaFCl, BaFI, SrFCl, etc. The visualized structure of the BaFBr crystal is shown in Figure 1.

Raman spectra of BaFBr have already been reported in [12-14], and the bands were related to specific modes of lattice vibrations. However, consideration of Raman modes after exposure to ionizing radiation has not been given due attention.

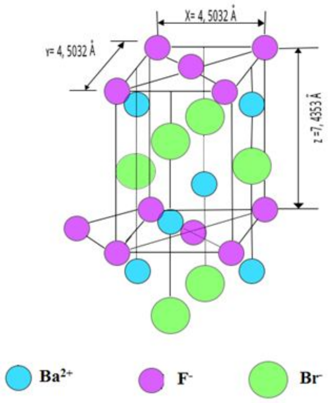

Figure 1 – BaFBr crystal structure

Experimental. BaFBr single crystals were obtained on an OKB-8093 setup (Redmet-8). Graphite was used as a material for manufacturing heating units and crucibles. Work on obtaining BaFBr single crystals was carried out by the Steber method. The starting reagents for single crystal growth were BaF_2 and $BaBr_2$. Samples prepared for the experiment in the form of plates were made in plane-parallel plates from the grown crystals of the BaFBr phosphor by cleavage along the cleavage plane.

The structure of BaFBr crystals was studied by X-ray diffraction using a MiniFlex 600 Rigaku setup with a copper cathode. X-ray spectra were recorded in the range from 3 to 90 degrees in the 2θ-θ mode.

Irradiation was carried out at the DC-60 ion accelerator (Nur-Sultan, Kazakhstan) with ^{131}Xe ions with a specific energy of 1.75 MeV/nucleon. Irradiation was carried out at room temperature, up tofluence 10^{10} to 10^{12} ion/cm^2.

The photoluminescence spectra of the irradiated crystals were measured using an SM 2203 spectrofluorimeter. The spectrofluorimeter is used for the near-ultraviolet and visible region of the spectrum and provides susceptible and stable measurements of excitation and luminescence spectra.

The Raman spectra were measured on a Solver Spectrum (NT-MDT) spectrometer. The spectra were excited by a laser beam with a wavelength of 473 nm and a spectral resolution of 1 cm^{-1}. The laser was focused using an objective with a magnification of 100x, forming a spot 2 μm in diameter on the sample surface.

Results and discussion. The X-raydiffractogram of the BaFBr sample is shown in Figure 2. In our diffractogram, BaFBr was detected, and the lattice parameters agree with the diffractogram of a stoichiometric composition.

Photoluminescence of impurity centers. The curves show the excitation and luminescence spectra of BaFBr crystals (Fig. 3, 4) irradiated with ^{131}Xe ions to various fluences at T = 300 K. It is fair to note that with an increase of the fluence from 10^{11} to 10^{12} ion/cm^2, the photoluminescence intensity increases.

With optical excitation of crystals, the decrease and slow increase in intensity may be due to topographic changes in the properties of the crystal. The luminescence band with a peak at 473 nm upon excitation with λ = 280 nm in BaFBr crystals is associated with centers containing O^{2-}_{F} impurity ions.

The EPR study in [15-16] showed various forms of oxygen impurity, which are assigned to $O^{2-}_{Br^-}$, $O^{2-}_{F^-}$, and O^{2-}_{2} in an indefinite region of the lattice. A study of the generation of F color, O, and V_k centers at low temperatures using magneto-optical and EPR methods showed that these vacancies are Br vacancies [15-16]. At temperatures above 120 K, the V_k (Br^-_2) centers become mobile and react with the centers, forming O^-_F hole traps. At temperatures above 200 K, F(Br^-) centers can

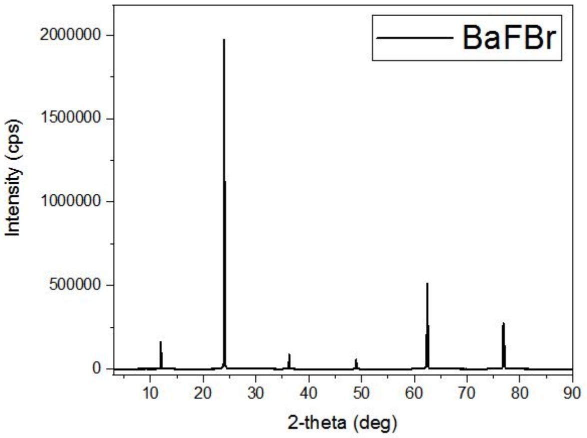

Figure 2 – X-ray diffractogram of BaFBr samples

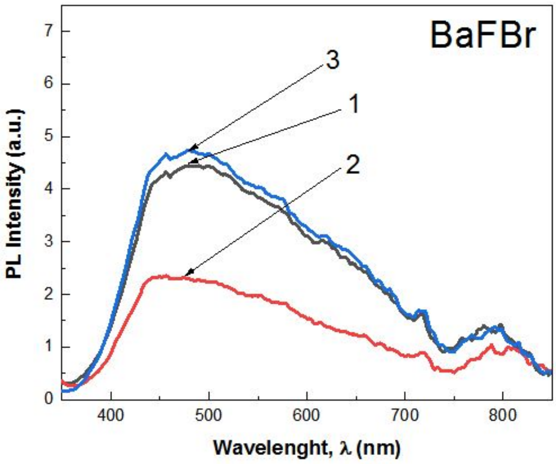

Figure 3 – Luminescence spectrum of BaFBr crystals irradiated with ^{131}Xe ions, $\Phi=10^{10}$-10^{12} ion/cm^2 at a temperature of 300 K, where 1 – $\Phi = 1 \times 10^{10}$ ion/cm^2; 2 - $\Phi = 1 \times 10^{11}$ ion/cm^2, 3 - $\Phi = 1 \times 10^{12}$ ion/cm^2

scatter and isolate [15], [17]. The study carried out in [15] proved that the defect center formed upon X-ray irradiation of BaFBr is due to oxygen impurity. The creation of F(Br$^-$) centers, according to [18], becomes clear from the processes:

$$V_k(Br_2^-) + O_{F^-}^{2-} \xrightarrow{ionizing\ \ irr.} F(Br^-) + V_k(Br_2^-) + O_{F^-}^{2-} \xrightarrow{T\geq 120K} F(Br^-) + O_{F^-}^{-}$$

Raman spectra. According to the literature data, obtained using calculations, barium fluorobromide crystals have 12 Raman vibrational modes, of which: six are active Raman modes with 2A$_{1g}$, B$_{1g}$, and 3$_{Eg}$ phonon symmetry, four active IR modes with 2A$_{2u}$ and 2E$_u$ phonon symmetry, and two acoustic modes with phonon symmetry 2A$_{2u}$ and 2E$_u$ [19-20].

It should be noted that at fluences above $\Phi = 1 \times 10^{12}$ ions/cm^2, the crystal began to delaminate and break down. Perhaps this is due to the distribution of bonding forces in crystals of the MeFX type (Me - Ca, Sr, Ba and X = Cl, Br, I), where there is a strong bond between the metal and halogen ions and between them, there is a double layer of halogens with more weak coupling [21].

Fig. 5 shows the Raman scattering (RS) spectrum of BaFBr crystals after ion irradiation, recorded at room temperature. Raman spectra measurements reveal well-defined bands with phonon frequencies of approximately 106 cm^{-1}, 122 cm^{-1}, 210 cm^{-1}, and 236 cm^{-1} and symmetry (E$_g$),

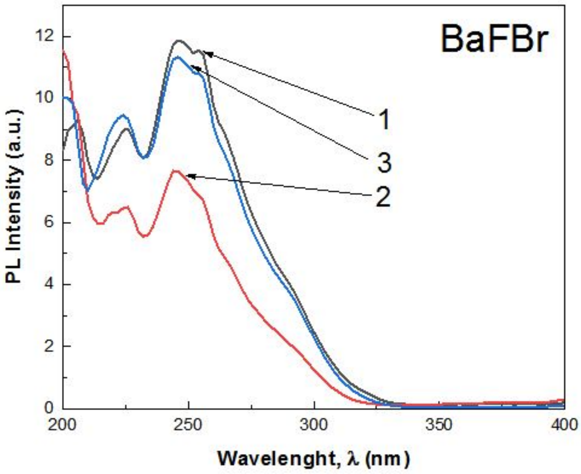

FIGURE 4 – Excitation spectra of BaFBr crystals irradiated with ^{131} Xe ions, 1.75 MeV/nucleon at 300K, where 1 – $\Phi = 1 \times 10^{10}$ ion/cm^2 ; 2 - $\Phi = 1 \times 10^{11}$ ion/cm^2 , 3 - $\Phi = 1 \times 10^{12}$ ion/cm^2

(A_{1g}),(B_{1g}), (E_g), respectively.These mode parameters obtained from experiments agree with the calculated data [12].

According to the data obtained from our measurements, we can observe the symmetry of oscillations regardless of the fluence of ion irradiation.

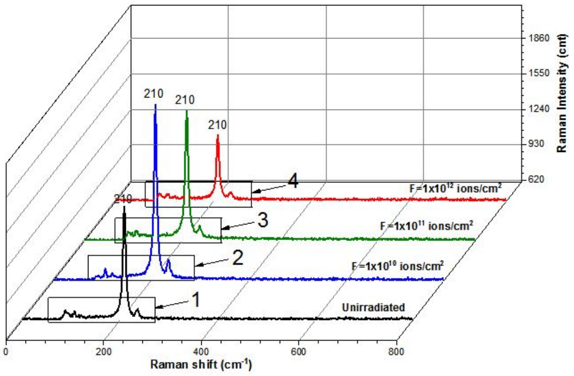

FIGURE 5 – Raman spectra of BaFBr crystals unirradiated and irradiated with xenon ions at room temperature as a function of fluence

According to [12-13], phonons with frequencies of 210 cm^{-1} and 236 cm^{-1} in BaFBr crystals can be attributed to the vibrational motions of fluorine ions. Phonons with a frequency of about 108 cm^{-1}, 121 cm^{-1} can be attributed to the vibrational motions of bromine ions [12-13]. It can be assumed that the degradation of the intensity of the Raman spectra as the fluence increases from 10^{10}-10^{12} ions/cm^2 is associated with the creation of F(F$^-$) centers. According to [22], there is a monotonic increase in the concentration of defect centers F(F$^-$) depending on the increase in X-ray irradiation at room temperature.The increase in the intensity of samples irradiated with $\Phi = 1 \times 10^{10}$ ion/cm^2 may be due to the overall increase in the number of defects in the crystal.

More detailed information on vibrational modes in BaFBr crystals irradiated with xenon ions is shown in Figure 6.

In [23], the possibility of local reduction of BaFBr ions with D_{4h} type point symmetry to impurity ions with C_{4v} spatial symmetry is mentioned in addition to the local destruction of the pure

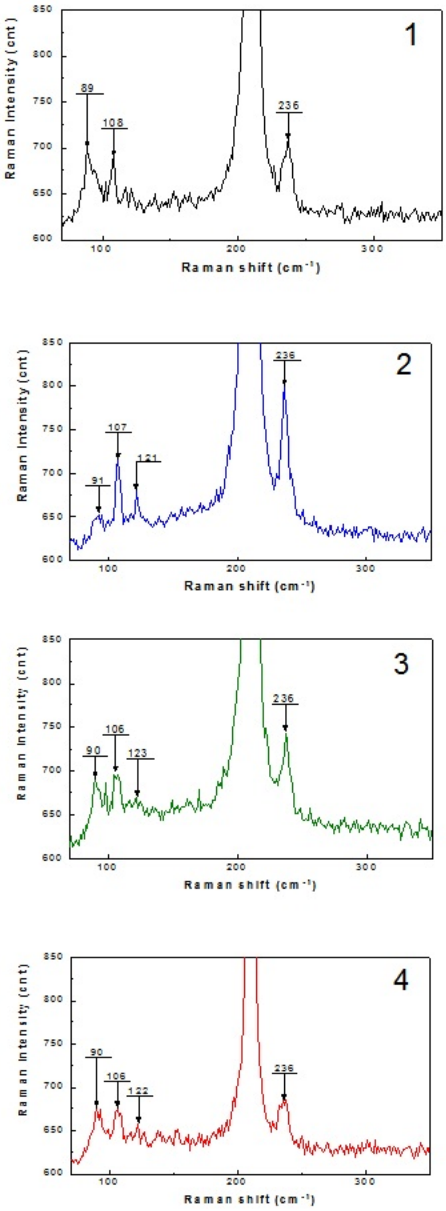

Figure 6 – Raman scattering spectra of BaFBr crystals unirradiated and irradiated with xenon ions at room temperature as a function of fluence: 1 – unirradiated sample; 2 – $\Phi = 1 \times 10^{10}$ ion/cm^2; 3 - $\Phi = 1 \times 10^{11}$ ion/cm^2, 4 - $\Phi = 11 \times 10^{12}$ ion/cm^2

translation group. F(Br$^-$) centers have a similar structure of point symmetry in [24-25], which may be consistent with the substitution mechanisms described in [14-15].

Conclusion. The model for the formation of bromine and oxygen centers under ion irradiation is comparable with the literature data. The luminescence band in the objects under study is due to oxygen centers, next to which there are bromine F centers. The decrease in the intensity of fluorine phonons in the Raman scattering spectra as the fluence increases, starting from $\Phi = 1 \times 10^{10}$ - 1×10^{12} ions/cm^2, can be associated with the creation of F(F$^{\breve{}}$) defect centers.

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Ксенон иондарымен BaFBr кристалдарының рамандық спектрлеріне сәулеленудің әсері

Аннотация. Осы жұмыста энергиясы E=229 МэВ-кетең ксенон иондарымен сәулеленген барий фторобромид кристалдарының флюенстері $\Phi=1\times10^{10}$ ион/см ^{2}, $\Phi=1\times10^{11}$ ион/см ^{2}, $\Phi=1\times10^{12}$ ион/см ^{2} дейінгі люминесценттік қасиеттері зерттелген. Фотолюминесценция спектрлері ақау қалыптастыру механизмдеріне қатысатын бақыланбайтын BaFBr қоспасының болуын көрсетеді. BaFBr монокристалдарының бетінен комбинациялық шашырау спектрлерінің көмегімен флюенске байланысты тордың тербеліс жиілігі анықталады. Фтор иондарына жататын жиіліктерде флюенс жоғарылаған сайын раман спектрлерінің интенсивтілігі төмендеулі байқалады.

Түйін сөздер: BaFBr, жылдам ауыр иондар, люминесценция, фотолюминесценция, раман спектрлері, ақаулар.

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Влияние облучения ионами ксенона на рамановские спектры кристаллов BaFBr

Аннотация. В работе исследованы люминесцентные свойства кристаллов фторобромида бария облученных ионами ксенона с энергией E=229 МэВ до флюенсов (1×10^{10} - 1×10^{12}) ион/см ^{2}. Спектры фотолюминесценции показывают наличие неконтролируемой примеси в BaFBr, которая участвует в механизмах дефектообразования. С помощью спектров комбинационного рассеяния с поверхности монокристаллов BaFBr определены частоты колебаний решетки в зависимости от флюенса. Прослеживается уменьшение интенсивности комбинационного рассеивания по мере увеличения флюенса на частотах принадлежащим ионам фтора.

Ключевые слова: кристалл BaFBr, быстрые тяжелые ионы, люминесценция, фотолюминесценция, спектры комбинационного рассеивания, дефекты.

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