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Deformation-stimulated luminescence of a KBr crystal matrix

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Radiative relaxations of electronic excitations - self-trapped excitons (STE) in regular lattice sites (intrinsic luminescence) and exciton-like formations (ELF) in the field of homologous cations (exciton-like luminescence). It has been found that the maximum effect of luminescence enhancement occurs upon uniaxial deformation along the <110> crystallographic direction (compared to <100>), which coincides with the direction of the self-trapped anion exciton (<110>) in the KBr crystal matrix. The exciton mechanism was estimated from the increase in the intensity of the intrinsic $\sigma(4,42eV)$ - and $\pi(2,3eV)$ - luminescences of STE, and the enhancement of luminescence intensity of near-single Na^{+} (2.85 eV), pair ions Na^{+} , Na^{+} (3.1 eV) and Na^{+} Pb^{++} (3.4 eV)- centers - recombination mechanism of radiative relaxation of electronic excitations.

Keywords: alkali halide crystal (AHC); X-ray luminescence; uniaxial deformation; exciton; electron-hole assembly; exciton-like luminescence

Introduction

Alkali halide crystals (AHC) systematize the fundamental issues of relaxation of electronic excitations, such as migration, self-trapping, radiative annihilation, as well as their decay into primary radiation defects [1–3]. As a result of many years of experimental research in AHC, the spectral characteristics and structures of radiation defects, as well as the nature of luminescence, both in regular lattice sites and in the field of various point defects and impurities, which create local deformations of the crystal matrix, have been reliably established [1–5].

In recent years, AHC relaxation processes have been actively studied using comprehensive [6, 7], uniaxial [8–12], and local [13–16] deformations created by light homologue cations [17]. In AHC under the action of ultrahigh pressures, completely new structures that differ from classical structures have been discovered and studied [7].

The study of relaxation processes of electronic excitations (EE) in AHC in the field of additional perturbing factors is very useful for the development of materials with desired physical characteristics and the search for optical materials that retain a wide spectral range of transparency, even under the influence of ionizing radiation.

In this paper, we study the deformation-stimulated effect leading to an increase in exciton-like luminescences due to an increase in the probability of self-trapping of free excitons in regular lattice sites and recombination assembly of electron-hole pairs in a local deformation field caused by light homologue cations in the KBr crystal matrix.

The aim of the study is to establish the effect of low-temperature (85 K) uniaxial deformation (<100>, <110>) on the luminescence of exciton and electron-hole recombination in KBr, KBr-Na and KBr-Pb,Na crystals.

Objects of research and technique of experiment

The KBr crystals with a low concentration of impurity sodium ions (0.01–1.0 ppm) grown at the final stage of zone melting on the basis of raw materials that underwent preliminary purification in a halogen stream were taken as a reference sample [8–10]. The concentrations of impurity ions are indicated in brackets, taking into account the embedding coefficient in the samples KBr-Na(100 ppm) and KBr-Pb(600 ppm), Na(100 ppm). KBr and KBr:Na crystals were grown at the Institute of Physics of the University of Tartu (Estonia) by the Stockbarger method in an evacuated ampoule. Doped KBr:Na crystals were grown on the basis of zone-purified KBr raw materials with additions of NaBr powder preliminarily dried in vacuum [2, 3, 8, 10]. The concentration of sodium ions in KBr:Na samples, taking into account the coefficient of incorporation of sodium into KBr lattice, was 100 ppm or 1000 ppm. The Pb^{++} concentration was also 600 ppm.

To study the X-ray luminescence (XRL) spectra of crystals, bremsstrahlung from a RUP-120 setup with a W anticathode operating in the 3 mA and 100 kV mode was used as ionizing radiation.

The deformation-stimulated luminescence of crystals was studied using a special cryostat [18], which allows to carry out uniaxial deformation in one of two directions (<100>, <110>) at 85 K and to register radiation both under constant stress and after its removal on the crystal.

The emission spectra of the crystals were scanned using an MSD-2 high-aperture monochromator and an H 8259-01 type photomultiplier (Hamamatsu) operating in the photon counting mode in the range of 6.0–1.5 eV, which were controlled by special programs SpectraScan and ThermoScan [19, 20].

Experimental results

For pure KBr crystals, the fundamental (intrinsic) radiation is the luminescence of a self-trapped exciton (STE) with maxima at 4.42 eV (σ- polarized) and 2.28 eV (π- polarized), recorded [21] at low temperatures (4.2 K).

With an increase in temperature, these luminescences are extinguished quickly enough, and as a result, at 80 K, the intensity of σ-luminescence decreases by 30 times, and π-luminescence decreases by 300 times. Nevertheless, highly sensitive luminescent equipment operating in photon counting mode with a high-power monochromator makes it possible to register both radiation bands at 80 K (Figure 1, curve 1) as a reference signal for studying the effect of deformation.

The experimental results presented in Figure 1 by recording X-ray luminescence (XRL) spectra show that with an increase in the degree of relative uniaxial deformation (<100>, <110>) in KBr crystals, an increase in the intensity of the intrinsic σ- and π-luminescences of STE and a new luminescence band with maxima at 3.6 eV.

At the same time, it was registered that the maximum increase in the radiation intensity at the same degree of uniaxial deformation ($\varepsilon \approx 1.0\%$) occurs in the case of action along the <110> crystallographic direction compared to <100> (Figure 1, compare curves 2 and 3).

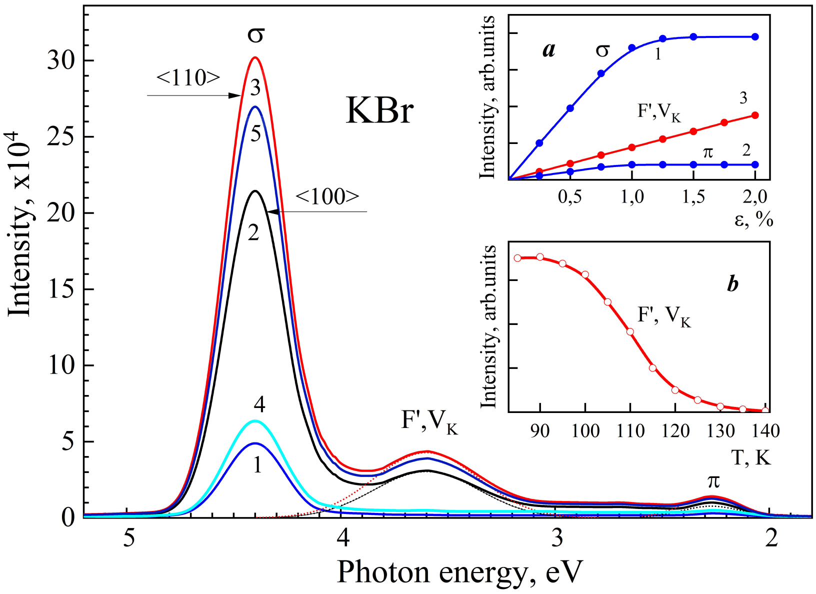

Figure 1. X-ray luminescence spectra of a zone-purified (benchmark) KBr crystal measured at 85 K before deformation (1) and under constant exposure at 85 K to uniaxial deformation ($\varepsilon \approx 1.0\%$) along the <100> (2) and <110> (3) crystallographic directions, as well as after removal (4) and during repeated deformation (5) to the initial level of compression. The elementary Gaussians obtained by decaying the XRL spectra are indicated by dotted lines. Inset: a – intensity dependence of σ (1)- and π (2)- STE luminescences (4.42 eV and 2.3 eV, respectively) and F', V_K (3) – luminescence (3.6 eV) on degree of uniaxial deformation; b – temperature dependence of F', V_K -luminescence (3.6 eV).

An analysis of the directional effect of low-temperature uniaxial deformation (85 K, <100>, <110>) on the XRL spectra of the KBr crystal shows that the intensities of σ- and π-luminescences increase linearly up to approximately 1% of the deformation (Figure 1, inset a, curves 1 and 2), after which they gradually pass to saturation, and the luminescence intensity at 3.6 eV increases linearly up to 2% deformation without saturation (Figure 1, inset a, curve 3), as evidenced by the accumulation of radiation defects, probably F' and V_K- centers. It can be seen from the dependence plot $I \sim f(\varepsilon)$ that the effect of a linear increase in the intensity of σ- and π-luminescences of STE occurs in the elastic part of the uniaxial deformation of the KBr crystal (Figure 1, inset a, curves 1 and 2). This is also evidenced by the experimental fact that after the removal of the voltage acting on the crystal, it is followed by an instantaneous weakening (up to 90%) of the intensity of the σ- and π-luminescence of the STE and the disappearance of the detected new luminescence band with a maximum at 3.6 eV (curve 4 in Figure 1). The intensities of the above luminescence bands are restored almost completely upon repeated application of uniaxial deformation to the crystal to the same extent as the previous time (curve 5 in Figure 1).

The temperature dependence of the X-ray spectra shows that luminescence annealing at 3.6 eV occurs in the temperature range (110–120 K) corresponding to the maximum destruction of F'-centers in KBr crystals (inset b, Figure 1). This means that the luminescence with a maximum at 3.6 eV is the result of tunneling processes between the ground states, primarily the F' and V_K-centers [22, 23]. The analysis of the energy scheme of the tunneling transition between the main levels of the F' and V_K centers was carried out by analogy with the band scheme considered in [24] for tunneling transitions Tl^0 - V_K and Ag^0 - V_K in KCl-Tl and KCl-Ag crystals, respectively.

Another rare feature of permanently acting uniaxial elastic deformation was experimentally discovered to detect uncontrolled light impurities of homologue cations (for example, Na^+, Li^+), which is demonstrated for a KBr single-crystal matrix (Figure 2).

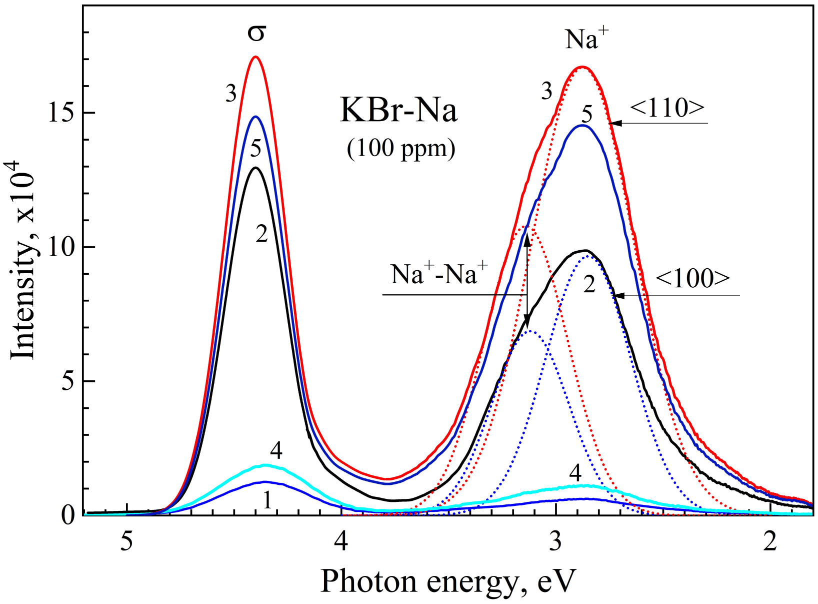

Figure 2. X-ray luminescence spectra of a KBr-Na(100ppm) crystal measured at 85 K before deformation (1) and under constant exposure at 85 K to uniaxial deformation ($\varepsilon \approx 1.0\%$) along the <100> (2) and <110> (3) crystallographic directions, as well as after removal (4) and when re-deformed (5) to the original level of compression. The elementary Gaussians obtained by decaying the XRL spectra are indicated by dotted lines.

With an increase in the degree of uniaxial deformation at 85 K, luminescence of an exciton-like formation appears with maxima at 2.8 eV and 3.1 eV, which are characteristic of single Na^+ and pair Na^+, Na^+ ions, respectively (Figure 2, curves 2 and 3). The figure shows that these bands in the absence of deformation practically do not appear as structural luminescence bands (Figure 1, curve 1). It should be noted that in KBr-Na crystals, the maximum enhancement of the luminescence intensity at the same degree of uniaxial deformation ($\varepsilon \approx 1.0\%$) occurs with deformation along the <110> crystallographic direction than <100> (Figure 2, compare curves 2 and 3).

Dependence of the intensity of σ- and π-luminescences of STE (4.42 eV and 2.3 eV, respectively) and luminescence near single Na^+ (2.85 eV) and pair Na^+, Na^+ (3.1 eV) on the degree of uniaxial deformation shows that in KBr-Na crystals, up to approximately 1.0%, the linear dependence of the intensity for all luminescence bands is preserved (Figure 3).

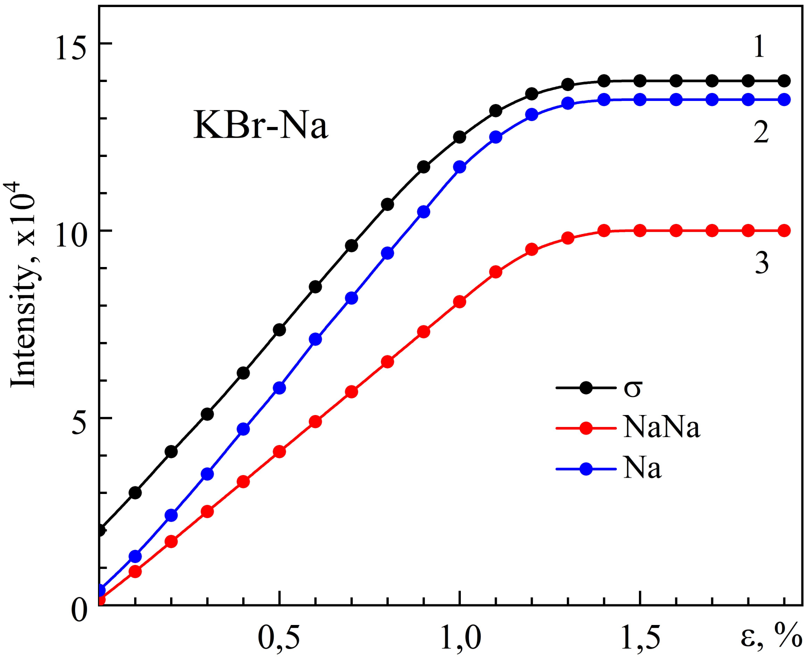

Figure 3. Dependence of the intensity σ - STE luminescence (1) and exciton-like luminescence near Na^+ (2) or Na^+ - Na^+ (3) on the degree of uniaxial deformation – $I = f(\varepsilon)$ at 85 K.

The dependence $I = f(\varepsilon)$ shows (Figure 3) that the detected effect of radiation intensity amplification occurs in the elastic part of the uniaxial deformation, which is also established for other similar crystals [8–12].

The linear dependence of the luminescence intensity on the degree of uniaxial deformation – $I = f(\varepsilon)$, according to the classical representation of the fluidity of materials, characterizes the elastic part of the deformation. To obtain an exhaustive answer that the increase in the luminescence intensity depends on the degree of elastic deformation of the KBr-Na crystal, unique experiments were carried out related to the removal and repeated application of uniaxial deformation at low temperature (85 K) without sealing the cryostat. After removing the voltage acting on the KBr-Na crystal, the intensity of the above luminescence bands decreases almost to the level in the absence of deformation (Figure 2, compare curves 1 and 4), as in the case of KBr (Figure 1, compare curves 1 and 4). Subsequently, with repeated deformation (Figure 2, curve 5) to the initial level of compression ($\varepsilon \approx 1.0\%$) up to 90%, the intensity values are restored, both σ-luminescence (4.42 eV), and luminescence of near-single Na^+ (2.85 eV) and pair ions Na^+, Na^+ (3.1 eV) (Figure 2, curve 5), which is unambiguously associated with elastic deformation.

Studies of the X-ray spectra of KBr-Na crystals with different sodium ion concentrations at the same degree of uniaxial elastic deformation at 85 K show that the intensity of σ-luminescence (4.42 eV) does not depend on the sodium concentration (Figure 4, curve 1), as in the reference KBr crystal, has a constant value, and the ELF luminescence intensities near single Na^+ (2.85 eV) and pair Na^+, Na^+ (3.1 eV) ions obey the growth curve characteristic of single and pair impurity luminescence centers (Figure 4, curves 2 and 3).

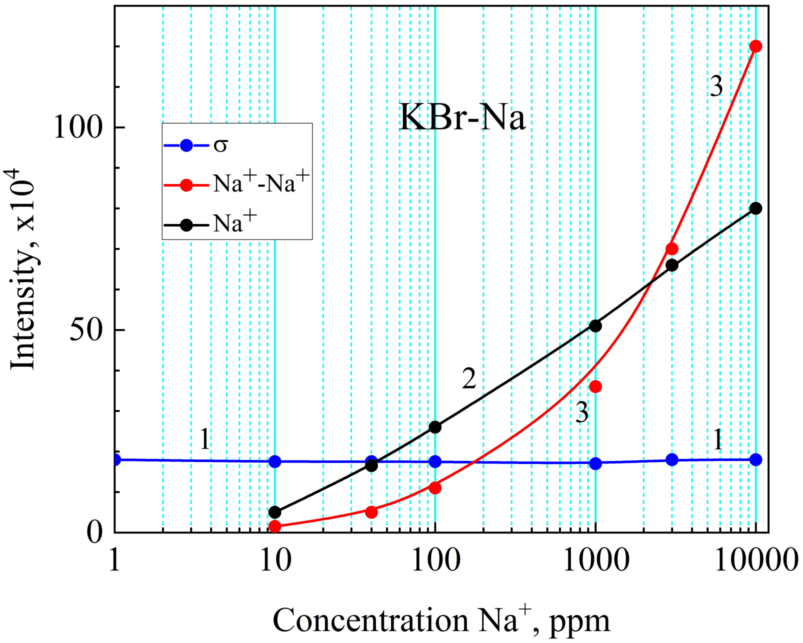

Figure 4. Dependence of the intensity (1) σ-STE luminescence (4.42 eV) and ELF luminescence near single (2) Na^+ (2.85 eV) and pair (3) Na^+ - Na^+ ions (3.1 eV) at the same degree ($\varepsilon \approx 1.0\%$) of uniaxial deformation (<100>) applied at 85 K.

In KBr-Na crystals, the effect of a correlated increase in the intensity of σ (4.42 eV),- and π (2.3 eV)- STE luminescences with an increase in the degree of deformation (Figure 2, curves 2-4) can be quite explained by analogy with the KI-Tl [13], with an increase in the probability of self-trapping of free excitons in regular lattice sites. We note that the effect of increasing the intensity of the intrinsic luminescence of KBr and KBr-Na crystals up to 1% deformation (Figures 1 and 2) can be associated with an increase in the probability of self-trapping of free excitons in regular lattice sites [13].

In KBr-Na crystals, the correlation effect of the increase in the intensity of exciton-like luminescence near Na^+ (2.85 eV) was also found, which cannot be explained by the exciton mechanism of energy transfer to impurities [13]. Most likely, we are dealing with a recombination mechanism for the formation of an exciton-like formation in the field of an impurity sodium (Na^+) ion, as is the case in KCl-Na crystals [8–10].

To detect the recombination assembly of electron-hole pairs with the formation of ELF in the impurity field, a special experiment was carried out with KBr-Pb(600ppm),Na(100ppm) crystals to study the effect of uniaxial elastic deformation simultaneously on two emission bands − 2.8 eV and 3.44 eV. A low sodium concentration (100 ppm) provides an elementary emission band (2.8 eV) characteristic of single sodium ions.

The uniqueness of the choice of the KBr-Pb(600ppm),Na(100ppm) crystal lies in the fact that Pb^{++} is an optically active impurity with absorption, excitation, and emission spectra, while the Na^{+} impurity is activated only under conditions of uniaxial elastic deformation as a probe for the assembly of electron-hole couple next to you. Figure 5 shows the X-ray spectrum of the KBr-Pb(600 ppm)Na(100 pmm) crystal at 85 K in order to show the dynamics of the influence of the increasing degree of uniaxial deformation on the STE emission intensity with maxima at 4.42 eV (σ), 2.3 eV (π), Pb^{++} (3.44 eV) and exciton-like formation (ELF) in the Na^{+} field (2.85 eV). In the absence of deformation (curve 1), the dominant emission bands are σ (4.42 eV) and Pb^{++} (3.44 eV) luminescence, less intense π-luminescence (2.3 eV), and "traces" of Na^{+} ions (2.85 eV) are also noticeable. With an increase in the degree of uniaxial deformation (up to 1.0%), the intensities of the above emission bands in KBr-Pb(600 ppm)Na(100 ppm) increase (Figure 5, curves 2-4), as is the case for the KBr-Na crystal (see Figure 2, curves 2-4).

Of particular note is the experimental fact that with an increase in uniaxial strain (<100>), along with other emission bands, the ELF luminescence intensities in the field of Na^{+} (2.85 eV) and Pb^{++} (3.44 eV) - centers, are indicative of the effective energy transfer of electronic excitations (free excitons or electron-hole pairs) to Na^{+} and Pb^{++} impurities. It is known from the literature data [11–13] that increasing uniaxial deformation leads to an increase in the probability of self-trapping of free excitons in regular lattice sites, after which the intensities of the matrix luminescence increase, like the luminescence of a self-trapped exciton (STE).

At the same time, it was experimentally established [13] that with an increase in the degree of uniaxial elastic deformation in KI-Tl crystals, the intensity of Tl^{+}-luminescence (2.85 eV) is redistributed in favor of the intensity σ (4.1 eV),- and π (3.3 eV)- STE luminescences by reducing the free path of the exciton to self-trapping.

In KBr-Pb,Na crystals, the effect of a correlated increase in the intensity of the intrinsic σ (4.42 eV),- and π (2.3 eV)- luminescence of STE with an increase in the degree of deformation (Figure 5, curves 2-4) can be quite explained by an increase in the probability of self-trapping of free excitons in regular lattice sites by analogy with the KI-Tl crystal [13].

Figure 5 shows that in the KBr-Pb,Na crystals there is also an increase in the intensity of all luminescence bands (4.42 eV, 3.44 eV, 2.85 eV and 2.3 eV) depending on the increasing degree of deformation and has a similar character, as in Figure 3, consisting of two branches: linear − characterizing the elastic part of the deformation; constant (independent of deformation) − characterizing the plastic part of the deformation.

For KBr-Pb,Na crystals, the absorption spectrum of the Pb^{++} center with a

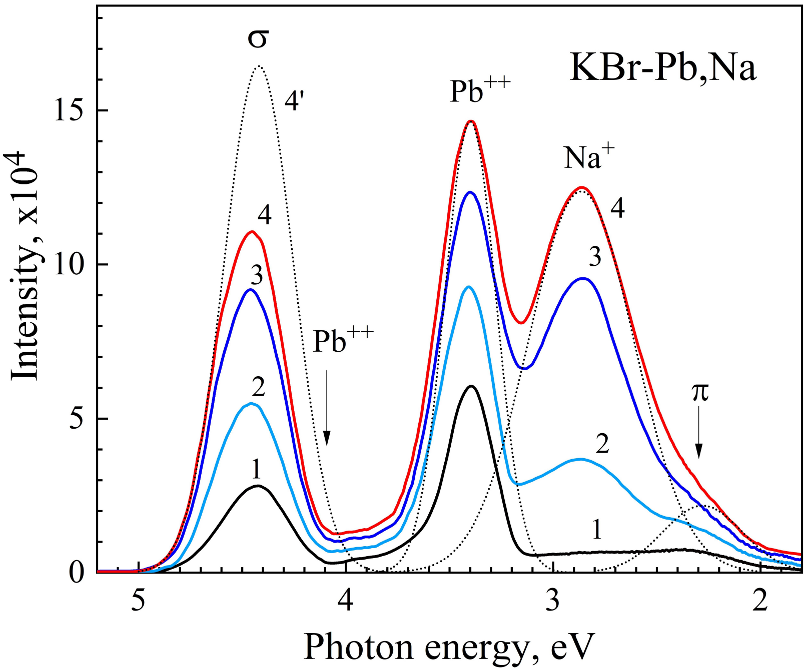

Figure 5. XRL spectra XRL spectra of a KBr-Pb(600 ppm),Na(100 ppm) crystal, measured at 85 K before deformation (1) and under constant exposure to an increasing value of uniaxial deformation (2 − 0.2%, 3 − 0.4%, 4 − 0.6%) along the <100> crystallographic direction at 85 K. $4'$ - spectral position of σ - luminescence in the absence of reabsorption of Pb^{++} - center.

maximum at 4.15 eV was recorded by absorption spectroscopy on the basis of the Evolution-300 spectrophotometer at 85 K, and then the emission spectra with maximum at 3.4 eV at photoexcitation of 4.1 eV, characteristic of Pb^{++} centers. Due to the absorption spectrum of the Pb^{++} center, the reabsorption effect is observed in the X-ray luminescence spectra of the KBr-Pb,Na crystal, due to which the emission spectrum is distorted, where it is indicated by the down arrow in Figure 5.

The true spectral position of the σ-STE luminescence in the KBr-Pb,Na crystal, taking into account the half-width (0.34 eV), maximum (4.42 eV) and intensity under elastic deformation, according to Figure 4, curve 1, is represented by a dotted band (curve $4^{'}$).

Thus, in KBr-Na and KBr-Pb,Na crystals, it was clearly experimentally demonstrated that with an increase in the degree of low-temperature uniaxial elastic deformation, the intensities of luminescence of an impurity nature increase, associated with the luminescence of the Pb^{++} center (3.4 eV) and ELF in the field of Na^{+} (2.8 eV), which testify to the efficiency of energy transfer of electronic excitations to Pb^{++}, Na^{+} impurities.

The observed effect of enhancement of the luminescence intensity of an impurity nature opens up the possibility of using for the interpretation of its electron-hole assembly mechanism with the formation of an exciton-like formation near cation impurities by analogy found in [8–10].

Discussion

In the KBr crystal matrix, both in the reference and doped with cationic impurities (Na^+, Pb^{++}), an extremely saturated increase in the intensity of its intrinsic luminescence (especially σ-luminescence) is observed at the same degree of uniaxial low-temperature (85 K) deformation ($\varepsilon \approx 1.0\%$) along the <100> and <110> crystallographic directions. This experimental result clearly shows that the probability of exciton self-trapping at regular lattice sites in a uniaxial strain field does not depend on the presence of a local perturbation from point defects (Na^+, Pb^{++}).

The experimentally found effects of luminescence enhancement by the exciton or recombination mechanism with the formation of an exciton-like formation in the field of cation impurities are best realized with uniaxial deformation in the <110> direction than in the <100> direction. The model representation is shown in Figure 6. The reason for the increase in the intensity of σ- and π-luminescences of STE with an increase in the degree of deformation in the KBr crystal matrix is apparently associated with an increase in the probability of self-trapping of free excitons in regular lattice sites due to a reduction in the free path of excitons to self-trapping, as was found for crystals KI [12–13].

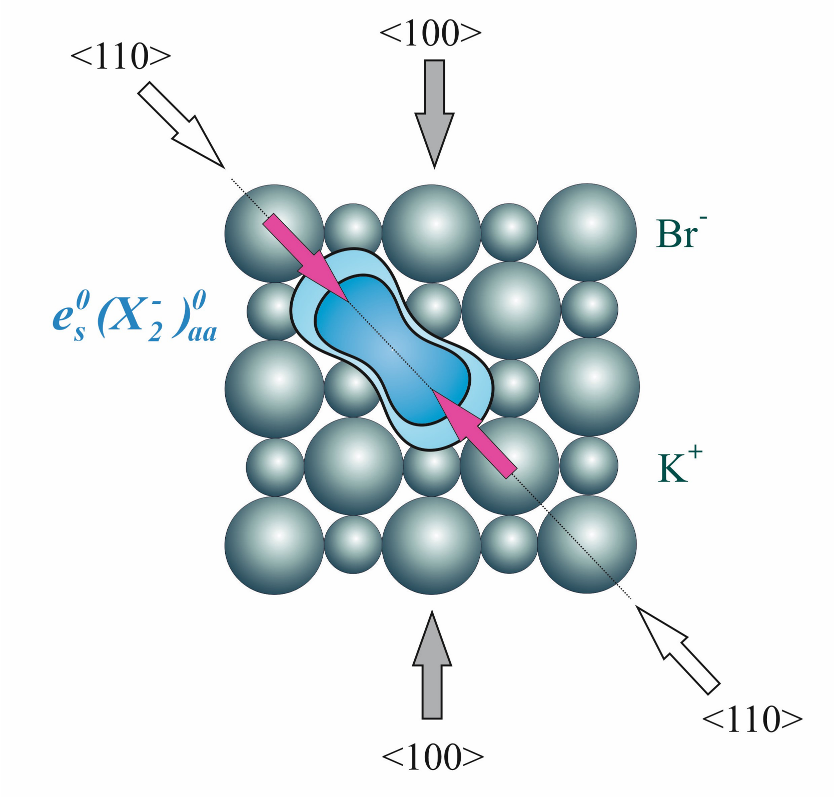

Figure 6. Model representation of the interaction of uniaxial strain along the <110> and <100> directions of the KBr crystal matrix, taking into account the STE structure at regular lattice sites. The outer arrows of the figure indicate the directions of uniaxial compression of the crystal along the <110> and <100> directions, carried out using a special cryostat [18]. The inner pink-colored arrows in the figure indicate the directions of anion glide during uniaxial deformation.

In KI-Tl crystals, a direct effect of low-temperature uniaxial deformation on the exciton mean free path before self-trapping has been experimentally demonstrated. At the same time, it was established [13] that with an increase in the degree of compression, a redistribution of intensities occurs: the attenuation of Tl^{+} - radiation (2.85 eV) and the simultaneous increase in the intensity of the intrinsic σ (4.42 eV),- and π (2.3 eV)- luminescences of the STE. The effect is interpreted as a direct effect of elastic deformation on the shortening of the free path of an exciton to self-trapping at a regular lattice site.

So, according to the exciton mechanism of energy transfer of electronic excitations in alkali halide crystals, the intensity of intrinsic luminescence should rise with an increase in the degree of elastic deformation ($I \sim f(\varepsilon)$) , while the intensity of luminescence of an impurity nature, on the contrary, should decrease significantly due to the deterioration of the energy transfer of free excitons to impurities associated with an increase in the probability of their self-trapping at regular lattice sites.

In KBr-Na, KBr-Pb,Na crystals, the correlation effect of the increase in the intensity of Pb^{++} (3.44 eV)- and exciton-like luminescence near Na^{+} (2.85 eV) was also found, which cannot be explained by the exciton mechanism of energy transfer to impurities, leading to a decrease in the luminescence intensity impurity nature with an increase in the degree of uniaxial deformation [10–13]. The enhancement of the luminescence of an impurity nature in KBr-Na, KBr-Pb,Na crystals with an increase in the degree of elastic deformation can be explained by the mechanism of formation of an exciton-like formation by recombination of electron-hole pairs in the field of cation impurities (Pb, Na), as in the case of KCl-Na crystals [8–10].

In fact, during low-temperature deformation (85 K) in the <100> direction from the end of the AHC crystal, the actual lattice compression occurs along the <110> crystallographic direction, which corresponds to the STE direction in the KBr crystal matrix. In other words, the mechanical external load will act in the <100> direction of the crystal, and the effective compression of the lattice will occur along the length of the STE, since the best slip will correspond to the <110> direction (Figure 6). If we transfer the mechanical external load along the <110> direction, which coincides along the length of the STE and ELF structure, into the field of the cationic impurity, then it is reasonable to expect the maximum effect of luminescence enhancement upon low-temperature deformation of AHC.

In this work, using low-temperature (85 K) uniaxial deformation (<100>, <110>), for the first time, it was possible to fix the exciton and electron-hole recombination mechanisms of radiative relaxation of electronic excitations, both in regular lattice sites and in the field of cation impurities, respectively.

Conclusion

For KBr and KBr-Na crystals, by recording X-ray luminescence spectra under the influence of low-temperature (85 K) uniaxial deformation (<100>, <110>), it was established:

1. The maximum luminescence enhancement effect was found upon uniaxial deformation along the <110> crystallographic direction (compared to <100>),

which coincides with the direction of the self-trapped anionic exciton (<110>) in the KBr crystal matrix, at which the best sliding along the anion lattice row occurs.

2. Exciton mechanism of radiative relaxation of electronic excitations - by increasing the intensity of intrinsic σ (4.42 eV) - and π (2.3 eV) - luminescences of STE (KBr and KBr-Na).

3. Recombination mechanism of radiative relaxation of an exciton-like formation formed by assembling electron-hole pairs in the field of a sodium ion - by increasing the luminescence intensity near single Na^+ (2.85 eV) and pair Na^+, Na^+ (3.1 eV) ions (KBr-Na).

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