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**Paper title:** *RECOMBINATION RADIATION IN  
BaSO<sup>4</sup>*

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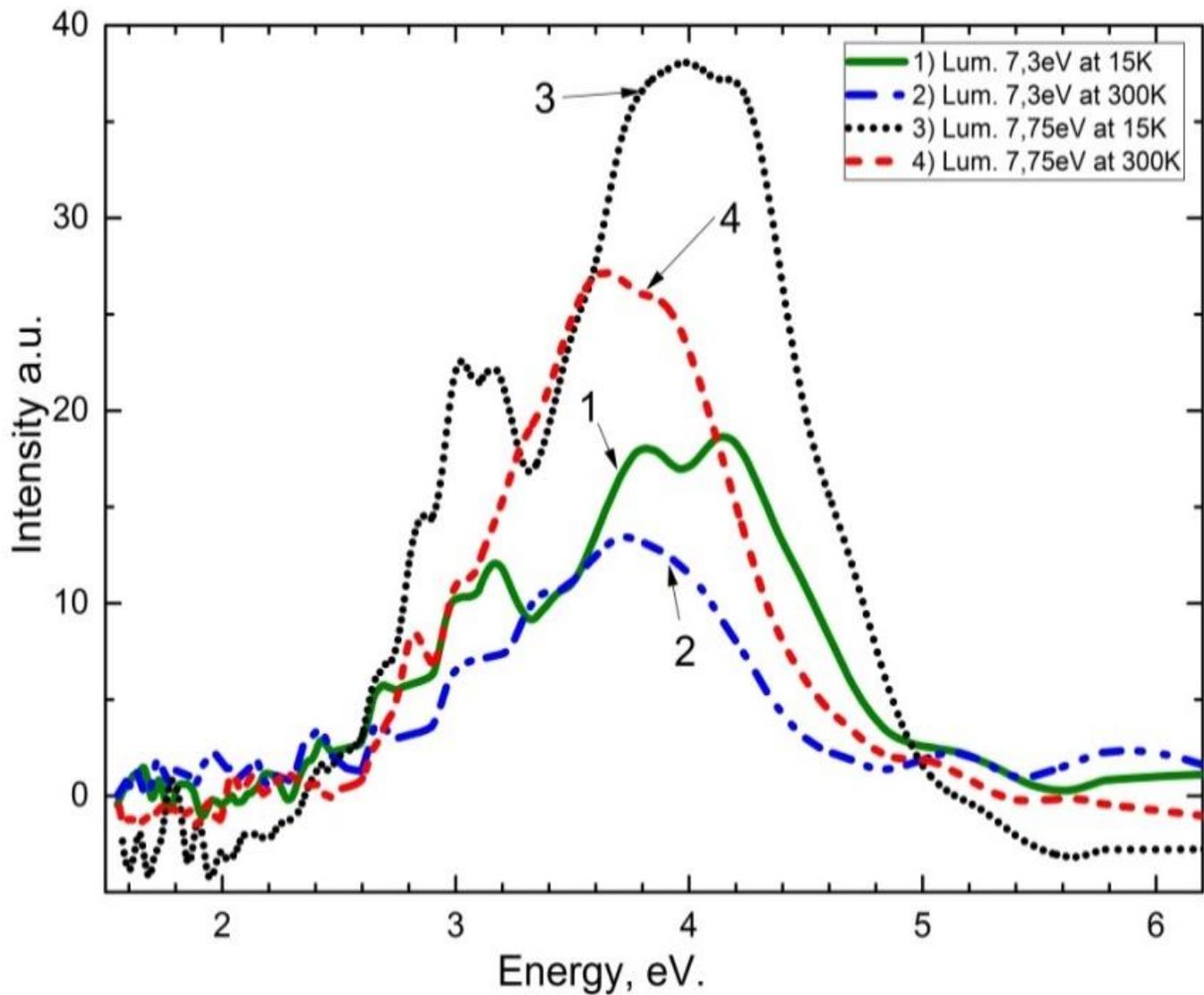
# Introduction

- Alkaline earth metal sulfates with impurities are used as thermoluminescent dosimeters. In the irradiated pure  $\text{BaSO}_4$  crystals in the temperature range from 15 K to 300 K, intrinsic luminescence appears. In the studies of the authors of [1] it was shown that in the X-ray luminescence of natural crystals  $\text{BaSO}_4$ ,  $\text{CaSO}_4$ ,  $\text{SrSO}_4$ ,  $\text{PbSO}_4$ , the emission band at 360 nm is detected. The authors associated the emission bands at 360 nm with the recombination of electrons with localized holes  $\text{SO}_4^-$  during the irradiation of crystals.
- It was assumed that  $\text{BaSO}_4$  crystals as well as  $\text{CaSO}_4$  crystals will be used as thermoluminescent dosimeters. In this regard,  $\text{BaSO}_4$  crystals were mainly used to create dosimetric peaks of TSL. The authors of [2] showed that in the irradiated  $\text{BaSO}_4 - \text{Eu}^{(2+)}$  crystal, the light sum in the dosimetric peak of TSL at 513 K is 3-4 times greater than in the commercial  $\text{CaSO}_4 - \text{Dy}^{(3+)}$  crystal. The appearance of the TSL peak is associated with the recombination formation of the decomposition of the reaction  $\text{Eu}^{(2+)} \leftrightarrow \text{Eu}^{(3+)}$ . Emission of  $\text{Eu}^{(2+)}$  - centers is detected at 373 nm, and the induced emission of  $\text{Eu}^{(3+)}$  is at 612 nm. The light sum under the TSL peak should be proportional to the emission intensity of the  $\text{Eu}^{(2+)}$  impurity

- The authors' articles [3] compared the intensity or the light sum under the TSL band at 227 ° C in a pure BaSO<sub>4</sub> crystal activated with impurities Mn<sup>2+</sup> and Cu<sup>+</sup>. It was experimentally shown that the intensity of the TSL peak at 227 ° C in BaSO<sub>4</sub> - Cu is 9 times stronger than the similar TSL peak in pure BaSO<sub>4</sub>. It is assumed that an increase in the light sums under the TSL band at 227 ° C is associated with the formation of electron-hole trap centers based on Cu<sup>+</sup> impurity in irradiated BaSO<sub>4</sub> - Cu. The authors of [4] assumed that the introduction of isovalent impurities into the BaSO<sub>4</sub> - Eu<sup>2+</sup> crystal creates anion and cationic vacancies that stimulate the creation of defects that increase the light sum under the band of the TSL dosimetric peak. In this paper, we study the nature of intrinsic short-wave radiation excited by photons with an energy of 6.2–12.4 eV at 15 K and 300 K; also the formation of electron-hole trap centers in irradiated BaSO<sub>4</sub> crystals. The intrinsic recombination radiation of BaSO<sub>4</sub> in the form of a powder with a purity of 99.99% was studied upon excitation by photons with an energy of 6.2–12.4 eV in a wide spectral region, in the temperature range 15–300 K.

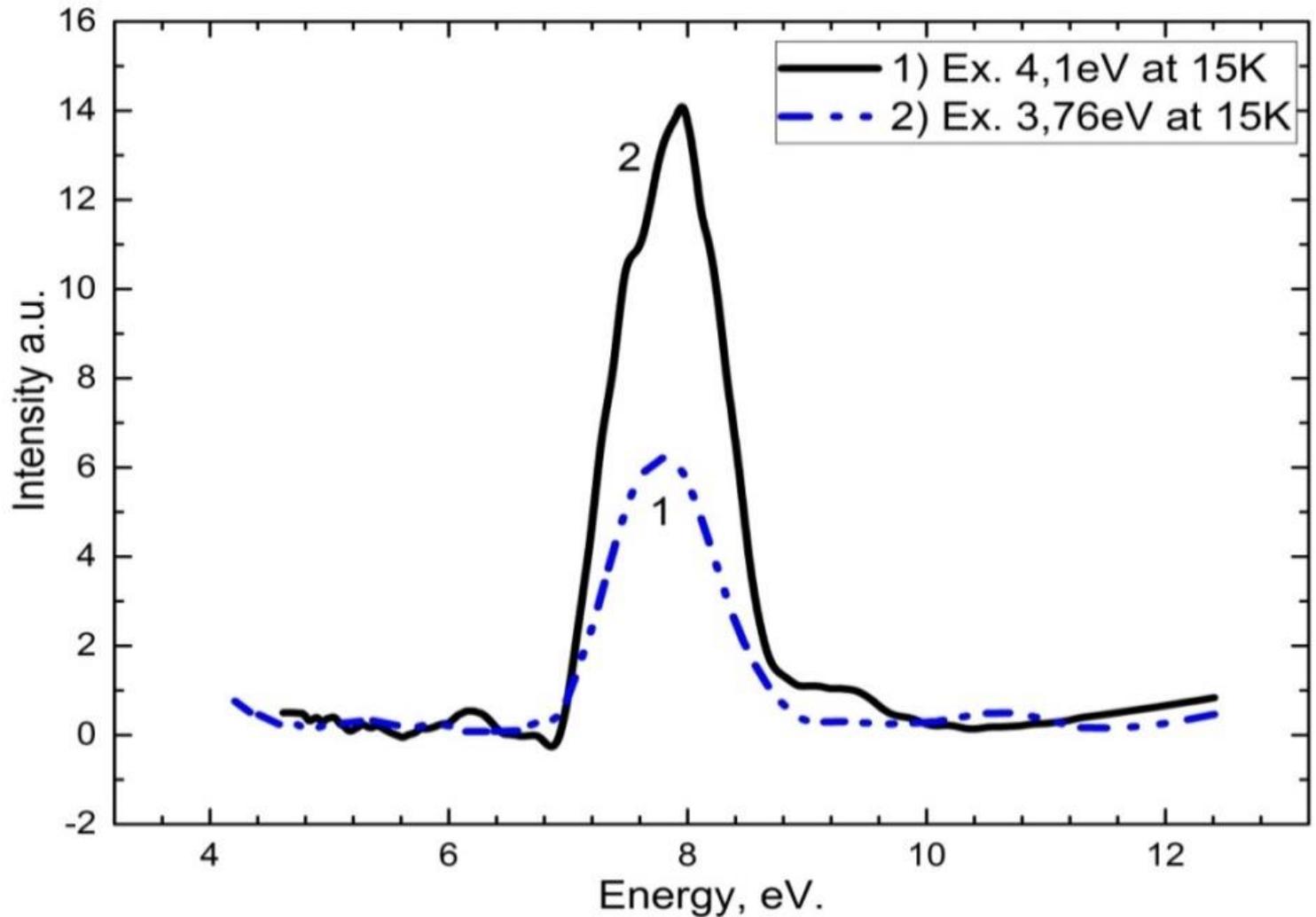
# THE RESULTS OF THE EXPERIMENTS AND THEIR DISCUSSION

- **Figure 1** shows the emission spectrum of BaSO<sub>4</sub> powder excited by photons with an energy of 7.3 eV at a temperature of 15 K (curve 1) and at a temperature of 300 K (curve 2). Figure 1 shows that the main emission bands of 4.1 eV, 3.8 eV and 3.64 eV appear, and long-wave radiation appears in the range of 3.0-3.2 eV, 2.7-2.8 eV and 2.3-2.4 eV at 15 and 300 K
- In figure 1 shows the emission spectrum of BaSO<sub>4</sub> excited by photons with an energy of 7.75 eV at 15 and 300 K (curves 3.4). The same emission bands of irradiated BaSO<sub>4</sub> appear at a photon energy of 7.3 eV. The intensity of all emission bands upon excitation by photons with an energy of 7.75 eV is 2 times more intense than the radiation arising upon excitation by photons with an energy of 7.3 eV and individual bands are clearly distinguished. In fig. 2. The excitation spectrum for the emission band of 3.75 eV (curve 1) and for the emission band at 4.13 eV (curve 2) are presented.



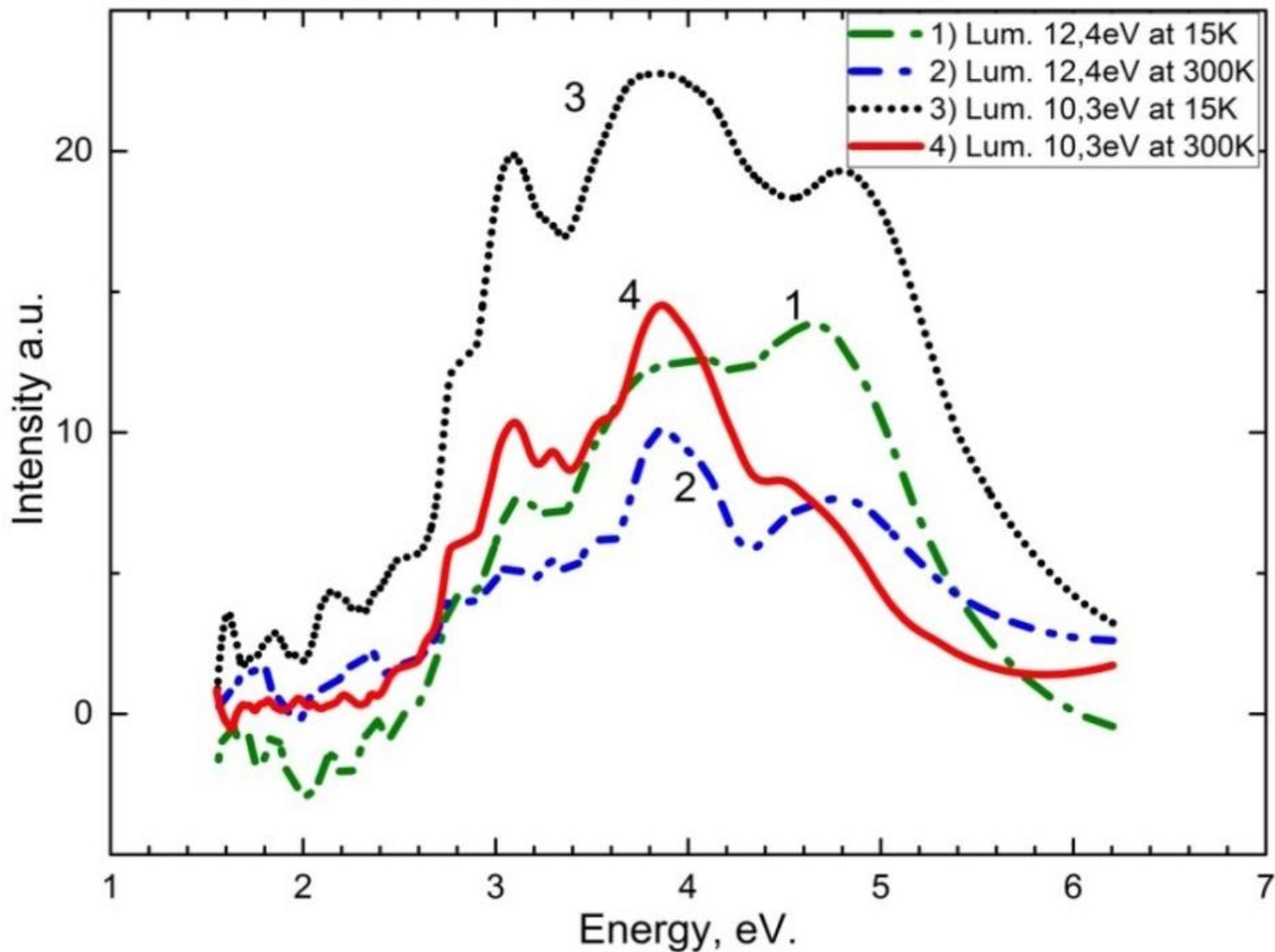
**Figure 1.** Luminescence of BaSO<sub>4</sub> excited by photons with an energy of 7.3 eV at 1) 15 K 2) 300 K and 7.75 eV 3) 15 K 4) 300 K

- **Figure 2** shows that the emission band at 3.75 eV is excited at a photon energy of 7.9 eV at 15 K. The emission band at 4.14 eV is excited at a photon energy of 8.0 eV (Fig. 2, curve 2). The excitation intensity for the emission band at 7.9 eV is 2 times greater than the excitation intensity for the emission band at 3.75 eV.



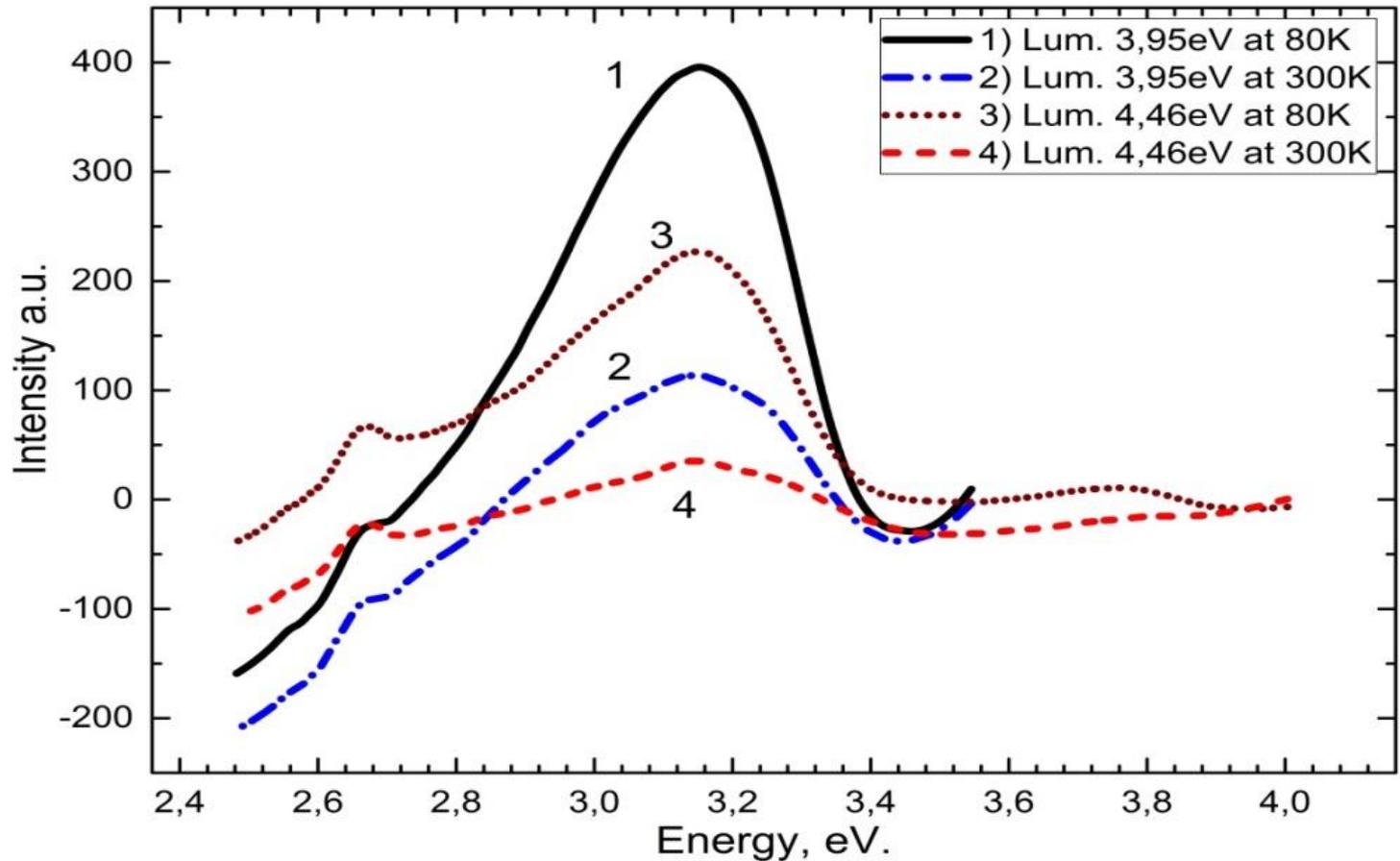
**Figure 2.** The excitation spectrum for the emission band of 4.1 eV and 3.75 eV at temperature 15 K

- In **Figure 3**. The emission spectrum of  $\text{BaSO}_4$  is presented upon excitation by photons with energies of 10.3 eV and 12.4 eV at 15 K and 300 K. From Fig. 3. It is seen that short-wavelength emission bands appear at 5.0 eV, 4.77 eV, 3.8 eV, and 3.64 eV, and long-wavelength emission bands appear at 3.0–3.1 eV, 2.6–2.75 eV, 2.3–2.4 eV, at 15 K and 300 K. Short-wavelength emission bands at 5.0 eV appear more efficiently when excited by high-energy photons with an energy of 10.3 eV. The remaining emission bands of 4.77 eV, 4.1 eV, 3.8 eV, 3.64 eV appear the same when excited by photons with energies of 12.4 eV and 10.3 eV. At room temperature of 300 K, the intensity of all emission bands decreases by a factor of 2–2.5.



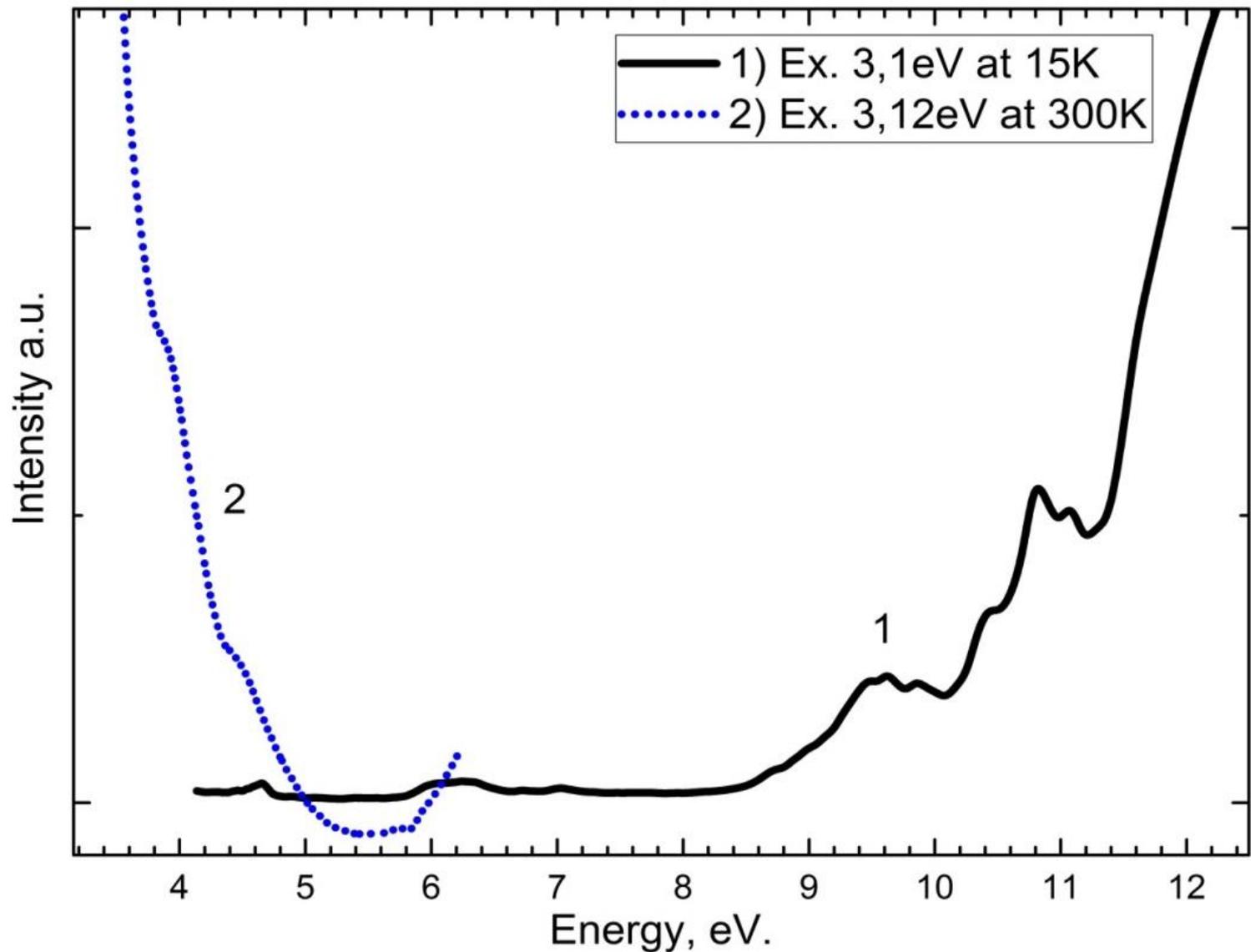
**Figure 3.** Luminescence BaSO<sub>4</sub> excitation by photons with energies of 10.3 eV and 12.4 eV at 15 K and 300 K.

In **Figure 4.** (curves 1,2) the BaSO<sub>4</sub> emission spectrum is presented upon excitation by 3.95 eV photons at 80 K and 300 K (curves 1,2). In this figure (curves 3,4) shows the BaSO<sub>4</sub> emission spectrum when excited by photons with an energy of 4.46 eV at 80 K and 300 K. From Figure 4 (curves 1,2) shows that the same emission bands appear with maxima at 3.14 eV and 2.6 eV at 80 K and 300 K.



**Figure 4.** Luminescence of BaSO<sub>4</sub> at energies of 3.95 eV at 80 K and 300 K (curves 1.2).

**In Figure 5** (curve 1) shows the creation spectrum for recombination radiation at 3.1 eV and at 15 K. Figure 5 shows that the recombination radiation band of 3.1 eV is formed at photon energies of 6.2 eV, 9.54-9.9 eV, and 10.5-10.8 eV, where BaSO<sub>4</sub> free electron-hole pairs. Figure 5 (curve 2) also shows the excitation spectrum for the recombination radiation band of 3.14 eV at 300 K. Figure 5 (curve 2) shows that the recombination radiation of 3.14 eV appears at the center of trap at the trap centers at photon energies of 3.9 eV and 4.46 eV at 300 K. Thus, upon excitation by photons with energies of 6.2 eV, 7.3 eV and 7.75 eV, the main short-wavelength emission bands appear at 3.64 eV, 3.7-3.8 eV and 4.1 eV in BaSO<sub>4</sub> powder samples. These emission bands appear more efficiently at temperatures of 15 K. Measurement of the excitation spectrum of these emission bands shows that these bands are more efficiently created at photon energies of 6.2 eV and 7.8 eV. When BaSO<sub>4</sub> is excited by high-energy photons at 10.3 eV and 12.4 eV at temperatures of 15 K and 300 K, in addition to the known emission bands of 3.64 eV, 3.7 eV, 3.8 eV and 4.1 eV, new emission bands appear at 4.77 eV and 5.9 eV. The appearance of short-wavelength emission bands of 5 eV and 4.77 eV was discovered by us for the CaSO<sub>4</sub> crystal in our latest publications [5]. These short-wavelength emission bands at 5.0 eV, 4.77 eV were associated by recombination of electrons from the conduction band with localized holes formed upon the transition of an electron from the third subband of the valence band upon excitation by high-energy photons with energies of 10.3 eV and 12.4 eV at 15 K in CaSO<sub>4</sub>. The study of long-wavelength emission bands at 3.0–3.14 eV, 2.6–2.7 eV, and 2.3–2.4 eV was attributed by many authors to induced defects without experimental proof. For the BaSO<sub>4</sub> crystal, the nature of these bands was studied by us for the first time.



**Figure 5.** The excitation spectrum of 3.1 eV peaks at temperatures of 15 K and 300 K

**In fig. 5** (curve 1), it was shown that the recombination radiation band at 3.1 eV is produced at photon energies of 6.2 eV, 9.54–9.9 eV, and 10.3–10.8 eV, where free electron – hole pairs are generated in the fundamental region of the matrix. The electron-hole trap centers are formed upon capture of free electrons by the anionic complexes  $\text{SO}_4^{2-}$ , and the hole is localized in the form of  $\text{SO}_4^-$  - radical.

In accordance with the aim of this work, we studied the nature of intrinsic short-wave radiation and the laws governing the formation of electron-hole trap centers upon irradiation with high-energy photons with energies from 6.2 eV to 12.4 eV. As a result of an experimental study, the intrinsic short-wave radiation was measured upon excitation by photons:

Own short-wave radiation was measured upon excitation by photons with energies of 7.3 eV and 7.75 eV at 15 K and 300 K. The measurement results showed that at 15 K an intense emission peak appears at 4.1 eV, 3.8 eV, 3.64 eV and all long-wavelength emission bands at 3.0 -3.2 eV, 2.7-2.8 eV, 2.3-2.4 eV. At a temperature of 300 K, less intense emission peaks appear at 3.8 eV and 3.64 eV, as well as all long-wavelength emissions. Measurement of the excitation spectra for the emission bands of 3.76 eV and 4.1 eV showed that the emission bands at 4.1 eV are 2 times more efficiently excited at a photon energy of 8 eV. It was also shown that these emission bands are less efficiently excited at photon energies of 6.2 eV and 8.5–9.0 eV.

# Conclusion

- Based on the obtained experimental results on measuring the excitation spectra of long-wavelength recombination radiation at 3.14 eV, it is assumed that the present radiation is excited in the transparency region of BaSO<sub>4</sub> at 3.9 eV and 4.43 eV. It is assumed that the excitation spectrum for the emission band at 3.14 eV and 2.65 eV is inside the center at the electron-hole trap centers. <sup>LiKSO<sub>4</sub></sup> Electron trap centers are formed upon electron trap by the anionic complex  $[(SO_4)]^{(2-)}$  and hole capture centers are associated with a localized hole in the form of a  $[(SO_4)]^-$  radical. Electronic  $[(SO_4)]^{(3-)}$  centers and  $[(SO_4)]^-$ -capture centers in BaSO<sub>4</sub> are formed.

# REFERENCES

- 1 M.L. Gaft, L.V. Bershov, A.R. Krasnaya and V.T. Yoskolka Phys Chem Minerals (1985). 255-260.
- 2 O. Annalakshmi, M.T. Jose and Modhusooddan Radiation Protection Dosimetry (2012). Pp. 1-7.
- 3 J. Manam, S. Das, Journal of Physics and Chemistry of solids 70 (2009). 379-387.
- 4 R. Sangeetha Rani, Arunachalam Lashannan journal of Luminescence 124 (2016). 63-69.
- 5 T.N. Nurakhmetov, Zh.M. Salikhodzha, A. Akilbekov, A.M. Zhunusbekov, D.A.Z. Kainarbay, B.M. Sadykova, D.Kh. Daurenbekov, B.M., K.B. Zhanglysov Radiation Measurements, Vol. 125, p. 19-24, June 2019
- 6 V. Ramoswamy, R.M. Vimolathithan, V. Ponnusamy, M.T. Jose 134 (2013). 791-797.
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