A low-temperature vacuum ultraviolet luminescence spectroscopy study of $K_2Al_2B_2O_7$ crystals

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The paper presents the results of a study of the electronic excitation dynamics, fast luminescence and energy transport in the undoped $K_2Al_2B_2O_7$ (KABO) single crystals. Recent developments in medical, lithographic, spectroscopic, and optical storage applications require all-solid-state laser sources in the ultraviolet (UV) spectrum, especially at the wavelength of 266 and 193 nm. The KABO, a newly developed nonlinear optical borate crystal, is considered to be one of the best candidates for this purpose due to its wide range of optical transparency down to 180 nm and very suitable structural characteristics for UV harmonic generation [1, 2].

The present study was carried out by the means of the low-temperature luminescence VUV spectroscopy with the time-resolution. Time-resolved and steady-state photoluminescence (PL) spectra in the energy range from 1.2 eV to 6.2 eV, PL excitation spectra and reflectivity in the broad energy range from 3.7 eV to 21 eV, and the decay kinetics of PL were measured at 8 K for these crystals at the SUPERLUMI experimental station of HASYLAB using synchrotron radiation (SR). The time-resolved spectra were recorded within a time window (length Δt) correlated with the arrival of SR pulses (delayed δt). In the present experiments the delay and length were $\delta t_1 = 0.5$ ns, $\Delta t_1 = 2.3$ ns for the first window (fast component), and $\delta t_2 = 14$ ns, $\Delta t_2 = 58$ ns for the second window (slow components). The 0.3 m ARC Spectra Pro-300i monochromator and either R6358P (Hamamatsu) photomultiplier, or CCD camera were used as a registration system. The PL excitation spectra were corrected. All the examined single crystals were grown at the Institute of Geology and Mineralogy of SB RAS (Novosibirsk, Russia).

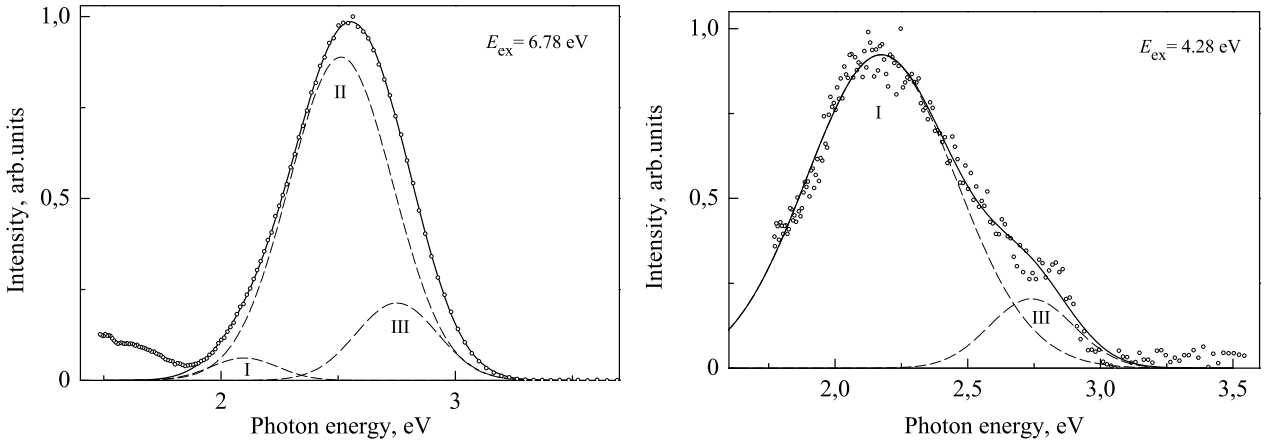

Figure 1: The PL emission spectra of KABO at 8 K under selective photoexcitation at E_{ex}

Figure 1 shows the luminescence spectra of KABO at 8 K under selective photoexcitation with E_{ex}=6.78 and 4.28 eV. The PL emission spectra consists of three elementary Gauss-shape bands at 2.15 (I), 2.50 (II) and 2.75 eV (III). The band II can be excited only in the fundamental optical absorption band of KABO, and tentatively it can be assigned to the intrinsic emission of KABO. In contrast to that the bands I and III can be excited at the photon energies from the optical transparency band of KABO, and they should be assigned to photoluminescence of defects. There was revealed an additional PL band at 1.46 eV, which is more pronounced on recording with CCD

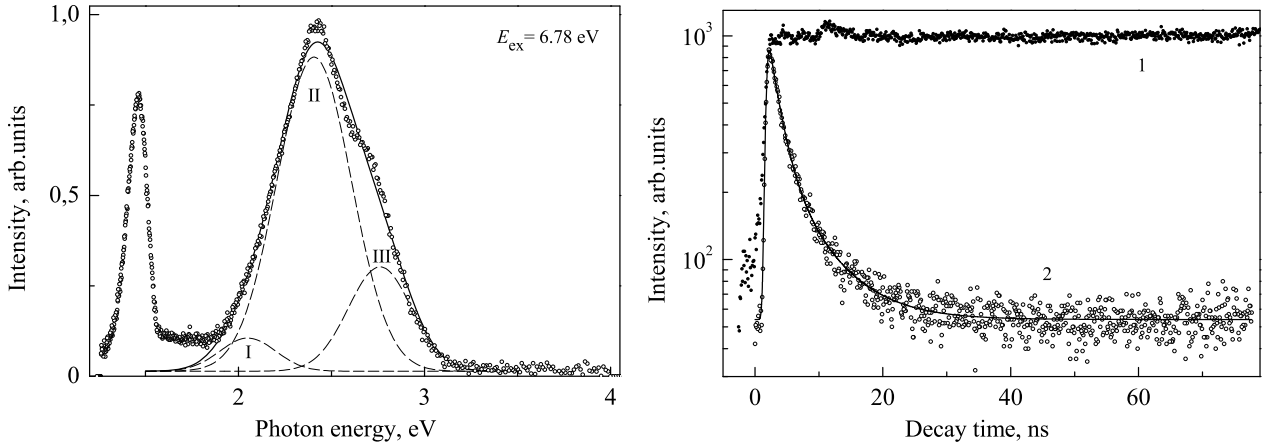

Figure 2: The PL emission spectra recorded with CCD camera for KABO at 8 K (left). The 2.3 eV PL decay kinetics of KABO at 8 K and E_{ex}=6.8 (1) and 4.1 eV (2) (right)

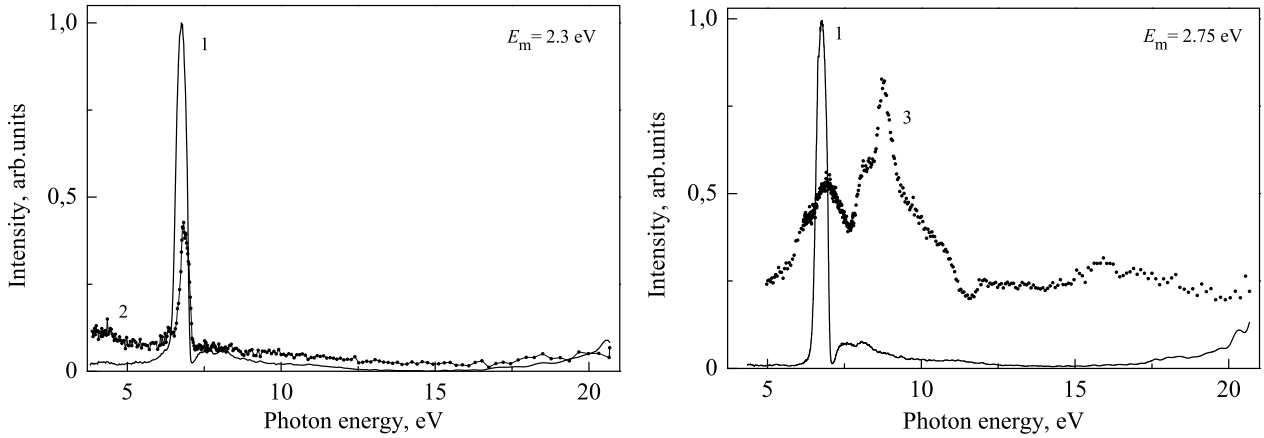

Figure 3: The PL excitation (1,2) and reflection (3) spectra of KABO at 8 K measured for the time-integrated (1) and fast (2) components

camera, Fig. 2. Figure 2 shows also a time-response of the 2.3 eV PL under selective photoexcitation at two different energies. In doing so, the slow PL decay kinetics with a time-constant from the micro- and millisecond time-range, represented at our measurements as a pedestal, occurs under photoexcitation at 6.8 eV. In contrary to that, an excitation at 4.1 eV leads to the fast two-exponential decay kinetics with the time-constants of 1.8 and 6.5 ns and the ratio of their intensities of 100:30. This luminescence can be excited in the whole range of the investigated host absorption (Fig. 3), starting from the edge at 6.6 eV where the exiting radiation penetrates as deep as 0.5 mm into the crystal. Similar broad-band intrinsic emissions in other alkali metal borates LiB_3O_5, $Li_2B_4O_7$ at 3.6–3.8 eV have been attributed to the radiative annihilation of self-trapped exciton, which arose from the self-trapping of molecular exciton in these crystals. Analogously, we assign tentatively the 2.5 eV emission in the KABO to the radiative annihilation of self-trapped exciton resulting from the self-trapping of molecular type exciton created by 6.88 eV photons. An origin of the 2.15 and 2.75 eV emission band is not yet understood, but it was revealed also in the KABO phosphorescence spectrum at 8 K, indicating that it is due to the electron recombination.

References

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