Combined luminescence and X-ray emission study of self-trapped excitons in Al$_2$BeO$_4$ crystal

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In oxides at low temperatures the process of formation of anion self-trapped excitons (ASTE) and their radiative decay is known as ASTE luminescence. At the same time, there are only solitary reports about probable formation of cation self-trapped excitons (CSTE) in wide-gap compounds, revealed by resonant inelastic X-ray scattering method [1,2]. ASTEs we discovered in Al$_2$BeO$_4$ crystals in [3]. Our work is devoted to combined research of properties of ASTE and CSTE using methods of luminescence and X-ray emission spectroscopy at the selective XUV excitation in Al$_2$BeO$_4$ crystal.

Experimental work was carried out in HASYLAB (Hamburg) and in MAX-lab (Lund). At temperatures 10 and 300 K, the luminescence spectra (2 – 8 eV), the luminescence excitation spectra (5-20 eV, 50-600 eV) was recorded at the SUPERLUMI station (beam-line I) or using synchrotron radiation (SR) from the BW3 beam-line (HASYLAB, DESY). The 0.3 m ARC SpectraPro-308i monochromator equipped with the R6358P (Hamamatsu) photomultiplier was used to analyse luminescence in the 2-6 eV region at the SUPERLUMI station. The 2 m primary monochromator (Al gratings) were applied in measurements of the luminescence excitation spectra (LES) over the 5-20 eV energy range. A typical spectral resolution was 0.32 nm. At the BW3 the SR from the undulator was further monochromatized by a Zeiss SX700 monochromator. The luminescence spectra (LS) were measured by a 0.4 m vacuum monochromator (Seya-Namioka scheme) equipped with a microchannel plate-photomultiplier (MCP 1645, Hamamatsu). The LES were corrected for the equal number of the exciting photons but the LS are presented as they are measured. The measurements of the X-ray fluorescence in the vicinity of the Al 2p and Be 1s photoabsorption edges were performed at the beam-line 1511-3 at MAX-lab (Lund University) with energy resolution better than 0.1 eV.

Accordingly [3] LS is characterized by three bands of intrinsic luminescence. Emission bands with the maximums at 3.5 and 4.1 eV dominate in LS at the excitation near the fundamental absorption edge (figure 1). The intensity of 5.2 eV luminescence increases after the excitation in the range of inner-shell transitions. The value of Stokes shifts can be used to estimate the energy loss during self-trapping process. Both minimums corresponding to 1s Be and 2p Al inner-shell transitions are clearly expressed in LES presented in figure 2. So, we could suppose the both cation’s sublattice take part in the exciton’s self-trapping process.

A wide long-wave shoulder of the elastic scattering peak in the resonant X-ray emission spectra of wide-gap compounds was firstly reported in [1]. It was supposed that such profile of the observed spectra connects with lattice’s distortion and in fact reflects formation of CSTE. In the case of Al$_2$BeO$_4$ crystal wide long-wave shoulders of the elastic scattering peak were observed in the resonant X-ray emission spectra measured in the range of both 1s Be and 2p Al inner-shell transitions (figure 3 and 4). It should be noted two experimental facts. Firstly, the widths of long-wave shoulders differ in the cases of inner-shell transitions of different cations. Secondary, the widths of long-wave shoulders in principal coincide with Stokes shifts values. Joint analysis of UV-luminescence and resonantly excited X-ray emission spectra leads to assumption that self-trapping of both ASTE and CSTE results in formation of short-living defects at the same local structural units of crystal lattice. The value of energy loss taking for lattice distortion differs for different structural units and depends on tendency of this structural units to the short-term distortion.
Figure 1: Luminescence spectra (1-3) and excitation luminescence spectrum of 4.0 eV emission in Al$_2$BeO$_4$ crystal, T=10K. Energy of excitation: 9.5 eV for curve 1, 130 eV for curve 2 and 525 eV for curve 3.

Figure 2: Excitation spectrum of 5.2 eV luminescence in Al$_2$BeO$_4$ crystal at T=10K.

Figure 3: Resonantly excited in the range of L-edge absorption of aluminum ion X-ray fluorescence spectra at T=300K.

Figure 4: Resonantly excited in the range of K-edge absorption of beryllium ion X-ray fluorescence spectra at T=300K.

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References