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Inner-shell excitation of intrinsic luminescence and resonantly excited X-ray fluorescence at Be 1s edge in oriented BeO crystals

V. Pustovarov[a,*], V. Ivanov[a], M. Kirm[b], A. Kikas[b], K. Kooser[b], T. Käämbre[b], A. Kruzhalov[a], E. Zinin[c]

[a]Experimental Physics Department, Ural State Technical University, Mira Street 19, 620002, Ekaterinburg, Russia
[b]Institute of Physics, University of Tartu, Riia 142, Tartu 51014, Estonia
[c]Budker Institute of Nuclear Physics SB RAS, Novosibirsk, Russia

Abstract

The results of studies of the time-resolved luminescence of self-trapped excitons (STE) with different multiplicity as well as the X-ray emission in oriented BeO crystals under excitation in the vicinity of the 1 s Be photoabsorption edge are presented. The branching of the electronic excitations relaxation depends strongly on the crystal orientation after the inner-shell excitation. The common features between the processes of the STE luminescence and resonantly excited X-ray fluorescence have been discussed.

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Keywords: Exciton; Time-resolved luminescence; Inner-shell excitation

1. Introduction

The anisotropy of optical constants and luminescence properties are typical of crystals with low lattice symmetry. In particular, one can observe that luminescence bands with different polarization degree according to the selected optical axis under any kind of excitation, can reveal different paths in the relaxation of electronic excitation. We have detected both the mentioned effects in BeO crystals. In our earlier work [1], we found in luminescence spectra (LS) the broad emission bands peaked at 4.9 and 6.8 eV with different polarization degrees. These bands were attributed to the radiative decay of self-trapped excitons (STE) of two kinds having different configuration of a hole nucleus (a small-radius polaron of the O^--ion type in a local fragment of crystal lattice). Our recent investigations, using VUV-excitation in the range of 8–35 eV [2], revealed that the excited states of STE are characterized by a multiplicity of different factors depending on the excitation energy and sample orientation. In the present work, we investigate the radiation relaxation of core excitations generated by soft X-ray synchrotron radiation (SR) in oriented BeO crystals, and we compare the STE luminescence yield and resonantly excited X-ray fluorescence.

2. Experimental technique and methods

Prism-shaped BeO crystals with natural growth faces were investigated. The time-resolved luminescence spectra (LS) (2.5–10.6 eV), the luminescence excitation spectra (50–200 eV), as well as the decay kinetics of luminescence at $T = 10$ and 295 K, were recorded using SR from the BW3 beam-line (HASYLAB, DESY, Hamburg). SR from the undulator was monochromatized by a Zeiss SX700 monochromator. The LS were measured by a 0.4 m vacuum monochromator equipped with a microchannel plate-photomultiplier. The time resolution of the detection system was ~250 ps (FWHM). Simultaneously with the time-integrated LS, the spectra in the two time window, delayed relative to the exciting SR-pulse, were registered: the fast component: a delay time $\delta t_1 = 0.2$ ns, a length of

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*Corresponding author. Tel.: +7 343 3754711; fax: +7 343 3743884.
E-mail address: pva@dpt.ustu.ru (V. Pustovarov).

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time window $\Delta t_1 = 3.4$ ns; the slow component: $\delta t_2 = 34$ ns, $\Delta t_2 = 118$ ns.The measurements of the X-ray fluorescence in the vicinity of the Be 1s photoabsorption edge were performed at the beam-line 1511-3 at MAX-lab (Lund University, Sweden) with energy resolution better than 0.1 eV.

3. Experimental results

3.1. Exciton luminescence

The measured LS and luminescence excitation spectra (LES), as well as time decay, are shown in Figs. 1–3. We found that the spectral position of the minimum, corresponding to the 1s Be absorption edge in the 4.9 eV LES, sensitively depends on the orientation of the crystal C-axis and **E**-vector of SR, Fig. 1. Consequently, the primary electronic excitations of different energy (probably the core excitons) differ in their symmetry, and we wanted to find the difference in the relaxation parts of such excitations as well. Our study revealed a new fast decay component of the UV-luminescence of BeO crystals in the orientation of $E\perp C$ (perpendicular) (Fig. 2). This component is absent in another orientation. The LS of the new fast component ($\tau = 0.7$ ns) has a maximum at 5.2 eV (Fig. 3). This component was observed under photon excitation at the Be 1s absorption edge only (Fig. 2). Earlier [2], we found the fast luminescence peaked at 4.0 eV at $E\parallel C$ (parallel) under excitation in the energy range near the fundamental absorption edge. This luminescence was interpreted as the radiative decay of singlet type of STE. In this work, we discovered that new fast luminescence excited in alternative orientation. These experimental results point to the manifestations of singlet STE, but in another geometrical configuration of the hole component.

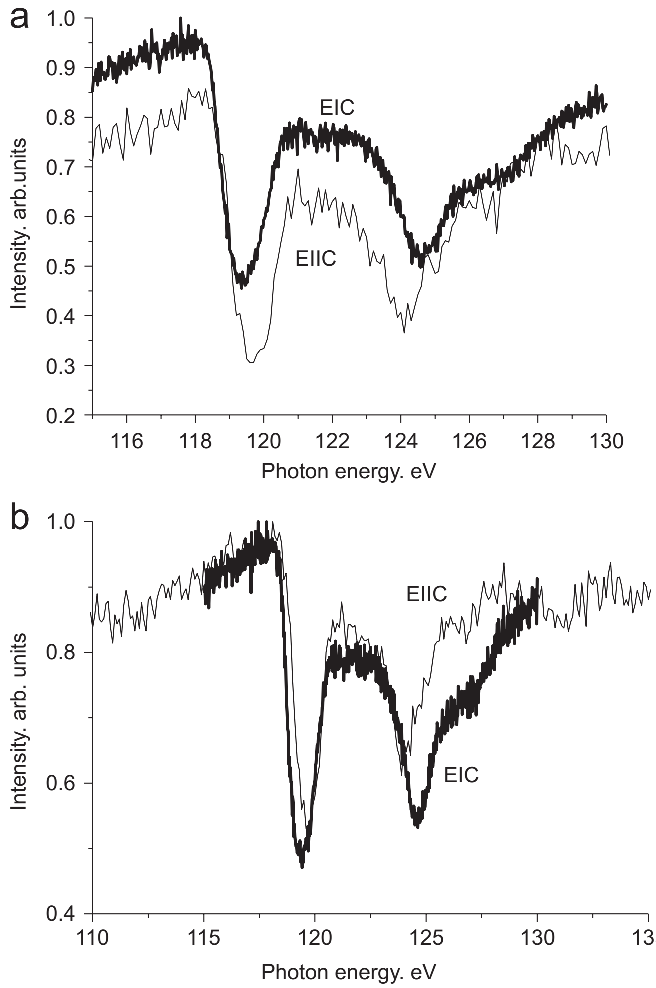

Fig. 1. Luminescence excitation spectra of 4.9 (a) and 6.8 eV (b) emission band in BeO crystals for the different orientation at $T = 8$ K.

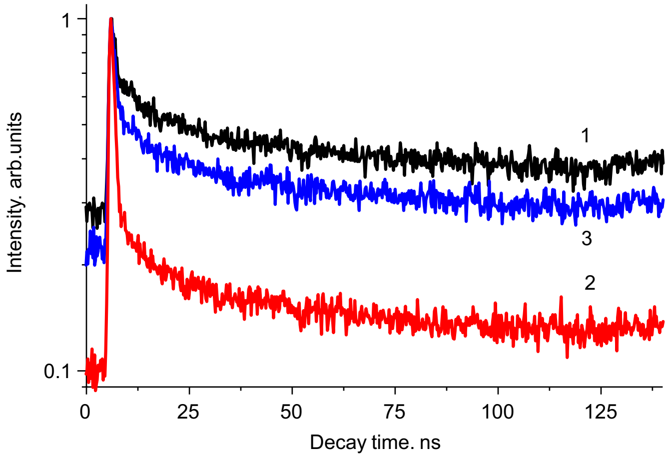

Fig. 2. The luminescence decay kinetics of BeO crystals for $E\perp C$ orientation at $T = 8$ K: $E_{\mathrm{emis}} = 5.16$ eV, $E_{\mathrm{exc}} = 117$ or 150 eV (1), 119.5 or 124.5 eV (2) and 121 or 135 eV (3). The decomposition of curve 2 into the exponential components resulted in decay times 0.7, 10, ~120 ns.

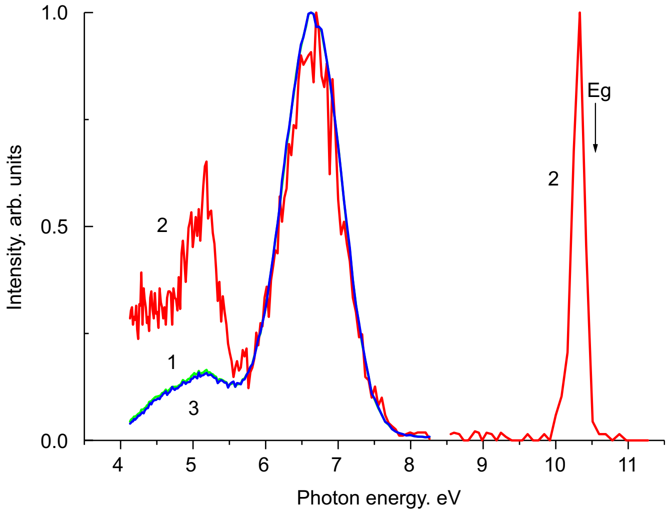

Fig. 3. Normalized luminescence spectra of BeO crystals for E⊥C orientation at $T = 8$ K. $E_{\mathrm{exc}} = 119.5$ eV: time-integrated (1), fast (2), slow (3) time windows.

Another narrow band of fast luminescence (10.32 eV, $\tau \leqslant 0.3$ ns) is observed in the region of fundamental absorption edge (Fig. 3). This emission is excited at the