Optical properties of lead carbonate

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Fast nanosecond fluorescence of lead compounds combined with their high density make them attractive materials for scintillator applications which require high counting rates. In [1,2] lead carbonate was proposed as a new fast, heavy scintillator. Here we present the results of the measurements performed at the SUPERLUMI station of the reflectivity, luminescence emission, excitation and kinetics of single crystal $PbCO_3$ as well as of crystal powders in the VUV spectral range.

Fig. 1 presents the luminescence spectra of the single crystal of $PbCO_3$ at 6 K for the excitation energies 5 and 20 eV. The spectra consist of two broad bands peaking at ~320 nm and 440 nm (the spectra were not corrected for the apparatus function) with their relative intensities depending on the excitation energy. The maximum efficiency of the excitation of the 320 nm band was at ~ 5 eV. At RT the high energy band was observed only in the single crystal but not in powder samples when excited in the fundamental absorption range. With temperature increase from 10 to 30 K the yield of the 320 nm luminescence dropped by 2 orders of magnitude. A decrease in the intensity of the 450 nm band was observed as steps at 30 K, 80 K and 150 K. The changes in luminescence kinetics with temperature are shown in the right hand pannel of Fig.1. The presented curves were measured at the excitation energy of 5 eV, at 20 eV the tendencies were similar with a slightly higher contribution of the slow microsecond component. At 13 K a fast component with τ ~1ns and a slow one of the microsecond order were observed in the decay kinetics of both luminescence bands. At 30 K the yield of the slow component of the 320 nm band substantially decreased while the kinetics of the 450 nm band remained the same. Starting with 150 K a component with τ ~ 30 ns was manifested in the decay of the 320 nm band while the amplitude of the fastest one decreased. Kinetics of the 450 nm band manifested temperature quenching resulting in the reduction of the yield and acceleration of the decay which proceeded up to the room temperature. With further temperature elevation the dominant decay component of the 320 nm band remained unchanged and its characteristic time was ~ 30 ns. Comparison of the decay kinetics of two luminescence bands indicate that there is energy transfer from the 320 nm band to the 450 nm one. These results differ from those of [2] where only the lower energy band was observed in the luminescence spectrum. In addition it had a dip at 450 nm coinsiding with the absorption maximum reported in [1]. This discrepency can be caused by different geometry of the experiment (the luminescence light in [2] was detected in the transmission mode) as well as different excitation energy (511 KeV) and hence different penetration depth (increasing the probability of the reabsorption of the 320 nm emission).

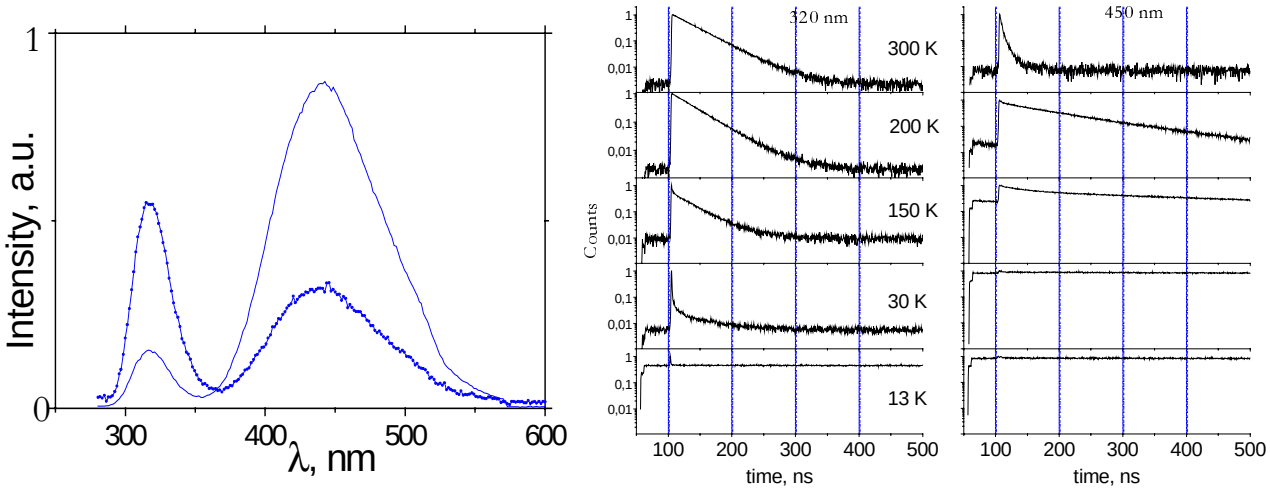

Figure 1: PbCO3 luminescence spectra at 6 K, excitation energies 20 eV (solid curve) and 5 eV (dotted curve), and temperature dependence of the luminescence decay kinetics at 320 and 450 nm excited by 5 eV photons.

Luminescence excitation spectra of 320 and 450 nm emission measured at 6 K are presented in Fig. 2 together with the reflectivity spectrum. The position of the first reflectivity peak allows to suggest that the energy bandgap of $PbCO_3$ is larger than 5 eV. The main excitation peak of the 320 nm luminescence at 4.8 eV at 6 K coincides with the Urbach tale of the fundamental absorption. By 7 eV its yield drops almost to zero and the onset of the increase at ~ 12 eV can be considered to be due to photon multiplication. The profile of the excitation curve is similar to that of the exciton-type luminescence. The excitation spectrum of the 450 nm band at low energies reproduces the course of the 320 nm yield in confirmation of the energy transfer between corresponding luminescence centres discussed above. In the region of fundamental absorption the yield of 450 nm luminescence remains non-zero indicating efficient energy transfer by separated electrons and holes. Comparison between the excitation and emission spectra of single crystal and powder samples allows to suggest that 450 nm band can be of defect origin, however, crystals of improved optical quality are required to establish the origin of the luminescence centres.

In conclusion we would like to note that such different crystals as $PbWO_4$,$PbSO_4$ and $PbCO_3$ all manifest fast nanosecond emission the intensity of which increases by 2 orders of magnitude with cooling down to LHeT. Comparative analysis of the optical properties of these system of different structure and degree of covalency should assist in establishing the origin of luminescence in complex lead oxide compounds.

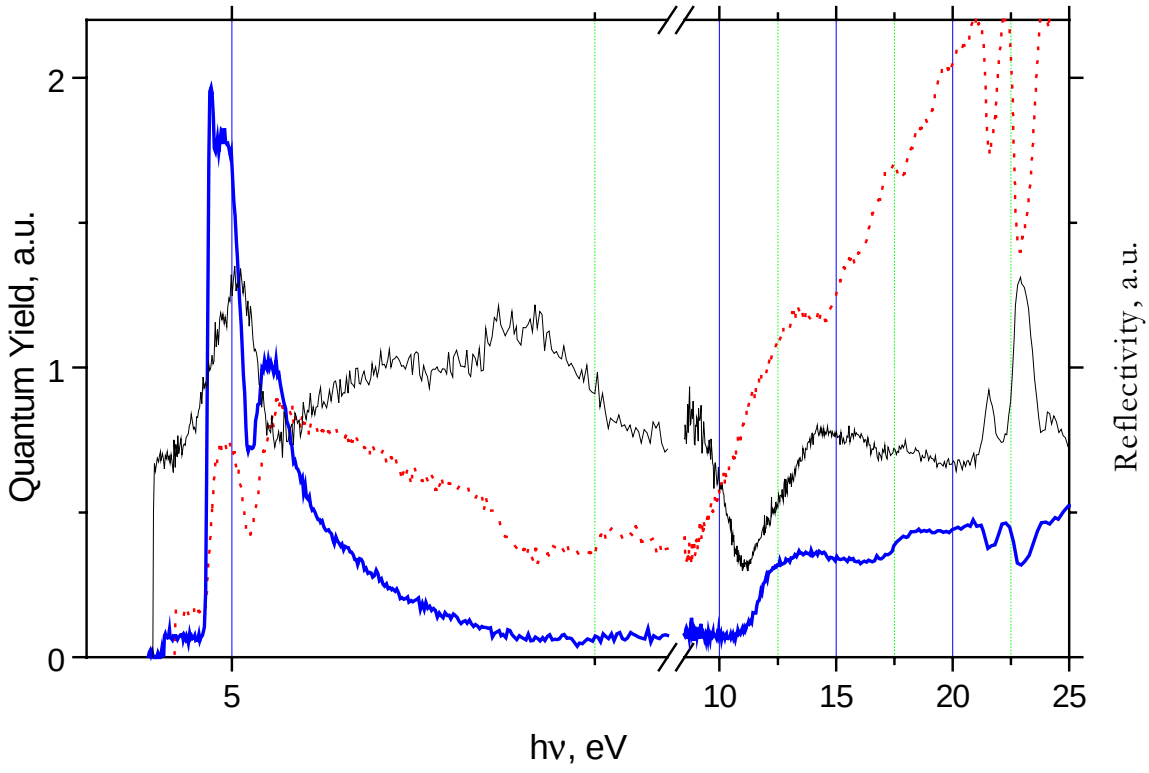

Figure 1: $PbCO_3$ reflectivity (RT, thin curve) and luminescence excitation spectra of 320 nm (thick curve) and 450 nm (dotted curve) luminescence at 6 K.

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References

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[2] W.W. Moses et al., IEEE Trans. Nucl. Sci., 38, 648 (1991).