Intrinsic and Ce^{3+} -Related Luminescence of Single Crystals and Single Crystalline Films of YAP Perovskites: New Results

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Abstract—Intrinsic emission of undoped and Ce^{3+} doped single crystal (SC) and single crystalline films (SCF) of YAlO_3 (YAP) was studied using the time-resolved luminescence spectroscopy under excitation by pulsed X-ray and synchrotron radiation. In contrast to YAP SCF, the YAP bulk SC are characterized by the presence of Y-in-Al-positions antisite defects (AD) and large concentration of vacancy-type defects (VD) of different types, which contribute to the intrinsic emission of YAP SC. Such type defects are centers of the luminescence in the UV (6.0–4.75 eV) and near UV (4.75–2.2 eV) ranges, overlapping with the Ce absorption and emission bands. Furthermore, they can give rise to shallow trapping centers as well. For this reason the AD and VD can strongly influence the scintillation properties of YAP:Ce SC. Specifically, the decay kinetics of the Ce emission in YAP:Ce SC, besides the fast component (23.3 ± 3.5 ns) related to radiative 5d-4f transition of Ce ions, contains also two slow components in the hundred ns and μs ranges, which are due to the AD and VD-related traps. The advantages of scintillators based on YAP:Ce SCF, namely faster scintillation decay and lower contribution of slow components in the total light yield, can be achieved due to the absence of above mentioned bulk crystal defects.

Index Terms—Antisite and vacancy type defects, intrinsic luminescence, scintillators, YAP single crystals and single crystalline films.

I. INTRODUCTION

SINGLE crystals (SC) of Ce-doped YAlO_3 (YAP) and LuAlO_3 (LuAP) perovskites grown from melt by Czochralski or Bridgman methods are the well-known scintillators for different fields of applications [1], [2]. Due to high melting temperature (1870°C) and existence of two cation sublattices, a variety of point defects can occur in these SC [3],

which decrease the figure of merit of these scintillation materials [4]. Among them, the Y or Lu -in-Al-positions Y_{Al} and Lu_{Al} anti-site defects (AD) and oxygen vacancy-type defects (VD) are of particular importance. Contrary to SC, in YAP:Ce and LuAP:Ce single crystalline film (SCF) scintillators grown by liquid phase epitaxy (LPE) from PbO-based melt-solution at rather low ($< 1050^\circ\text{C}$) temperatures the AD are absent and the VD concentration is substantially lower [5], [6]. This has a strong effect on the characteristics of the phosphors based on YAP SC and SCF. For instance, the Ce-doped YAP and LuAP SCF show faster decay kinetics of the Ce^{3+} emission and lower contribution of slow components in the scintillation decay due to absence of the emission and trapping centers, related with AD and VD, which are always present in the bulk SC of these materials [3].

Comparison of SC and SCF appears to be useful also for studying the intrinsic luminescence of these compounds [6], as well as for investigation of the energy transfer processes in these scintillators [5]. The comparative investigation of intrinsic luminescence of YAP SC and SCF under synchrotron radiation (SR) excitation has been fulfilled in our previous works [5], [6]. The intrinsic UV luminescence of YAP SC in the high-intensity complex bands peaked at 5.68 and 4.1 eV was tentatively interpreted by us as emission of excitons localized around Y_{Al} AD and VD (LE (AD) and LE (VD) centers, respectively). The luminescence of YAP SCF is determined by the low-intensity UV self-trapped exciton (STE) emission peaked approximately at 5.7 eV arising at the regular sites of perovskite lattice and the emission of excitons localized around Pb^{2+} trace impurity in the band peaked at 4.185 (LE (Pb) centers). Nevertheless, the structure of the mentioned complex emission bands in YAP SC has not been yet investigated in details. In paper [4] we also found that the luminescence of STE, LE (AD) and LE (VD) centers is characterized by the complex emission decay. However, due to the narrow observation time interval (up to 200 ns) under high repetition rate SR excitation the decay times of longer components of STE, LE (AD) and LE (VD) emission could not be well evaluated in [6]. The same holds for the study of the influence of LE (AD) and LE (VD) centers on the decay kinetics of Ce^{3+} emission in YAP:Ce SC [5].

The aim of this work is to continue comprehensive comparative investigation of the intrinsic and Ce^{3+} related luminescence characteristics of YAP SC and SCF in order to clarify in the details the role of AD and VD in the energy transfer process between the host and impurity luminescence centers. For this

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purpose the comparative time-resolved luminescence characterization of the YAP SC and SCF was performed under the selective VUV SR excitation: characteristic wavelengths were chosen from the YAP excitation spectra at 10 K. Furthermore, time-resolved emission spectra and the decay kinetics of the YAP and YAP:Ce SC were measured in the broader time interval (up to 3000 ns) using the pulsed (1.5 ns) X-ray radiation in the 77–300 K range.

II. SAMPLES AND EXPERIMENTAL TECHNIQUE

The undoped and Ce^{3+} doped (0.1 at. %) YAP SC was grown by Czochralski method in Crytur Ltd (Turnov, Czech Republic) from the charge containing Y_2O_3 and Al_2O_3 oxides of 4–5N purity. The undoped and Ce^{3+} doped (0.11 at. %) YAP SCF analogues with thickness in 25–35 μm range were grown by LPE from melt-solution based on $\text{PbO-B}_2\text{O}_3$ flux onto undoped YAP substrates with (100) orientation from the charge of the same purity and origin.

Time-resolved luminescence investigations of the YAP SC and SCF samples with similar $3 \times 5 \times 1$ mm dimension were performed under excitation by SR (pulse duration of 0.126 ns) at the Superlumi station (HASYLAB at DESY) at 10 K. Excitation spectra were corrected for the spectral dependence and intensity of the excitation energy in the 3.8–25 eV range. The emission spectra of the SC and SCF were measured with monochromator ARC and PMT Hamamatsu R6358P, and were not corrected. Temperature dependence of the shape of the YAP SC luminescence spectra and YAP and YAP:Ce SC emission decay kinetics (up to 3000 ns time interval) was studied under pulsed (pulse duration of 1.5 ns) X-ray excitation in the 77–300 K range on a set-up based on MDR-2 grating monochromator and PMT FEU-100.

III. EXPERIMENTAL RESULT AND DISCUSSION

The intrinsic luminescence of YAP SC in the range of 6.5–4.75 eV is given in Figs. 1 and 2 under SR excitation with different energies in the exciton region [6] at 10 K and pulsed X-ray excitation at different temperatures in the 77–300 K range, respectively. We interpret it as the superposition of the STE and LE (AD) emission bands peaked at 5.69 and 5.28 eV at 10 K, and 5.63 and 5.22 eV at 233 K (see decomposition of curve 1 in Fig. 1 and curve 3 in Fig. 3, respectively). Relative intensity of the mentioned bands depends on the Y_{Al} AD concentration in YAP SC and the temperature of measurement (Fig. 2). In AD-free YAP SCF only the STE emission is observed in this spectral range (Fig. 1, curve 5). The spectrum of STE emission in YAP SCF (curve 5) is slightly high-energy shifted with respect to the analogous spectrum of YAP SC (curves 1 and 2) due to the absence of the LE (AD) emission band. Another important feature of the SCF luminescence is the lower intensity (at least by one order of magnitude) of the STE emission (curve 5) with respect to the intensity of the STE and LE (AD) emission band in YAP SC (curve 2).

The host luminescence of YAP SC in the 260–470 nm range (Fig. 1) presents the superposition of the emission of excitons localized around the vacancy-type defects (VDs) of different types peaked at 4.08 eV (LE (VD1) emission band) [6]. Additionally

Fig. 1. Emission spectra of YAP SC (1–4) and SCF (5) at 10 K under selective excitation by SR in the exciton range with an energy of 7.935 (1), 7.74 (2) and 7.155 eV (3), as well as in the range of F-center absorption at 5.63 eV (4). The positions of all host emission bands and their possible nature are indicated. Dashed lines represent the Gaussian decomposition of curve (1) above 5 eV, see also the text.

to this band another two emission bands peaked at 3.70 and 3.29 eV (LE (VD2) and (LE (VD3) bands, respectively) are well resolved in the luminescence spectra of YAP SC (Figs. 1 and 2). Such a quantity of the LE (VD) bands is consistent with the existence of two oxygen sites in YAP structure [7] as well as with the existence of four different configurations of F^+ centers found by EPR [9]. Furthermore, the emission of the F^+ and F-centers (oxygen vacancy with one and two electrons) peaked at 3.54 and 2.91 eV, respectively [6], [8], [9] are also present in the emission spectra of YAP SC (Fig. 1, curves 1 and 4). It is worth noting that these intrinsic emission bands are well overlapped with the Ce^{3+} emission band peaked at 3.72 and 3.42 eV corresponding to the $5d-4f$ ($^2F_{5/2,7/2}$) radiative transition (two arrows on the top in Fig. 1) which can result in certain contribution of the LE (VDs) luminescence to the total scintillation output of YAP:Ce SC under high-energy excitation [5].

Temperature-induced change in the shape of the emission spectra of YAP SC in the range of 77–300 K under pulsed X-ray excitation is shown in Fig. 2(a). The temperature dependence of the intensity of the STE and LE (AD) emission bands monitored at 5.8 and 5.3 eV, respectively, as well as the intensity of the LE (VD1) and LE (VD2) emission bands registered respectively at 4.07 eV and 3.7 eV are shown in Fig. 2(b). As can be seen from Fig. 2, temperature quenching of the STE emission gives rise to some increase in the intensity of the LE (AD) emission band (curves 1 and 2, respectively). Decreasing intensity of the LE (AD) emission above 170 K is accompanied with the burning of the LE (VD1) and LE (VD2) centers emission, where the intensity of the latter center increases more quickly with respect to that of the former one (curves 3 and 4, respectively).

Thus, the temperature dependence of luminescence spectra and intensity of the components of the complex UV emission bands of YAP in the UV (6–5 eV) and near UV (4.7–2.7 eV) ranges shown in Fig. 2(a) and 2b, respectively, reflects the chain

Fig. 2. Emission spectra of YAP SC under pulsed X-ray excitation at different temperatures in the range of 77–300 K (a) and temperature dependence of intensity of different emission bands (b): STE (1), LE (AD) (2), LE (VD1) (3) and LE (VD2) (4) monitored at 5.80, 5.30, 4.07 and 3.7 eV, respectively. Dashed lines in Fig. 2(a) represent the Gaussian decomposition of curve (3) above 5 eV, see also the text.

processes of thermal delocalization, migration and another capture of the charge carriers from the shallow centers to the deepest ones in the order STE $\rightarrow$ LE (AD) $\rightarrow$ LE (VD1) $\rightarrow$ LE (VD3) $\rightarrow$ LE (VD2) with the corresponding emission bands peaked at 5.68, 5.30, 4.08, 3.35 and 3.7 eV, respectively. It is worth noting that the temperature dependence of different bands of the YAP SC intrinsic emission is similar to the temperature dependence of the intrinsic UV emissions of YAG and LuAG garnets [10].

The excitation spectra of STE emission in YAP SC and SCF are shown in Fig. 3(a) (curves 1 and 2, respectively). At 9 K the STE emission band in the YAP SCF, monitored at 5.67 eV, is excited in the complex bands located in the exciton range with the main maxima at 7.905 eV and shoulder at 8.32 eV (Fig. 3(a), curve 2). The excitation spectra of YAP SC in this region is more complicated and most probably present superposition of the STE excitation band and the LE (AD) excitation band with two maxima at 7.89 and 8.13 eV (Fig. 3(a), curve 1). The fine structure of STE and LE (AD) excitation bands (two peaks at 7.905, 8.32 eV and 7.89, 8.13 eV, respectively) presumes an existence of two excited levels of exciton-like states in YAP [6]

Fig. 3. (a)—excitation spectra at 10 K of STE emission (1, 2) in YAP SC (1) and SCF (2) monitored at 5.68 eV, and LE (VD1) emission in YAP SC (3) monitored at 4.27 eV; (b)—excitation spectra of LE (VD2) emission (1) monitored at 3.7 eV as well as LE (VD2) and F-center luminescence (2) monitored at 2.91 eV in YAP SC at 10 K.

similarly to the garnet compounds [10], and points to possible formation of the singlet and triplet states of these centers.

The LE (VD1) and LE (VD2) emission monitored at 4.26 eV (Fig. 3(a), curve 3) and 3.7 eV (Fig. 3(b), curve 2) in the YAP SC is excited predominantly in the complex bands peaked at 7.67 and 7.62 eV at the low-energy side of the YAP SC exciton absorption. The luminescence of YAP SC monitored at 2.91 eV (Fig. 3(b), curve 1), caused by superposition of the LE (VD3) band and mainly the F-center emission band (Fig. 1), is excited in the bands peaked at 7.47 eV and 7.07 eV in the exciton absorption range, as well as in the bands peaked at 5.67 and 5.24 eV related to the intrinsic $1S \rightarrow 1S$, $3P$ transitions of F-centers [8]. All the mentioned peaks within the 7–8 eV range correspond to the energy of formation of the bound exciton states related to the respective centers.

The temperature dependences of the emission decay kinetics in the STE, LE (AD) and LE (VD1) and LE (VD2) bands under excitation by pulsed X-ray radiation in the range of 77–300 K in YAP SC are shown in Fig. 4. It has been shown in Figs. 1 and 2 that due to the significant overlapping of the corresponding STE and LE (AD) bands, as well as the LE (VDs)—related emission bands, the emission decay monitored at 5.63 eV (a), 3.87 eV (b) and 3.35 eV (c) consists of at least two superposed components with different fitting parameters related to the subbands. Nevertheless, using the wavelength-resolved luminescence measurements at the mentioned energies, and taking into account the

Fig. 4. Decay kinetics of X-ray excited emission of YAP SC at different temperatures monitored at energies of 5.63 eV (a), 3.87 eV (b) and 3.35 eV (c).

shape of decay curves as well as the temperature dependence of components of the complex emission band of the intrinsic luminescence of YAP SC (Fig. 2), the characteristic temperatures at 77 K and 237 K were determined (Fig. 4(a)), where the STE and LE (AD) emission decay dominates, respectively. Similarly chosen temperatures (170 K in Fig. 4(b); 237 K in Fig. 4(c) and 282 K in Fig. 4(b) and (c)) allow the observation of the prevailing emission decay of LE (VD1), LE (VD3) and LE (VD2) centers, respectively.

The decay kinetics of all the bands (Fig. 4) clearly demonstrates their complex character. Three-component approximation $I(t) = \sum A_i \exp(-t/\tau_i) + \text{background}$ ($i = 1, 2, 3$) of the corresponding decay curves at given temperatures (displayed in **bold**) allows us to determine the fast (τ_1), middle (τ_2) and slow

TABLE I
FAST (τ_1), MIDDLE (τ_2) AND SLOW (τ_3) DECAY CONSTANTS OF LUMINESCENCE OF STE, LE (AD) AND LE (VD)—RELATED CENTERS AT THE CORRESPONDING TEMPERATURES; THE SYMBOLS DENOTES THE COMPONENTS WITH SMALL CONTRIBUTIONS

Centers	T [K]	τ_1 [ns]	τ_2 [ns]	τ_3 [ns]
STE	77	39	915	11417
	86	28	655	7372
	107	20	226	5628
LE (AD)	176	20	1063s	10765
	237	15,7	807	4668
LE VD1	170	65	710s	10765
	237	26	180s	6019
LE VD2	285	15,5	162	5724
	176	64	680s	2924
LE VD3	237	33	320s	1912

(τ_3) decay constants of the luminescence of the STE, LE (AD) and LE (VD)- related emission centers in tens ns, hundred ns and μs ranges, respectively (see Table I). The temperature dependence of the shape of decay curves and calculated parameters for these centers indicate qualitatively their different nature (Fig. 4, Table I). This supports the conclusion about the existence of several types presumable of triplet excitons, related to the STE, LE (AD) and LE (VDs) states, with different energies of their formation. At the same time, due to excitation by X-rays, the existence of decay components caused by the carrier re-trapping also is not excluded. However, the calculated de-trapping times of the leading 105 K and 150 K thermoluminescence glow curve peaks of YAP:Ce [11] point to the possible interconnection with τ_3 at the highest temperatures only.

It is worth noting that the concentration of Y_{Al} AD and VD and related with them the emission centers is significantly larger in YAP SC than that in YAP SCF [6]. This has a strong effect on the characteristics of the phosphors based on YAP:Ce SC and SCF [5]. The host emission bands peaked at 5.68 eV and 4.08 eV in YAP:Ce SC are strongly overlapped with the Ce^{3+} absorption bands peaked at 5.65 eV and 5.18 eV and around 4.47–4.25–4.13 eV, respectively (Fig. 2(a)). As a result of such an overlap, the Ce^{3+} emission in YAP:Ce SC can be partly excited via the host emission centres. The scintillation decay in Ce-doped YAP SC can be also influenced by the re-trapping the migrating charge carriers at the Y_{Al} AD and VDs of different types, which act as shallow traps during the energy transfer from the perovskite host to the Ce^{3+} ions [4], [11].

The decay kinetics of Ce^{3+} luminescence measured at 350 nm in YAP:Ce SC under pulsed X-ray excitation in 77–293 K range is shown in Fig. 5(a). Three-component exponential approximation of the corresponding decay curves makes possible to determine the decay times of components τ_i and their contribution A_i ($i = 1, 2, 3$) to the amplitude of scintillation pulse. Temperature dependences of τ_i and A_i values are presented in Fig. 5(b) and (c), respectively. As is seen from these figures, the decay kinetics of the Ce^{3+} luminescence in the temperature range 77–295 K, besides the main fast component caused by the radiative 5d-4f transitions of Ce^{3+} ions with the lifetime $\tau_1 = 23.3 \pm 3.5$ ns, contains also two temperature—dependent slow components in the ranges $\tau_2 = 103 - 212$ ns and $\tau_3 = 0, 915 - 3, 05$ μs . The latter two components arise most

Fig. 5. (a)—decay kinetics of Ce^{3+} luminescence at 350 nm in YAP:Ce SC under pulsed x-ray excitation in 77–293 K range; (b, c)—temperature dependence of the decay times (b) and contributions A_i ($i = 1, 2, 3$) (c) of fast (τ_1 , intrinsic 5d-4f transition), middle (τ_2) and slow (τ_3) components of Ce^{3+} luminescence.

probably due to the intrinsic luminescence and trapping centers involved in the Ce^{3+} excitation processes in YAP:Ce SC. This assumption is supported by certain correlation between the temperature dependences of τ_i and A_i ($i = 1, 2, 3$) (c) of fast (τ_1 , intrinsic 5d-4f transition), middle (τ_2) and slow (τ_3) components of Ce^{3+} luminescence. In particular, an increase of τ_3 magnitude in the 77–180 K temperature range can be explained by “burning” of the LE (AD) luminescence in this temperature region resulting in an increase of the contribution of these centers into the processes of Ce^{3+} centers excitation. Decreasing τ_3 value in the 180–295 K temperature range is correlated with the temperature quenching of LE (AD) luminescence and prevailing contribution of LE (VDs) centers luminescence into the total light yield of intrinsic emission in this temperature region (Fig. 2(b)). Nature of the component with lifetime τ_2 requires more detailed experimental investigation. Tentatively, it might be caused by the participation

of one or more types of LE (VDs) centers with faster than LE (AD) center emission decay (Table I) in the excitation process of the Ce^{3+} luminescence.

Thus, the decay kinetics of the Ce^{3+} luminescence, besides the radiative 5d-4f transitions of Ce^{3+} ions, contains, at least, two slow components in the range of hundred of ns and μs , which are caused by the emission and trapping centers formed by intrinsic defects of YAP:Ce SC, specifically AD and VD of different types.

In YAP:Ce SCF scintillators the emission and trapping centers related to AD and VD are absent [3]. Consequently, YAP:Ce SCF demonstrates faster decay kinetics of the Ce^{3+} emission and significantly lower contribution of slow components in scintillation pulse in comparison with SC analogues ($K_S = 0.07\%$ and 0.42% , respectively) [5].

IV. CONCLUSION

We have demonstrated that the UV band of the intrinsic emission of YAP single crystals in the 6.5–4.75 eV range is of complex character and consists of the self-trapped exciton (STE) emission band peaked at 5.69 eV and the emission band of excitons localized around Y_{Al} antisite defects (LE (AD) centers) peaked at 5.28 eV at 10 K. The complex intrinsic emission band of YAP single crystals in the 4.75–2.2 eV range includes the luminescence of excitons localized at vacancy-type defects of different types in the bands peaked at 4.08, 3.70 and 3.29 eV (LE (VD1), LE (VD2) and LE (VD3) centers, respectively) as well as the emission of the F and F^+ centers in the bands peaked at 2.91 eV and 3.54 eV, respectively. Contrary to YAP single crystals, in YAP single crystalline films only the STE emission is observed in the unperturbed sites of perovskite lattice.

The fine structure of excitation bands in exciton range (specifically, two peaks at 7.905 and 8.13 eV or 7.89 and 8.13 eV for STE and LE (AD) emission, respectively) and the complex three-component character of the STE, LE (AD) and LE (VDs) centers luminescence decay with the decay times of corresponding components in tens ns, hundred ns and μs range presume an existence of two excited levels of exciton-like states in YAP, and point to the possible formation of the singlet and triplet relaxed excited states of mentioned centers. The excitation bands peaked at 7.67, 7.62 and 7.56 eV correspond to the energy of formation of the bound exciton states of the LE (VD1), LE (VD2) and LE (VD3) centers in YAP SC, respectively.

Temperature dependence of the luminescence spectra and intensity of components of the intrinsic emission bands of YAP SC reflect the processes of thermal delocalization, migration and subsequent capture of the charge carriers starting from the shallow centers towards the deeper ones in the sequence $\text{STE} \rightarrow \text{LE (AD)} \rightarrow \text{LE (VDs)}$ which is similar to behavior of the intrinsic luminescence of YAG and LuAG garnets [10].

Thus, scintillation properties of the YAP:Ce SC scintillators are strongly influenced by the presence of the Y_{Al} AD and VDsof different types, which give rise to the luminescence bands in UV (6.0–4.75 eV) and near UV (4.75–2.2 eV) ranges, overlapping with the corresponding Ce^{3+} absorption and emission bands. Furthermore, these defects give rise to shallow trapping centers as well [9], [11]. As a result, the scintillation response is formed not only by the fast component

(23.2 ± 3.5 ns) due to Ce^{3+} luminescence, but it contains also two slower components in the range of hundreds of ns—units μs , which arise due to the AD and VD-related emission centers and traps. Elimination of these defects in YAP:Ce SCF scintillators results in the simpler and faster decay kinetics and the lower content of slower components in the Ce^{3+} luminescence under excitation in the region of interband transitions [5].

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