SPECTROSCOPY OF YTTRIUM SCANDATE DOPED WITH YTTERBIUM ION O.K. Alimov, M.E. Doroshenko, E.A. Dobretsova, K.A. Pierpoint, S.Ya. Rusanov, V.V. Kashin, V.B. Tsvetkov

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Abstract

The spectral-kinetic properties of Yb³⁺ optical centers in YScO₃ crystal fiber were studied using selective laser spectroscopy. Various spectral characteristics of the Yb³⁺ optical centers were determined, including homogeneous and inhomogeneous broadening values, Stark level splitting, and the lifetime of Yb³⁺ ion optical centers. Three distinct types of Yb³⁺ optical centers were identified. The first optical center, with a lifetime of $\tau^{III}_{77K} = 4.05$ ms, was formed due to the substitution of Y³⁺ for Yb³⁺ ions in the local site of C_{3i} . The other two Yb³⁺ optical centers, with lifetimes of $\tau^{I}_{77K} = 1.0$ ms ($C^{II}_2(Y^{3+})$ center) and $\tau^{II}_{77K} = 0.620$ ms ($C^{II}_2(Sc^{3+})$), respectively, were formed as a result of the substitution of Y³⁺ and Sc³⁺ for Yb³⁺ in the local site of C_2 .

Key words: bixbyite, yttrium scandate, optical center, rare-earth ion, selective laser spectroscopy.

1. Introduction

Rare-earth doped sesquioxide materials have several advantageous properties, including high thermal conductivity [1–3], strong Stark-splitting [4–6], low phonon energies [7], as well as a wide transparency window ranging from 0.25 μ m to 9.6 μ m [8]. As a result, they are highly suitable for deployment in high-power solid-state lasers, particularly within the infrared wavelength range. Additionally, these materials can serve as alternatives to the widely used YAG crystals.

Yttrium oxide (Y₂O₃) and scandium oxide (Sc₂O₃) have been found to possess a cubic bixbyitetype structure [9,10]. These positions include the 8*b* site, which exhibits C_{3i} symmetry, and the 24*d* site, which has C_2 symmetry. Within the unit cell, there are three cations occupied the C_2 site, and one cation at the C_{3i} site. The overall crystal structure is formed by the arrangement two different 6vertex polyhedra that share corners and edges. As a result, the crystal structure can be described as a chessboard packing arrangement, derived from the fluorite structural type.

Various rare-earth dopants in Y₂O₃ crystals have been studied, revealing that RE^{3+} ions can substitute of Y³⁺ in both the C_2 and C_{3i} structural sites [4,11–15]. Sc₂O₃ is an equally promising host material due to its high crystal field resulting from lattice distortions caused by the substitution of relatively small Sc³⁺ for large RE^{3+} ions [16–18].

Rare-earth sesquioxides have been found to exhibit different crystal structures [19–21], leading to variation in their laser properties depending on the specific rare-earth ion and host crystal structure. One of the YScO₃ phase modification, similar to Y_2O_3 and Sc_2O_3 , possesses a bixbyite-type crystal structure [22]. In our works [10,23–25], YScO₃ (Y:Sc ratio is about 1:1) crystals were grown in the form of fibers using the laser-heated pedestal growth (LHPG) technique. This involved heating and melting the powder preform under CO₂ laser irradiation, followed by rapid cooling [26].

The luminescence and absorption spectra of bixbyite-like YScO₃ media doped with rare-earth ions include lines that are inhomogeneously broadened due to disorder in the crystal structure [27– 30]. This characteristic makes these media highly promising for applications in ultrafast lasers and chirped pulse amplification. There are some publications on lasers based on single crystal and ceramic mixed sesquioxides doped by Nd³⁺ [31], Tm³⁺ [32–35], Ho³⁺ [36]. However, Yb³⁺-doped materials can be an alternative to Nd³⁺-doped active media for lasing in the range of 1 μ m. Ytterbiumdoped materials have a smaller quantum defect compared to neodymium materials, as well as a wider emission band [37–39]. Therefore, ytterbium-doped matrices are more suitable as active media in femtosecond lasers. Yb³⁺ ion with electronic configuration ⁴*f*₁₃ possesses one ²*F*_{5/2} excited state with an energy range of 10200-11100 cm⁻¹, which is ideal for pumping with commercially available laser diodes emitting in the 915-980 nm range. This electronic level structure of Yb³⁺ excludes possible absorption from the ²*F*_{5/2} excited state, as there are no higher energy levels available. Successful implementation of Yb lasers has been achieved in mixed sesquioxide ceramic hosts such as Yb³⁺:(Sc_xY_{1-x})₂O₃ [40] and Yb³⁺:(Lu_xY_{1-x})₂O₃ [41].

The investigation of RE^{3+} :YScO₃ mixed sesquioxide requires modern methods of selective laser spectroscopy. These methods enable the determination of quantitative features of optical centers and the identification of channels for both radiative and non-radiative energy [42]. Especially this applies to the transfer of excitation energy between the RE^{3+} optical centers, which are formed in the YScO₃ crystal fiber by substituting the positions of yttrium and scandium. The utilization of selective laser spectroscopy methods will allow for obtaining a complete characterization of the optical properties of RE^{3+} impurity center in YScO₃ crystal fiber and assessing the potential of this matrix as a laser-active medium.

Applying the laser-heated pedestal growth (LHPG) technique to obtain YScO₃ crystal fiber have been described in details earlier [10,24]. Features of the bixbyite-like crystal structure of YScO₃ have also been discussed in previous our research [10]. Additionally, the spectroscopic features of Nd³⁺ and Tm³⁺ dopants in YScO₃ crystal fibers have also been extensively investigated and published [10,23–25]. This current study aims to specifically focus on the spectral and kinetic properties of Yb³⁺:YScO₃ crystal fibers using advanced selective laser spectroscopy methods.

2. Materials and Methods

Crystals of 0.1 at. % Yb³⁺:YScO₃ were successfully grown using the laser-heated pedestal growth (LHPG) technique [10]. Commercial powders of Y₂O₃ and Sc₂O₃ of a purity of more than 99.999% (Sigma Aldrich, Burlington, MA, USA) were used as precursors, while Yb₂O₃ was used as activators. The crystals of Yb³⁺:YScO₃ have been obtained in the form of a fiber with a diameter of 0.6 mm and a length of 50 mm. The crystal observed belongs to the bixbyite structural type and has a cubic structure with a space group of $Ia\overline{3}$.

The time-resolved luminescence excitation and luminescence spectra, as well as decay kinetics, were measured on the MDR-23 monochromator at temperatures of 77 K and 300 K. The FEU-83 µ Hamamatsu R5108 photomultipliers were used as detectors. The Tektronix - TDS3052B broadband oscilloscope was used to record the time-resolved luminescence spectra and decay curve at the Solar LP-604 parametric generator excitation. The double spectral selection technique was used to record the decay curves accurately [42].

3. Results and Discussions

Figure 1 shows absorption spectra of YScO₃ crystal fiber doped with Yb³⁺ ions being measured at temperatures of 300 K and 77 K. The decrease in temperature from 300K to 77K results in narrowing of the absorption bands of Yb³⁺ ions, their structuring, and the emergence of new lines that were not visible at the higher temperature (300 K). Surprisingly, the number observed absorption lines (six ones) exceeds the expected number of three lines. This suggests the presence of additional Yb³⁺ optical centers within the crystal matrix, which contribute to the observed spectral overlap and the splitting and population of Stark components in the ${}^{2}F_{7/2}$ ground state. The existence of distinct optical centers of Yb³⁺ ions in other crystals like Y₂O₃ and Sc₂O₃ has been previously observed in [43].

The absorption spectrum of Yb³⁺:YScO₃ crystal fiber was measured on the ${}^{2}F_{7/2} \rightarrow {}^{2}F_{5/2}$ transition of Yb³⁺ ion at a temperature of 77K. In Figure 1, curve 1 shows one narrow peak with a maximum at $v_{max} = 10246$ cm⁻¹. Additionally, there are three broad bands with weakly resolved structure and maxima at 11111, 10753, and 10582 cm⁻¹. This spectrum is compared to the energy levels of Yb³⁺ ions with local symmetries of C_2 and C_{3i} in Y₂O₃ and Sc₂O₃ crystals as a reference [43]. The optical centers of Yb³⁺ ions in the Sc₂O₃ crystal exhibit the highest values of Stark splitting of the ${}^{2}F_{5/2}$ level being measured as $\Delta E = 856$ cm⁻¹ and $\Delta E = 1126$ cm⁻¹ for Yb³⁺ centers with local symmetries of C_2 (Figure 1*b*) and C_{3i} (Figure 1*d*), respectively. In Yb³⁺:Y₂O₃, the Stark splitting values (ΔE) are equal to 726 cm⁻¹ and 1062 cm⁻¹ for Yb³⁺ optical centers with local symmetries of C_2 (Figure 1*a*) and C_{3i} (Figure 1*c*), respectively. Furthermore, in YScO₃ crystal fiber spectra, the Stark splitting of ${}^{2}F_{5/2}$ transition is determined to have a value of $\Delta E = 864 \text{ cm}^{-1}$ (Figure 1, a curve 1).

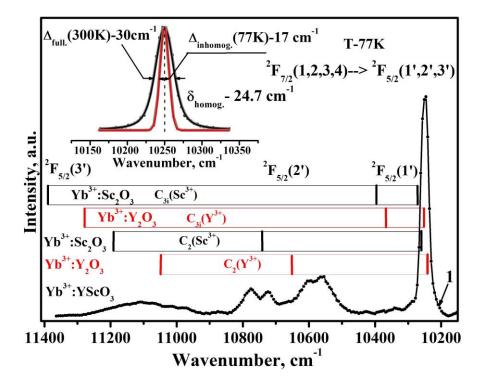


Figure 1. Absorption spectrum of Yb³⁺-doped YScO₃ crystal fiber being measured at 77 K. Stark splitting of the ${}^{2}F_{5/2}$ transition of Yb³⁺ optical centers with a local symmetry of C_{2} in Y₂O₃ (*a*) and Sc₂O₃ (*b*). Stark splitting of the ${}^{2}F_{5/2}$ transition of Yb³⁺ optical center with a local symmetry of C_{3i} in Y₂O₃ (*c*) and Sc₂O₃ (*d*) [43]. In the inset, absorption spectrum of the ${}^{2}F_{7/2}(1) \rightarrow {}^{2}F_{5/2}(1')$ resonance transition of Yb³⁺ ion in YScO₃ crystal fiber.

The analysis of the Stark splitting values for the ${}^{2}F_{5/2}(1', 2', 3')$ level in the sesquioxide materials reveals that the YScO₃ crystal fiber doped with Yb³⁺ ions exhibits splitting intermediate between the Yb³⁺:Y₂O₃ and Yb³⁺:Sc₂O₃ crystals. This observation is consistent with an intermediate lattice parameter value of the YScO₃ crystal fiber, as it is a solid solution. However, determining of the precise position of the Stark level components is complicated due to the presence of multiple types of Yb³⁺ optical centers in the YScO₃ crystal fiber, similar to those formed in Y₂O₃ and Sc₂O₃ crystals [43]. It is noteworthy that the absorption bands corresponding to the Stark components of the Yb³⁺ optical centers in YScO₃ exhibit broadening, which is presumably related to disorder in the crystal structure and the statistical distribution of Y³⁺ and Sc³⁺ ions in two structural positions, where they are partially substituted by Yb³⁺. The spin-orbit interaction within the ${}^{2}F_{J}$ multiplet remains unchanged. In Figure 1, the Stark sublevels associated with the ${}^{2}F_{5/2}$ excited state denoted by bars, and the sublevels of the ${}^{2}F_{7/2}$ ground state are represented by unbarred numbers. The numbering of levels within the multiplet follows a bottom-to-top sequence.

To analyze the shape of the absorption band of the ${}^{2}F_{7/2}(1) \rightarrow {}^{2}F_{5/2}(1')$ transition and evaluate the value of homogeneous and inhomogeneous broadening in the Yb³⁺:YScO₃ crystal, the ${}^{2}F_{7/2}(1) \rightarrow$ ${}^{2}F_{5/2}(1')$ transition was singled out from the overall spectrum taken at 77 K and 300K. Figure 1 shows the optical absorption spectrum (curve 1) of the Yb³⁺:YScO₃ crystal fiber being measured at 77K. The absorption line according to the ${}^{2}F_{7/2}(1) \rightarrow {}^{2}F_{5/2}(1')$ transition of Yb³⁺ ions in the crystal fiber is shown in the inset of Figure 1 where the half-width of the transition line is indicated by horizontal arrows. The absorption line of the ${}^{2}F_{7/2}(1) \rightarrow {}^{2}F_{5/2}(1')$ transition being measured at 300K appears symmetrical and exhibits a peak at a frequency of $v_{max} = 10246 \text{ cm}^{-1}$. The full width of the transition at half height, Δ_{full} (300 K), was determined to be 30 cm⁻¹ by approximating the line with a Voigt distribution function [44]. Lowering the temperature to 77 K causes the absorption lines of the ${}^{4}F(1) \rightarrow {}^{4}F_{5/2}(1')$ transition were fitted using a Gaussian distribution function. Using the equation derived in in the work [44], the value of the homogeneous broadening of the ${}^{4}F_{7/2}(1) \rightarrow {}^{4}F_{5/2}(1')$ transition was calculated to be $\delta(300\text{K}) = [\Delta^{2}_{\text{full}}(300\text{K}) - (\Delta')^{2}_{\text{inhomog}}(77\text{K})]^{1/2} = 24.7 \text{ cm}^{-1}$.

To gain a more comprehensive understanding of the mechanism behind the formation of Yb3+ optical centers in YScO₃, we measured luminescence decay kinetics at a temperature of 77K (Figure 2). The excitation of ytterbium ions was performed specifically on the ${}^{2}F_{7/2}(1) \rightarrow {}^{2}F_{5/2}(1')$ resonance transition. Subsequently, the resulting fluorescence signals were analyzed by determining the fluorescence lines with peak values at vdet. = 9737, 9634, 9320, and 8985 cm⁻¹.

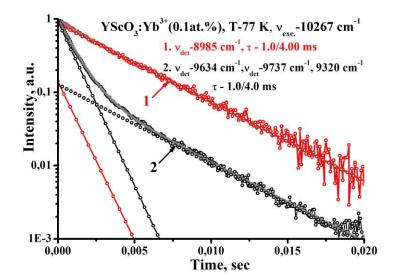


Figure 2. Luminescence decay kinetics of Yb³⁺ ions in YScO₃. Excitation of Yb³⁺ ions was carried out on the ${}^{2}F_{7/2}(1) \rightarrow {}^{2}F_{5/2}(1')$ transition at $v_{\text{exc.}} = 10267 \text{ cm}^{-1}$. The detection was then performed at frequencies of $v_{\text{det.}} = 8985 \text{ cm}^{-1}$ (curve 1) and $v_{\text{det.}} = 9737, 9634, 9320 \text{ cm}^{-1}$ (curve 2).

The decay luminescence curves (Fig. 2, curves 1, 2) can be better characterized by considering them as a sum of two exponentials with lifetimes of 1.0 and 4.0 ms. The relative contribution of the long-lived or short-lived components in the total intensity of Yb³⁺ luminescence varies depending on the detection frequency. In the YScO₃ crystal structure, Yb³⁺ optical centers with a lifetime of $\tau = 1.0$ ms are formed when Yb³⁺ substitutes of Y³⁺ in the C_2 site. The intensity of the short-lived component increases when registering the decay kinetics of Yb³⁺ luminescence on the line belonging to the long-lived center ($v_{det.} = 9737$ cm⁻¹). This increase is caused by the overlap of the luminescence bands with peaks of $v_{max.} = 9737$ cm⁻¹ and 9625 cm⁻¹. This overlapping leads to a combined effect on the luminescence intensity, resulting in a change in the relative contribution of the short-lived component. On the other hand, when the detection frequency is $v_{det} = 8985$ cm⁻¹, the decay kinetics of Yb³⁺ luminescence can be accurately described by a single exponential decay law (Fig. 2, curve 1). This detection band corresponds to a long-lived optical center with a lifetime of $\tau = 4.0$ ms. This long-lived center is formed when Yb³⁺ substitutes with Y³⁺ in the C_{3i} site.

Figure 3 shows luminescence decay kinetics being measured on the ${}^{2}F_{7/2}(1) \rightarrow {}^{2}F_{5/2}(1')$ transition in Yb³⁺:YScO₃ at excitation of $v_{\text{exc.}}$ = 10235 cm⁻¹ and a temperature of 77 K. The luminescence of the Yb³⁺ ion in YScO₃ was recorded on the ${}^{2}F_{5/2}(1') \rightarrow {}^{2}F_{5/2}(2, 3, 4)$ transition at frequencies of 9891, 9775, and 9653 cm⁻¹. The luminescence decay curve being measured at 77K was described by a single exponential decay law with a lifetime of $\tau = 0.620$ ms, indicating the presence of a second short-lived optical center with a local symmetry of C_2 in the crystal fiber. This center is formed in the matrix through the substitution of scandium ions with ytterbium ions.

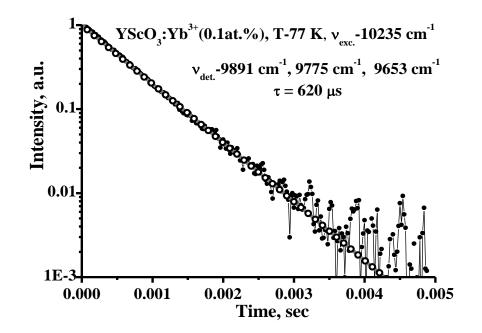


Figure 3. Luminescence decay kinetics being measured on the ${}^{2}F_{7/2}(1) \rightarrow {}^{2}F_{5/2}(1')$ transition of Yb³⁺ ions in YScO₃ at an excitation of 10235 cm⁻¹. The luminescence decay curve was registered at frequencies of $v_{det.} = 9891$, 9775, and 9653 cm⁻¹.

The difference in lifetimes of the two short-lived centers is due to the difference in ionic radii of Y^{3+} (0.87 Å) and Sc^{3+} (0.75 Å), which are replaced by Yb^{3+} (1.01 Å) in the C_2 site. The substitution of yttrium or scandium ions in the lattice with ytterbium ion leads to a change in the distance between the cation and its oxygen environment in the distorted octahedron [45–47]. Decreasing the distance between the impurity center and its oxygen environment lead to changes in the crystal field parameters that are responsible for the probabilities of radiative transitions[42]. Therefore, the optical center with a lifetime of 0.620 ms is attributed to the substitution of Sc^{3+} with C_2 site symmetry in the YScO₃ crystal lattice for Yb^{3+} , and the optical center being formed due to the substitution of Y^{3+} with C_2 site symmetry in the YScO₃ crystal lattice for Yb^{3+} has a longer lifetime (1.0 ms).

To confirm the existence of at least two types of Yb³⁺ optical centers in YScO₃, the timeresolved luminescence spectra were measured (Figure 3). By increasing the time delay (t_{del}) from 40 µs to 1.58 ms, the intensities of the Yb³⁺ ion fluorescence lines with maxima at $v_{max.} = 9775$, 9632, 9320 cm⁻¹ were redistributed. Specifically, an increase in intensity was observed for the luminescence line with maxima at $v_{max.} = 9737$ and 9320 cm⁻¹, while a decrease in intensity was observed for the line with maxima at $v_{max.} = 9625$ cm⁻¹.

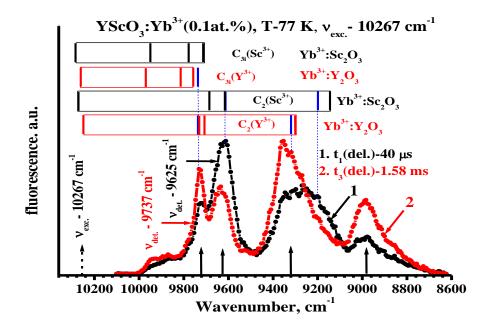


Figure 4. Time-resolved luminescence spectra of ytterbium ions in YScO₃ being measured at time delays of $t_{1(del.)} = 40 \ \mu s$ (curve 1) and $t_{3(del.)} = 1.58 \ ms$ (curve 2). The observed luminescence decay kinetics were recorded at specific frequencies, as indicated by the arrows. Stark splitting of the ${}^{2}F_{7/2}$ level of Yb³⁺ optical centers with local symmetry of C_{2} and C_{3i} in Y₂O₃ (red one) and Sc₂O₃ (black one) crystals [43].

The number of luminescence lines being obtained on the ${}^{2}F_{5/2} \rightarrow {}^{2}F_{7/2}$ transition of Yb³⁺ is five, that exceeds the calculated value (four), therefore, there are at least two types of optical centers of the ytterbium ion in the YScO₃ crystal fiber. As shown in the works [43], two types of optical centers exist in both Y₂O₃ and Sc₂O₃ crystals doped with Yb³⁺ ions: a) low-symmetric in the *C*₂ site, and b) high-symmetric one in the *C*_{3*i*} site. The Stark component energies for these two types of Yb³⁺ optical centers in both Y₂O₃ and Sc₂O₃ crystals are obtained at T=10K in [40] and shown in Figure 4. The largest splitting of the ${}^{2}F_{7/2}$ level is observed for the center with *C*₂ local symmetry in the Sc₂O₃ crystal. Calculations on crystal field theory have been performed in [41] to determine the Stark component energies and the the ${}^{2}F_{7/2}$ and ${}^{2}F_{5/2}$ for Yb3+ ions in the Y₂O₃, yielding results that are in good agreement with previous studies [40].

Figure 5 shows the luminescence spectra being measured on the ${}^{2}F_{7/2}(1) \rightarrow {}^{2}F_{5/2}(1')$ transition of Yb³⁺ ion at an excitation of $v_{\text{exc.}} = 10235 \text{ cm}^{-1}$ and a temperature of T = 77 K. When the time delay is changed from $t_{1(\text{del})} = 40 \text{ } \mu\text{s}$ to $t_{3(\text{del})} = 1.6 \text{ } \text{ms}$, there is no significant change observed in the luminescence spectra, particularly in the spectral line with a maximum at $v_{\text{max}} = 9653 \text{ cm}^{-1}$. Weak lines with maxima at $v_{\text{max}} = 9891$, 9775, and 9653 cm⁻¹ are observed on the short-wavelength side of the luminescence spectrum. Increasing the time delay up to $t_{\text{del}} = 1.6 \text{ } \text{ms}$ results in a slight shift of the luminescence line with a maximum at $\lambda_{\text{max}} = 1036 \text{ } \text{nm}$, a decrease in the intensity of the band with a maximum at $v_{\text{max}} = 9225 \text{ cm}^{-1}$, and appearance of a weak luminescence line with a maximum at $v_{\text{max}} = 8834 \text{ cm}^{-1}$.

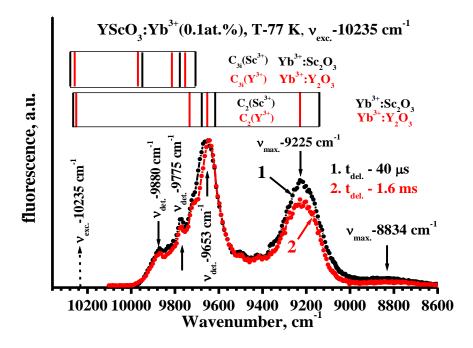


Figure 5. Time-resolved luminescence spectra of Yb³⁺ ions in YScO₃ being measured at temperature of T = 77 K and time delays of $t_1 = 40 \ \mu s$ (curve 1) and $t_3 = 1.6 \ m s$ (curves 2). Stark component energies of the ${}^2F_{7/2}$ level for Yb³⁺ optical centers with local symmetries of C_2 and C_{3i} in both Y₂O₃ (red one) and Sc₂O₃ (black one) crystals [43].

These factors suggest the presence of a dominant short-lived Yb³⁺ optical center in YScO₃. This is confirmed by the kinetic studies shown in Figure 3. The energies of Stark components of Yb³⁺ ion in both Y₂O₃ and Sc₂O₃ crystals, corresponding to two types of optical centers with a local symmetry of C_2 and C_{3i} , were determined in previous work [43] at a temperature of T = 10 K are shown in Figure 5. Here, the red lines indicate the positions of Yb³⁺ ion Stark components in Y₂O₃, while the black lines represent positions of Yb³⁺ in Sc₂O₃. Based on the kinetic and spectral measurements carried out for Yb³⁺:YScO₃ it has been determined that there are three types of Yb³⁺ optical centers in the YScO₃ crystal fiber. Two of them with a local symmetry of C_2 labeled as C_2^{II} (Y³⁺) and C_2^{II} (Sc³⁺) correspond to substitution of Y³⁺ and Sc³⁺ cations with Yb³⁺, respectively. The third one with a high symmetry of C_{3i} labeled as C_{3i} (Y³⁺) corresponds to substitution of Y³⁺ with Yb³⁺. The position of the Stark components of the Yb³⁺ optical centers in YScO₃ crystal have been determined based on the obtained results.

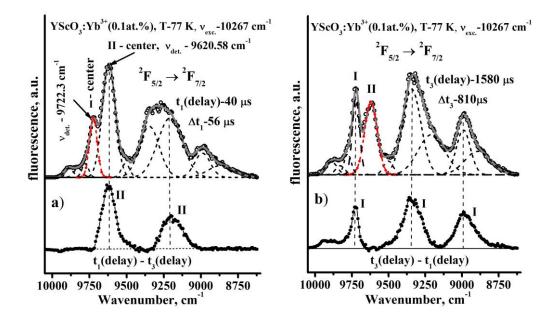


Figure 6. Low-temperature (77 K) time-resolved luminescence spectra being measured on the ${}^{2}F_{5/2}$ $\rightarrow {}^{2}F_{7/2}$ transition of Yb³⁺ ions in the YScO₃ crystal fiber at two different time delays of: (*a*) $t_{1(del.)} =$ 40 µs (*a*) and $t_{3(del.)} = 1.58$ ms (*b*). The luminescence spectra of Yb³⁺ ions were normalized to the lines with maxima at 9737 and 9625 cm⁻¹ being obtained at the time delays of $t_{1(del.)} = 40$ µs and (*b*) $t_{3(del.)} = 1.58$ ms, respectively. The difference spectra of luminescence [t1(del.) - t3(del.)] and [t3(del.) - t1(del.)] are shown at the bottom of (*a*) and (*b*), respectively.

The luminescence spectra (Figure 4, curves 1 and 2) have been normalized based on the spectral lines at frequencies oof $v_{\text{max}} = 9737 \text{ cm}^{-1}$ and $v_{\text{max}} = 9625 \text{ cm}^{-1}$ (Figure 6*a* and *b*, highlighted in red on the spectra). Time delays of $t_{1(\text{del.})} = 40 \text{ }\mu\text{s}$ and $t_{3(\text{del.})} = 1.58 \text{ }m\text{s}$ were used to measure the luminescence spectra. To analyze the spectra, a Gaussian distribution function was employed to separate the spectral components. Surprisingly, six spectral lines were observed instead of the expected three (excluding the main inter-Stark ${}^{2}F_{5/2}(1) \rightarrow {}^{2}F_{7/2}(1')$ transition).

The luminescence spectrum obtained at a short time delay ($t_{1(del.)} = 40 \ \mu s$) was subtracted from the spectrum taken with a long-time delay ($t_{3(del.)} = 1.58 \ ms$). So, the positions of two lines associated with the short-lived ytterbium center can be identified (Figure 6a, bottom). Similarly, the spectrum obtained at a long time delay was subtracted from the spectrum taken with a short time delay, that reveals the positions of the spectral lines corresponding to the long-lived center (Figure 6a, bottom). The number of luminescence lines corresponds to the calculated number of the Stark components of the ${}^{2}F_{7/2}(1, 2, 3, 4)$ level of the long-lived center. The position of the Stark components of Yb³⁺ ions in the YScO₃ crystal is shown in Figure 4 and 5 (blue line), serving as a basis for assigning the luminescence lines to optical centers with local symmetries of C_2 and C_{3i} . According to Figure 4, the luminescence spectral lines of Yb³⁺ ions belong to optical centers with local symmetries of C_2 (Y³⁺), C_{3i} (Y³⁺), and C_2 (Sc³⁺). In the case of a single low-symmetry center II (Yb³⁺), the luminescence spectra of the ytterbium ion can be attributed to optical centers with local symmetries of C_2 (Sc³⁺) and C_2 (Y³⁺), which lines are overlapped (see Figure 5).

Figure 7 shows the impact of excitation frequency (curves 1, 2, 3) on the time-resolved luminescence spectra being measured on the ${}^{2}F_{5/2}(1) \rightarrow {}^{2}F_{7/2}(1', 2', 3')$ transition of Yb³⁺ short-lived optical centers (II(Y³⁺) and II(Sc³⁺)). Analysis reveals that the luminescence spectra exhibit distinctive shift at various excitation frequencies. Notably, the maximum observed shift, denoted as $\Delta E \sim 30$ cm⁻¹, can be observed.

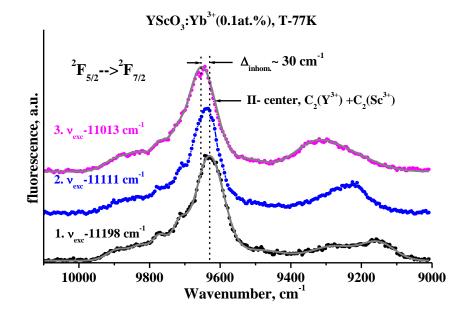


Figure 7. Time-resolved luminescence spectra being measured on the ${}^{2}F_{7/2} \rightarrow {}^{2}F_{5/2}$ transition of ytterbium ions in the YScO₃ crystal fiber (curves 1, 2, 3). These spectra were observed at a time delay of $t_{1(del.)} = 40 \ \mu s$ and a temperature of 77 K. The luminescence measurements were performed using excitation frequencies of 11198 cm⁻¹, 11111 cm⁻¹, and 11013 cm⁻¹, corresponding to curves 1, 2, and 3, respectively.

The observed shift in the maxima of the short-lived center and the dependence of the luminescence spectra on excitation suggest the presence of disorder in the YScO₃ crystal fiber matrix of the crystal fiber. The degree of disorder in the optical center II of Yb3+ ions is influenced by the inhomogeneous broadening of the luminescence and absorption lines. In the long-wavelength wing of the Yb³⁺ ion luminescence spectrum (Figure 7), the bands associated with C^{II}_2 (Y³⁺, Sc³⁺) symmetrical centers (Fig. 3a, b) exhibit varying intensity and spectral position on the ${}^{2}F_{5/2}(1', 2') \rightarrow {}^{2}F_{7/2}(2, 3, 4)$ inter-Stark transitions, depending on the excitation wavelength. The spectral shift and intensity change of these Yb³⁺ bands are determined by the efficient excitation of the C^{II}_2 (Y³⁺) and C^{II}_2 (Sc³⁺) centers. It is worth noting that simultaneous excitation of Yb³⁺ optical centers (C^{II}_2 (Y³⁺)

and $C^{II_2}(Sc^{3+})$), which have a heterogeneous distribution within a single optical center, can also occur (Figure 7, curve 3). This observation can be attributed to the close proximity and spectral overlap of the Stark energy levels associated with the $2F7/2 \rightarrow 2F5/2$ absorption transition of two distinct optical centers, II(Y3+) and II(Sc3+), with the C_2 local symmetry.

4. Conclusions

The spectral-kinetic properties of Yb³⁺ optical centers in YScO₃ crystal fiber were investigated using selective laser spectroscopy methods. The quantitative spectral characteristics of Yb³⁺ optical centers were determined: homogeneous broadening ($\delta(300K) = 24.7 \text{ cm}^{-1}$), inhomogeneous broadening ($\Delta'_{inhom.}(77K) = 17 \text{ cm}^{-1}$), Stark splitting, and lifetimes of Yb³⁺ optical centers.

Three types of Yb³⁺ optical centers of ions have been found in YScO₃. The first one is a highly symmetric optical center with a local symmetry $C_{3i}(Y^{3+})$ and a lifetime of $\tau^{I}_{77K} = 4.0$ ms, which is formed as a result of substitution of Y³⁺ ions for ytterbium ions in the crystal lattice. The second center has local symmetry $C_2^{II}(Y^{3+})$ and a lifetime of $\tau_{77K}^{II} = 1.0$ ms. The third one is a low-symmetry $C_2^{II}(Sc^{3+})$ center with lifetimes of $\tau_{77K}^{II} = 0.620$ ms.

It has been shown that the broadening of the absorption and luminescence spectra of the Yb³⁺ optical centers with local symmetry C_2^{II} (Y³⁺) and C_2^{II} (Sc³⁺) in YScO₃ is due to the disorder in the YScO₃ crystal structure.

Funding: This research was funded by the Russian Science Foundation, grant number 22-22-00968.

Conflicts of Interest: The authors declare no conflict of interest.

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