

Polarized spectroscopy of Ho:YAlO₃ crystals for 2 μm and 3 μm lasers

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ABSTRACT

We report on polarized spectroscopic properties of Ho³⁺ ions in orthorhombic (sp. gr. *Pnma*) yttrium orthoaluminate YAlO₃ crystals for laser development at 2 μm and 3 μm. This includes polarized Raman, absorption and luminescence spectra, fluorescence lifetime measurements and Stark energy-level study. The transition intensities for Ho³⁺ ions are calculated using the Judd-Ofelt theory. The peak stimulated-emission cross-sections are 2.01×10⁻²⁰ cm² at 1977 nm (⁵I₇ → ⁵I₈) and 2.31×10⁻²⁰ cm² at 2918 nm (⁵I₆ → ⁵I₇) for light polarization $\mathbf{E} \parallel \mathbf{b}$. For both transitions, pump-induced polarization-switching is expected. The fluorescence lifetimes of the ⁵I₇ and ⁵I₆ Ho³⁺ manifolds are 7.27 and 0.36 ms, respectively (for 1 at.% Ho³⁺-doping).

Keywords: yttrium aluminate, holmium ions, laser crystal, optical spectroscopy, Judd-Ofelt analysis.

1. INTRODUCTION

The Holmium ion (Ho³⁺), with an electronic configuration of [Xe]4f¹⁰, plays a crucial role in solid-state laser technology due to its appealing emission properties. Its main emission, corresponding to a transition from the first excited-state (⁵I₇) to the ground-state (⁵I₈), is slightly above 2 μm, which is relatively eye-safe [1]. Another emission channel slightly below 3 μm is due to the ⁵I₆ → ⁵I₇ transition, Fig. 1(a). 2-μm Ho-lasers find applications in remote sensing, wind mapping [2,3], and laser surgery, as their emission aligns with a local maximum of water absorption in bio-tissues and the atmosphere [4]. This wavelength range is also ideal for pumping mid-IR optical parametric oscillators (OPOs) and optical parametric amplifiers (OPAs) based on non-oxide nonlinear crystals [5]. Emission near 3 μm on the other side coincides with the absolute maximum of water absorption which corresponds to minimum penetration depth [4]. All emerging applications related to the development of continuous-wave (CW), Q-switched, and mode-locked (ML) bulk or thin-disk lasers operating in these wavelength ranges, require a thorough characterization of the applicable Ho-doped crystals.

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Yttrium aluminium perovskite (YAIO₃ or YAP) is a crystalline material with remarkable optical, thermal, and mechanical properties, which, similar to the well-known yttrium aluminium garnet (Y₃Al₅O₁₂ or YAG), is a compound in the Al₂O₃-Y₂O₃ system [6]. However, in contrast to the cubic YAG, YAP belongs to the orthorhombic symmetry class (space group $D_{2h}^{16} - Pnma$), which renders it biaxial and anisotropic in nature.

The intrinsic birefringence of YAIO₃ crystals and the related anisotropy of the Ho³⁺ emission in this host determine the linearly polarized emission from such solid-state lasers [6,7]. This is especially valuable for applications in nonlinear optical, THz and Raman frequency conversion, where linearly polarized laser sources are needed. It also suppresses thermally induced birefringence effects typical for cubic crystals such as YAG. Additionally, YAIO₃ has a relatively low phonon energy compared to other oxide laser hosts (551 cm⁻¹), see Fig. 1(b), due to the absence of tetrahedral molecular groups in its lattice. The low phonon energies diminish the rates of multiphonon non-radiative relaxation, which is particularly relevant for the development of lasers emitting in the wavelength range between 2 and 3 μm, with the latter considered to be a rough upper limit for any oxide based solid-state laser [8]. The ⁵I₆ → ⁵I₇ Ho³⁺ laser transition in YAIO₃ was known from Xe flash lamp pumping [9] but resonant laser pumping with a tuning element resulted in emission up to 3.017 μm [10].

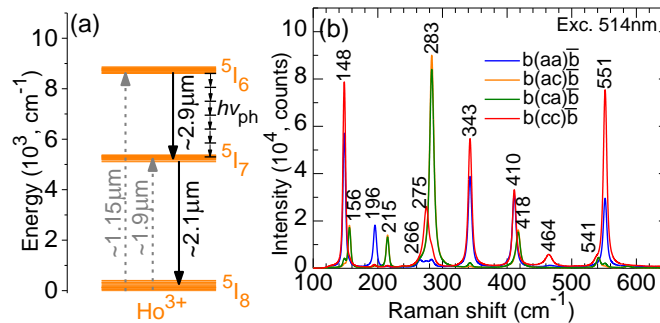


Figure 1. (a) A simplified energy-level diagram of the Ho³⁺ ion showing pump and laser transitions, $h\nu_{ph}$ – phonon energy; (b) polarized Raman spectra of undoped YAIO₃ for the $b(ij)\bar{b}$ geometry ($i, j = a, c$); Porto's notations are used, and the excitation wavelength is $\lambda_{exc} = 514$ nm.

Reliable spectroscopic data are instrumental to optimize the design of Ho:YAIO₃ lasers. In the present work, we report on a comprehensive spectroscopic study of Ho:YAIO₃ crystals bearing in mind their potential for laser development around 2 and 3 μm. In particular, we focus on the polarization anisotropy of the Ho³⁺ emission properties in this optically biaxial material which can explain the polarization switching behavior often observed in Ho:YAIO₃ lasers.

2. RESULTS AND DISCUSSION

2.1 Optical absorption

The optical indicatrix frame for YAIO₃ coincides with the crystallographic axes. With the convention $n_x < n_y < n_z$ for the principal refractive indices the correspondence is $xyz \equiv bac$. We adopt here the $Pnma$ space group for YAIO₃ (if the $Pbnm$ space group is used instead, the correspondence would be $xyz \equiv cba$ [6]). In this work, we denote the principal light polarizations as $E \parallel a, b, c$. A cube from Ho:YAIO₃ was well oriented in this frame and all six faces were polished. Using a single sample rules out any dependence of the results on quality or doping concentration variation. The absorption spectrum in the range of 350 – 2150 nm was acquired using a spectrophotometer (Lambda 1050, Perkin Elmer, spectral resolution: 0.3 – 1 nm) and a Glan-Taylor polarizer.

The polarized absorption spectrum of Ho:YAIO₃ is shown in Fig. 2. It is expressed in absorption cross-sections ($\sigma_{abs} = \alpha_{abs}/N_{Ho}$, α_{abs} – absorption coefficient). The ion density for the 1 at.% Ho³⁺-doped YAIO₃ crystal was 1.97×10^{20} at/cm³. Ho³⁺ has an electronic configuration of [Xe]4f¹⁰ with a ground-state ⁵I₈. In the absorption spectrum, the bands are assigned to electronic transitions from the ⁵I₈ Ho³⁺ manifold to the excited states from ⁵I₇ to ⁵G₄. The absorption spectra are strongly polarized. They are structured suggesting an insignificant temperature broadening for this material owing to a weak electron-phonon interaction.

The ${}^5I_8 \rightarrow {}^5I_7$ transition around 1.9 μm is used for resonant (in-band) pumping of Ho lasers operating at $\sim 2 \mu\text{m}$ [4]. It can be addressed by Tm-fiber lasers or GaSb-based laser diodes. For this transition, the maximum σ_{abs} is $1.29 \times 10^{-20} \text{ cm}^2$ at 1977 nm, corresponding to an absorption bandwidth (full width at half maximum, FWHM) of 7 nm, for light polarization $E \parallel b$. For the other two polarizations, the absorption is weaker, and the dominant absorption peak appears at a shorter wavelength of 1929 nm, $\sigma_{\text{abs}} = 0.98 \times 10^{-20} \text{ cm}^2$ ($E \parallel a$) and $\sigma_{\text{abs}} = 1.18 \times 10^{-20} \text{ cm}^2$ ($E \parallel c$). The absorption bandwidth is similar, 6 and 7 nm, respectively.

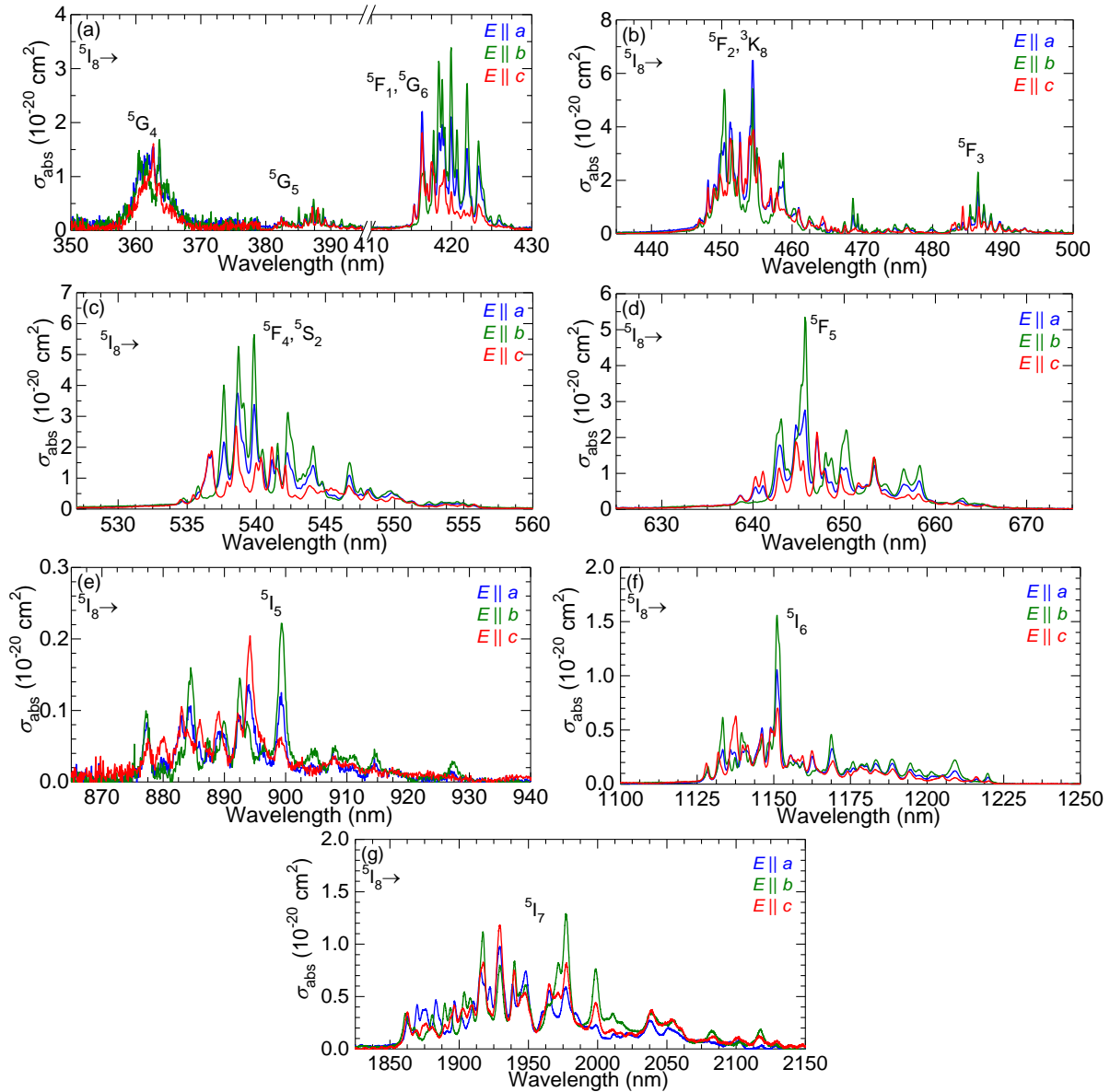


Figure 2. (a)-(f) Polarized absorption spectrum of $\text{Ho}^{3+}:\text{YAlO}_3$, light polarizations: $E \parallel a, b, c$ (space group $Pnma$).

The ${}^5I_8 \rightarrow {}^5I_6$ transition around 1.15 μm is relevant for resonant pumping of Ho lasers emitting at $\sim 2.9 \mu\text{m}$. It matches the emission of Raman fiber lasers [11]. In this spectral range, the maximum σ_{abs} amounts to $1.56 \times 10^{-20} \text{ cm}^2$ at 1151 nm (for light polarization $E \parallel b$) with a FWHM of 2 nm. At the same peak wavelength, the absorption cross-sections for the other two polarizations are lower, $\sigma_{\text{abs}} = 1.06 \times 10^{-20} \text{ cm}^2$ ($E \parallel a$) and $\sigma_{\text{abs}} = 0.70 \times 10^{-20} \text{ cm}^2$ ($E \parallel c$).

2.2 Judd-Ofelt analysis

The transition intensities of Ho³⁺ ions in the YAlO₃ crystal were calculated using the Judd-Ofelt (J-O) formalism by applying both the standard J-O theory [12,13], and its modification accounting for intermediate configuration interaction with the excited configuration of the opposite parity 4fⁿ⁻¹5d¹ (ICI) [14,15]. The set of squared reduced matrix elements U^(k) (k = 2, 4, 6) was calculated using the free-ion parameters reported by Tanner *et al.* [16]. The contribution of magnetic dipole (MD) transitions, following the selection rules ΔS = 0, ΔL = 0, ΔJ = 0, ±1, was computed within the Russell-Saunders approximation based on the wave functions of the Ho³⁺ free ion.

The experimental absorption oscillator strengths <f_{exp}> are listed in Table 1. Here, the polarization averaging is performed as <..> = 1/3(a + b + c). The absorption oscillator strengths calculated using both the J-O and ICI models, f^Σ_{calc}, are also included in Table 1, where the superscript “Σ” indicates the total (ED + MD, ED - electric dipole) value. The intensity parameters are as follows: Ω₂ = 1.280, Ω₄ = 3.708, Ω₆ = 2.066 [10⁻²⁰ cm²] (the standard J-O theory) and Ω₂ = 1.328, Ω₄ = 3.074, Ω₆ = 4.424 [10⁻²⁰ cm²] and R₂ = 0.026, R₄ = -0.031 and R₆ = 0.074 [10⁻⁴ cm] (the ICI model). Here, the R_k (k = 2, 4, 6) parameters describe the configuration interaction.

The root-mean-square (r.m.s.) deviation between the experimental and calculated absorption oscillator strengths is notably reduced when employing the ICI model (0.209), compared to the standard J-O theory (0.362). In addition, the application of the standard model underestimates the radiative lifetime of the lower excited state (⁵I₇). Consequently, the ICI model was adopted for subsequent calculations.

Table 1. Experimental and calculated absorption oscillator strengths for the 1 at.% Ho:YAlO₃ crystal. E_J – energy range; <Γ> = ∫<σ_{abs}(λ)>dλ – integrated absorption cross-section; <f_{exp}>, f^Σ_{calc} – experimental and calculated absorption oscillator strengths, respectively; <..> indicates polarization averaging; ED, MD – electric dipole and magnetic dipole contributions, respectively.

Transition ⁵ I ₈ → ^{2S+1} L _J	E _J , cm ⁻¹	<Γ>×10 ²⁰ , cm ² nm	<f _{exp} >, 10 ⁻⁶	f ^Σ _{calc} , 10 ⁶ J-O	f ^Σ _{calc} , 10 ⁶ ICI
⁵ I ₇	4894 - 5340	73.082	2.155	2.271 ^{ED} +0.579 ^{MD}	1.637 ^{ED} +0.579 ^{MD}
⁵ I ₆	8313 - 8786	16.858	1.412	1.612 ^{ED}	1.281 ^{ED}
⁵ I ₅	11004 - 11398	1.812	0.256	0.308 ^{ED}	0.271 ^{ED}
⁵ F ₅	15214 - 15615	19.491	5.231	5.112 ^{ED}	5.128 ^{ED}
⁵ F ₄ + ⁵ S ₂	18242 - 18610	17.297	6.655	6.388 ^{ED}	6.762 ^{ED}
⁵ F ₃ + ⁵ F ₂ + ³ K ₈ + ⁵ G ₆	20422 - 22265	31.076	16.789	16.707 ^{ED} +0.123 ^{MD}	16.675 ^{ED} +0.123 ^{MD}
⁵ G ₅	23614 - 24018	8.417	5.401	5.606 ^{ED}	5.451 ^{ED}
³ K ₇ + ⁵ G ₄	25625 - 26127	1.848	1.379	0.891 ^{ED} +0.005 ^{MD}	1.016 ^{ED} +0.005 ^{MD}
⁵ G ₂ + ³ H ₆	27377 - 27868	7.138	6.135	5.966 ^{ED}	6.106 ^{ED}
<i>r.m.s. deviation</i>				0.362	0.209

The probabilities of spontaneous radiative transitions A^Σ_{calc}(JJ'), the luminescence branching ratios β(JJ'), and the radiative lifetimes of the excited states τ_{rad} are compiled in Table 2, as determined using the ICI approximation. Here, we considered only the transitions from the ⁵I₇ and ⁵I₆ upper laser levels. Their radiative lifetimes are 6.19 ms and 2.77 ms, respectively, and the luminescence branching ratio for the ⁵I₆ → ⁵I₇ transition around 2.9 μm is as high as 16.3%.

Table 2. Calculated probabilities of emission transitions from the ⁵I₇ and ⁵I₆ manifolds of Ho³⁺ ions in the YAlO₃ crystal: <λ_{em}> - mean emission wavelength, A^Σ_{calc} – probability of spontaneous radiative transitions (ED + MD), β(JJ') – luminescence branching ratio, A_{tot} and τ_{rad} – total probability of radiative spontaneous transitions and radiative lifetime of the excited state, respectively.

J →	J'	<λ _{em} >, nm	A ^Σ _{calc} (JJ'), s ⁻¹	β(JJ')	A _{tot} , s ⁻¹	τ _{rad} , ms
⁵ I ₇	⁵ I ₈	1947.0	119.2 ^{ED} + 42.40 ^{MD}	1	161.6	6.19
⁵ I ₆	⁵ I ₇	2892.7	37.15 ^{ED} + 21.57 ^{MD}	0.163	360.41	2.77
⁵ I ₆	⁵ I ₈	1163.7	301.7 ^{ED}	0.837		

Weber *et al.* reported on the transition intensities for Ho:YAlO₃ based on unpolarized absorption spectra [17]. Their analysis employing the standard J-O formalism yielded $\tau_{\text{rad}}(^5I_7) = 6.41$ ms, $\tau_{\text{rad}}(^5I_6) = 3.00$ ms and $\beta(^5I_6 \rightarrow ^5I_7) = 15.6\%$ which agrees with our refined data.

2.3 Stimulated-emission cross-sections

The luminescence spectra were acquired using an optical spectrum analyzer (Yokogawa AQ6376) purged with N₂ gas to reduce the impact of structured water vapor absorption which spectrally overlaps with both Ho³⁺ ion emissions that we study. A large mode area zirconium fluoride (ZrF₄) fiber was employed for collecting the luminescence. A metal-grid polarizer was used for recording polarized spectra. For excitation, a Ti:Sapphire laser tuned to ~900 nm was employed.

The stimulated-emission (SE) cross-sections, σ_{SE} , were calculated employing the Füchtbauer–Ladenburg (F-L) formula [18]:

$$\sigma_{\text{SE}}^i(\lambda) = \frac{\lambda^5}{8\pi(n)^2\tau_{\text{rad}}c} \frac{\beta(JJ')W_i(\lambda)}{1/3 \sum_{j=a,b,c} \int \lambda W_j(\lambda) d\lambda}, \quad (1)$$

where, λ denotes the wavelength of light, $\langle n \rangle$ is the mean refractive index at the average emission wavelength, c is the speed of light, τ_{rad} stands for the radiative lifetime of the emitting state (⁵I₆ or ⁵I₇), $W_i(\lambda)$ signifies the luminescence spectrum calibrated for the spectral response of the setup, and $i = a, b, c$ indicates the light polarization.

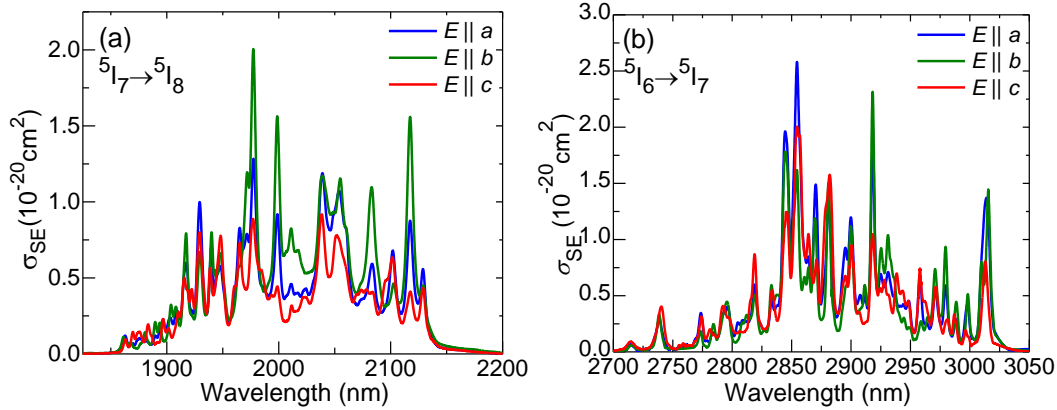


Figure 3. Stimulated-emission (SE) cross-sections, σ_{SE} , of Ho³⁺ in YAlO₃: (a) the ⁵I₇ → ⁵I₈ transition; (b) the ⁵I₆ → ⁵I₇ transition. Light polarizations: $\mathbf{E} \parallel a, b, c$.

The polarized σ_{SE} spectra for Ho³⁺:YAlO₃ are depicted in Fig. 3. For the quasi-three-level ⁵I₇ → ⁵I₈ transition giving rise to 2 μm lasing, in the spectral range where laser emission is expected, the maximum SE cross-section is observed for light polarization $\mathbf{E} \parallel b$, with a value of 2.01×10^{-20} cm² at 1977 nm. Ho³⁺ ions exhibit strong polarization anisotropy of their emission properties as for the other two polarizations the SE cross-sections are lower, $\sigma_{\text{SE}} = 1.28 \times 10^{-20}$ cm² at 1977 nm ($\mathbf{E} \parallel a$) and $\sigma_{\text{SE}} = 0.92 \times 10^{-20}$ cm² at 2038 nm ($\mathbf{E} \parallel c$). This suggests natural selection of linearly polarized emission from Ho:YAlO₃ lasers, along the *b*- or *a*-axis, depending on the chosen cut.

For the ⁵I₆ → ⁵I₇ transition responsible for laser emission near 2.9 μm, the maximum σ_{SE} is determined to be 2.58×10^{-20} cm² at 2854 nm for $\mathbf{E} \parallel a$ light polarization. Due to the long luminescence lifetime of the terminal level for this transition and the occurrence of resonant excited-state absorption on the same transition, ⁵I₇ → ⁵I₆, 2.9-μm Ho-lasers typically operate at longer wavelengths than their zero-phonon lines. The corresponding peak σ_{SE} values are $\sigma_{\text{SE}} = 2.58 \times 10^{-20}$ cm² at 2854 nm ($\mathbf{E} \parallel a$), $\sigma_{\text{SE}} = 2.31 \times 10^{-20}$ cm² at 2918 nm ($\mathbf{E} \parallel b$) and $\sigma_{\text{SE}} = 2.00 \times 10^{-20}$ cm² at 2855 nm ($\mathbf{E} \parallel c$).

2.4 Luminescence decay

The luminescence dynamics study involved a nanosecond optical parametric oscillator (Horizon, Continuum), a 1/4 m monochromator (Oriel 77200), a fast InGaAs detector, and an 8 GHz digital oscilloscope (DSA70804B, Tektronix). The decay from the ⁵I₇ and ⁵I₆ states of Ho³⁺ ions was studied under resonant excitation: $\lambda_{\text{exc}} = 1930$ nm and 895 nm, and

detected at 2020 nm and 1204 nm, respectively. The studied crystal, 1 at.% Ho:YAlO₃, was finely ground into a powder to reduce the effect of radiation trapping (reabsorption) on the measured lifetimes.

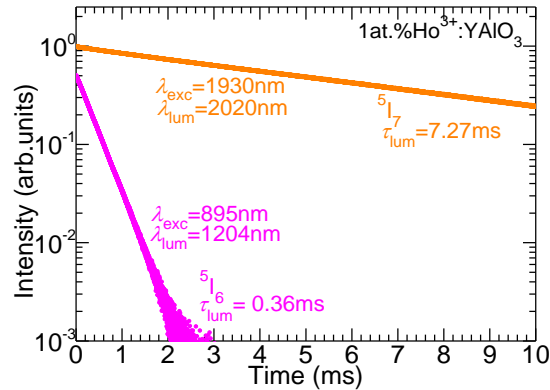


Figure 4. Luminescence decay curves from the 5I_6 and 5I_7 Ho³⁺ states measured under resonant excitation for a 1 at.% Ho:YAlO₃ crystal (a finely powdered sample), λ_{exc} and λ_{lum} – excitation and luminescence wavelengths, respectively, τ_{lum} – luminescence lifetime.

The measured luminescence decay curves, plotted on a semi-logarithmic scale, are presented in Fig. 4. The luminescence decay times τ_{lum} are 7.27 ms for the 5I_7 state and 0.36 ms for the 5I_6 state. The τ_{lum} value for the 5I_7 manifold is longer than the radiative lifetime calculated using the ICI model (6.19 ms) most probably due to the residual reabsorption. Šulc *et al.* reported a 5I_7 luminescence lifetime of $\tau_{lum} = 7.1$ ms for a 0.34 at.% Ho³⁺-doped YAlO₃, in line with our result [19]. The luminescence lifetime for the 5I_6 manifold is much shorter than the calculated radiative lifetime (2.77 ms) due to a notable multiphonon non-radiative relaxation from this level: the energy-gap to the lower-lying state is 3310 cm⁻¹, and the maximum phonon energy of the YAlO₃ host crystal is 551 cm⁻¹, so that a 6-phonon process is expected.

2.5 Crystal-field splitting

To resolve the Stark splitting of the 5I_8 to 5I_6 Ho³⁺ multiplets in YAlO₃, polarized absorption and luminescence spectra were measured at 12 K where virtually all the ions rest in the lowest-energy Stark sub-level of a given manifold, as illustrated in Figure 5. The low-temperature (LT) absorption spectra were plotted versus the photon energy and the LT emission spectra - versus E_{ZPL} – photon energy (both expressed in cm⁻¹) revealing the splitting of the excited-states (5I_7 , 5I_6) and the ground-state (5I_8), respectively. An APD DE-202 closed-cycle cryo-cooler equipped with an APD HC 2 Helium vacuum cryo-compressor and a Laceshore 330 temperature controller were used for the LT studies.

Table 3. Experimental crystal-field splitting of the three lowest Ho³⁺ multiplets in the YAlO₃ crystal.

$^{2S+1}L_J$	Energy E , cm ⁻¹	Number		ΔE , cm ⁻¹
		Theor.	Exp.	
5I_8	0, 21, 52, 68, 85, 141, 156, 164, 169, 203, 234, 272, 335, 370, 386, 421, 446	17	17	446
5I_7	5141, 5180, 5185, 5217, 5221, 5228, 5268, 5287, 5311, 5319, 5335, 5350, 5355, 5368, 5373	15	15	232
5I_6	8683, 8688, 8728, 8734, 8758, 8763, 8770, 8778, 8793, 8834, 8838, 8865, 8869	13	13	186

In YAlO₃, the dopant Ho³⁺ ions replace for the Y³⁺ host-forming cations in a single type of sites of C_s symmetry with a distorted XII-fold oxygen coordination. For C_s symmetry sites, each $^{2S+1}L_J$ multiplet with an integer J is split into a total of $2J + 1$ sub-levels (17 for 5I_8 , 15 for 5I_7 and 13 for 5I_6). All Stark sub-levels were successfully identified. The assignment of the LT spectra followed the early work of Antonov *et al.* [20] considering that the authors performed their studies at 77 K where a noticeable thermal population of the lower-lying Stark sub-levels takes place, so that not all the Stark sub-levels were previously resolved. The experimental energies of the Stark sub-levels of the 5I_8 to 5I_6 Ho³⁺ multiplets are listed in Table 3. The total Stark splitting ΔE of the 5I_8 and 5I_7 terminal laser levels is relatively large: 446 cm⁻¹ and 232 cm⁻¹, respectively, and is thus responsible for the relatively broadband emission of Ho³⁺ in YAlO₃.

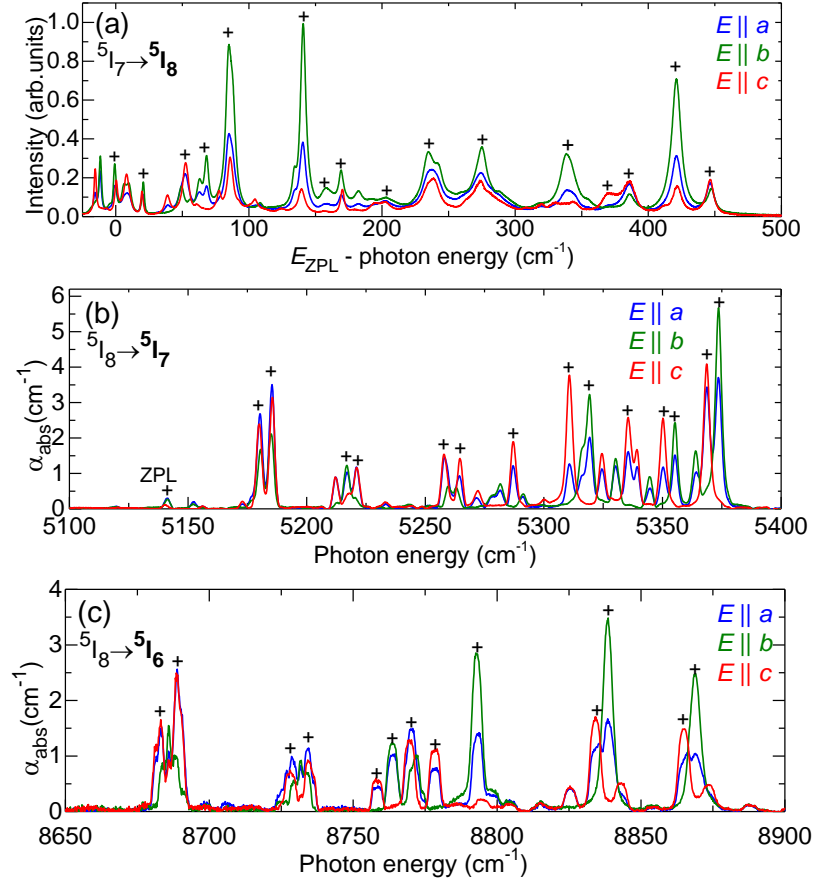


Figure 5. Low-temperature (12 K) spectroscopy of Ho^{3+} ions in YAlO_3 : (a) LT luminescence spectra, the ${}^5\text{I}_7 \rightarrow {}^5\text{I}_8$ transition; (b),(c) LT absorption spectra: (b) the ${}^5\text{I}_8 \rightarrow {}^5\text{I}_7$ transition; (c) the ${}^5\text{I}_8 \rightarrow {}^5\text{I}_6$ transition; “+” indicates peaks assigned to electronic transitions of Ho^{3+} ions in C_s sites.

3. CONCLUSIONS

To conclude, orthorhombic YAlO_3 crystals doped with Ho^{3+} ions exhibit attractive spectroscopic properties for the development of 2 μm and 3 μm lasers, namely (i) relatively broad and intense emission bands due to the ${}^5\text{I}_7 \rightarrow {}^5\text{I}_8$ and ${}^5\text{I}_6 \rightarrow {}^5\text{I}_7$ transitions for polarized light, (ii) large Stark splitting of the terminal laser manifolds (${}^5\text{I}_8$ and ${}^5\text{I}_7$), and (iii) a relatively long ${}^5\text{I}_7$ luminescence lifetime and a reasonably long lifetime of the ${}^5\text{I}_6$ manifold (considering a notable non-radiative path from this state in oxides). Coupled with the good thermal and thermo-mechanical properties of YAlO_3 as a host matrix and its moderately high phonon energy, $\text{Ho}:\text{YAlO}_3$ is of great interest for power-scalable operation around 2 μm both in bulk and thin-disk laser geometries. The long luminescence lifetime of the ${}^5\text{I}_7$ state (~ 7 ms) and the large Stark splitting of the ground-state (446 cm^{-1}) support low-threshold laser operation, while the anisotropy of the laser gain underlines linearly polarized emission. However, also two deficiencies can be predicted: (i) due to the close SE cross-sections spectra for orthogonal eigen-polarization states and expected anisotropy of thermal lensing properties, $\text{Ho}:\text{YAlO}_3$ may be prone to polarization switching upon changing the pump level, output coupling or while tuning the wavelength; (ii) the suggested weak electron-phonon coupling in this matrix results in structured emission spectra with narrow individual emission peaks possibly limiting the generation of ultrashort (femtosecond) pulses from mode-locked lasers; it is difficult to predict the natural selection of the polarization from the present results in the case of broadband oscillation as in mode-locked lasers. The latter can be overcome by employing compositionally “mixed” (solid-solution) $\text{Ho}:(\text{Y,Gd,Lu})\text{AlO}_3$ crystals or by combining two of the principal polarizations into an eigen-polarization in a special polarization-selective geometry of the active element.

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