

Investigation of spectroscopic properties of $\text{LiNbO}_3:\text{Ho}^{3+}$ crystals

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Abstract. In this paper the Stark problem for Ho^{3+} ion ($4f^{10}$ electronic configuration) in LiNbO_3 crystal is solved. Main spectroscopic parameters induced by inter-Stark transitions are determined. Based on analysis of experimental and theoretical data of the main spectroscopic characteristics of $\text{LiNbO}_3:\text{Ho}^{3+}$ crystals, its perceptiveness as a material for optical cooling devices is shown.

1. Introduction

Lithium Niobate crystals LiNbO_3 (LN), doped by Rare Earth (RE^{3+}) are perspective materials for compact multifunction lasers in the infrared and visible regions of the spectrum [1,2], as well as for optical cooling systems and self-cooled lasers [3,4]. Optical spectra of impurity absorption and emission in LN- Ho^{3+} crystal are investigated in [5]. Particularly, the energetic scheme of Stark levels of $^5\text{I}_8$, $^5\text{I}_7$, $^5\text{I}_6$ and $^5\text{I}_5$ multiplets, standard Judd-Ofelt analysis is provided. At the same time, optical spectra of LN- Ho^{3+} crystal in 1.8 -2.2 μm wavelength range, caused by inter-Stark transitions of ground ($^5\text{I}_8$) and first excited ($^5\text{I}_7$) multiplets are little studied experimentally. Meanwhile, these transitions are interesting for LN- Ho^{3+} optical cooling systems. Theoretical investigation of spectroscopic properties of LN- Ho^{3+} crystal in 1.8 -2.2 μm wavelength range is given in [6].

In this work spectroscopic properties of LN- Ho^{3+} crystal in 1.5-2.0 μm wavelength range in order to identify the possibility of establishing optical cooling systems and self-cooled lasers based on LN- Ho^{3+} crystal are discussed.

2. Spectroscopic characteristics of LN- Ho^{3+} crystal

It is well known, that in the crystal field (CF) of LN (point symmetry C_{3v}) ground ($^5\text{I}_8$) and first excited ($^5\text{I}_7$) multiplets of Ho^{3+} ion are splitting to 11th and 10th Stark sublevels respectively [6], transitions between which are defined the spectroscopic properties of the LN- Ho^{3+} crystal.

As known, the intensive lines of impurity absorption and emission spectra of RE^{3+} doped crystals are caused by indirect electro-dipole (IED) and magneto-dipole (MD) transitions, thus the line straight of transition is determined by expression $S_{i \rightarrow f} = \chi_{ed} \cdot S_{i \rightarrow f}^{(ied)} + \chi_{md} \cdot S_{i \rightarrow f}^{(md)}$ (where $S_{i \rightarrow f}^{(ied)}$ and $S_{i \rightarrow f}^{(md)}$ are line straight of IED and MD transitions, χ is local field correction). Un-polarized line straight of inter-stark transitions are [6]: $S_{i,f}^{(ied)} = \sum_t \Omega_t A_t(i, f) |\langle J_f || U_t || J_i \rangle|^2$ and $S_{i,f}^{(md)} = A_{md}(i, f) S_{md}$. Here $\Omega_2 = 4.3 \times 10^{-20}$, $\Omega_4 = 5.11 \times 10^{-20}$, $\Omega_6 = 1.89 \times 10^{-20} \text{ cm}^2$ are Judd - Ofelt's parameters [5],



$\langle {}^5I_7 \| U_2 \| {}^5I_8 \rangle = -0.1477$, $\langle {}^5I_7 \| U_4 \| {}^5I_8 \rangle = 0.3338$, $\langle {}^5I_7 \| U_6 \| {}^5I_8 \rangle = -1.2852$ are reduced matrix elements of the irreducible unit operator U_t , $S^{(md)} = 51\mu^2/e^2$ is line straight of multiplet-to-multiplet MD transition (μ is Bohr's magneton) [6]. Coefficients

$$A_t^{(\text{ied})}(i, f) = \sum_{m=-t}^t \left| \sum_{M_i, M_f} (-1)^{J_f - M_f} a_{J_f M_f}^{*(f)} a_{J_i M_i}^{(i)} \begin{pmatrix} J_f & t & J_i \\ -M_f & m & M_i \end{pmatrix} \right|^2 \quad (1)$$

$$A_{\text{md}}(i, f) = \sum_{m=-1}^1 \left| \sum_{M_i, M_f} (-1)^{J_f - M_f} a_{J_f M_f}^{*(f)} a_{J_i M_i}^{(i)} \begin{pmatrix} J_f & 1 & J_i \\ -M_f & m & M_i \end{pmatrix} \right|^2 \quad (2)$$

determine distribution of intensities of IED and MD transitions by Stark states. Here J is the total angular momentum, M is its projection, $a_{J_i M_i}^{(i)}$ are numerical coefficients in the wave functions of v^{th} Stark state [6], $\begin{pmatrix} J_f & t & J_i \\ -M_f & m & M_i \end{pmatrix}$ is 3j-symbols. Values of calculated line straights of actual transitions and main spectroscopic characteristics, such as absorption coefficients (α), probabilities of spontaneous transitions (A), luminescence branching ratio (β), absorption efficiencies (α) are given in tables 1 and 2.

Table 1. Parameters of ASF spectra of $\text{LiNbO}_3:\text{Ho}^{3+}$ at $T = 300\text{K}$.

transition	$\lambda_{\text{lum}}, \text{nm}$	$S \times 10^{-21} \text{ cm}^2$	$A, \text{ s}^{-1}$	$\delta = N_i/N_0$	$\beta, \%$
$\mu_3 \rightarrow \nu_5$	2020	4.78368	41.8486	0.1244	1.5
$\mu_4 \rightarrow \nu_5$	2013	4.3078	38.08	0.1146	1.3
$\mu_5 \rightarrow \nu_5$	2008	4.93298	43.933	0.1082	1.4
$\mu_6 \rightarrow \nu_6$	2021	7.64323	66.7652	0.1026	2.0

Table 2. Parameters of absorption spectra of $\text{LiNbO}_3:\text{Ho}^{3+}$ at $T = 300\text{K}$.

transition	λ_p, nm	$S, 10^{-21} \text{ cm}^2$	$\alpha, 10^{-26} \text{ cm}^3$	N_i/N_0	$\eta_{\text{abs}}, \%$
$\nu_5 \rightarrow \mu_1$	2035	3.6653	1.4216	0.0754	0.8
$\nu_6 \rightarrow \mu_1$	2053	5.1332	2.0086		1.0
$\rightarrow \mu_2$	2042	6.5145	2.5354	0.0617	1.2
$\rightarrow \mu_3$	2037	4.0845	1.5858		0.8

3. $\text{LiNbO}_3:\text{Ho}^{3+}$ crystal as perspective optical cold material

It is known that for determining the spectral region of expected optical cooling and evaluation of the efficiency of this effect by means of the anti-Stokes fluorescence (ASF) method it is useful to use the following expression [7]

$$\gamma = \eta \left[1 - e^{-(\alpha_p + \alpha_b)L} \right] \frac{\lambda_p - \langle \lambda_f \rangle}{\langle \lambda_f \rangle}. \quad (3)$$

Here γ is the efficiency of the optical cooling effect, η is the quantum yield of fluorescence from the excited level, α_p is the absorption coefficient at the optical excitation wavelength λ_p , α_b is the background absorption of the matrix, L is the thickness of the sample along optical excitation, and $\langle\lambda_f\rangle$ is the mean wavelength of fluorescence: $\langle\lambda_f\rangle = \int_0^\infty \lambda I_f(\lambda) d\lambda / \int_0^\infty I_f(\lambda) d\lambda$, (I_f is the spectral intensity of fluorescence). By using values of spectroscopic characteristics calculated in [6], it is easy to obtain $\langle\lambda_f\rangle = 2002 \text{ nm}$. In figure 1 the absorption spectra of LN - Ho^{3+} at 1.8 - 2.2 μ wavelength [5], the possible implementation of the optical cooling with CW pumping at wavelengths 2035–2053 nm are shown as well. Thus, the effect of optical cooling, on the basis of crystal LN - Ho^{3+} , can be obtained by ASF method at wavelengths $\lambda > 2002 \text{ nm}$. For estimation of efficiency of the optical cooling effect we assume that, first off, intensity of pumping are so far from saturation, second, $\alpha_b \ll \alpha_p$, and third, the probability of zero-phonon transition from $^5\text{I}_7$ multiplet is negligibly small, so the quantum yield from the excited Stark sublevel is mainly determined by the luminescence branching ratio. Then, assuming $L=1\text{cm}$ and $N \approx 10^{21} \text{ cm}^{-3}$, according to Tables 1 and 2, it is easy to estimate the value of γ for different schemes of pumping and emission wavelengths. Where in value of absorption coefficient $\alpha_p(\lambda)$ at λ wavelength is connected with value of α (Table 2) by expression $\alpha_p(\lambda) = \alpha(\lambda)N_i/\Delta\lambda$, where $N_i = \delta N$ is population of ν_i^{th} level (δ is Boltzmann factor). In numerical estimations we use the following values: $\Delta\lambda \approx 10 \text{ nm}$, $N \approx 10^{21} \text{ cm}^{-3}$. Results of evaluation are given in Table 3. In Table 3 is given expected values of temperature decrease $\Delta T = \gamma(\varepsilon_{em} - \varepsilon_p)/k$, (ε_{em} is energy of anti-Stokes luminescence, ε_p is pump energy) as well. It is seen, that temperature of cooling of separate channel is varies in the range of 0.8 - 2.6 K and efficiency is 0.014 - 0.032%, which is comparable with the corresponding characteristics of other materials [8]. Since channels are independent to each other, the total efficiency will be: for 2035nm $\gamma = 0.154\%$ ($\Delta T = 10.83 \text{ K}$) and for 2050 nm $\gamma = 0.17\%$ ($\Delta T = 17.87 \text{ K}$).

Table 3. Temperature and efficiency of optical cooling at $T = 300 \text{ K}$.

transition	$\nu_5 \rightarrow \mu_1$		$\nu_6 \rightarrow \mu_1$		$\nu_6 \rightarrow \mu_2$		$\nu_6 \rightarrow \mu_3$	
	$\Delta T, \text{ K}$	$\gamma, \%$	$\Delta T, \text{ K}$	$\gamma, \%$	$\Delta T, \text{ K}$	$\gamma, \%$	$\Delta T, \text{ K}$	$\gamma, \%$
$\mu_3 \rightarrow \nu_5$	0.83	0.016	2.02	0.018	1.75	0.023	1.27	0.021
$\mu_4 \rightarrow \nu_5$	1.07	0.014	2.05	0.015	2.11	0.021	1.53	0.018
$\mu_5 \rightarrow \nu_5$	1.40	0.015	2.46	0.016	2.6	0.022	2.04	0.020
$\mu_6 \rightarrow \nu_6$	1.08	0.022	2.52	0.023	2.35	0.032	1.61	0.028

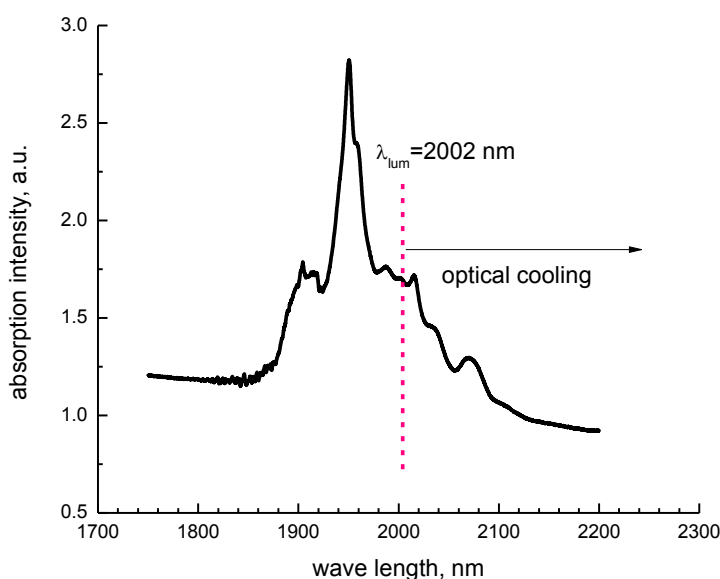


Figure 1. Absorption spectra of LN-Ho³⁺ at 1.8-2.2 μ [5]

4. Conclusion

Estimations show that the LN - Ho³⁺ crystal is perspective material for optical cooling systems. At the same time, it should be noted, that these estimations are rough enough. To get accurate values of efficiency of cooling, it is necessary to do more detailed experimental and theoretical investigations of spectroscopic characteristics of LN - Ho³⁺ crystal including the widths and shifts of spectral lines, as well as examination of their temperature dependences.

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