Photoluminescence and polariton dispersion law in terbium nitrate hydrate crystals

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Abstract. The article examines the dispersion law of polariton waves in terbium nitrate hydrate crystals. We have calculated the values of the unitary polaritons frequencies corresponding to the refractive index equal to unit as well as the velocity and the effective mass of the unitary polaritons. The diagram of terms splitting in the low symmetric crystalline field is constructed. It is shown that the abnormal decrease of the polariton waves’ group velocity occurs due to the splitting of the terbium ions spectral terms in the crystalline field. We consider the conditions for the essential increase of the photoluminescence intensity and decrease of the lasing threshold in terbium nitrate hydrate crystals.

1. Introduction
Electromagnetic radiation in the dielectric medium interacts with the crystal lattice polar vibrations as well as with the exciton states of a semiconductor or a dielectric. These interactions are the most efficient when the frequencies and the wave vectors of the electromagnetic and exciton waves are close, that is when the resonance takes place. Therefore, in such conditions the hybrid (excitonic polaritons) waves with the typical dispersion law \( \omega(k) \) are formed \[1, 2\]. The dispersion of polaritons has found numerous applications in many fields of science and engineering. By changing the polaritons velocities it is possible to design new devices such as filters \[3\], delay lines \[4-7\], scintillators \[8\] etc. varying the crystal absorption by means of injecting the active ions. The recent researches show the most perspective fields to use the polaritons effects for designing optical sources based on the dielectric crystals \[9-12\]. The polaritons theory allows calculating the refractive index and some characteristics of the electromagnetic waves such as the group velocity, the effective mass of polaritons, the spectral thermal energy density and others. The rare earth is interesting for investigating because of its properties. The photoluminescence spectra of the rare earth ions are based on the electron transitions between the levels of the inner 4f-shell, which is screened from the external influences of outer electrons (usually p- and s-shells). It allows the appearance of the exciton terms and excitonic polaritons in bulky materials even at room temperature. The inner-shell 4f-4f transitions comprise both visible and near-infrared ranges \[13\]. They are sharp and easily recognizable. Since these transitions are forbidden by the selection rules, the lifetimes of the excited states are long \[14\]. In
addition, they are almost independent of the chemical environment. They have a high photoluminescence quantum yield [15]. Only a limited number of researches include the information about the polaritons dispersion characteristics of rare earth ions in the dielectric medium. In our earlier research we have already investigated one group of rare earth ions: erbium, ytterbium and thulium [16, 17]. In this step of our research we have obtained information about terbium ions, which are prospective for the laser generating. The examination of paramagnetic terbium ions Tb$^{3+}$ is particularly important because it is necessary to develop the models of solid laser in the visible (green) spectral range with high efficiency and power of laser emission together with its high spatial characteristics. Terbium ions are mostly used as the impurity centers in dielectric mediums. This allows creating population inversion of levels, which leads to laser transitions. Thereto, terbium ions have a set of terms which will be used to realize a three-level scheme of laser generating in the visible (green) range that has not yet been realized. So far there was a tendency in the solid laser technology to focus so much on the research near the infrared range. It is partially related to the fact that for a long time powerful radiation sources capable to pump the active medium in ultraviolet and visible ranges were not developed. Only for the last few decades the new types of diodes have been created in line with the mentioned goals. Moreover, the investigation of terbium optical properties is significantly important in the case of creating the scintillator. This paper presents a method for assessing the optical properties of terbium nitrate hydrate. The excitonic polaritons spectrum of terbium ions was investigated. As a result, the refractive index and the dispersion characteristics of terbium nitrate hydrate were obtained in the wide frequency range. We provide theoretical and experimental analysis by using Maxwell's equations with regard to the external field and oscillations. In particular, we have calculated the velocity of the unitary polaritons, for which the refractive index is unity. It allowed us to present a principal scheme of the terbium laser. Our main goal was to provide the possibility of lowering the lasing threshold by reducing the group velocity when splitting in the crystal field.

2. Experimental research
The research involved using the polycrystalline samples of terbium nitrate hydrate ($\text{Tb(NO}_3)_3\cdot6\text{H}_2\text{O}$). The analysis of the samples optical characteristics was described by recording the spectra of transmission and reflection. The scheme of the experimental setup for observing the photoluminescence spectra is illustrated in Figure 1. As the sources of excitation radiation we used the laser Nd$^{3+}$: YAG, the nitrogen laser and the ultraviolet (UV) light-emitting diodes (1) with the wavelengths of $\lambda=266$, 337, 369 and 385 nm correspondingly. This radiation was directed with the help of the fiber (3) into the cuvette (6) with the samples placed on the table (4) by means of fastening (5). The absorption and photoluminescence spectra were registered on the spectrometer (2). The focusing lens system (7) was used to prevent the divergence of the beam. The experimental data was input to the analog-to-digital converter of the computer for the final processing. Figure 2 illustrates the photoluminescence spectra excited by different light sources. In Figure 2a the photoluminescence spectrum of terbium nitrate hydrate for the diodes pump radiation is given. It should be noted that these relations are normalized according to the maximum experimentally obtained data (curve 1 – pump radiation, curve 2 – photoluminescence spectrum). In Figure 2b the photoluminescence spectrum of terbium nitrate hydrate for the nitrogen laser pump radiation ($\lambda=266$ nm) is given. These relations are normalized according to the maximum experimentally obtained data as well. The low-symmetry crystal field resulted in the appearance of splitting and, consequently, the sub-levels $^5\text{F}_2$ and $^5\text{D}_4$ of the paramagnetic ion Tb$^{3+}$ multiplets (see Figure 3). In this case the nitrogen laser ($\lambda=337$ nm) is used as the source of light.
Figure 1. The schematic of the experimental setup for observing the photoluminescence spectrum of erbium nitrate hydrate: 1 – light-emitting diodes or laser; 2 – spectrometer; 3 – fiber; 4 – table, 5 – adapter; 6 – cuvette, 7 – lens system.

Figure 2. The spectra of the experimental setup for observing the photoluminescence spectrum of terbium nitrate hydrate with the wavelengths of pump radiation a) 385 nm (diode) b) 266 nm (laser).
Figure 3. The photoluminescence spectrum of terbium nitrate hydrate at room temperature under the nitrogen laser excitation (λ = 337 nm).

When excited by different sources a number of lines corresponding to the transitions between different energy levels in terbium ions were revealed. Pumping by different sources, however, has shown the same results: the most intensive peak was at the wavelength of λ=550 nm; the second intensive peak corresponded to the wavelength of 488 nm. In our experimental setup we used different diodes. All of these diodes had the same capacities. However, when we used the λ=385 nm diode the intensity of the photoluminescence spectrum was the highest (see Figure 2a). The photoluminescence spectra of terbium ions in the optical range are based on the electron transitions between the levels of the inner 4f-shell, which is screened from the external influences of outer electrons (usually p- and s-shell). This screening leads to the appearance of the fluorescence narrow spectral lines.

3. Discussion
The free ion of erbium Tb³⁺ has 11 electrons in its 4f shell. The ground-state manifold for terbium ions is ⁷F₆. After the excitation a transition between the ground-state manifold and another manifold is realized, which can be nonradiative or fluorescent. The most complete description of the free ions spectrum was obtained in [18-20]. These researches provide the information about a wide spectral range – from the ultraviolet wavelength region to the infrared one, with the additional calculation of splitting the levels in the magnetic field. All energy levels present in the photoluminescence spectrum are accounted in Table 1. By comparing the data from [18-20] with the data obtained from our experiment, we defined the wavelengths corresponding to the quantum transitions in the 4f-shell free terbium ions in terbium nitrate hydrate. The wavelength λ = 488 nm corresponds to the transition from ⁵D₄ to ⁷F₆ ground state level. The lines at wavelengths λ = 550 nm, 587 nm, 621 nm and 655 nm correspond to the transitions from ⁵D₄ level to the closely placed levels of Tb³⁺. The corresponding transitions between the energy levels are shown in Figure 4. Thus, by using the diode and laser we realized the pumping scheme for the inverse population between ⁵D₄ and ⁷F₆ levels. This diode is prospective for realizing laser generation with the wavelength of 550 nm corresponding to the green spectral range. When we use the diode (λ = 385 nm) the resonant pumping of ⁵D₁ term takes place.
with relaxation to $^5D_4$. As a result the superluminescent emission from $^5D_4$ level to the first excited state $^7F_3$ may be observed (see Figure 2a). The same thing happens if a pulsed UV laser (266 nm) is used (see Figure 2b). Using the pulsed nitrogen laser ($\lambda = 337$ nm) as the exciting source of light results in more complicated photoluminescence spectrum (see Figure 3). Thus, in all these cases the superluminescence and lasing at fairly strong pumping is observed.

**Table 1.** Calculated and experimentally observed energy levels for terbium nitrate hydrate.

| Energy level | Free ions [18] | Terbium nitrate hydrate |
|--------------|----------------|-------------------------|
|              | $\text{Tb}^{3+}$ | $\text{Tb(NO}_3)_3\cdot6\text{H}_2\text{O}$ |
|              | $E$, cm$^{-1}$ | $\lambda$, nm | $E$, cm$^{-1}$ | $\lambda$, nm |
| $^7F_6$      | 0              | $\infty$        | 0              | $\infty$        |
| $^7F_3$      | 2137           | 4679            | 2037           | 4909            |
| $^7F_4$      | 3457           | 2893            | 3256           | 3071            |
| $^7F_3$      | 4525           | 2210            | 4362           | 2293            |
| $^7F_2$      | 5120           | 1953            | 5044           | 1983            |
| $^7F_1$      | 5672           | 1763            | 5478           | 1825            |
| $^7F_0$      | 5244           | 1907            | -              | -               |
| $^5D_4$      | 20634          | 485             | 20488          | 488             |
| $^5D_3$      | 26383          | 379             | -              | -               |

**Figure 4.** The energy diagram of terbium nitrate hydrate pumping at the wavelengths of: a) $\lambda = 385$ nm, b) 266 nm.

4. Theoretical considerations
The theoretical modeling of the electromagnetic wave propagation in erbium nitrate hydrate relies on the Maxwell’s equation [6, 7]:

\[
\begin{align*}
\text{rot} E &= -\frac{\partial B}{\partial t}; \\
\text{div} B &= 0; \\
\text{rot} H &= \frac{\partial B}{\partial t}; \\
\text{div} D &= 0.
\end{align*}
\]

(1)

The material equations are needed to close the Maxwell’s equations. The material equations link the electromagnetic quantities with each other:

\[
\begin{align*}
\dot{D} &= \varepsilon_0 \varepsilon E = \varepsilon_0 \varepsilon \dot{E} + \dot{P}; \\
\dot{B} &= \mu_0 \mu \dot{H} = \mu_0 \mu \dot{H} + \mu_0 \dot{M}.
\end{align*}
\]

(2)

If we want to solve (1), we have the following conditions and materials:

\[\rho(r) = 0, \, j(r) = 0\]

Here, the electric charge density and electric current density are assumed to be zero.

After applying a few vector operations we get the wave equation for the electromagnetic field \(E\):

\[
\text{rot rot} E - \text{grad div} E = -\mu_0 \mu \frac{\partial}{\partial t} \text{rot} \dot{H} = -\mu_0 \mu \varepsilon_0 \varepsilon \frac{\partial^2 \dot{E}}{\partial t^2}.
\]

(3)

The simplest possible solution to the Maxwell’s equations is the plane wave. For monochromatic plane waves in the dielectric medium:

\[
(\nabla^2 - \frac{\varepsilon \mu}{c^2}) \dot{E} = 0.
\]

(4)

Finally, the dispersion law for polaritons waves has the following form:

\[
\omega^2 = \frac{c_0^2 k^2}{\varepsilon(\omega) \mu(\omega)}; \quad \mu(\omega) = 1; \quad \omega^2 = \frac{c_0^2 k^2}{\varepsilon(\omega)};
\]

\[i \varepsilon_0 \varepsilon \ddot{E}_0 \exp \left[ i \vec{k} \cdot \vec{r} - \omega t \right] = 0.
\]

(5)

For the dipole moment and the polarization vector we obtain:

\[
\bar{p}_0 = \frac{e^2 F}{m(\omega_0^2 - \omega^2)} \dot{E}_0;
\]

(7)

\[
\bar{P}_0 = \frac{e^2 F}{m V_0 (\omega_0^2 - \omega^2)} \dot{E}_0.
\]

(8)
The equation of motion for the polarization vector is represented as:

$$\ddot{P} = -\omega_0^2 P + \frac{e^2 F}{mV_0} \ddot{E}; \quad \ddot{P} = \frac{e^2 F\ddot{u}}{V_0} \dot{P}_0 \exp(i(kr - \omega t)).$$

(9)

Introducing the plasma frequency $\omega_p$, from the equation (9) we obtain the relation:

$$\ddot{P} = -\omega_0^2 P + \omega_p^2 \dot{E}; \quad \omega_p^2 = \frac{e^2 F}{mV_0}.$$  

(10)

For the electric displacement we have:

$$\ddot{D}_0 = \varepsilon_0 \ddot{E}_0 + \ddot{P}_0 = \varepsilon_0 [1 + \frac{e^2 F}{mV_0(\omega_0^2 - \omega^2)}] \ddot{E}_0 = \varepsilon_0 \varepsilon(\omega) \ddot{E}_0.$$  

(11)

Therefore, the dispersion of the electromagnetic waves can be described as:

$$\varepsilon(\omega) = 1 + \frac{e^2 F}{mV_0(\omega_0^2 - \omega^2)} = 1 + \frac{\omega_p^2}{\omega_0^2 - \omega^2} = \frac{\omega_0^2 - \omega^2}{\omega_0^2 - \omega^2};$$

$$\omega_p^2 = \omega_0^2 + \omega^2;$$

$$\omega_0^2 = \frac{e^2 F}{mV_0}.$$  

(12)

With regard for the electronic polarizability the dielectric permittivity equation becomes:

$$\varepsilon(\omega) = \varepsilon_\infty \frac{\omega_0^2 - \omega^2}{\omega_0^2 - \omega^2}; \quad \varepsilon_\infty = n_\omega^2.$$  

(13)

Here $\varepsilon_\infty = n_\omega^2$ is the high-frequency permittivity, $\omega_i$ and $\omega_0$ are the frequencies of the corresponding longitudinal and transverse waves. Accordingly, the dispersion law of polaritons in the dielectric media is written as:

$$\omega^2 = \frac{c_0^2 k^2}{\varepsilon(\omega)\mu(\omega)} = \frac{c_0^2 k^2(\omega_0^2 - \omega^2)}{\varepsilon_\infty(\omega_0^2 - \omega^2)} = \frac{c_0^2 k^2(\omega_0^2 - \omega^2)}{(\omega_0^2 - \omega^2)}; \quad c^2 = \frac{c_0^2}{\varepsilon_\infty}.$$  

(14)

Solving (14) gives two frequency-dependent solutions for the wave vector $k$ as a function of $\omega$ since the left-hand side is quadratic in $k$. Close to the absorption two solutions exist. This means that near the resonance the dispersion of the light cannot be considered independently from the dispersion of the excitations. They both form a composite new entity propagating in the medium. The corresponding quantum excitations may be classified as polaritons, resemble to lattice polaritons found in such crystals as NaCl, LiNbO$_3$ and others.

In this case the dielectric function may be presented as:

$$\varepsilon(\omega) = \varepsilon_\infty + \sum_{j=1}^{n_\infty} \frac{q_j N}{m_j \varepsilon_0(\omega_{0j}^2 - \omega^2)}.$$  

(15)

Finally, we obtain the permittivity description known as the Kurosawa relation:
\[ \mathcal{E}(\omega) = \varepsilon_0 \prod_{j=1}^{\omega_n} \frac{\omega_{1j}^2 - \omega^2}{\omega_{0j}^2 - \omega^2}. \]  

(16)

With regard to the solution of the wave equation we come to the polariton dispersion law written as:

\[ \omega^2 = \frac{\varepsilon_0^2 k^2}{\varepsilon_0 \prod_{j=1}^{\omega_n} \frac{\omega_{1j}^2 - \omega^2}{\omega_{0j}^2 - \omega^2}}. \]  

(17)

Here, for simplicity we consider the magnetic permeability to be equal to unity. For the unitary polaritons, for which the refraction index \( n \) is unity, we obtain the following expression:

\[ n^2 = \varepsilon \mu = \frac{c_0^2 k^2}{\omega^2} = 1 \]  

(18)

\[ \frac{c_0^2 k^2}{\omega^2} = \frac{\varepsilon_0^2 c_0^2 k^2}{\varepsilon_0 \prod_{j=1}^{\omega_n} \frac{\omega_{1j}^2 - \omega^2}{\omega_{0j}^2 - \omega^2}} = \varepsilon_0 \prod_{j=1}^{\omega_n} \frac{\omega_{1j}^2 - \omega^2}{\omega_{0j}^2 - \omega^2} = 1 \]  

(19)

The group velocity of polaritons waves in the material can be found from the known relation:

\[ \mathbf{V}(\omega) = \frac{d\omega}{dk} = \left( \frac{dk}{d\omega} \right)^{-1} \]  

(20)

For the effective mass of quasi-particles in the isotropic media we use the formula:

\[ m(\omega) = \left( \frac{d^2E}{dp^2} \right)^{-1} = \hbar^2 \left( \frac{d^2\mathcal{E}}{dk^2} \right)^{-1} = \hbar \left( \mathbf{V}(\omega) \frac{d\mathbf{V}(\omega)}{d\omega} \right)^{-1} \]  

(21)

The work [11] shows that the longitudinal frequencies are of the same order as the transverse frequencies. To make a more exact calculation that will agree more closely with the results of the experiment we must consider \( \omega_{1\omega} = (1,1 \pm 1,2)\omega_{1\omega} \) for the high frequency and \( \omega_{2\omega} = (1,01 \pm 1,02)\omega_{1\omega} \) for the low frequency. Figures 5 and 6 illustrate the calculation dispersion law \( \omega(k) \) for the electromagnetic waves in the visible range with and without splitting.
Figure 5. a) The dispersion curves for the terbium nitrate hydrate in the visible range. The corresponding wavelengths: A1 – 488 nm, A2 – 444 nm, U1 – nm. b) The corresponding transition between the energy levels.

Figure 6. The dispersion curves for the terbium nitrate hydrate in the visible range with splitting.

The parameters of the electromagnetic waves at the points corresponding to the unitary polaritons, for which the refractive index is unity, are shown in Tables 2 and 3. In those points the reflection coefficient R at normal incidence is zero, i.e. the material is characterized by high transparency for the incident radiation.

Table 2. The parameters of the electromagnetic waves at the singular points in the visible range without splitting.

| Singular points | $\omega, 10^{15}$ rad/s | $\lambda, \text{nm}$ | $k, 10^7 \text{m}^{-1}$ | n  | R  | $v, 10^5 \text{m/s}$ |
|-----------------|------------------------|---------------------|------------------------|----|----|---------------------|
| U1              | 4.69                   | 401                 | 1.57                   | 1.00 | 0.00  | 1.75                |
| A1              | 3.86                   | 488                 | -                      | $-\infty$ | 1.00  | 0.00                |
| Singular points | $\omega$, $10^{15}$rad/s | $\lambda$, nm | $k$, $10^3$ m$^{-1}$ | n | R | $v$, $10^3$ m/s |
|----------------|--------------------------|-------------|-----------------|---|---|----------------|
| U1             | 3.81                     | 494         | 1.27            | 1.00 | 0.00 | 0.09           |
| U2             | 3.84                     | 491         | 1.28            | 1.00 | 0.00 | 0.17           |
| U3             | 3.87                     | 487         | 1.29            | 1.00 | 0.00 | 0.77           |

Table 3. The parameters of the electromagnetic waves at the singular points in the visible range with splitting.

5. Applications
Nowadays one of the main applications of the rare earth elements is to use them as the active medium in fiber optics and lasers. Nevertheless, industrial lasers based on terbium do not currently exist. As it has already been mentioned this is because the first high-power UV LED, allowing for pumping of the active medium in the visible range, appeared only at the end of the last century. Due to its high sensitivity and intensity the phosphors based on the rare earth elements have a high quantum yield. It is possible to create a more efficient high-energy radiation detector even at low intensity. The experimental and theoretical works presented in this paper allow using terbium as the phosphors in such detectors. The experimental setup presented in Figure 1 may be used for the superluminescence and lasing with $\lambda = 550$ observation [16, 17] in the dielectrics doped with the terbium ions. The high efficiency inverse population pumping is reached due to the “stopping of light” when the frequency of the exciting emission coincides with the spectral positions of the unitary polariton excitation (points U1, U2 and U3 in Figure 6). The group velocities of the corresponding polariton waves are very small and the probability of the exciting emission absorbance by the dielectric crystals becomes very high. For the unitary polaritons (points U1, U2, U3 in figure 6) the energy density of the laser electric field inside the discussed dielectrics essentially increases (on several orders) and lasing may be reached at fairly small exciting light intensity.

6. Conclusions
Summarizing, in this paper we have presented and analyzed the photoluminescence spectrum of terbium nitrate hydrate. The theoretical and experimental results for the energy level of free ions of Tb$^{3+}$ and terbium nitrate hydrate levels have been obtained. The dispersion dependencies of the polaritons waves in terbium nitrate hydrate have been established. The positions of the unitary polaritons, for which the refraction index is unity, have been determined. It was shown that the group velocities of the unitary polaritons are thirty times less than the speed of light in the vacuum. The increase of the efficiency of the laser generating is justified. The experimental schemes for the terbium laser have been proposed. The possibility of laser generating at the wavelength of 550 nm has been shown.

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