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Er³⁺-doped tellurofluorophosphate glasses for lasers and optical amplifiers

M Jayasimhadri¹, L R Moorthy^{1,3}, K Kojima², K Yamamoto²,
Noriko Wada² and Noriyuki Wada²

¹ Department of Physics, Sri Venkateswara University, Tirupati-517502, India

² Department of Applied Chemistry, Faculty of Science and Engineering, Ritsumeikan University, Kusatsu, Shiga 525-8577, Japan

E-mail: lrphysics@yahoo.co.in

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Abstract

Absorption, fluorescence and decay properties of erbium-doped alkali tellurofluorophosphate glasses (RTFP) with the molar compositions of 50 (NaPO₃)₆–10 TeO₂–20 AlF₃–19 RF–1 Er₂O₃ (R = Li, Na or K) have been investigated in order to utilize these materials for the development of fibre amplifiers and lasers. The Judd–Ofelt (JO) model was applied to the absorption intensities of Er³⁺ (4f¹¹) transitions to determine the JO intensity parameters Ω_λ ($\lambda = 2, 4$ or 6). These evaluated intensity parameters are used to calculate the spontaneous emission probabilities and branching ratios from the excited state J manifolds to the lower lying J' manifolds of Er³⁺ emission transitions. The radiative lifetimes of these excited states are determined from their respective spontaneous emission probabilities. Visible and NIR emission spectra have been studied for all three Er³⁺-doped tellurofluorophosphate glasses. In particular, for the emission at 1.5 μm corresponding to the $^4\text{I}_{13/2} \rightarrow ^4\text{I}_{15/2}$ transition, radiative parameters like the full width at half maximum (FWHM), emission cross-sections (σ_e) and figure of merit ($\text{FWHM} \times \sigma_e$) were determined and compared with other hosts. The measured lifetimes of the $^4\text{S}_{3/2}$ level are compared with predicted radiative lifetimes calculated from the JO theory. The potentiality of Er³⁺-doped tellurofluorophosphate glasses for devices such as lasers and optical amplifiers is discussed.

1. Introduction

In recent years, the incorporation of rare-earth ions into solids has attracted more attention due to potential applications such as planar waveguide amplifiers, short wavelength laser sources, optical amplifiers for telecommunications and solid-state frequency upconverters [1–5].

³ Author to whom any correspondence should be addressed.

Er³⁺-doped glasses are especially attractive for numerous applications such as microchip lasers [6, 7], erbium-doped fibre amplifiers (EDFAs) in wavelength division multiplexing (WDM) systems [8, 9], near infrared telecommunication windows [10], eye safe laser systems [11] and lidar transmitters [12]. In addition, the Er³⁺-doped fluoride fibre laser at 3 μm radiation acts as a compact and efficient source for applications in medical and micro-surgery [13, 14]. In the current trend, wavelength division multiplexing systems require amplifier materials with increased transmission capacity in order to obtain broader gain bandwidth for the development of computer networks and other data transmitting services. A wide bandwidth optical amplifier is vital for the development of multichannel transmission systems. The amplification bandwidth of the conventional SiO₂-based EDFA is limited [15, 16]. Host glasses based on tellurium oxide doped by Er³⁺ ions exhibit large stimulated emission cross-section and broader fluorescence half-width at 1.5 μm [17]. The low phonon energies of these glasses reduce the non-radiative transition rates of the rare-earth ions when compared to silica and borate glasses [18]. Moreover, wide infrared transmittance, good chemical durability and also the large refractive index values influence the emission cross-sections of rare-earth ion transitions in tellurite-based glasses [15, 19]. With the addition of fluorides into the tellurite-based rare-earth-doped glasses, one can obtain broad amplifier bandwidth with an intrinsically flat gain required for the development of optical transmission networks [20]. These special optical properties as well as their potential applications motivated us to undertake the present study.

In the present work, a Judd–Ofelt (JO) analysis [21, 22] for the absorption bands of Er³⁺ ions in tellurofluorophosphate glass host was performed in order to determine the optical intensity parameters, spontaneous emission probabilities, branching ratios and radiative lifetimes for various excited manifold-to-manifold transitions. Furthermore, complete knowledge of various radiative spectroscopic properties is essential in order to design and develop optical glass materials for device applications. From the fluorescence spectra, the emission cross-sections, bandwidths and lifetimes for certain luminescent levels of Er³⁺ in tellurofluorophosphate glasses are estimated. In particular, the fluorescence properties of the $^4\text{I}_{13/2} \rightarrow ^4\text{I}_{15/2}$ transition located around 1534 nm are discussed. The measured lifetimes and non-radiative relaxation rates are also reported for the excited $^4\text{S}_{3/2}$ level in all the tellurofluorophosphate glasses.

2. Experimental procedure

The alkali tellurofluorophosphate (RTFP) glasses used in the present study were prepared by doping 1 mol% of Er with the chemical compositions of 50 (NaPO₃)₆–10 TeO₂–20 AlF₃–19 LiF–1 Er₂O₃ (LiTFP); 50 (NaPO₃)₆–10 TeO₂–20 AlF₃–19 NaF–1 Er₂O₃ (NaTFP); and 50 (NaPO₃)₆–10 TeO₂–20 AlF₃–19 KF–1 Er₂O₃ (KTFP). Batches of 10 g starting materials of Er₂O₃, (NaPO₃)₆, TeO₂, AlF₃ and RF (R = Li, Na and K) were fully mixed and melted in a platinum crucible at 960 °C for 30 min in an electrical furnace. Then the melt was poured into a preheated brass mould and annealed for 10 h at a temperature close to the softening point of the glasses (350 °C). These samples were polished to flat and parallel faces for optical measurements. The refractive indices were measured on an Abbe refractometer by using the sodium lamp as the light source. The density measurements were made by the Archimedes method with water as the immersion liquid. The Er³⁺ concentrations were calculated from the initial glass composition and the sample densities. The refractive indices, densities and the Er³⁺ concentrations of the three glass samples are shown in table 1.

Electronic absorption spectra were recorded with a Varian Cary 5E UV–VIS–NIR spectrophotometer over a spectral range of 340–1650 nm. The visible fluorescence spectra in

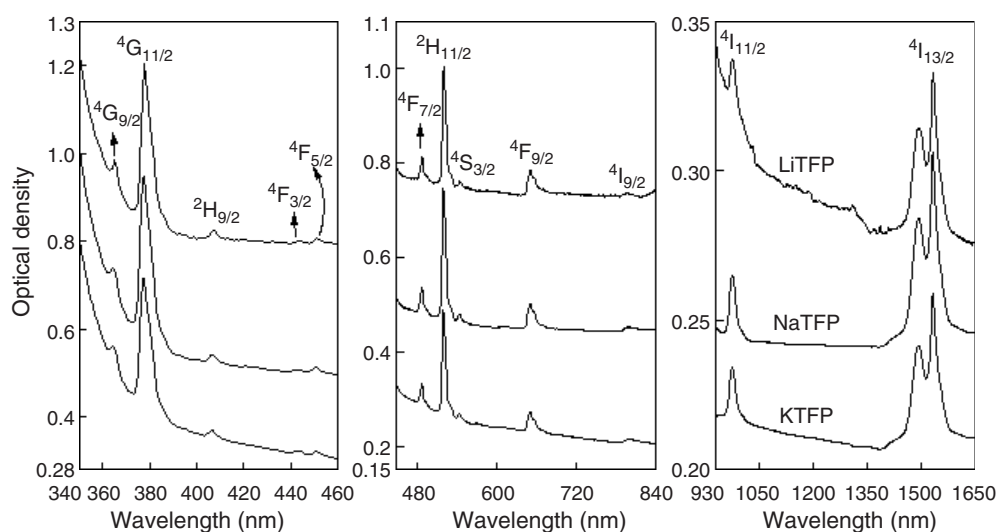


Figure 1. UV-VIS-NIR absorption spectra of Er³⁺-doped RTFP glasses.

Table 1. Measured physical properties for Er³⁺:RTFP glasses.

Physical quantities	LiTFP	NaTFP	KTFP
Sample thickness (mm)	2.50	2.37	2.26
Refractive index	1.585	1.586	1.582
Density (g cm ⁻³)	2.62	2.62	2.60
Concentration (mol l ⁻¹)	0.151	0.150	0.147
Concentration (ions cm ⁻³ × 10 ²⁰)	0.907	0.902	0.885

the range 500–700 nm were measured with a Hitachi F-3010 fluorescence spectrophotometer by exciting the samples at 380 nm, using a xenon lamp as an excitation source. The near-infrared fluorescence spectra in the wavelength range 1400–1700 nm were measured by using a computer controlled 970 nm diode laser (SDL-6362-P1) as an excitation source, InGaAs photo diode detector, lock-in-amplifier (Li-570A) and monochromator (Ni Kon G-250). Room-temperature decay lifetimes for the ⁴S_{3/2} level were measured with a pulsed tunable laser (Lambda Physics, Scan Mate OPPO) under 540 nm excitation by monitoring the emission at 546 nm, and the signal was collected by using a digital oscilloscope (Le Croy LS 140 100 Mz).

3. Results and discussion

3.1. Absorption spectra and Judd–Ofelt analysis

The recorded optical absorption spectra of Er³⁺-doped alkali tellurofluorophosphate glasses are shown in figure 1. A total of 12 absorption bands are identified and the assignment of the transitions from the ground state ⁴I_{15/2} to various excited states are indicated. The most intense absorption bands at 378 and 520 nm are attributed to the ⁴I_{15/2} → ⁴G_{11/2} and ⁴I_{15/2} → ²H_{11/2} hypersensitive transitions, respectively [23].

The experimental oscillator strengths f_{exp} can be determined from the room-temperature absorption spectra by [24]

$$f_{\text{exp}} = \frac{mc^2}{N\pi e^2} \int \varepsilon(\nu) d\nu \quad (1)$$

where the term before the integral is represented by atomic constants (m and e are the mass and charge of the electron, c is the velocity of light). The integral itself corresponds to the area under the absorption curve. $\varepsilon(\nu)$ is the molar absorption coefficient and f is a dimensionless quantity. These f_{exp} values have been used to determine the JO intensity parameters Ω_λ by a least squares fit. According to the JO theory [21, 22], the calculated oscillator strengths from the initial state to an excited state are

$$f_{JJ'} = \frac{8\pi^2 m c \nu}{3 h e^2 (2J + 1)} [\chi S_{\text{ed}}(JJ') + n S_{\text{md}}(JJ')] \quad (2)$$

where $\chi = \frac{(n^2+2)^2}{9n}$ is a factor for the effective field at a well-localized centre in a medium with an isotropic refractive index and S_{ed} and S_{md} are the electric and the magnetic dipole line strengths which are given by

$$S_{\text{ed}}(JJ') = e^2 \Sigma \Omega_\lambda (\psi J || U^\lambda || \psi' J') \quad (3)$$

$$S_{\text{md}}(JJ') = \left(\frac{e \hbar}{4 \pi m c} \right) (\psi J || L + 2S || \psi' J')^2 \quad (4)$$

where $||U^\lambda||$ are the double reduced matrix elements of the unit tensor operator of rank $\lambda = 2, 4, 6$ calculated from the intermediate coupling approximation [25]. In our JO analysis we have utilized the reduced matrix elements reported by Ravi Kanth Kumar *et al* [26], since these matrix elements depend only on the rare-earth ion and not on the host material. A measure of the accuracy of the fit between the experimental and calculated oscillator strengths is given by the root mean square (rms) deviation

$$\sigma_{\text{rms}} = \left[\frac{\Sigma (f_{\text{exp}} - f_{\text{cal}})^2}{N} \right]^{1/2} \quad (5)$$

where N is the number of levels included in the fit. The small rms deviations of ± 0.299 , ± 0.224 and ± 0.247 ($\times 10^{-6}$) shown in table 2 indicate good fit between the experimental and calculated oscillator strengths. However the effect of the host on the JO parameters influences the radiative transition probabilities, since they contain the crystal-field parameters, inter-configurational radial integrals and the interaction between the central ion and the environment [27]. In table 3, the magnitudes of the three JO intensity parameters evaluated in the present work increase in the order $\Omega_2 > \Omega_4 > \Omega_6$ and are comparable with other reported values for different hosts [27–30]. Generally, the Ω_2 parameter is sensitive to the symmetry of the rare-earth site and is strongly affected by covalency between rare-earth ions and ligand anions, whereas Ω_4 and Ω_6 are related to the rigidity of the host medium in which the ions are situated [31]. In the present work, the Ω_2 values are larger than that of Er^{3+} -doped fluorophosphate [23], YAG [28], silicate and fluoride [32] host materials, which indicates that Er^{3+} -doped tellurofluorophosphate glasses are more covalent in character, with higher symmetry. The spectroscopic quality factor ($X = \Omega_4/\Omega_6$) [33] values determined for Er^{3+} in LiTFP, NaTFP and KTFP are found to be 0.934, 0.743 and 0.477 respectively, which are within the range of 0.5–1.0 for Er^{3+} ions in different hosts [27–30].

The magnetic dipole line strength (S_{md}) exists only for certain transitions between the states which obey the transition selective rules $\Delta S = \Delta L = 0$, $\Delta J = 0, \pm 1$ [21, 22]. In order to obtain broadband and flat $1.5 \mu\text{m}$ emission spectra, it is more essential to increase the relative contribution of the electric dipole transition (S_{ed}) [24]. But, S_{md} is independent of the ligand fields and is a characteristic of the particular transition determined by the quantum numbers, whereas S_{ed} is a function of glass structure and composition and can be calculated according to equation (3), by using JO theory. However, it is possible to increase the electric dipole contribution (S_{ed}) by modifying the composition and the structure of the host glass. The

Table 2. Experimental (f_{exp}) and calculated (f_{cal}) oscillator strengths ($\times 10^{-6}$) for the absorption bands of Er³⁺ in RTFP glasses.

Transition	LiTFP			NaTFP			KTFP		
	Energy	f_{exp}	f_{cal}	Energy	f_{exp}	f_{cal}	Energy	f_{exp}	f_{cal}
$^4I_{15/2} \rightarrow$									
$^4I_{13/2}$	6 517	1.4892	1.3183	6 517	1.5868	1.4550	6 517	1.5804	1.4232
$^4I_{11/2}$	10 233	0.2658	0.6346	10 243	0.5445	0.7258	10 243	0.5173	0.7054
$^4I_{9/2}$	12 559	0.1502	0.2448	12 528	0.2814	0.2186	12 512	0.3841	0.1456
$^4F_{9/2}$	15 333	2.0065	1.8787	15 357	1.8793	1.8674	15 357	1.4791	1.5621
$^4S_{3/2}$	18 344	0.2747	0.5120	18 377	0.3016	0.5707	18 377	0.2875	0.5736
$^2H_{11/2}$	19 189	7.0682	7.3971	19 226	8.7936	8.9613	19 226	7.6577	7.5408
$^4F_{7/2}$	20 486	1.4308	1.9634	20 486	1.7379	2.1043	20 486	1.5114	2.0034
$^4F_{5/2}$	22 167	0.3451	0.6176	22 167	0.3155	0.6872	22 167	0.3761	0.6907
$^4F_{3/2}$	22 516	0.1790	0.3545	22 567	0.1515	0.3953	22 567	0.1171	0.3973
$^2H_{9/2}$	24 563	0.4130	0.7315	24 624	0.5245	0.7999	24 563	0.5239	0.7798
$^4G_{11/2}$	26 448	13.3940	13.0256	26 448	15.9365	15.7534	26 448	13.1320	13.2567
$^4G_{9/2}$	27 390	0.9177	1.2199	27 465	1.1034	1.1894	27 465	1.0584	0.9568
rms ($\times 10^{-6}$)		± 0.299			± 0.224			± 0.247	

Table 3. A comparison of evaluated Judd–Ofelt intensity parameters Ω_λ (10^{-20} cm²) of Er³⁺ in different hosts.

Glass	Ω_2	Ω_4	Ω_6	Order	Ω_4/Ω_6	Reference
LiTFP	4.697	1.210	1.296	$\Omega_2 > \Omega_6 > \Omega_4$	0.934	This work
NaTFP	5.918	1.070	1.441	$\Omega_2 > \Omega_6 > \Omega_4$	0.743	This work
KTFP	5.086	0.693	1.453	$\Omega_2 > \Omega_6 > \Omega_4$	0.477	This work
Phosphate	6.280	1.030	1.390	$\Omega_2 > \Omega_6 > \Omega_4$	0.740	[27]
YSGG	0.920	0.480	0.870	$\Omega_2 > \Omega_6 > \Omega_4$	0.550	[28]
LSS	7.610	1.850	2.090	$\Omega_2 > \Omega_6 > \Omega_4$	0.880	[29]
PSS	4.990	1.510	2.320	$\Omega_2 > \Omega_6 > \Omega_4$	0.650	[29]
CaLiBO	3.680	0.760	1.520	$\Omega_2 > \Omega_6 > \Omega_4$	0.500	[30]

calculated line strength ratios of $S_{\text{ed}}/(S_{\text{ed}}+S_{\text{md}})$ for the $^4I_{13/2} \rightarrow ^4I_{15/2}$ transition of Er³⁺-doped LiTFP (0.752), NaTFP (0.769) and KTFP (0.766) glasses are found to be larger than those of fluoride (0.683), silicate (0.675), phosphate (0.652), germanate (0.568) glasses [34, 35], thereby indicating that Er³⁺:RTFP glasses are more preferable for device applications.

The JO intensity parameters presented in table 3 are used to estimate the radiative transition probabilities between an excited state (J) and the lower-lying terminal levels (J') using the following expression:

$$A_{\text{R}}(J, J') = \frac{64\pi^4\nu^3}{3h(2J+1)} \left[\frac{n(n^2+2)^2}{9} S_{\text{ed}} + n^3 S_{\text{md}} \right]. \quad (6)$$

The total radiative transition probability $A_{\text{T}}(J) = \sum_{J'} A_{\text{R}}(J, J')$ of any excited state can be obtained by adding the radiative transition probabilities $A_{\text{R}}(J, J')$ calculated for all the terminal states. The radiative lifetimes τ_{R} of an excited state (J) can be extracted from the total radiative transition probability using the expression

$$\tau_{\text{R}}(J) = \frac{1}{\sum_{J'} A_{\text{R}}(J, J')} = [A_{\text{T}}(J)]^{-1}. \quad (7)$$

The predicted radiative lifetimes for various excited luminescent levels of Er³⁺:RTFP glasses shown in table 4 are comparable with those obtained in other different hosts [27, 30, 36].

Table 4. Comparison of predicted radiative lifetimes (τ_R) for various excited levels of Er^{3+} :RTFP glasses with different hosts.

Excited states	LiTFP (μs)	NaTFP (μs)	KTFP (μs)	Phosphate (μs) [27]	LiBO (μs) [30]	GPTC (μs) [36]
$^4\text{G}_{11/2}$	43	36	43	—	78	—
$^2\text{H}_{9/2}$	319	283	309	—	—	—
$^4\text{F}_{5/2}$	362	337	357	—	686	—
$^4\text{F}_{7/2}$	288	275	301	290	534	—
$^2\text{H}_{11/2}$	153	127	151	120	—	67
$^4\text{S}_{3/2}$	575	513	514	560	1 111	340
$^4\text{F}_{9/2}$	753	747	888	880	1 333	364
$^4\text{I}_{9/2}$	6076	6340	8046	7780	10 694	3150
$^4\text{I}_{11/2}$	5565	4896	5070	6140	10 342	3300
$^4\text{I}_{13/2}$	6836	6345	6488	9960	11 320	3640

In all three glasses the radiative lifetimes of the excited states decreases in the order $^4\text{I}_{13/2} > ^4\text{I}_{9/2} > ^4\text{I}_{11/2} > ^4\text{F}_{9/2} > ^4\text{S}_{3/2} > ^4\text{F}_{5/2} > ^2\text{H}_{9/2} > ^4\text{F}_{7/2} > ^2\text{H}_{11/2} > ^4\text{G}_{11/2}$ with an exception of the $^4\text{I}_{9/2}$ level in the KTFP glass sample. This may be due to the decrease of Ω_4 in KTFP glass, when compared with LiTFP and NaTFP glasses. The fluorescence branching ratios β_R from the relaxation state (J) to a particular final level (J') can be determined from the radiative decay rates by

$$\beta_R(J, J') = \frac{A_R(\psi J; \psi' J')}{A_T(J, J')} = A_R(J, J')\tau_R. \quad (8)$$

The fluorescence branching ratio is a critical parameter for the laser designer, because it characterizes the possibility of attaining stimulated emission on any specific transition.

3.2. Visible fluorescence spectra

The partial level energy diagram of Er^{3+} in tellurofluorophosphate glasses is shown figure 2, with visible and near-infrared emissions indicated by dotted and solid lines. The visible fluorescence spectra of Er^{3+} -doped tellurofluorophosphate glasses are shown in figure 3. Four emission transitions $^2\text{H}_{11/2} \rightarrow ^4\text{I}_{15/2}$, $^4\text{S}_{3/2} \rightarrow ^4\text{I}_{15/2}$, $^4\text{F}_{5/2} \rightarrow ^4\text{I}_{13/2}$, and $^4\text{F}_{9/2} \rightarrow ^4\text{I}_{15/2}$, centred nearly at 520, 550, 625 and 674 nm respectively, are detected for Er^{3+} :RTFP glasses upon 380 nm excitation. The peak stimulated emission cross-section $\sigma_e(\lambda_p)$, which is essential in predicting the performance of lasing action, can be determined from the following expression:

$$\sigma_e(\lambda_p) = \frac{\lambda_p^4}{8\pi cn^2 \Delta\lambda_{\text{eff}}} A_R(J, J') \quad (9)$$

where λ_p is the peak wavelength and $\Delta\lambda_{\text{eff}}$ is the effective linewidth of the emission band. From table 5, it is observed that the two emission levels corresponding to $^2\text{H}_{11/2} \rightarrow ^4\text{I}_{15/2}$ and $^4\text{F}_{9/2} \rightarrow ^4\text{I}_{15/2}$ transitions possess large emission cross-sections and high branching ratios, thereby suggesting that these transitions have the most potential for visible laser emission.

3.3. NIR fluorescence spectra

The near-infrared fluorescence spectra of Er^{3+} -doped RTFP glasses measured at 1.5 μm corresponding to the $^4\text{I}_{13/2} \rightarrow ^4\text{I}_{15/2}$ transition upon 970 nm laser excitation are shown in

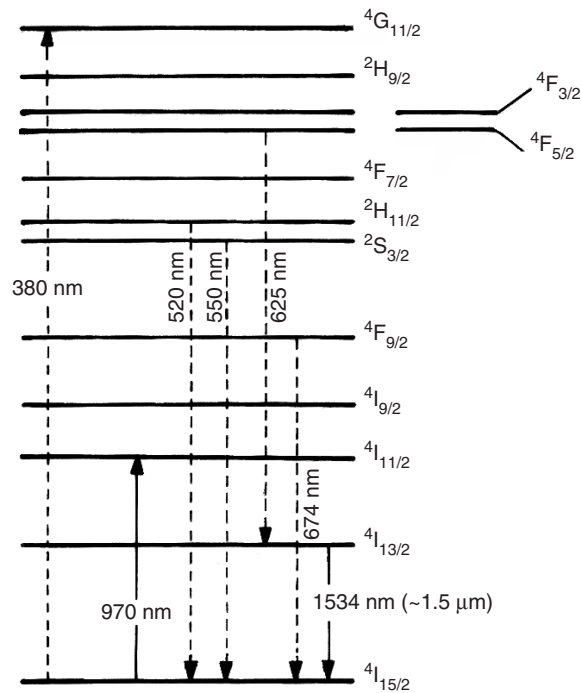


Figure 2. Partial energy level diagram showing visible and near-infrared emissions in Er³⁺-doped tellurofluorophosphate glasses.

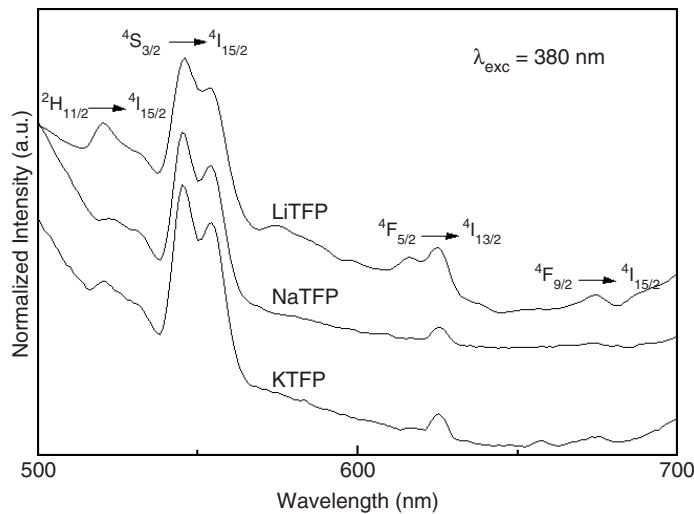


Figure 3. UV-excited visible fluorescence spectra of Er³⁺-doped RTFP glasses.

figure 4. The 970 nm laser excitation wavelength corresponds to the ${}^4I_{11/2} \leftarrow {}^4I_{15/2}$ state, which in turn non-radiatively relaxes to ${}^4I_{13/2}$. From this state, only the radiative emission to the ${}^4I_{15/2}$ ground state is possible, since the energy gap is nearly $\sim 6500 \text{ cm}^{-1}$. The peak emission wavelengths, radiative transition rates, stimulated emission cross-sections (σ_e) and

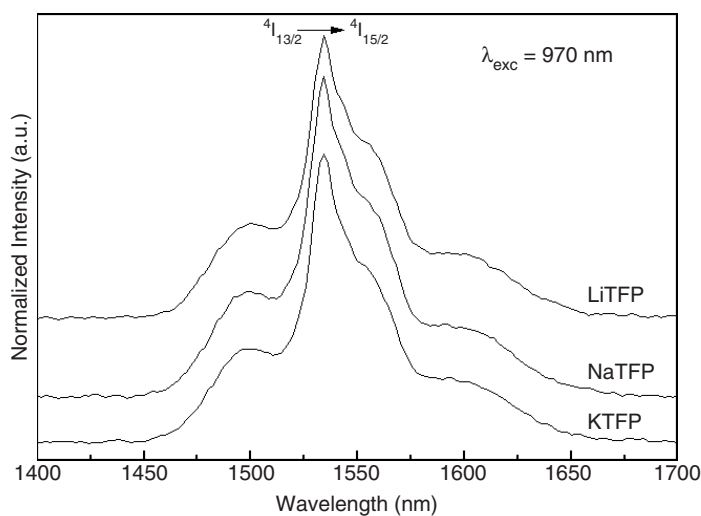


Figure 4. Near-infrared fluorescence spectra of the ${}^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition centred at 1534 nm in Er^{3+} -doped tellurofluorophosphate glasses under 970 nm excitation.

Table 5. Emission peak wavelengths (λ_p nm), radiative transition probabilities (A , s^{-1}), stimulated emission cross-sections ($\sigma_e \times 10^{-22}$) and branching ratios (β) for Er^{3+} :RTFP glasses.

Level	λ_p	LiTFP			NaTFP			KTFP		
		A	σ_e	β_{cal}	A	σ_e	β_{cal}	A	σ_e	β_{cal}
${}^2H_{11/2} \rightarrow {}^4I_{15/2}$	520	6085.20	345.63	0.933	7409.86	426.87	0.939	6203.82	450.12	0.935
${}^4S_{3/2} \rightarrow {}^4I_{15/2}$	550	1169.13	34.09	0.673	1309.52	40.53	0.673	1309.54	40.88	0.674
${}^4F_{5/2} \rightarrow {}^4I_{13/2}$	625	1094.27	100.88	0.400	1137.17	188.50	0.384	1023.73	142.90	0.366
${}^4F_{9/2} \rightarrow {}^4I_{15/2}$	674	1184.92	166.20	0.893	1183.34	219.75	0.884	985.39	158.56	0.876
${}^4I_{13/2} \rightarrow {}^4I_{15/2}$	1534	146.27	112.92	1.000	157.59	117.02	1.000	154.12	124.21	1.000

radiative branching ratios (β_R) for the ${}^4I_{13/2} \rightarrow {}^4I_{15/2}$ near-infrared emission of Er^{3+} -doped tellurofluorophosphate glasses are also included in table 5. On combining equations (6) and (9), one can observe that the stimulated emission cross-section (σ_e) is directly proportional to the refractive index of the glass host ($\sigma_e \sim (n^2 + 2)^2/n$) [18] and inversely proportional to the half-width of the emission band. Moreover, it is apparent that Er^{3+} :RTFP glasses possess large stimulated emission cross-sections to utilize these glasses for broadband and high-gain amplification devices. A figure of merit (FOM) for the bandwidth is the product $\text{FWHM} \times \sigma_e$, which indicates the gain bandwidth of an amplifier. The ${}^4I_{13/2} \rightarrow {}^4I_{15/2}$ emission properties like FWHM, σ_e and FOM of Er^{3+} ions in different glass hosts [37–39] are shown in table 6 along with the refractive indices. Generally transitions with large stimulated emission cross-sections exhibit low threshold and high gain laser operation. From table 6, it can be realized that Er^{3+} :RTFP glasses are good host materials for broadband amplifiers and also for laser applications in the near-infrared region.

3.4. Luminescence decay

The radiative decay from the ${}^4S_{3/2}$ level of Er^{3+} ions in tellurofluorophosphate glasses is fitted with a nearly single exponential function as shown in figure 5. The lifetime (τ_{meas}) of any

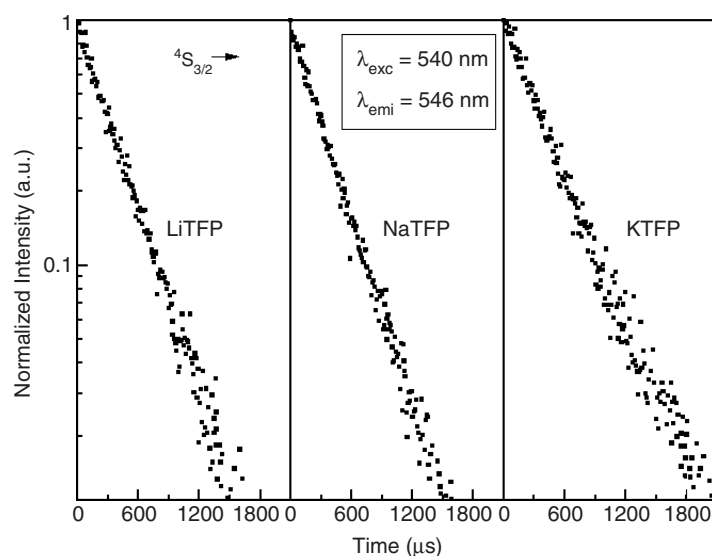


Figure 5. Luminescence decay profile of the $^4S_{3/2}$ level in Er^{3+} :RTFP glasses.

Table 6. NIR emission parameters of the $^4I_{13/2} \rightarrow ^4I_{15/2}$ transition for Er^{3+} in various glass hosts.

Glasses	Refractive index	FWHM	σ_e ($\times 10^{-21}$ cm ²)	FOM ($\sigma_e \times$ FWHM)	Reference
LiTFP	1.585	37.9	11.29	427.9	This work
NaTFP	1.586	39.3	11.70	459.8	This work
KTFP	1.582	36.4	12.42	452.1	This work
Silicate	1.585	40	5.50	220	[37]
Phosphate	1.569	37	6.40	236.8	[38]
Germanate	1.625	53	5.68	301	[39]

excited state is defined as the $1/e$ decay time of the fluorescence intensity. The measured lifetimes of the $^4S_{3/2}$ level in LiTFP, NaTFP and KTFP are 330, 314 and 403 μs , respectively. These values are comparable to those reported for BaF_2 - ThF_4 (300 μs) [40], oxyfluoroborate (239 μs) [41] and fluorozirconate (398 μs) [42] glasses. The predicted radiative lifetimes for the $^4S_{3/2}$ level using JO theory are 575, 513 and 514 μs for LiTFP, NaTFP and KTFP respectively. From the measured and predicted radiative lifetimes, the quantum efficiency $\eta = \tau_{meas}/\tau_R$ for the transition $^4S_{3/2} \rightarrow ^4I_{15/2}$ at 0.5 μm of Er^{3+} :RTFP glasses are found to be approximately 57%, 61% and 78% respectively. The non-radiative relaxation rate ($W_{NR} = 1/\tau_{meas} - 1/\tau_R$) for Er^{3+} -doped LiTFP, NaTFP and KTFP obtained are 1291, 1235 and 536 s^{-1} , respectively. The luminescence decay properties such as lifetimes, quantum efficiencies, and non-radiative relaxation rates are given in table 7. However, we were not able to measure the decay times and in turn the quantum efficiencies for the emission transition $^4I_{13/2} \rightarrow ^4I_{15/2}$ at 1534 nm due to the lack of experimental facilities in the near-infrared region.

4. Conclusion

Optical absorption, fluorescence and luminescence decay analyses of Er^{3+} -doped tellurofluorophosphate glasses have been performed. The Judd–Ofelt model has been applied

Table 7. Quantum efficiencies, lifetimes and non-radiative relaxation rates for the ${}^4S_{3/2}$ level of Er^{3+} :RTFP glasses.

Host	η (%)	τ		W_{NR} (s^{-1})
		τ_{meas} (μs)	τ_{cal} (μs)	
LiTFP	57.4	330	575	1291
NaTFP	61.2	314	513	1235
KTFP	78.4	403	514	536

to calculate the JO intensity parameters from the measured oscillator strength of the absorption spectra of Er^{3+} :RTFP glasses. The spectroscopic quality factors ($X = \Omega_4/\Omega_6$), which are critically important in predicting the stimulated emission of any laser active medium, are determined. The spectroscopic quality factors for Er^{3+} in LiTFP, NaTFP and KTFP are found to be 0.934, 0.743 and 0.477, which are higher than that of Er^{3+} :YAG (0.32) [28]. The line strength ratios ($S_{ed}/S_{ed} + S_{md}$) calculated for the ${}^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition shows that these glasses possess larger gain bandwidth at $1.5 \mu m$. The radiative lifetimes, spontaneous emission probabilities and branching ratios for all the excited ${}^4G_{11/2}$, ${}^2H_{9/2}$, ${}^4F_{5/2}$, ${}^4F_{7/2}$, ${}^2H_{11/2}$, ${}^4S_{3/2}$, ${}^4F_{9/2}$, ${}^4I_{9/2}$, ${}^4I_{11/2}$ and ${}^4I_{13/2}$ states are predicted theoretically. From the visible emission spectra, the emission cross-sections for the ${}^2H_{11/2} \rightarrow {}^4I_{15/2}$, ${}^4S_{3/2} \rightarrow {}^4I_{15/2}$, ${}^4F_{5/2} \rightarrow {}^4I_{13/2}$ and ${}^4F_{9/2} \rightarrow {}^4I_{15/2}$ transitions are reported. For the near-infrared emission transition at $1.5 \mu m$, the emission parameters such as FWHM, σ_e , FOM are determined and compared with other hosts. Measured lifetimes and quantum efficiencies for the ${}^4S_{3/2}$ level in LiTFP, NaTFP and KTFP have been reported at room temperature. The results of these investigations indicate that Er^{3+} -doped tellurofluorophosphate glasses may be useful for the development of lasers and fibre amplifiers.

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References

- [1] Vermelho M V D, Gonveia-Neto A S, Amorim H T, Cassanjes F C, Ribeiro S J L and Messaddeq Y 2003 *J. Lumin.* **102/103** 755
- [2] Voronoko Y K, Sobol A A, Karasik A Y, Eskov N A, Rabochkina P A and Ushakov S N 2002 *Opt. Mater.* **20** 197
- [3] Lazaro J A, Valles J A and Rebolledo M A 1999 *IEEE. J. Quantum Electron.* **35** 827
- [4] Cheng C 2004 *Opt. Laser Technol.* **36** 607
- [5] Hattori K, Kitagawa T, Oguma M, Okazaki H and Ohmori Y 1996 *J. Appl. Phys.* **80** 5301
- [6] Liu Z, Qi C, Dai S, Jiang Y and Hu L 2003 *Opt. Mater.* **21** 789
- [7] Laporta P, Taccheo S, Longhi S, Svelto O and Svelto C 1999 *Opt. Mater.* **11** 269
- [8] Jing H and Jianquan Y 2005 *Opt. Commun.* **251** 132
- [9] Yeh C H, Lee C C and Chi S 2004 *Opt. Commun.* **241** 333
- [10] Van Deun R, Nockemann P, Gorller-Walrand C and Binnemans K 2004 *Chem. Phys. Lett.* **397** 447
- [11] Mierczyk Z, Kwasny M, Kopczynski K, Gietka A, Lukasiewicz T, Frukacz Z, Kisielowski J, Stepień R and Jedrzejewski K 2000 *J. Alloys Compounds* **300/301** 398
- [12] Dragic P D 2005 *Opt. Commun.* **250** 403

- [13] Hibst R 1992 *Lasers Surg. Med.* **12** 125
- [14] Nagel D 1997 *Lasers Surg. Med.* **21** 79
- [15] Ohishi Y, Mori A, Yamada M, Ono H, Nishida Y and Oikawa K 1998 *Opt. Lett.* **23** 274
- [16] Atkins C G, Massicott J F, Armitage J P, Wyatt R, Ainslie B J and Craig-ryan S P 1989 *Electron. Lett.* **25** 910
- [17] Mori A, Ohishi Y and Sudo S 1997 *Electron. Lett.* **33** 863
- [18] Wang J S, Vogel E M and Snitzer E 1994 *Opt. Mater.* **3** 187
- [19] Resfield R 1975 *Struct. Bonding* **22** 123
- [20] Sidebottom D L, Green P F and Brow R K 1997 *J. Non-Cryst. Solids* **222** 354
- [21] Judd B R 1962 *Phys. Rev.* **127** 750
- [22] Ofelt G S 1962 *J. Chem. Phys.* **37** 511
- [23] Vand Deun R, Binnemans K, Gorller-Walrand C and Adam J L 1999 *J. Alloys Compounds* **283** 59
- [24] Rama Moorthy L, Srinivasa Rao T, Janardhnam K and Radhapaty A 2000 *Spectrochim. Acta A* **56** 1759
- [25] Carnall W T, Crosswhite H and Crosswhite H M 1978 *Energy Level Structure and Transition Probabilities of Trivalent Lanthanides in LaF₃* (Aronne, IL: Argonne National Laboratories)
- [26] Ravi Kanth Kumar V V, Jayasankar C K and Jagannathan R 1996 *Phys. Status Solidi b* **195** 287
- [27] Sardar D K, Gruber J B, Zandi B, Hutchinson J A and Trussell C W 2003 *J. Appl. Phys.* **93** 2041
- [28] Sardar D K, Bradley W M, Perez J J, Gruber J B, Zandi B, Hutchinson J A, Trussell C W and Kokta M R 2003 *J. Appl. Phys.* **93** 2602
- [29] Lakshman S V J and Ratnakaram Y C 1987 *J. Non-Cryst. Solids* **94** 222
- [30] Renuka Devi A and Jayasankar C K 1996 *J. Non-Cryst. Solids* **197** 111
- [31] Weber M J 1982 *J. Non-Cryst. Solids* **47** 117
- [32] Tanabe S 1999 *J. Non-Cryst. Solids* **259** 1
- [33] Kaminskii A A 1981 *Laser Crystals* (Berlin: Springer)
- [34] Pauling L 1929 *J. Am. Chem. Soc.* **51** 1010
- [35] Porque J, Jiang S, Hwang B C, Fuflyigin V, Salley E, Zhao J and Peyghambarian N 2000 *Proc. SPIE* **3942** 60
- [36] Pan Z, Morgam S H, Dyer K, Ueda A and Liu H 1996 *J. Appl. Phys.* **79** 8906
- [37] Zou X and Izumitani T 1993 *J. Non-Cryst. Solids* **162** 68
- [38] Jiang S, Luo T, Hwang B C, Smekatala F, Seneschal K, Lucas J and Peyghambarian N 2000 *J. Non-Cryst. Solids* **263/264** 364
- [39] Lin H, Pun E Y B, Man S Q and Liu X R 2001 *J. Opt. Soc. Am. B* **18** 602
- [40] Yeh D C, Pertin R R, Sibley W A, Madigou V, Adam J L and Suscavage M J 1989 *Phys. Rev. B* **39** 80
- [41] Kumar A, Rai D K and Rai S B 2002 *Spectrochim. Acta A* **58** 3067
- [42] Tsuda M, Soga K, Inoue H and Makishima A 2000 *J. Appl. Phys.* **88** 1900