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Author(s): Markus P. Hehlen, Timothy R. Gosnell, Nigel J. Cockroft, Allan J. Bruce, W. H. Grodkiewicz, Gerry Nykolak, Joseph Shmulovich, Ruby Gosh, and M. R. X. Barros

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Yb^{3+},Er^{3+} Codoped Silica-Based Glasses for Planar Optical Waveguide Amplifiers

Markus P. Hehlen, Timothy R. Gosnell, Nigel J. Cockroft
Los Alamos National Laboratory, Mailstop E535, Los Alamos, NM 87545.

Allan J. Bruce, W.H. Grodkiewicz, Gerry Nykolak, Joseph Shmulovich, Ruby Gosh, M.R.X. Barros
AT&T Bell Laboratories, 600 Mountain Ave., Murray Hill, NJ 07974.

Abstract

The spectroscopic properties of Er^{3+}-doped and Yb^{3+},Er^{3+}-codoped sodium- and alumino-silicate glasses, potential planar optical waveguide amplifier materials for 1.55 μm, are investigated. With the exception of $^4I_{13/2}$, multiphonon relaxation is dominant for all Er^{3+} excited-state multiplets, allowing efficient population of the emitting $^4I_{13/2}$ state by $^4I_{11/2}$ excitation around 980 nm. In both glasses, multiphonon relaxation from $^4I_{13/2}$ is negligible up to 678 K, and the respective 300 K absolute quantum yields are ~0.9 for 0.05 mol% Er^{3+}-doped samples under low-power $^4I_{11/2}$ excitation. OH^- impurities are able to efficiently quench $^4I_{13/2}$ excitation. Absolute $^4I_{13/2}$ quantum yields are used to predict an optimum Yb^{3+},Er^{3+} concentration range for sensitization. Yb^{3+} codotation strongly enhances pump-light absorption and increases $^4I_{13/2}$ population by almost two orders of magnitude in some samples. Relative $^2H_{11/2}/^4S_{3/2}$ upconversion luminescence intensities are shown to be a sensitive measure for the substantial internal sample heating observed in Yb^{3+} codoped glasses.

Keywords: Rare earth doped materials, Glass and other amorphous materials, Spectroscopy-fluorescence and luminescence, Integrated optics-components-materials

Introduction

The superb performance of silica optical fibers and Er^{3+} doped fiber amplifiers (EDFA) led to a revolution of high-speed telecommunication in the last decade [1]. In long-distance optical networks, amplification is required to compensate for signal attenuation and losses in the silica fiber, and with the use of Er^{3+} all-optical rather than electro-optical amplification can be achieved [2]. The Er^{3+} energy-level structure (Figure 1a) permits pumping of the $^4I_{11/2}$ excited state around 980 nm by semiconductor lasers, and the $^4I_{13/2}$ state is populated by subsequent multiphonon relaxation. The $^4I_{13/2} \rightarrow ^4I_{15/2}$ emission may then provide gain around 1.55 μm, a wavelength which matches the attenuation minimum of silica fibers. Significant inhomogenous broadening of the optical transitions in the disordered glass host allows wavelength multiplexing, and it is possible to operate several channels with different wavelengths at multi-gigabit per second data transmission rates in a single fiber link.

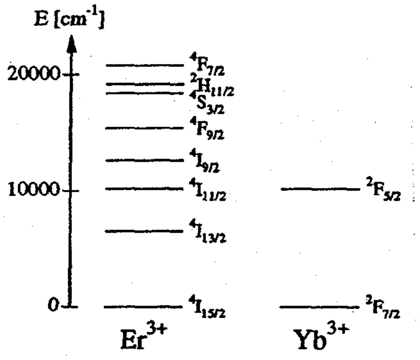

Figure 1. Relevant $^{2S+1}L_J$ multiplets of a) Er^{3+} and b) Yb^{3+}.

EDFA's consist of various components including several meters of doped fiber, splitters, multiplexers, filters, pump laser, and electronics, typically resulting in a bulky and costly device. Although not inhibitive for main optical data networks these drawbacks hinder the realization of fiber-to-the-home. Today, individual consumers are still linked by conventional copper-wire technology which involves low-speed, service-intensive, electronic components creating bottlenecks in the overall data capacity.

The highly mature silicon optical bench technology offers the alternative of integrating planar optical waveguides, semiconductor diode lasers, and a variety of passive optical elements on a single silicon chip (Figure 2). This potential for significant miniaturization of optical amplifiers has created a considerable research interest in new Er^{3+} based amplifier materials. The restriction to short device lengths in planar optical waveguide amplifiers (POWA) requires much higher Er^{3+} concentrations than the typical ~100-300 ppm doping used in EDFA's in order to achieve sufficient pump-light absorption and net gain. Recently, the feasibility of POWA's has been demonstrated [4-6], and the highest net gain of 15 dB was observed in a 14600 ppm Er^{3+}-doped 4.5 cm long sodium silicate planar waveguide pumped at 980 nm with 280 mW [6]. At these high Er^{3+} concentrations, energy-transfer processes such as upconversion and energy migration to impurity sites followed by non-radiative relaxation become important and degrade the $^4I_{13/2}$ quantum yield. In all POWA's demonstrated so far, high pump powers, usually beyond typical outputs of semiconductor diode lasers, were required to produce useful net gain [4-6].

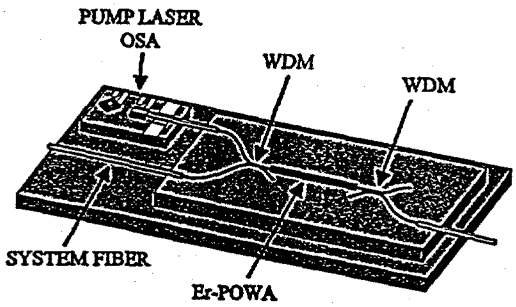

Figure 2. Conceptual design of an Er^{3+} planar optical waveguide amplifier (POWA) module which integrates all of the optical components on a small silicon chip [3].

The Yb^{3+} ion has been known for a long time as an efficient sensitizer for Er^{3+} [7]. The energies of $^2F_{5/2}$ of Yb^{3+} and $^4I_{11/2}$ of Er^{3+} are very similar (Figure 1), and excitation created in $^2F_{5/2}$ can be transferred to $^4I_{11/2}$ almost resonantly. In addition, both the oscillator strength and the peak absorption of Yb^{3+} around 980 nm are more than an order of magnitude higher than those for Er^{3+} in this wavelength region. Potentially, strong pump-light absorption by Yb^{3+} can be achieved while maintaining Er^{3+} concentrations at relatively low levels. In this paper, we present results of a spectroscopic investigation of an extensive series of Er^{3+}-doped and Yb^{3+},Er^{3+}-codoped sodium-silicate and alumino-silicate glasses. Compared to standard silica, these glasses offer a 10 to 20 times greater solubility for rare-earth ions without clustering and therefore provide a wide range for optimization of Er^{3+} and Yb^{3+} concentrations for potential codoped POWA materials.

Results and Discussion

The single-ion excited-state dynamics of Er^{3+} in silica-based glasses is mainly governed by the competition of radiative and multiphonon relaxation. The oscillator strengths of near-infrared and visible Er^{3+} absorptions of a variety of sodium and alumino silicates with different glass compositions have been analyzed in terms of the Judd-Ofelt theory. Table 1 shows $^4I_{11/2}$ and $^4I_{13/2}$ radiative relaxation rate constants k_{rad} calculated from this analysis for two representative samples. The oscillator strengths in alumino silicates are systematically higher, resulting in higher values for k_{rad}. The highest-energy vibrational Si-O modes $\hbar\omega_{max}$ are the most likely acceptors in multiphonon relaxation. Their energies, 1100 and 1015 cm^{-1} in sodium- and alumino-silicate glass, respectively [8], are much higher than kT at 300 K, and the multiphonon relaxation rate constant k_{mp} can therefore be approximated by the low-temperature energy-gap law [9]

$$k_{mp} = \beta e^{-\alpha(\Delta E - 2\hbar\omega_{max})} \tag{1}$$

Multiphonon relaxation rate constants, calculated from Eq.(1) using 6600 and 3650 cm^{-1} for the $^4I_{13/2}$ and $^4I_{11/2}$ energy gaps, respectively, and the material parameters α=4.7·10^{-3} cm and β=9.0·10^7 Hz [9], are compared with k_{rad} in Table 1. For both glasses, multiphonon relaxation from $^4I_{13/2}$ can be neglected

Table 1. Comparison of calculated radiative (Judd-Ofelt theory) and multiphonon (energy-gap law Eq.(1)) relaxation rate constants in [s^{-1}] for Er^{3+} doped sodium and alumino silicates at 300 K.

$^{2S+1}L_J$	Sodium silicate		Alumino silicate	
	k_{rad}	k_{mp}	k_{rad}	k_{mp}
$^4I_{13/2}$	74	1.10^{-1}	150	$4\cdot10^{-2}$
$^4I_{11/2}$	79	1.10^{5}	186	$5\cdot10^{4}$

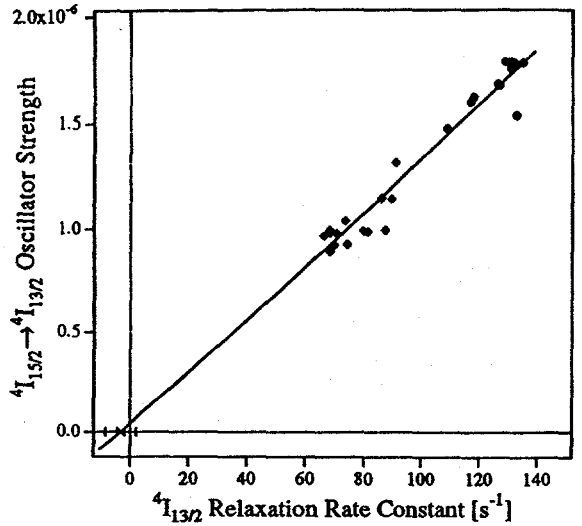

Figure 3. $^4I_{15/2}\rightarrow{}^4I_{13/2}$ oscillator strengths versus $^4I_{13/2}$ relaxation rate constants (300 K) for 0.4 mol% Er^{3+}-doped sodium (squares) and alumino (circles) silicates. The linear fit intersects the x-axis at -3.1±5.1 s^{-1}.

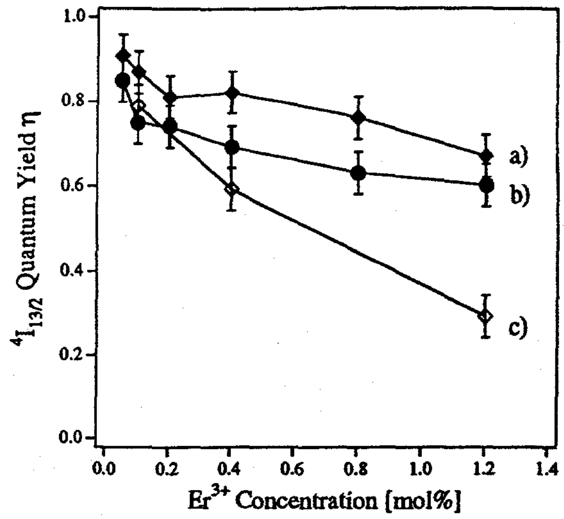

Figure 4. Absolute $^4I_{13/2}\rightarrow{}^4I_{15/2}$ luminescence quantum yields at 300 K for a series of Er^{3+}-doped sodium silicates (a), alumino silicates (b), and high-OH^- sodium silicates (c) using $^4I_{11/2}$ excitation around 980 nm.

whereas more than 99.9% of the $^4I_{11/2}$ population decays non-radiatively to $^4I_{13/2}$. Figure 3 shows $^4I_{15/2}\rightarrow{}^4I_{13/2}$ oscillator strengths for several 0.4 mol% Er^{3+} doped sodium- and alumino-silicate glasses as a function of measured $^4I_{13/2}$ relaxation rate constants. The linear fit intersects at the origin within the experimental accuracy, indicating the absence of non-radiative relaxation for $^4I_{13/2}$. This behavior is in sharp contrast to low-phonon materials such as fluorides, chlorides, and bromides, which have strongly suppressed multiphonon decay and $^4I_{11/2}$ lifetimes of typically more than 3 ms.

Depending on the glass preparation procedures, traces of OH^- impurities can be incorporated in these materials. The highest-energy vibrational OH^- mode has been measured at 3850 cm^{-1}, and the respective calculated $^4I_{13/2}$ multiphonon relaxation rate constant is $1.6\cdot10^{10}$ s^{-1} [Eq.(1)]. The effect of such high-energy OH^- modes in the coordination sphere of Er^{3+} can be seen in the absolute $^4I_{13/2}$ luminescence quantum yields η shown in Figure 4. A series of sodium silicates intentionally prepared with elevated OH^- concentration shows a much more pronounced drop of η with increasing Er^{3+} concentration than the low-OH^- sample series. With the onset of $^4I_{13/2}$ excitation migration for Er^{3+} concentrations >0.2 mol%, the minority of Er^{3+} ions coordinated to OH^- impurities can be accessed, resulting in significant quenching of $^4I_{13/2}$ population. Minimizing OH^- levels, *e.g.* by remelting glass preforms under sub-ppm H_2O inert atmosphere, is therefore one essential step in the manufacturing procedures of glasses optimized for Er^{3+} POWA applications.

The overall efficiency of Yb^{3+} sensitization of $^4I_{13/2}$ emission relies on several factors: (i) the quantum yield of the $^2F_{5/2}$ Yb^{3+} state, (ii) the energy migration among Yb^{3+} ions, (iii) the energy-transfer step from Yb^{3+} to Er^{3+}, and (iv) the $^4I_{11/2}$ relaxation properties. For a given host the efficiencies of processes (iii) and (iv) are characteristic for the material and cannot be varied in a wide range. The Yb^{3+} energy-migration

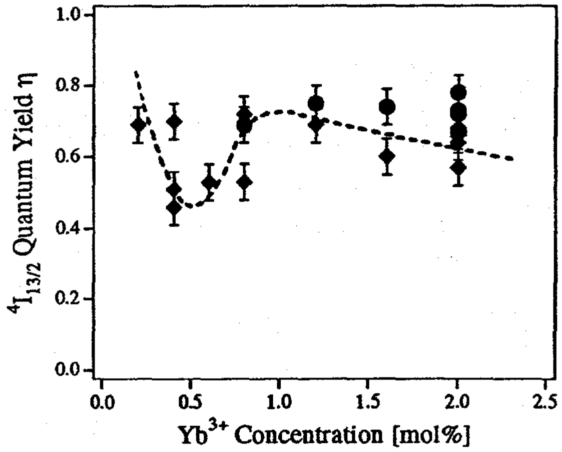

Figure 5. Absolute $^4I_{13/2}\rightarrow{}^4I_{15/2}$ luminescence quantum yields at 300 K for a series of 0.4 mol% Er^{3+}-doped sodium silicates (squares) and alumino silicates (circles) codoped with Yb^{3+} and excited around 980 nm.

properties on the other hand can be controlled through variation of the Yb^{3+} concentration. Absolute $^4I_{13/2}$ luminescence quantum yields η are a direct measure for the overall efficiency of Yb^{3+} sensitization and are suited to predict favorable Er^{3+} and Yb^{3+} concentration regimes. Figure 5 presents η values for a series of 0.4 mol% Er^{3+}-doped sodium and alumino silicates codoped with Yb^{3+}. Three regimes are found: At very low Yb^{3+} concentration the samples exhibit the Er^{3+}-like behavior (Figure 4) with high η for both glass types. At 0.4..0.8 mol%, Yb^{3+} ions are on average separated by ~17..14 Å, and Yb-Yb energy migration is not efficient yet. In combination with the absorption occurring mainly on Yb^{3+} this leads to a decrease of the $^4I_{13/2}$ quantum yield. At >0.8 mol%, the average Yb^{3+} ion separation is smaller than <14 Å, a regime typical for the onset of electric-dipole induced energy transfer [10]. $^2F_{5/2}$ excitation on Yb^{3+} can therefore access Er^{3+} which act as efficient traps, restoring η to relatively high values. Neglecting saturation effects, the absorption rate of a 0.4 mol% Er^{3+} and a 0.4 mol% Er^{3+}, 2 mol% Yb^{3+} codoped sample differs by a factor of ~80, and the almost indentical quantum yields (~0.8) for these samples indicate a $^4I_{13/2}$ population increase of almost two orders of magnitude. Figure 5 suggests 1 to 2 mol% Yb^{3+} codotation of 0.4 mol% Er^{3+} samples to be a useful regime. At higher Yb^{3+} concentrations, quenching sites for Yb^{3+} become relevant and degrade the $^2F_{5/2}$ and thus the overall quantum yield.

Each multiphonon relaxation from $^4I_{11/2}$ to $^4I_{13/2}$ releases ~3650 cm^{-1} of energy as heat into the host This, together with multiphonon relaxation following upconversion, leads to internal sample heating particularly for Yb^{3+} codoped samples in which absorption rates and consequently multiphonon relaxation rates are strongly enhanced. The relatively weak Er^{3+} upconversion luminescence in the visible provides quantitative information on the extent of internal sample heating. The two close-lying states $^4S_{3/2}$ and $^2H_{11/2}$ (Figure 1a) can be considered a closed system, and the luminescence intensities are a measure for the respective thermal populations and thus for the internal sample temperature. Figure 6 shows a 0.4 mol% Er^{3+} alumino silicate with and without 4.0 mol% Yb^{3+} codotation under 1 W excitation into a 50 µm spot size. The significant increase of the $^2H_{11/2}/^4S_{3/2}$ intensity ratio reflects the increasing internal sample temperature upon Yb^{3+} codotation. An internal temperature of 663 K was found for the 4.0% Yb^{3+} codoped sample shown in Figure 6. Measurements of the $^4I_{13/2}$ lifetime for sample temperatures up to 678 K however have shown no significant increase of $^4I_{13/2}$ multiphonon relaxation even at these high temperatures. Therefore, the pronounced sample heating observed in Yb^{3+} codoped glasses is more likely to affect thermomechanical rather than spectroscopic properties.

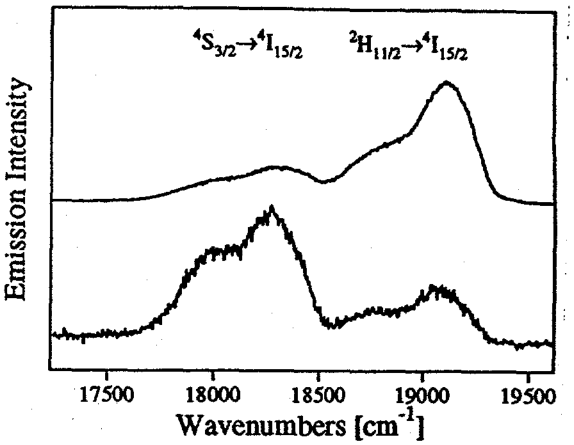

Figure 6. Room temperature $^4S_{3/2}\rightarrow{}^4I_{15/2}$ and $^2H_{11/2}\rightarrow{}^4I_{15/2}$ upconversion luminescence spectra of a 0.4 mol% Er^{3+} (bottom) and a 0.4 mol% Er^{3+},4.0 mol% Yb^{3+} (top) doped alumino silicate with high-power excitation around 980 nm.

Conclusions

Sodium and alumino silicates are favorable glass host to incorporate rare-earth ions for POWA applications. The vibrational energies of Si-O modes are sufficiently high to make multiphonon relaxation of the pumped $^4I_{11/2}$ state to the emitting $^4I_{13/2}$ state efficient, but still low enough to observe almost pure radiative relaxation from $^4I_{13/2}$. Glass manufacturing techniques exist to minimize the amount of OH^- impurities at which $^4I_{13/2}$ excitation is readily quenched. The sensitization of $^4I_{13/2}$ emission by Yb^{3+} requires a minimum Yb^{3+} concentration to be efficient. In the range of 1 to 2 mol% Yb^{3+} codoped into 0.4 mol% Er^{3+} glasses, energy migration among Yb^{3+} ions followed by energy transfer to Er^{3+} seems to be efficient, and the observed quantum yields indicate a strong increase of $^4I_{13/2}$ population. The significant sample heating associated with Yb^{3+} codotation does not affect the excited-state relaxation of $^4I_{13/2}$. These materials are promising candidates for further maturation into thin films and planar optical waveguides. In addition, a further reduction of Er^{3+} concentration seems to be possible using optimized Yb^{3+} codotation.

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