Vol 12 No 9, September 2003
1009-1963/2003/12(09)/0997-04

Chinese Physics

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and IOP Publishing Ltd

Spectral properties of erbium-doped heavy metal oxyfluoride silicate glasses for broadband amplification*

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(Received 12 December 2002; revised manuscript received 18 March 2003)

Erbium-doped glasses showing a wide 1.55μm emission band are reported in a novel heavy metal oxyfluoride glass system SiO_2–PbO–PbF_2 and their optical properties such as emission spectra, fluorescence lifetime and the refractive index have been investigated. The broad and flat $^4I_{13/2} \rightarrow {}^4I_{15/2}$ emission of Er^{3+} ions around 1.55μm can be used as host materials for potential optical amplifiers in wavelength-division-multiplexing network system. We find that with increasing PbF_2 content in the glass composition, the fluorescence full width at half maximum and fluorescence lifetime of the $^4I_{13/2}$ level of Er^{3+} increase, while refractive index and density decrease.

Keywords: heavy metal oxyfluoride silicate glasses, spectral properties, rare earth
PACC: 4280S, 3350D, 2320C

1. Introduction

Due to the rapid increase of information capacity and the need for flexible networks, there exists urgent demand for optical amplifiers with a wide and flat gain spectrum in the telecommunication window, to be used in the wavelength-division-multiplexing (WDM) network system.[1−4] Although the performance of the present silica-based erbium-doped fibre amplifier (EDFA) can be used in WDM but with fewer channels, there exists the requirement for a larger number of channels, which could be realized by using an EDFA with a wider gain spectrum.[2] From a practical standpoint, the flatness of the gain is also critical because the light intensity for different channels would be varied by the multistep amplification if the gain of the amplifiers are not independent of wavelengths. So far, although much effort has been spent on phosphate glass, fluorophosphates glass, tellurite glass and bismuth glass,[1,5,6], etc., the chemical and mechanical stability and fiberizability of these glasses as practical materials are still open problems.[7] However, silicate glasses are the most chemically and mechanically stable and also more easily fabricated into various shapes such as rods and optical fibres.[8] Therefore, the design of a new silicate host for Er^{3+} for wide and flat spectra of the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition at 1.55μm is a target at present.

In this paper, the new heavy metal oxyfluoride silicate glasses of Er^{3+}-doped $50SiO_2$-(50–x)PbO - $xPbF_2$ are prepared. The emission spectra, fluorescence lifetime of the $^4I_{13/2}$ level, refractive indices, densities and the Judd–Ofelt intensity parameters Ω_t (t=2,4,6) are investigated for various PbF_2 content.

2. Experiments

The glass samples with a composition (mol%) $50SiO_2$-(50–x) PbO-$xPbF_2$-$0.5Er_2O_3$ (x=0,10,20,30, 40,50) were prepared from high purity SiO_2, Pb_3O_4, PbF_2 and Er_2O_3 reagents. The 20g batches of the raw materials were melted at 1000–1100°C for 20–30min in covered silicate crucibles in a SiC Globar furnace with an air atmosphere. The glasses were subsequently annealed at the glass transition temperature. The glass samples were then cut and optically polished. Densities were measured according to the Archimedes'

*Project supported by the National Natural Science Foundation of China (Grant No 60207006), and the Shanghai Foundation of Optical Science and Technology, China (Grant No 022261046).

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principle using distilled water as the medium. Refractive indices of the samples were measured on a prism by minimum deviation method. UV/VIS/NIR absorption spectra were recorded between 300 and 1700nm using a spectrophotometer. Emission spectra measurements were carried out with a TRIAX550 spectrofluorimeter using a 980nm laser diode (LD) as pumping source. The fluorescence lifetimes of the $^4I_{13/2}$ level of Er^{3+} were measured with light pulses of the 980nm LD and an HP546800B 100MHz oscilloscope.

3. Results and discussion

Figure 1 shows that densities and refractive indices of the glass samples decrease with increasing PbF_2 content. Such a feature can be explained as follows: due to the gradual replacement of the Pb═O double bonds by two Pb—F single bonds, the structure of glasses becomes looser, which results in decreasing densities. Correspondingly, the refractive indices also decrease.

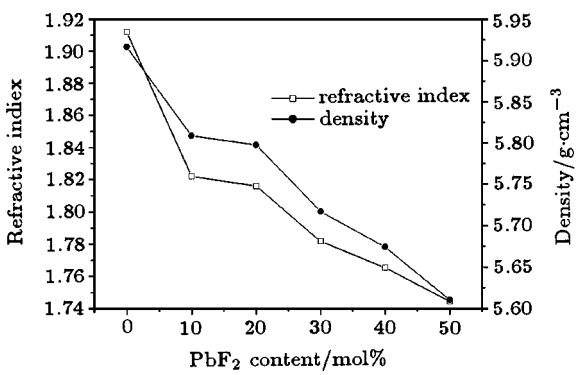


Fig.1. Compositional dependence of refractive indices and densities in Er^{3+}-doped $50SiO_2$-(50–x) PbO-$x$$PbF_2$ glasses.

The fluorescence full width at half maximum (FWHM) is very important for optical amplifiers to realize broadband and the gain amplification, which can be used to evaluate the gain properties of the optical amplifiers. The gain bandwidth of an amplifier is determined largely by the width of the emission spectrum.[9,10] Fig. 2 shows the normalized emission spectra in Er^{3+}-doped heavy metal oxyfluoride silicate glasses. The highest emission intensities are normalized to 1 for all the spectra. The FWHM of the glass samples are shown in Fig.3. It can be seen that FWHM in Er^{3+}-doped heavy metal oxyfluoride silicate glasses increases with increasing PbF_2 content. The emission bandwidth at 1.55μm band for the $50SiO_2$-$50PbF_2$-$0.5Er_2O_3$ glass is 56nm, which is smaller than that for the lead tellurite glass, ~67nm, but is wider than that for the phosphate (~50nm), the silicate glass (~36nm) and the germanate glass (~53nm).[11,12] This means that Er^{3+}-doped heavy metal oxyfluoride silicate glasses can be used as host materials for potential 1.55μm broadband amplifiers.

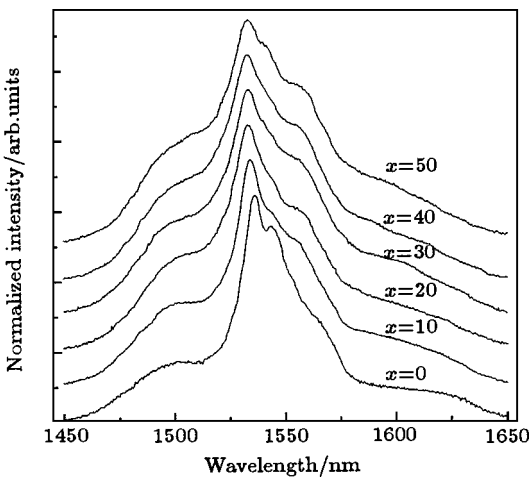


Fig.2. Compositional dependence of the normalized fluorescence spectra in Er^{3+}-doped $50SiO_2$-(50–x) PbO-$x$$PbF_2$ glasses.

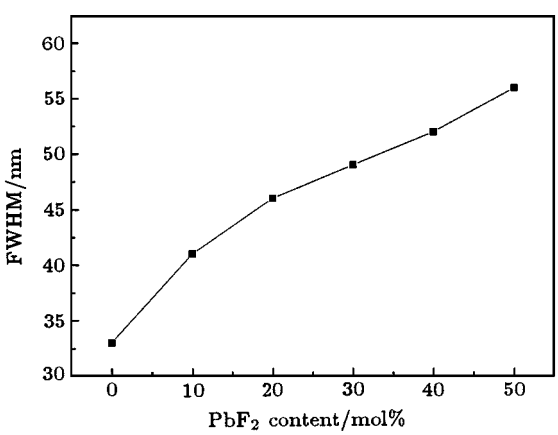


Fig.3. Compositional dependence of FWHM in Er^{3+}-doped $50SiO_2$-(50–x) PbO-$x$$PbF_2$ glasses.

In the case of the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition of Er^{3+}, because the difference of total angular momenta ΔJ equals 1, there is a contribution of magnetic dipole transitions (S_{md}).[13,14] In order to obtain broadband and flat emission spectrum, it will be effective to increase the relative contribution of electric dipole transition (S_{ed}).[2] S_{md} is independent of the ligand fields and is characteristic of the transition determined by quantum numbers, while S_{ed} is a function of ligand fields; and it is possible to increase S_{ed} by modifying the structure and compositions of the glass hosts. According to the Judd–Ofelt theory, the line strength of

S_{ed} of the 1.55μm $^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition of Er^{3+} is given by[15]

$$S_{ed}[^4I_{13/2};\ ^4I_{15/2}] = 0.0188\Omega_2 + 0.1176\Omega_4 + 1.4617\Omega_6, \quad (1)$$

where Ω_2, Ω_4 and Ω_6 are Judd–Ofelt intensity parameters. Consequently, for S_{ed} of the $^4I_{13/2} \rightarrow {}^4I_{15/2}$ transition of Er^{3+}, parameter Ω_6 is dominant and consequently is also dominant for 1.55μm emission. Therefore, increasing the parameter Ω_6 is helpful in increasing the bandwidth of 1.55μm emission. According to the Judd–Ofelt theory, the three intensity parameters Ω_t (t=2,4,6) can be obtained experimentally from the measured absorption spectra and refractive indices of the glass hosts. The values of Ω_2, Ω_4 and Ω_6 for the glass samples are calculated using the procedure provided in Ref.[16,17]. Figure 4 shows the compositional dependence of Ω_t (t=2,4,6) of the glass samples. Apparently, with increasing PbF_2 content, the values of Ω_4 and Ω_6 increase monotonically, and the value of Ω_2 decreases monotonically. According to previous studies,[2] Ω_2 is related to the symmetry of the glass hosts while Ω_6 is inversely proportional to the covalency of Er—O bond, which is assumed to be related to the local basicity around the rare-earth sites, which can be adjusted by the composition or structure of the glass hosts. On the basis of the electronegativity theory,[18] the smaller the difference of electronegativity between cation and anion ions, the stronger the covalency of the bond. The values of electronegativity, for Pb, O, and F elements, are 1.8, 3.5, and 4.0, respectively. As a result, the covalency of Pb—F bond is lower than that of Pb—O bond. Consequently, the value of Ω_6 increases with the substitution of PbF_2 for PbO.

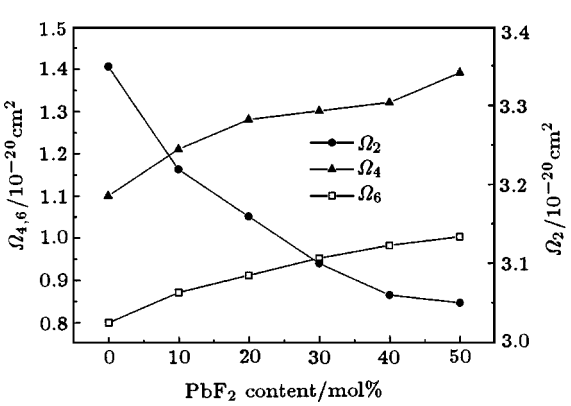


Fig.4. Compositional dependence of intensity parameters Ω_t (t=2,4,6) in Er^{3+}-doped $50SiO_2$-(50–x) PbO-$x$$PbF_2$ glasses.

Figure 5 shows the relationship between FWHM and Ω_6 in Er^{3+}-doped heavy metal oxyfluoride silicate glasses. The parameter Ω_6 shows a strong dependence on FWHM of the 1.55μm emission band, and the glass sample with a large Ω_6 generally possesses a broad FWHM. This result demonstrates the validity of the above statement.

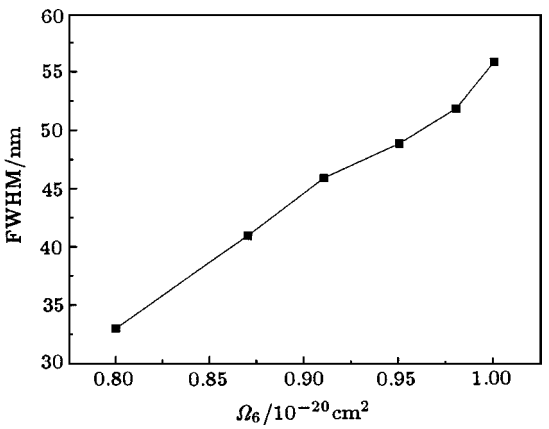


Fig.5. Relationship between FWHM and parameter Ω_6 in Er^{3+}-doped $50SiO_2$-(50–x) PbO-$x$$PbF_2$ glasses.

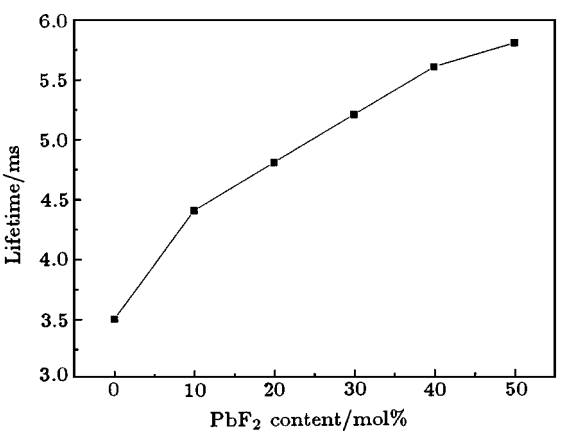


Fig.6. Compositional dependence of the lifetime of $^4I_{13/2}$ level of Er^{3+} in $50SiO_2$-(50–x) PbO-$x$$PbF_2$ glasses.

The lifetime of $^4I_{13/2}$ level of Er^{3+} is also an important parameter for optical amplifiers. A critical factor in the success of Er^{3+}-doped optical amplifiers in optical communications is the long lifetime of the metastable state that permits the required high population inversions to be obtained under steady-state conditions using modest pump powers.[19] The lifetime is related mainly to the phonon energy of the glass hosts. If neglecting the energy transfer process, in accordance with the simplified rate relation among total lifetime τ, radiative lifetime τ_r and the non-radiative lifetime τ_{nr}, $1/\tau_m = 1/\tau_r + 1/\tau_{nr}$, increasing the measured lifetime is due to low non-radiative

decay processes.[20] The larger the phonon energy of the glass hosts, the higher the probability for non-radiative decay, and consequently the lower the lifetime of the $^4I_{13/2}$ level of the Er^{3+}. On the contrary, the lower the phonon energy of the glass hosts, the higher the upconversion emission, which is also a disadvantageous property for optical amplifiers. Thus, the glass hosts should have medium phonon energy for Er^{3+}-doping to realize good amplification properties. Figure 6 illustrates the change of the lifetime of $^4I_{13/2}$ level of Er^{3+} in heavy metal oxyfluoride silicate glasses with different PbF_2 concentrations. From Fig. 6 it can be observed that the lifetime of $^4I_{13/2}$ level of Er^{3+} increases with increasing PbF_2 content. The increased lifetimes of the glass samples could be explained as follows: first, the non-radiative transition rate decreases due to the coupling of Er^{3+} with a large number of Pb—F bonds with low phonon energy, which leads to increasing the measured lifetimes; Second, the radiative lifetime calculated by using the Judd–Ofelt theory is inversely proportional to the refractive index of the glass host,[4] since refractive indices of the glasses decrease with increasing PbF_2 content, and so do the measured lifetimes.

4. Conclusion

The effects of the PbF_2 content on FWHM, fluorescence lifetime of the $^4I_{13/2}$ level, densities, refractive indices and Judd–Ofelt intensity parameters in Er^{3+}-doped heavy metal oxyfluoride glasses have been investigated. As the PbF_2 content increases, the FWHM and fluorescence lifetime of the $^4I_{13/2}$ level of Er^{3+} increase, while refractive index and density decrease. The FWHM value is related to Ω_6 and the line strength of S_{ed}; and the larger the parameter Ω_6 and the line strength S_{ed}, the broader the FWHM. The heavy metal oxyfluoride silicate glasses studied can be used as host materials for potential optical amplifiers in WDM.

Acknowledgements

We would like to express our thanks to Mr. S G Li for optical spectra tests.

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