

Persistent spectral hole burning studies in europium doped sodium silicate and borosilicate glasses

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Abstract

Spectroscopic studies confirmed the coexistence of Eu^{3+} and Eu^{2+} in sodium silicate and borosilicate glasses doped with europium. Persistent spectral hole burning was observed up to room temperature. Multiple hole burning was possible with moderate laser powers. When the glass composition was modified by adding 4–5% yttrium oxide, hole retention capability, hole burning efficiency and hole density also increased in both silicate and borosilicate glasses. Hole burning is due to a charge exchange between the Eu^{3+} and its environment. Certain defect centers, which were identified by electron spin resonance measurements, contributed to the formation of stable holes in the borosilicate glass.

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1. Introduction

Sodium borosilicate glasses have good optical and mechanical properties. The presence of alkali atoms in the glass network produces non-bridging oxygen atoms and helps to incorporate trivalent rare-earth ions in the glass matrix [1]. In general, rare-earths form trivalent compounds and they are useful for the development of luminescent and laser devices [2]. However, europium and samarium can be incorporated into solids in divalent form under certain circumstances [3,4]. Trivalent rare-earth oxides have sharp optical absorption and emission peaks whereas the transitions of divalent rare-earths are broad and very intense [5–7]. One of the recent applications of

rare-earth doped glasses is as optical hole burning materials [8]. Glasses are better than crystals in some aspects because they are easy to prepare and the inhomogeneous broadenings encountered in glasses are much higher than those of single crystals [9]. Also glass compositions may be tailored to create a large ratio of inhomogeneous to homogeneous broadenings, a critical requirement for high density optical hole-burning memory [8]. Persistent hole burning has been observed in europium doped silicate [10,11], borate [12,13], germanate [14] and tellurite [15] glasses. The hole burning mechanism and efficiency appear to depend on the glass composition and preparation methods used [10–21]. Multiple hole burning was demonstrated in a lanthanum-aluminosilicate glass [22] but higher laser power was required to burn a very shallow hole. There is a need to design materials with superior hole burning characteristics. Here, the compositional dependence of spectral hole burning was investigated in silica based glasses and some significant results were obtained.

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2. Experimental

We prepared europium doped sodium silicate and borosilicate glasses. The raw chemicals were obtained from Alfa-Aesar and their purity was better than 99.9%. Chemicals were mixed in 8 g batches for an hour in a porcelain mortar. The chemicals were heated in an alumina crucible using a box furnace at 1450 °C for borosilicate (or 1550 °C for silicate), in ambient air for 1 h and then the crucible was cooled to room temperature. These samples were not annealed. Some samples were melted using a tube furnace under flowing gas; N₂ (90%) + H₂ (10%) at 69 kPa. The resulting glass was yellowish in color but transparent in the visible region. The sample compositions are summarized in Table 1. The samples, after polishing to obtain the shape of a rectangular block, were cooled in a closed cycle cryostat down to 10 K. Low temperature transmission measurements were performed with light sources obtained from Ocean Optics (Model LS-1 and AIS Model UV-2). A Rhodamine 6 G continuous wave dye laser having a linewidth ~60 GHz was used for hole burning and excitation spectral recordings. A medium resolution spectrometer fitted with a photomultiplier tube was used to acquire the spectra. Some emission spectra were recorded using a charge coupled device (CCD) camera. The remaining experimental details are same as those described elsewhere [20]. The samples were cooled to 77 K to obtain electron spin resonance (ESR) measurements in the X-band region using a Varian E-12 spectrometer equipped with a rectangular cavity.

3. Results

3.1. Absorption spectra

Transmission of the samples was recorded using a tungsten halogen lamp and then normalized with the sample

thickness. The normalized transmission of the doped sample was subtracted from that of the undoped sample. The difference is plotted in Fig. 1 for the borosilicate samples (samples #6–8) and that of silicate glasses, samples #1–4, is not shown. The absorption due to defects [23], if any, is eliminated after subtraction. The types of defects are discussed in Section 3.4. The sharp peaks at 466, 530 and 580 nm in Fig. 1(a), are those of Eu³⁺ transitions (occur among the levels derived from 4f⁶ configuration), obtained for the glass melted in ambient air, and the broad absorption at 466 nm in Fig. 1(b) is due to Eu²⁺ [5–7]. Eu²⁺ transitions, 4f⁵5d → 4f⁶ are allowed transitions, because the parity of the initial level is different from that of the terminal level. In the (Eu, Y) co-doped glasses (samples #4 and #8), the Eu²⁺ concentration is higher. As a result the absorption is stronger in the co-doped sample (Fig. 1(c)).

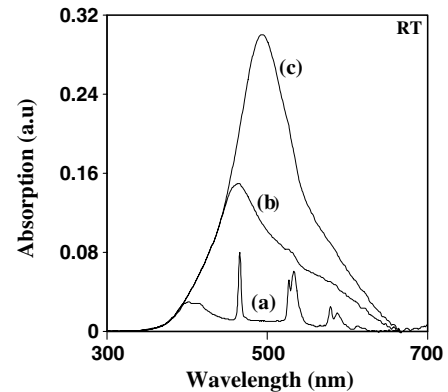


Fig. 1. Room temperature absorption spectra of sodium borosilicate glasses doped with (a) Eu (sample 6) (0.74 cm), (b) Eu (sample 7) (0.49 cm), (c) Eu, Y (sample 8) (0.78 cm); the corresponding sample thickness is given in parenthesis. The spectral plots are normalized with the respective sample thickness to account for the variations in thickness.

Table 1
Comparison of the salient features of different glasses

S#	Mol%					Atmos ^a	T_{\max}^b (K)	Γ_{inh}^c (nm)	Γ_h^d (nm)	HD ^e (%)	HN ^f	P^g (mw)
	B ₂ O ₃	SiO ₂	Na ₂ O	Y ₂ O ₃	Eu ₂ O ₃							
1	–	65	35	0	0	Air						
2	–	64	34	0	2	Air						
3	–	64	34	0	2	N ₂ + H ₂	150	2.1	0.07	27	3	1
4	–	61	33	4	2	N ₂ + H ₂	250 ^h	2.5	0.06	29	7	<1
5	34	34	32	0	0	Air						
6	34	34	30	0	2	Air						
7	34	34	30	0	2	N ₂ + H ₂	250 ^h	3.1	0.12	17	16	1
8	34	31.5	27.5	5	2	N ₂ + H ₂	295	2.9	0.1	18	18	<1

^a Refers to the melting atmosphere. N₂ (90%) + H₂ (10%) gases were flown at 69 KPa.

^b T_{\max} is the maximum temperature at which hole burning was observed.

^c Inhomogenous broadening (full width at half-maximum) of the zero phonon line at 10 K. The peak wavelength is 579 nm.

^d Refers to the observed hole width at 10 K for the peak at 579 nm.

^e HD is the hole depth at 10 K.

^f HN is the maximum number of holes burnt on the excitation spectrum at 10 K.

^g Refers to the minimum laser power required to burn a spectral hole in the sample at 10 K.

^h Intensity versus time plot revealed the effect of hole burning even at room temperature.

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