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Optical hole burning studies in europium doped oxide glasses

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Abstract

Europium doped sodium borate, silicate, borosilicate, germanate and tellurite glasses have been prepared by melt quenching technique. Optical spectroscopy techniques were used to characterize the divalent and trivalent ions. Optical hole burning technique was used to characterize rare-earth ion doped glasses for optical data storage. The hole burning mechanisms, which depend on the glass composition and the preparation methods used, are discussed.

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1. Introduction

Optical hole burning is a high resolution spectroscopy technique used to investigate zero-phonon transitions [1]. It has applications in high density optical data storage [2]. Though zero-phonon lines were investigated in a ruby crystal for the first time [3], an intense hole burning activity occurred on rare-earth ion doped materials, in particular on praseodymium [4] and europium doped crystals [5]. The technique requires a stabilized single frequency laser. In the seventies single frequency dye lasers were available and they were operated with rhodamine 6 G, one of the stable dyes, whose emission occurs at 570–620 nm. Pr^{3+} and Eu^{3+} ions have energy levels whose absorption wavelengths fall in this interval. So, most of the hole burning work was centered around these ions [6]. Zero-phonon transitions are inhomogeneously broadened even at liquid helium temperatures due to the random strain in the material, presence of dopants and the clustering of dopants [7]. The inhomogeneous broadening is of the order of gigahertz in single crystals [2–8], and several wavenumbers in glasses [9] and dye doped media [1]. However, clustering of the dopants also induce additional sites in the material [10,11].

Hole burning studies unravel some interesting fundamental physics at the microscopic level [6]. The nucleus of europium or praseodymium ion has a spin of 5/2. The crystal field components are split further due to second order hyperfine interaction. The hyperfine splittings are of the order of ~10 MHz for Pr^{3+} [8,12] and ~10^2 MHz in Eu^{3+} [5]. Population rearrangement among hyperfine levels produces transient hole burning [8]. The hole lasts for a second in certain crystals doped with Pr^{3+} [6,8] and several minutes in Eu^{3+} doped materials [2] at low temperatures. Such transient holes can be probed by rf-optical double resonance, also called optical detection of nuclear quadrupole resonance [8], sideband spectroscopy, etc. In addition to the transient phenomenon, hole burning may also occur due to photophysical [1,9] or photochemical processes [1,13]. Such phenomena were observed in organic systems [1] as well as inorganic systems [6,9,13]. For optical data storage a large ratio of inhomogeneous to homogeneous broadenings is required. However, hole width is more than that predicted by the lifetime of the levels, due to several other interactions that occur in the material at the microscopic level [1,6,14]. If the hole lasts for a long time (persistent spectral hole) then the material will retain

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Table 1
Compositions of europium doped glasses

B_2O_3 (mol%)	SiO_2 (mol%)	GeO_2 (mol%)	TeO_2 (mol%)	Na_2O (mol%)	Y_2O_3 (mol%)	Eu_2O_3 (mol%)	Melting atmosphere
65	0	0	0	33	0	2	H_2
66	0	0	0	27	5	2	H_2
0	64	0	0	34	0	2	H_2
0	61	0	0	33	0	2	H_2
34	34	0	0	30	0	2	H_2
34	31.5	0	0	27.5	5	2	H_2
0	0	78	0	21	0	1	H_2
0	0	75	0	21	3	1	H_2
0	0	78	0	21	0	1	Air
0	0	0	79	20	0	1	Air
0	0	0	69	27	3	1	Air
0	0	0	81	18	0	1	He

Melting atmosphere H_2 refers to 10% hydrogen and 90% nitrogen gas flow, at 10 PSI.

the information for a long time [15,16]. Such materials are useful for long-term data storage. For the last one decade several materials doped with europium or samarium have been investigated, which exhibited persistent hole burning [17–20]. The persistent hole burning in europium doped sol–gel glasses was due to rearrangement of the OH bonds at the photo-excited center [17] (photophysical process) and the exchange of charge between the photo-excited ion and its environment (Eu^{2+} or a charged defect) in the melt quenched glasses [18]. We investigated hole burning in several types of glasses to understand the hole burning phenomena. Our interest is to understand the hole burning mechanism. If we do understand the mechanism then we will be able to design materials with better characteristics for optical data storage.

2. New functionality in glasses

Optical hole burning is useful for the design of high density optical data storage media. The data storage capacity is directly proportional to the hole density. The glass functionality is to facilitate multiple hole burning at low laser powers and retain all the holes for a longtime. Glasses retain data for a very long time, if the holes are persistent. For any practical application hole burning should occur at temperatures higher than those achieved with cryogenics. The storage capacity increases if the holes are narrow and the inhomogeneous broadening is high. Therefore, the data storage capacity or the hole density depends on the hole burning mechanism and the glass composition. Our goal is to make glasses which satisfy the above requirements. In this article we will review some of the recent results obtained in our laboratory on a variety of glasses [9,13,21,23].

3. Experimental

We prepared several glasses doped with europium by melting in ambient air or in a reduced atmosphere as described in previous publications [9,13,21,23]. The samples and their compositions are given in Table 1. When the chemicals were melted under a controlled atmosphere, a tube furnace was used. In a glass that was melted in a reduced atmosphere more than 95% of the dopant ions were in divalent form. We could not make tellurite glasses in a reduced atmosphere because the samples turned opaque due to the high concentration of defects produced by the decomposition of tellurium oxide [21]. Room temperature absorption spectra of the samples were recorded using a CARY 3E spectrophotometer. For low temperature measurements the samples were cooled in a closed cycle cryostat. A block diagram of the experimental setup is shown in Fig. 1. A tunable single frequency dye laser irradiates the sample. A computer controlled spectrometer is used for data acquisition. The laser was tuned to resonantly excite the zero-phonon transition, ${}^7F_0 \rightarrow {}^5D_0$ of Eu^{3+} doped materials. For hole burning, the sample was exposed to a high power laser beam ($\sim 10^2$ mW). Excitation spectra were obtained by scanning a 1 mW laser beam. A comparison of the excitation spectra obtained before and after hole burning reveals the spectral hole. The remaining experimental details are same as those described previously [9,13,21].

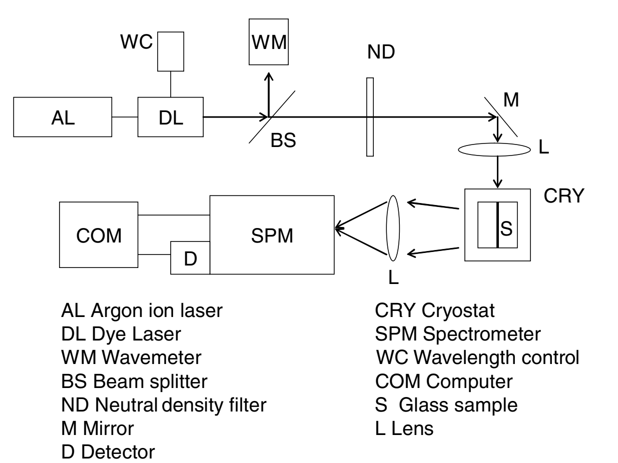

Fig. 1. Block diagram of the experimental setup.