Judd–Ofelt Analysis of Nd^{+3} Doped Sol–Gel Derived Silica Glass

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***Abstract*—Nd^{3+} doped Silica glass has been prepared successfully. The samples were prepared via a sol–gel process. The intensities of the f–f transitions in the absorption spectrum were analysed by application of the Judd–Ofelt theory. Sets of phenomenological Ω_λ intensity parameters have been extracted. This theory gives three intensity parameters Ω_2, Ω_4 and Ω_6, which are related to the local structure in the vicinity of rare-earth ions, and / or the covalency of the rare-earth ion sites.**

***Keywords*— Sol–gel process, silica glass, Judd–Ofelt theory, Absorption spectra, Rare earth doped Glass.**

I. INTRODUCTION

Sol-gel process is one of the most successful techniques for preparing nano crystalline metallic oxide materials due to low cost, easy of fabrication and low processing temperatures [1]-[4]. Generally, in a typical sol-gel process, a colloidal suspension or a sol is formed due to the hydrolysis and polymerization reactions of the precursors, which on complete polymerization and loss of solvent leads to the transition from the liquid sol into a solid gel phase. The wet gel can be converted into nanocrystals with further drying and hydrothermal treatment [5], [6]. The purpose of this paper is to report a new laser host material based on SiO_2. Major optical properties are determined from the absorption spectra to evaluate the performance of SiO_2 host material doped with Nd^{3+} for optical amplifier and laser application.

II. EXPERIMENTAL

Silica sols containing 1mol% Nd_2O_3 was prepared using tetraethyl orthosilicate, $NdNO_3$, deionized water, HCl and C_2H_5 OH. The chemicals used were all AR (analytical reagent) grade. Calculated amount of dopant salts were poured in TEOS under stirring condition at room temperature. The molar ratio of TEOS : C_2H_5OH : H_2O : HCl was 1: 4: 14: 0.01 respectively. TEOS and ethanol were magnetically stirred thoroughly till both were in well mixed state. To this well-mixed solution the remaining water was added in which the desired acid was mixed. Again the solution was magnetically stirred to get a clear solution. The sols were cast in polypropylene dishes and were sealed to avoid intercalation of external impurity. The gels were aged for one month at room temperature to obtain the sol-gel. The samples taken in silica crucibles were sintered in a Muffle furnace at 600 ºC for 3 h and then the furnace was cooled to room temperature at a rate of 0.5 ºC per minute. Transparent and bubble free glass was prepared reproducibly. The spectral measurements were carried out by spectrophotometer method. The Absorption spectra (Fig.1) in the spectral range 300-1000 nm was recorded on UV Visible double beam spectrophotometer model Perkin Elmer spectrophotometer model lambda 35.TheAbsorption spectra have been recorded in terms of wavelength (nm) vs .Absorbance (a.u.).The absorption spectra of Nd^{+3} doped Silica glass have been investigated. From these spectral data Judd-Ofelt parameters (Ω_λ) have been calculated to study the nature of bonding in these glasses. Intensities of the f-f transitions in the absorption spectra have been analyzed by the application of the Judd-Ofelt theory.
The Measured absorption Line Strengths of various absorption levels in Nd^{3+} doped Silica glass is presented in TABLE 1.

TABLE 1: Measured absorption Line Strengths of various Absorption levels in Nd^{3+} doped Silica Glass.

Absorption levels	λ (nm)	Sm (10^{-20} cm^{-1})
$^4I_{9/2} \rightarrow {}^4F_{3/2}$	867	0.3355
$^4F_{5/2}$, $^2H_{9/2}$	796	1.3931
$^4F_{7/2}$, $^4S_{3/2}$	740	1.1725
$^4F_{9/2}$	674	0.0872
$^2G_{7/2}$, $^4G_{5/2}$	577	0.8744
$^2K_{13/2}$, $^4G_{7/2}$, $^4G_{9/2}$	521	0.8844
$^2K_{15/2}$, $^2G_{9/2}$, $^4G_{11/2}$	461	0.2643

Ω_2=0.324, Ω_4=1.237 and Ω_6=1.756

III. RESULTS AND DISCUSSION

The Judd-Ofelt parameters are important for investigations of local structure and bonding in the vicinity of rare earth (RE) ions. The Ω_2 parameter is sensitive to both asymmetry and covalency at the RE sites [7]. Since the f→f transitions of RE ions are parity-forbidden, deviations from inversion symmetry give rise to Ω_2 [8].Changes of Ω_2 due to changes of asymmetry were confirmed [9]-[13]. In many cases, a dependence of Ω_2 on the covalency between RE ions and ligand anions is observed [7],[12]-[18].The effect of host glass on Ω_6 is explained by different models. Takebe and Nageno suggested that increasing ionic packing ratio causes an increase of Ω_6 for silicate and borate glasses [13], [19]-[21].
In the Absorption spectrum of Nd^{+3} doped SiO_2 Glass seven bands have been observed in spectral range 300-1000 nm. The

transitions from $^4I_{9/2}$ identified in the studied spectral range are considered as having superimposed, the barycentre of each band or group of bands, oscillator strengths and the matrix

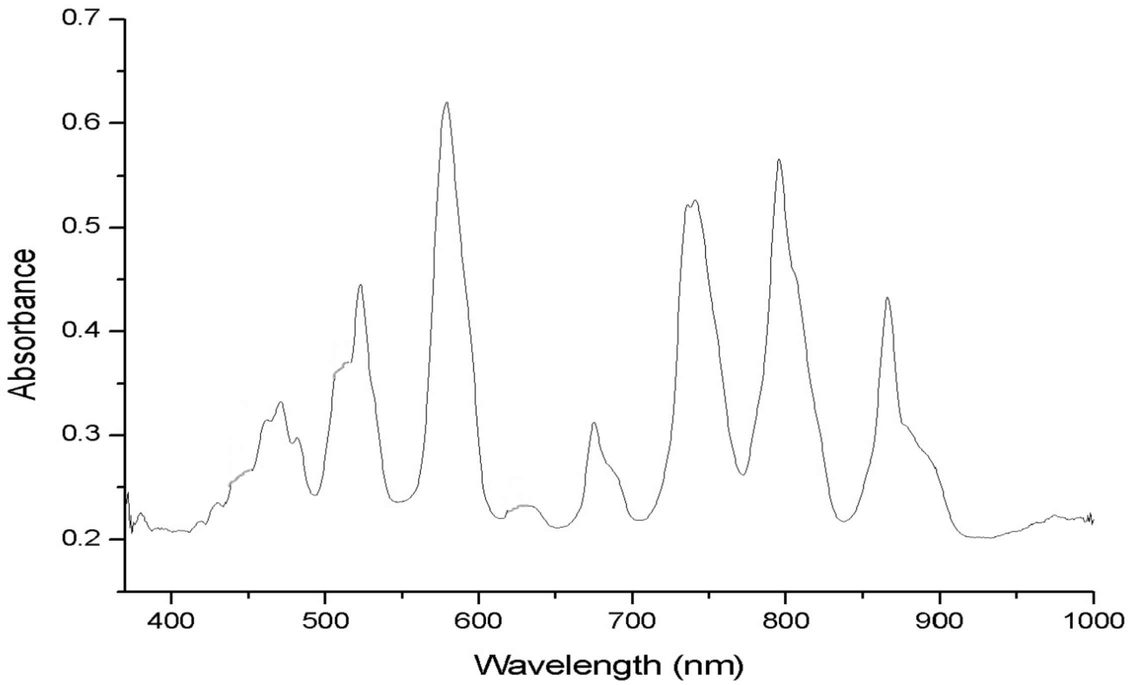

Figure 1. Absorption spectrum of Nd^{+3} doped sol-gel Silica glass

elements of Carnall [22], [23] are also included. The experimental intensity of the Absorption bands have been calculated in terms of measured absorption line strengths (S_m), by using the oscillator strength and matrix element, the values of Ω_2, Ω_4 and Ω_6 parameters of silica glass have been computed by partial regression and least square method. These parameters show the general tendency $\Omega_2<\Omega_4<\Omega_6$. The same tendency is also observed for Nd^{+3} ion in various glasses and crystals [24]. The small values of Ω_2 in glass specimen are associated with the micro-structural homogeneity [25] around theNd^{+3} ions. Ω_2 parameter is most sensitive to the local structural changes and involves the long-range terms in the crystal field potential [26], [27]. The achieved Judd-Ofelt parameters for Nd^{+3} doped sol-gel Silica glass are: Ω_2=0.324, Ω_4=1.237 and Ω_6=1.756.

IV. CONCLUSION

It is currently accepted that the Ω_2 parameter reveals the dependence of the covalency between Nd^{+3} ions and ligand anions, because Ω_2 is connected to the asymmetry of the local environment around the Nd^{+3} sites. In this way, the weaker the value of Ω_2, the more Centro symmetrical the ion site and the more ionic its chemical bond with the ligands. Ω_2 parameter is related to covalent chemical bonding between the central rare earth ion and the environment surrounding it and Ω_6 parameter is related to the rigidity and stability of the medium in which the ions are situated. This indicates that Nd^{+3} ions in Silica glass hosts present low covalence and symmetry.

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