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STUDY OF JUDD – OFELT PARAMETERS FOR RARE EARTH DOPED INORGANIC PHOSPHORS: - A MINI REVIEW

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ABSTRACT:
Luminescence is a widely used phenomenon in the field of research. It is being studied for the last 4-5 decades. Luminescence used for emission of light works mainly on phenomena of absorption (excitation and emission). There is various theoretical model used for the analysis of luminescence in rare earth-doped inorganic phosphors. But, in this review, will study Judd –Ofelt (JO) parameters used for quantitative determination of optical parameters, transition speed and emission quantum competency. Additionally, it is used to research the local structure and bonding around rare earth (RE) ions. Judd – Ofelt theory has shown to be useful equipment for locating certain transitions of choice. Its main application is to calculate the electric dipole oscillator strength between any states. This article is mainly a review of Judd –Ofelt study from examples published in last 5 years.

Keywords :- Judd-Ofelt; Inorganic Phosphors; Luminescence; Rare Earth; Quantum Efficiency.

INTRODUCTION :

Luminescence phenomena have enthralled human mankind always. The light produced from the glow worms, luminescent woods are all examples of natural luminescence phenomena. In daily life luminescence devices are so necessary that existence without them cannot be imagined. Examples include, LED TV's, signal displays, smartphone screens, etc. The most frequent methods of occurrence are luminescence and incandescence. Incandescence is phenomenon of light produced by heat energy; hence, anything heated to sufficiently high temperature will begin to glow [1]. A regular incandescent light bulb's tungsten filament shines brilliantly when heated more intensely. The stars and sun glow by the phenomena of Incandescence. Luminescence is a phenomenon that may happen at low and normal temperatures and occurs when light originates from another source of energy [2]. A little amount of energy is used to stimulate an atom's electron from its ground state, and the excited electron returns energy that manifests as visible light.

Luminescence Mechanism: The following figure shows Luminescence mechanism in:

Two different forms of recoil to ground state are depicted in figure; one is radiative and the other is non – radiative. The former is where the luminescence process takes place. The luminous material that favors radiative transitions over non- radiative one is said to be efficient [4]

Phosphors: Phosphors are solid inorganic substances with an impurity - doped host lattice. Energy is absorbed in this mechanism either through the host lattice or impurities [5]. The impurity concentrations are low because the luminescence process is mostly less efficient at higher concentrations. Inorganic phosphors are probable labels for time resolved luminescence staining and tests in aqueous environment [6-7]. These materials have the ability to undergo

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electronic transitions that motive light of higher frequency to a lower frequency (stokes shift).

Rare Earth doped Phosphors: Impurity doping is a method for changing properties of materials. Rare earth (RE) ion doping of phosphors confer optical, electrical, magnetic properties [8]. The group of 17 elements known as rare earth (RE) elements are group of 15 lanthanides (starting from lanthanum to lutetium) and scandium and yttrium. They were comparable in many chemical and physical properties as well as in their geographical deposits. Also the widespread utilization of rare earth elements establishes a strong basis for practical applications in today's generation [9-10].

Properties of Rare – Earth Elements:

Theoretical Parameter: Judd – Ofelt Theory

The concept of crystal field serves as the foundation for Judd –Ofelt (JO) Theory. It offers a model for simulating f-f transitions in solids and liquids. This theory solely takes into account $4f^n$ electronic configuration and ignores $4f^n$ –$4f^{n-1}5d^1$ interactions [15]. These parameters are tunable and can be computed using the absorption spectra. Additionally, it is built on single configuration, free- charge and static estimates. In this static pattern, the host charges that surround the center ion influence it via static electric surrounding known as crystal field [16]. This host atmosphere creates the static field of crystals and is looked upon as perturbation on the Hamiltonian of free- charges in the free ion pattern. This theory gives a expression for theoretical calculation of line strength, given as,

$S_{ED}(J;J') = \Sigma_{\lambda\text{-}2,4,6}\, \Omega_\lambda\, |\langle f^n [S L]J \| U^{(\lambda)} \| f^n [S'L']J' \rangle|$

Where Ω_λ is important JO parameters. The term given inside bracket shows double host matrix terms used in in between coupling. When this spin orbit coupling and the reciprocal repulsion of 4f ions are of the same sequence of magnitude, this is referred to as an "intermediate coupling "condition. Expanding the wave functions of the four states in an approximate Rusell – Saunders combination also known as LS –coupled states, can be used to integrate this effect.

The energy exchange in intra – caste 4f transitions are of magnetic dipole (MD) and electric dipole(ED) type. Since, this Magnetic dipole transitions has equal parity, they were permitted for 4f- 4f transitions. Also Electric Dipole transitions has even parity, due to which the Laporte rule forbids this [17]. The duration of an excited state is measured in terms of its population decaying to 1/e or 36.8%, of its starting inhabitant. Maximum emission probabilities and more frequent transitions from level, results in faster decay or shorter lifetimes. Inverse of total radiative lifetime are equivalent to computed radiative lifetime: $\tau_{rad}^{calc} = 1/A_r$ [18]. This makes further calculation easy.

Judd – Ofelt Theory in Practical Approach

The JO theory permits for calculation of radiative lifetime and branching ratio of emission, which allows for a computation of manifold - manifold transition probability. In this precise absorption data, especially a summed absorbed cross- section throughout a wavelength range of many manifold, which are necessary for the analysis of JO theory. It will be possible to mention line strength S_m using the integrated absorption cross – section.

$$Sm = \frac{3ch(2J+1)}{8\pi^3 e 2\bar{\lambda}}\; n\left(\frac{3}{n^2+2}\right)^2 \int_{manifold} \sigma(\lambda)d\lambda$$

In above expression

J is total angular momentum of basic ground manifold,

σ (λ) dλ is absorption of cross section as wavelength function

Average wavelength $\bar{\lambda}$ is obtained from first moment of absorbed cross - section data:

$$\bar{\lambda} = \frac{\Sigma\sigma(\lambda)}{\Sigma\lambda\sigma(\lambda)}$$

By fitting experimental absorption, the Judd – ofelt theory is utilized to establish set of

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phenomological parameters Ω_λ where λ =2, 4, 6 [20].

Procedure used in Judd – Ofelt Analysis

Application of Judd- Ofelt theory in Rare Earth

Praseodymium:

The Judd- Ofelt hypothesis has faced various issues when applied to praseodymium ions. The substantial RMS values obtained in Judd- Ofelt fitting are an indication that there are frequently significant discrepancies between computational and experimental measurements. Also, the transitions that were employed in the fit have a strong impact on the Judd- Ofelt parameters for Praseodymium (Pr) mixed materials. It is also being noted that Judd- Ofelt theory previous assumptions assumed that perturbing configurations were degenerate and widely splitup from f- f transition of $4f^n$ configuration. Stronger transitions are prevented from dominating fit by praseodymium [21].

Europium:

The conventional Judd – Ofelt hypothesis is again put to the test by ions of europium. Transitions like $^7F_0 \leftrightarrow {}^5D_{Jodd}$, and $^7F_0 \leftrightarrow {}^5D_0$ are prohibited transitions in this analysis. Several of this transitions are magnetic dipole in creation while some materials experience low intensity electric dipole transitions. Transitions in Eu^{3+} gives an idealistic testing ground for advancement to standard theory. Spin – forbidden transitions has been esteemed by Burdick and Downer according to which they shows that spin forbidden transition gets a most of the fraction of this intensities .It is observed that in $LaF_3:Er^{3+}$, analysis of oscillator strength was done using Judd – Ofelt theory and was inferred that least square intensity fit was minimized and yielded the JO properties as $\Omega_{(2)}$ = 5.449 * $10^{-2}cm^2$, $\Omega_{(4)}$ = 2.077 * 10^{-2} cm^2 and $\Omega_{(6)}$ = 6.873 * 10^{-21} cm^2.In this example of $LaF_3:Er^{3+}$, it was found that the oscillator strength for the multiple multiplet transitions at 300K depart on an mean of 13% from ideal scenario of equivalent thermal inhabitant of levels of crystal field $^4I_{5/2}$ ground state multiplets [22].This was the substantial source of inaccuracy that was inherent in this methodology.

Dysprosium:

Another study was about Dy^{3+}:B_2O_3–Ga_2O_3–Al_2O_3 in which 0.5% M Dy_2O_3 mixed glass recognized maximum emission intensity was estimated using Judd –Ofelt analysis to calculate radiative parameters using absorption spectrum. As per the observed absorption spectra, the highest peak was noted at wavelength of 1268nm which gave the maximum value of oscillator strength as $^6H_{15/2} \rightarrow {}^6F_{11/2}$ transition. The Judd- Ofelt parameter like Ω_2 was utilized for prediction of asymmetric atmosphere of Dy^{+3} and also for DyO covalence in glass in this study. The other two parameters i.e. Ω_4,Ω_6 to describe the viscosity and severity of glass simultaneously. Obtained values of Judd-Ofelt parameters in this study was Ω_2 = 6.59 * $10^{-20}cm^2$, Ω_4 = 0.19 * $10^{-20}cm^2$ and Ω_6 = 3.06 * $10^{-20}cm^2$.These theoretical values ensured the tendency of Judd- Ofelt intensity parameter as $\Omega_2 > \Omega_6 > \Omega_4$. [23]

CONCLUSIONS :

Over the past five decades, the Judd – Ofelt hypothesis has been the focal point of rare earth optical spectroscopy. It has sparked investigation into the theoretical underpinnings of the model and resulted in several adjustments throughout the time. The idea has parallel applications to a various rare- earth doped materials. Most of the materials researched under this have significant uses in phosphors for LED displays, solid state lasers, optical- amplifiers, up converting and quantum cutting materials. As per review, Judd- Ofelt theory has been found as useful method for transition intensity analysis, transition identification and quantum efficiency

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estimation. From the above example of Dysprosium, it was concluded that the created glass's quantum efficiency was observed as 63.75% by applying Judd – Ofelt theory and it also has the ability to serve as good quality laser. Judd – Ofelt theory's relative standard are liable for its prevalent used in various fields considering luminescence of rare earths (RE.)

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Table 1: Luminescence types, applications and their respective efficiencies given below [3]:

Luminescence type	*Typical application*	*Luminous efficiency*
Blackbody radiation	Tungsten filament lamp	~5%
Photoluminescence	Fluorescent lamp	~20%
Cathodoluminescence	Television screen	~10%
Electroluminescence	Light-emitting diode, flat panel display	0.1–50%

Table 2:

Atomic Number (Z)	**Name**	**Symbol**	**Ground State Configuration**	**Electronic Configuration**	**Application**
21	Scandium	Sc	$[Ar]3d^14s^2$	[Ar]	Catalysis, magnet, host material, ferroelectricity, piezoelectricity
39	Ytrium	Y	$[Kr]4d^15s^2$	[Kr]	Superconductivity, ferroelectricity, ferrimagnetism, host material, catalysis
57	Lanthanum	La	$[Xe]5d^16s^2$	$[Xe]4f^0$	Superconductivity, host material, catalysis
58	Cerium	Ce	$[Xe]4f^15d^16s^2$	$[Xe]4f^1$	Stimulator, photo catalysis, ferromagnetism
59	Praseodymium	Pr	$[Xe]4f^36s^2$	$[Xe]4f^2$	Quantum cutting(downconversion), ferromagnetism, photo catalysis
60	Neodymium	Nd	$[Xe]4f^46s^2$	$[Xe]4f^3$	Sensitizer , down conversion
61	Promethium	Pm	$[Xe]4f^56s^2$	$[Xe]4f^4$	Radioactive
62	Samarium	Sm	$[Xe]4f^66s^2$	$[Xe]4f^5$	Magnetism, catalysis
63	Europium	Eu	$[Xe]4f^76s^2$	$[Xe]4f^6$	Downconversion, catalysis, phosphor
64	Gadolinium	Gd	$[Xe]4f^75d^16s^2$	$[Xe]4f^7$	Matrix material, MRI, photo catalysis
65	Terbium	Tb	$[Xe]4f^96s^2$	$[Xe]4f^8$	Upconversion, ferromagnetism
66	Dysprosium	Dy	$[Xe]4f^{10}6s^2$	$[Xe]4f^9$	MRI, ferromagnetism, photo catalysis
67	Holmium	Ho	$[Xe]4f^{11}6s^2$	$[Xe]4f^{10}$	Upconversion, MR imaging
68	Erbium	Er	$[Xe]4f^{12}6s^2$	$[Xe]4f^{11}$	Upconversion, Photocatalysis,MR imaging ,NIR II imaging
69	Thulium	Tm	$[Xe]4f^{13}6s^2$	$[Xe]4f^{12}$	Upconversion, photocatalysis
70	Ytterbium	Yb	$[Xe]4f^{14}6s^2$	$[Xe]4f^{13}$	Stimulator,MRI, NIRI
71	Lutetium	Lu	$[Xe]4f^{14}5d^16s^2$	$[Xe]4f^{14}$	Host material

[12-14]

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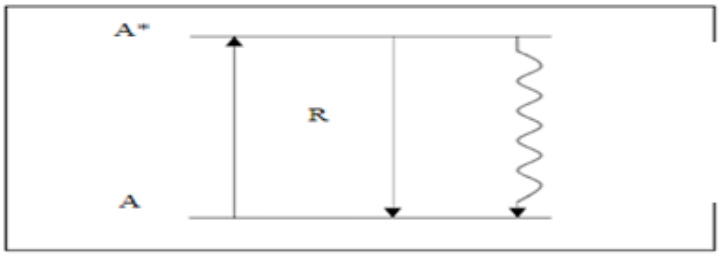

Figure1. In figure *represents the excited state, R is the radiative turn and NR is the non - radiative turn to ground state.

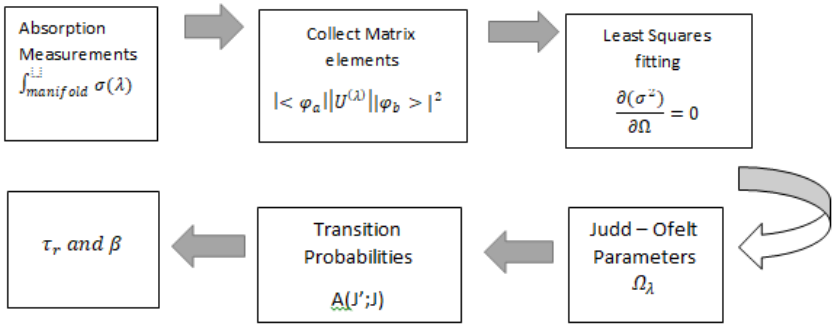

Figure 2: Step- wise procedure to calculate JO parameters

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