

Study of Judd-Oflet Parameters of the Er³⁺ doped systems in Alcoholic solution of some Oximes.

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Abstract : The ligand environment produced by various oximes in 60% ethanol around doped systems of Er³⁺ ions have been studied and characterized with respect to electronic spectral parameters. The parameters viz. Judd-Ofelt (T_λ), intensity of hypersensitive band, bonding parameters ($b^{1/2}$, $\delta\%$ & η) for doped in solution of organic oximes have been studied. The study provides useful information about interelectronic repulsion and spin interaction involved in metal-ligand bond.

Keywords: Hypersensitive transition, Doped systems.

Introduction

lanthanoides (Ln³⁺) are a unique class of materials due to their outstanding luminescent properties and numerous applications¹⁻⁶. Electronic spectral studies of lanthanoid ion complexes with reference to Judd-Ofelt parameters have been found to have due significance⁷.

In the present study six organic oximes viz. acetoxime (A), acetophenoneoxime (B), benzophenoneoxime (C), diacetylmonooxime (D), cyclooctanoneoxime (E) and camphoreoxime (F) have been used as ligands. The solution of each ligand was prepared in 60% aqueous ethanol solution (v/v) and a constant volume of ErCl₃.6H₂O salt solution (w/v) has been added to this solution. Er³⁺ ion has been doped in solutions of these oximes separately in the metal ligand ratio of 1:1 and 1:2. For Er³⁺-doped systems, we have observed nine peaks in the region of 390-1000 nm, these are due to ²G_{9/2}, ⁴F_{3/2}, ⁴F_{5/2}, ⁴F_{7/2}, ²H_{11/2}, ⁴S_{3/2}, ⁴F_{9/2}, ⁴I_{9/2} and ⁴I_{11/2} levels. The transition ⁴I_{15/2} → ²H_{11/2} is hypersensitive transition.

Results and discussion:-

The calculation of parameters viz. Oscillator strength (P), Judd-Ofelt (T_λ) bonding parameters ($b^{1/2}$, $\delta\%$ & η) have been computed by the programme developed by earlier workers¹³⁻¹⁹. The computed values of oscillator strength, Judd-Ofelt parameter, bonding parameters and nephelauxetic ratio have been tabulated (Tables 1-4). Here 'M : La' and 'M : Lb' represents metal to ligand ratio 1:2 and 1:1 respectively in the tables.

Spectral intensity parameters:

For Er³⁺-doped systems, we have observed nine peaks in the region of 390-1000 nm, these are due to ²G_{9/2}, ⁴F_{3/2}, ⁴F_{5/2}, ⁴F_{7/2}, ²H_{11/2}, ⁴S_{3/2}, ⁴F_{9/2}, ⁴I_{9/2} and ⁴I_{11/2} levels.

Oscillator strength :-

The computed values of oscillator strength (P_{obs} and P_{cal}) for Er³⁺-doped systems have been tabulated in Table -1 and 2. The observed values of oscillator strength for ²H_{11/2} band varies from 3.472×10⁻⁶ to 5.1291×10⁻⁶

Trend with respect to ²H_{11/2} level

Eb > Ea > Db > Ba > L1b > Ca > Da > Cb > L1a > Bb > Fa > Fb

The r.m.s. deviation between P_{obs} and P_{cal} varies from 2.71×10⁻⁷ to 6.23 ×10⁻⁷.

The low value of r.m.s. deviation proved validity of Judd-Ofelt theory.

Trend with respect to r.m.s. deviation

Da > Eb > Db > Ba > Ca > Cb > Bb > Fa > L1b > Ea > Fb > L1a

Judd-Ofelt parameters:-

The Judd-Ofelt parameters (T_2 , T_4 and T_6) for Er³⁺ doped systems have been tabulated in Table-3.

The values of T_2 , T_4 and T_6 are positive for present systems. The values of T_2 varies from 1.270 ×10⁻¹⁰ to 2.309×10⁻¹⁰. The values of T_4 varies from 6.542 ×10⁻¹¹ to 1.8040 ×10⁻¹⁰. The values of T_6 varies from 1.8590×10⁻¹⁰ to 2.6548 ×10⁻¹⁰. The values of T_4/T_6 varies from 0.3204 to 0.8750. The values of T_4/T_2 varies from 0.1420 to 0.8390.

The general sequence in the value of T_2 , T_4 and T_6 is

$T_6 > T_2 > T_4$

As per the values of T_4/T_6 and T_4/T_2 systems can be classified as –

The ratio T_4/T_6 indicates symmetry around the cation and varies from 0.3204 to 0.875.

On the basis of T_4/T_6 values, the doped Er³⁺ ion systems have been classified in the following CLASSES.

- (1) CLASS-1 : T_4/T_6 value is less than 0.5100. The ligand was Da.
- (2) CLASS-2 : T_4/T_6 values varying in between 0.5100 to 0.5850. The ligands were Ab, Ba, Bb, Ca, Ea, Fa & Fb.
- (3) CLASS-3 : T_4/T_6 values varying in between 0.6100 to 0.6620. The ligands were L1a, Cb & Eb.
- (4) CLASS-4 : T_4/T_6 value is greater than 0.8000. The ligand was Db.

These four CLASSES (1,2,3&4) reveal that on changing the metal to ligand ratio for ligands A, C, D & E, symmetry around the cation or symmetry of stereo environment around the doped Er³⁺ ion changes.

There is much variation in Judd-Ofelt parameter, shows that the sequence $T_4 < T_2 < T_6$ in most of the cases (except Cb and Db where it is $T_2 < T_6 < T_4$ and $T_2 < T_4 < T_6$ respectively), which is in good agreement with lanthanide metal ion characteristics.

Nephelauxetic ratio (β) and Bonding parameters ($b^{1/2}$, $\delta\%$ & η): -

The value of nephelauxetic ratio (β), for Er³⁺ -doped system varies from 0.9616 to 0.9729. Since the value of β is less than one for all the systems hence presence of covalent character in metal-ligand linkage is proved. Various bonding parameters have been tabulated in Table-4.

The value of bonding parameter ($b^{1/2}$) varies from 0.1164 to 0.1386. The value of $b^{1/2}$ represents the mixing of metal ion 4 f orbitals with the ligand orbitals. The positive value of bonding parameter indicates covalent character in metal-ligand bond. The value of Sinha's covalency parameter ($\delta\%$) varies from 3.040 % to 3.993% and value of covalency angular overlap parameter (η) varies from 0.0151 to 0.0198.

The values of bonding parameters ($b^{1/2}$, $\delta\%$ & η) and nephelauxetic ratio (β) have been tabulated in Table- 4. Since the value of β is less than one for all systems, indicative of covalency in metal-ligand bond. Trend with respect to β

$Ea > Ba = Eb > L1a > Fb > Cb > Db > Da > Bb > Ca > L1b > Fa$

From the above trend, Er^{3+} systems surrounded by camphor oxime having metal-ligand ratio 1:2 (Fa) have the highest covalent character in metal-ligand bond.

Hypersensitive transition and validity of Peacock's relation ($P \propto \sqrt{T_6}$):-

In Er^{3+} ion, the transition $^4I_{15/2} \rightarrow ^2H_{11/2}$, is regarded as hypersensitive transition. For hypersensitive transition, oscillator strength (P) is directly proportional to $\sqrt{T_6}$, this linear correlation has been proposed by R.D. Peacock. The values of proportionality constant K ($K = P / \sqrt{T_6}$) should be constant for the present systems. The value of proportionality constant K, for Er^{3+} doped systems varies from 0.8304 to 1.2118. Ligands Aa, Ba, Ca, Da, Db, 5b and Fb, exhibit constancy in the value of K, whereas Ab, Cb, Ea and Fa, show deviation from it.

Materials and method :-

All the chemicals and the solvent used were of analytical grade. $ErCl_3 \cdot 6H_2O$ having 99.1% purity was supplied by Indian Rare Earths Ltd. The ligands were dissolved to prepare 0.32 M and 0.16 M solutions in 60% aqueous ethanol (v/v) at room temperature (35 °C). Equal volume (10 ml) of each of these 0.32 M and 0.16 M ligand solution was added in 10 ml solution of 0.16M $ErCl_3 \cdot 6H_2O$ to get systems of having metal to ligand ratio 1 : 2 and 1 : 1 respectively. Solution spectra of these twelve systems were recorded by using standard spectrophotometer (Biomate UV-Visible spectro-photometer v7.07) in the range of 350 to 1000 nm.



Table 1:- Computed values of oscillator strength for Er³⁺ doped systems:-

SN	Levels	Er ³⁺ +solvent		Er ³⁺ +Aa		Er ³⁺ +Ab		Er ³⁺ +Ba		Er ³⁺ +Bb		Er ³⁺ + Ca		Er ³⁺ +Cb	
		P _{obs}	P _{cal}	P _{obs}	P _{cal}	P _{obs}	P _{cal}	P _{obs}	P _{cal}	P _{obs}	P _{cal}	P _{obs}	P _{cal}	P _{obs}	P _{cal}
1	² G _{9/2}	1.55293 X10 ⁻⁶	1.2473 X10 ⁻⁶	1.7614 X10 ⁻⁶	1.17473 X10 ⁻⁶	1.7946 X10 ⁻⁶	1.3836 X10 ⁻⁶	2.35803 X10 ⁻⁶	1.21192 X10 ⁻⁶	1.95418 X10 ⁻⁶	1.11834 X10 ⁻⁶	2.20875 X10 ⁻⁶	1.2102 X10 ⁻⁶	2.04861 X10 ⁻⁶	1.08745 X10 ⁻⁶
2	⁴ F _{3/2}	4.20577 X10 ⁻⁷	6.0441 X10 ⁻⁷	6.0738 X10 ⁻⁷	5.67662 X10 ⁻⁷	5.58234 X10 ⁻⁷	6.74304 X10 ⁻⁷	8.90351 X10 ⁻⁷	5.90189 X10 ⁻⁷	6.2847 X10 ⁻⁷	5.44001 X10 ⁻⁷	8.92809 X10 ⁻⁷	5.9036 X10 ⁻⁷	7.02458 X10 ⁻⁷	5.2663 X10 ⁻⁷
3	⁴ F _{5/2}	6.07281 X10 ⁻⁷	1.05814 X10 ⁻⁶	7.5256 X10 ⁻⁷	9.93394 X10 ⁻⁷	7.51413 X10 ⁻⁷	1.18065 X10 ⁻⁶	9.80062 X10 ⁻⁷	1.03273 X10 ⁻⁶	6.89393 X10 ⁻⁷	9.51831 X10 ⁻⁷	8.95294 X10 ⁻⁷	1.0327 X10 ⁻⁶	9.21016 X10 ⁻⁷	9.2142 X10 ⁻⁷
4	⁴ F _{7/2}	2.9117 X10 ⁻⁶	3.14375 X10 ⁻⁶	3.10073 X10 ⁻⁶	2.97987 X10 ⁻⁶	3.43067 X10 ⁻⁶	3.4371 X10 ⁻⁶	3.32393 X10 ⁻⁶	3.02387 X10 ⁻⁶	2.91413 X10 ⁻⁶	2.79506 X10 ⁻⁶	3.32994 X10 ⁻⁶	3.0095 X10 ⁻⁶	2.98108 X10 ⁻⁶	2.74754 X10 ⁻⁶
5	² H _{11/2}	3.87681 X10 ⁻⁶	4.09755 X10 ⁻⁶	3.55658 X10 ⁻⁶	3.6415 X10 ⁻⁶	3.7787 X10 ⁻⁶	3.98047 X10 ⁻⁶	3.80017 X10 ⁻⁶	3.7019 X10 ⁻⁶	3.55003 X10 ⁻⁶	3.57577 X10 ⁻⁶	3.75407 X10 ⁻⁶	3.6657 X10 ⁻⁶	3.56527 X10 ⁻⁶	3.50618 X10 ⁻⁶
6	⁴ S _{3/2}	3.75568 X10 ⁻⁷	8.7768 X10 ⁻⁷	4.60751 X10 ⁻⁷	8.23248 X10 ⁻⁷	4.33555 X10 ⁻⁷	9.77763 X10 ⁻⁷	5.18854 X10 ⁻⁷	8.55889 X10 ⁻⁷	4.09574 X10 ⁻⁷	7.88498 X10 ⁻⁷	5.91771 X10 ⁻⁷	8.5643 X10 ⁻⁷	4.77923 X10 ⁻⁷	7.6346 X10 ⁻⁷
7	⁴ F _{9/2}	2.38989 X10 ⁻⁶	2.5701 X10 ⁻⁶	2.35164 X10 ⁻⁶	2.48597 X10 ⁻⁶	2.43922 X10 ⁻⁶	2.68018 X10 ⁻⁶	2.41413 X10 ⁻⁶	2.38887 X10 ⁻⁶	2.14748 X10 ⁻⁶	2.21702 X10 ⁻⁶	2.35617 X10 ⁻⁶	2.3535 X10 ⁻⁶	2.27204 X10 ⁻⁶	2.27045 X10 ⁻⁶
8	⁴ I _{9/2}	1.68577 X10 ⁻⁷	3.36431 X10 ⁻⁷	2.2955 X10 ⁻⁷	3.36424 X10 ⁻⁷	1.93332 X10 ⁻⁷	3.2171 X10 ⁻⁷	2.60561 X10 ⁻⁷	2.94452 X10 ⁻⁷	2.13333 X10 ⁻⁷	2.77927 X10 ⁻⁷	2.73669 X10 ⁻⁷	2.8473 X10 ⁻⁷	2.43582 X10 ⁻⁷	3.01895 X10 ⁻⁷
9	⁴ I _{11/2}	7.9242 X10 ⁻⁷	9.25348 X10 ⁻⁷	5.8754 X10 ⁻⁷	8.61545 X10 ⁻⁷	7.56206 X10 ⁻⁷	1.02252 X10 ⁻⁶	6.89722 X10 ⁻⁷	8.99023 X10 ⁻⁷	6.481 X10 ⁻⁷	8.29408 X10 ⁻⁷	6.96693 X10 ⁻⁷	8.9817 X10 ⁻⁷	5.77111 X10 ⁻⁷	8.0302 X10 ⁻⁷
10	rms→ deviation s	σ = ± 2.91256X10 ⁻⁷		σ = ±2.71112X10 ⁻⁷		σ = ±307138X10 ⁻⁷		σ = ±4.30159X10 ⁻⁷		σ = ± 3.29129X10 ⁻⁷		σ =±3.84223X10 ⁻⁷		σ =± 3.57266X10 ⁻⁷	

Table2:- Computed values of oscillator strength for Er³⁺ doped systems:-

SN	Levels	Er ³⁺ +Da		Er ³⁺ +Db		Er ³⁺ +Ea		Er ³⁺ +Eb		Er ³⁺ +Fa		Er ³⁺ +Fb	
		P _{obs}	P _{cal}	P _{obs}	P _{cal}	P _{obs}	P _{cal}	P _{obs}	P _{cal}	P _{obs}	P _{cal}	P _{obs}	P _{cal}
1	² G _{9/2}	2.87668 X10 ⁻⁶	1.16863 X10 ⁻⁶	2.74547 X10 ⁻⁶	1.22378 X10 ⁻⁶	1.7995 X10 ⁻⁶	1.13217 X10 ⁻⁶	2.78574 X10 ⁻⁶	1.57082 X10 ⁻⁶	2.14373 X10 ⁻⁶	1.22676 X10 ⁻⁶	1.74463 X10 ⁻⁶	1.11966 X10 ⁻⁶
2	⁴ F _{3/2}	1.04657 X10 ⁻⁶	5.77986 X10 ⁻⁷	1.1 X10 ⁻⁶	5.83744 X10 ⁻⁷	7.66969 X10 ⁻⁷	5.51104 X10 ⁻⁷	1.38509 X10 ⁻⁶	7.61044 X10 ⁻⁷	7.66086 X10 ⁻⁷	5.96568 X10 ⁻⁷	5.05955 X10 ⁻⁷	5.4413 X10 ⁻⁷
3	⁴ F _{5/2}	1.06242 X10 ⁻⁶	1.01118 X10 ⁻⁶	1.12574 X10 ⁻⁶	1.02146 X10 ⁻⁶	1.06771 X10 ⁻⁶	9.6501 X10 ⁻⁷	1.64877 X10 ⁻⁶	1.33395 X10 ⁻⁶	1.0315 X10 ⁻⁶	1.04392 X10 ⁻⁶	7.79321 X10 ⁻⁷	9.52332 X10 ⁻⁷
4	⁴ F _{7/2}	3.17073 X10 ⁻⁶	2.8204 X10 ⁻⁶	3.39184 X10 ⁻⁶	3.19286 X10 ⁻⁶	3.18083 X10 ⁻⁶	2.83133 X10 ⁻⁶	4.78997 X10 ⁻⁶	3.95956 X10 ⁻⁶	3.12524 X10 ⁻⁶	3.06253 X10 ⁻⁶	2.80671 X10 ⁻⁶	2.81008 X10 ⁻⁶
5	² H _{11/2}	3.66846 X10 ⁻⁶	3.4337 X10 ⁻⁶	3.81501 X10 ⁻⁶	3.59293 X10 ⁻⁶	4.50823 X10 ⁻⁶	4.4565 X10 ⁻⁶	5.1291 X10 ⁻⁶	4.80463 X10 ⁻⁶	3.5063 X10 ⁻⁶	3.43752 X10 ⁻⁶	3.4727 X10 ⁻⁶	3.56345 X10 ⁻⁶
6	⁴ S _{3/2}	4.77012 X10 ⁻⁷	8.38259 X10 ⁻⁷	5.8156 X10 ⁻⁷	8.46431 X10 ⁻⁷	4.75422 X10 ⁻⁷	7.98486 X10 ⁻⁷	9.15942 X10 ⁻⁷	1.10332 X10 ⁻⁶	6.03905 X10 ⁻⁷	8.63367 X10 ⁻⁷	3.68544 X10 ⁻⁷	7.890 X10 ⁻⁷
7	⁴ F _{9/2}	2.14498 X10 ⁻⁶	1.97091 X10 ⁻⁶	3.14768 X10 ⁻⁶	2.91438 X10 ⁻⁶	2.20288 X10 ⁻⁶	2.2573 X10 ⁻⁶	3.320 ⁻⁷ X10 ⁻⁶	3.22807 X10 ⁻⁶	2.52042 X10 ⁻⁶	2.44514 X10 ⁻⁶	2.16589 X10 ⁻⁶	2.2655 X10 ⁻⁶
8	⁴ I _{9/2}	1.8305 X10 ⁻⁷	1.8193 X10 ⁻⁷	3.69617 X10 ⁻⁷	4.47633 X10 ⁻⁷	2.77333 X10 ⁻⁷	2.8038 X10 ⁻⁷	6.28055 X10 ⁻⁷	4.23445 X10 ⁻⁷	2.36201 X10 ⁻⁷	3.04357 X10 ⁻⁷	1.65148 X10 ⁻⁷	2.88177 X10 ⁻⁷
9	⁴ I _{11/2}	7.69498 X10 ⁻⁷	8.84211 X10 ⁻⁷	8.03394 X10 ⁻⁷	8.76652 X10 ⁻⁷	6.0577 X10 ⁻⁷	8.60775 X10 ⁻⁷	9.33333 X10 ⁻⁷	1.1541 X10 ⁻⁶	8.24276 X10 ⁻⁷	9.02024 X10 ⁻⁷	6.29551 X10 ⁻⁷	8.30568 X10 ⁻⁷
10	rms→ deviations	$\sigma = \pm 6.22844 \times 10^{-7}$		$\sigma = \pm 5.59552 \times 10^{-7}$		$\sigma = \pm 2.98081 \times 10^{-7}$		$\sigma = \pm 5.6702 \times 10^{-7}$		$\sigma = \pm 3.26949 \times 10^{-7}$		$\sigma = \pm 2.73348 \times 10^{-7}$	

Table3:- Computed values of Judd-Ofelt parameters for Er³⁺ doped systems:-

SN	Systems	M:L	T ₂	T ₄	T ₆	T ₄ /T ₆	T ₄ /T ₂
1	Er ³⁺ + Solvent	-	1.8957X10 ⁻¹⁰	1.3155X10 ⁻¹⁰	2.1330X10 ⁻¹⁰	0.6167	0.6939
2	Er ³⁺ +Aa	1:2	1.5802X10 ⁻¹⁰	1.3259X10 ⁻¹⁰	2.0027X10 ⁻¹⁰	0.6620	0.8390
3	Er ³⁺ +Ab	1:1	1.8292X10 ⁻¹⁰	1.2376X10 ⁻¹⁰	2.3819X10 ⁻¹⁰	0.5196	0.6765
4	Er ³⁺ +Ba	1:2	1.7245X10 ⁻¹⁰	1.1384X10 ⁻¹⁰	2.0833X10 ⁻¹⁰	0.5464	0.6601
5	Er ³⁺ +Bb	1:1	1.6886X10 ⁻¹⁰	1.0786X10 ⁻¹⁰	1.9201X10 ⁻¹⁰	0.5617	0.6387
6	Er ³⁺ +Ca	1:2	1.7260X10 ⁻¹⁰	1.0974X10 ⁻¹⁰	2.0850X10 ⁻¹⁰	0.5263	0.6358
7	Er ³⁺ +Cb	1:1	1.5862X10 ⁻¹⁰	1.1862X10 ⁻¹⁰	1.8590X10 ⁻¹⁰	0.6381	0.7478
8	Er ³⁺ +Da	1:2	1.8204X10 ⁻¹⁰	6.5426X10 ⁻¹¹	2.0414X10 ⁻¹⁰	0.3204	0.3593
9	Er ³⁺ +Db	1:1	1.2702X10 ⁻¹⁰	1.8040X10 ⁻¹⁰	2.0617X10 ⁻¹⁰	0.8750	0.1420
10	Er ³⁺ +Ea	1:2	2.3093X10 ⁻¹⁰	1.0866X10 ⁻¹⁰	1.9458X10 ⁻¹⁰	0.5584	0.4705
11	Er ³⁺ +Eb	1:1	2.1299X10 ⁻¹⁰	1.6592X10 ⁻¹⁰	2.6848X10 ⁻¹⁰	0.6180	0.7790
12	Er ³⁺ +Fa	1:2	1.5150X10 ⁻¹⁰	1.1808X10 ⁻¹⁰	2.1087X10 ⁻¹⁰	0.5599	0.7794
13	Er ³⁺ +Fb	1:1	1.6555X10 ⁻¹⁰	1.1225X10 ⁻¹⁰	1.9212X10 ⁻¹⁰	0.5842	0.6780

Table 4:- Computed values of bonding parameters for Er³⁺doped systems

SN	Systems	M:L	β	$b^{1/2}$	δ %	η
1	Er ³⁺ + Solvent	-	0.9729	0.1164	2.785	0.0138
2	Er ³⁺ +Aa	1:2	0.9696	0.1233	3.135	0.0156
3	Er ³⁺ +Ab	1:1	0.9661	0.1302	3.509	0.0174
4	Er ³⁺ +Ba	1:2	0.9699	0.1227	3.103	0.0154
5	Er ³⁺ +Bb	1:1	0.9673	0.1279	3.381	0.0168
6	Er ³⁺ +Ca	1:2	0.9665	0.1294	3.466	0.0172
7	Er ³⁺ +Cb	1:1	0.9686	0.1253	3.242	0.0161
8	Er ³⁺ +Da	1:2	0.9675	0.1275	3.359	0.0167
9	Er ³⁺ +Db	1:1	0.9682	0.1261	3.284	0.0163
10	Er ³⁺ +Ea	1:2	0.9705	0.1214	3.040	0.0151
11	Er ³⁺ +Eb	1:1	0.9699	0.1227	3.103	0.0154
12	Er ³⁺ +Fa	1:2	0.9616	0.1386	3.993	0.0198
13	Er ³⁺ +Fb	1:1	0.9691	0.1243	3.189	0.0158

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