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Absorption spectral studies of Ho^{3+} ions doped in some oximes

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Abstract: *Absorption spectralproduced by various oximesin 60% ethanol around doped systems of* Ho^{3+} *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-liand bond.*

Keywords*: Hypersensitive transition, Doped systems, Spectral intensities.*

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I. Introduction

Studies of spectral properties and coordination chemistry of lanthanoides have been widely carried out because of their outstanding luminescent properties and numerous applications.[1-8].Electronic spectral studies of lanthanoide ion complexes with reference to Judd-Ofelt parameters have been found to have due significance[9].

In the present study six organic oximes viz. acetoxime (A), acetophenoneoxime (B), bcnzophenoneoxime(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 $HoCI_3.6H_20$ salt solution (w/v) has been added to this solution. Ho^{3+} ion has been doped in solutions of these oximes separately in the metal ligand ratio of 1:1 and 1:2. In case of Ho^{3+}, we observed nine peaks in the visible region, these are due to $(^5G, ^3H)_5$, $^5G_4$, $(^5G, ^3G)_5$, $^5G_6$, $^3K_8$, $^5F_2$, $^5F_3$, $^5F_4$, $^5F_5$, levels[10-12]. The transition $^5I_8 \rightarrow {}^5G_6$ is hypersensitive transition.

II. 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 workers [13-19]. The computed values of oscillator strength, Judd-Ofeltparameter , 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. In case of Ho^{3+}doped systems, nine peaks have been observed in the visible region, in the range of 390 to 1000 nm, these are due to
$(^5G, ^3H)_5$, $^5G_4$, $(^5G, ^3G)_5$, $^5G_6$, $^3K_8$, $^5F_2$, $^5F_3$, $^5F_4$, and $^5F_5$ levels.

Oscillator strength

The computed values of oscillator strength (P_{obs} and P_{cal}) of various bands for Ho^{3+}-doped systems have been tabulated in Table 1-4. The observed values of oscillator strength for $^5G_6$ band varies from 1.27×10^{-5} to 1.70×10^{-5}.

Trend with respect to $^5G_6$ level
Aa >Ea> Ba > Eb > Ab > Fa > Ca > Da > Db >Cb> Fb > Bb
The r.m.s. deviation between P_{obs} and P_{cal} varies from 2.847×10^{-6} to 6.903×10^{-6}.
The low value of r.m.s. deviation proved validity of Judd-Ofelt theory.
Trend with respect to r.m.s. deviation
Ba > Ca > Da >Cb> Bb > Db > Aa >Ea> Ab > Eb > Fa > Fb
From the above trends, we can infer that acetoxime(Aa) with metal-ligand ratio 1:2 is stronger ligand for metal-ligand interaction.

Judd-Ofelt parameters

The Judd-Ofelt parameters (T_2 ,T_4 and T_6) for Ho^{3+} doped systems have been tabulated in Table 1&2.

The values of $T_{2,}$ T_4 and T_6 are positive for present systems. The values of T_2 varies from $2.17x10^{-10}$ to $3.28x10^{-10}$. The values of T_4 varies from $1.15x10^{-10}$ to $2.32\ x10^{-10}$. The values of T_6 varies from $8.221x10^{-11}$ to $1.940x10^{-10}$. The values of T_4 / T_6 varies from 0.9754 to 2.3699. The values of T_4 / T_2 varies from 0.4020to 0.7890. The general sequence in the value of $T_{2,}$ T_4 and T_6 is

$T_2>T_4>T_6$

On the basis of T_4/T_6 values, the doped Ho^{3+} ion systems have been classified in the following CLASSES.

(1) CLASS-1 T_4/T_6 values varying in between 0.9700 to1.0900. Ligands were Bb, Ea, Fa & Fb.

(2) CLASS-2 T_4/T_6 values varying in between 1.1200 to 1.4100. Ligands were Ba, Ca Cb, Da, Db & Eb.

(3) CLASS-3 T_4/T_6 values varying in between 1.9900 to 2.4000. Ligands were L1a & L1b.

These three CLASSES (1,2 &3) reveal that on changing the metal to ligand ratio for ligands L2 & L5, symmetry around the cation or symmetry of stereo environment around the doped Ho^{3+} ion changes.

Nephelauxetic ratio (β) and Bonding parameters ($b^{½}$, δ% &η):-

The values of nephelauxeticratio(β) and various bonding parameters for Ho^{3+} doped systems have been tabulated in Table 3.5.3. The value of nephelauxetic ratio (β), for Ho^{3+} doped system varies from 0.9947 to 1.0000. Since the value of β is less than one for all the systems hence presence of covalent character in metal-ligand linkage is proved.

The value of bonding parameter ($b^{½}$) varies from$4.21x10^{-3}$ to $2.14x10^{-2}$. 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 (δ%) varies from $3.54x10^{-3}$ % to $5.34x10^{-1}$ % and value of covalency angular overlap parameter (η) varies from $1.77x10^{-5}$ to $2.67x10^{-3}$.Trend with respect to β

L2b > L1b > L2a> L1a > L6b > L5b >Cb> L6a > Ca > L5a > L4b > L4a

From the above trend, Ho^{3+} systems surrounded by diacetylmonooxime having metal-ligand ratio1:2 (Da) have the highest covalent character in metal- ligand bond.

Hypersensitive transition and validity of Peacock's relation ($P\ \alpha\ \bar{\nu}\ T_6$):-

In Ho^{3+} ion, the transition $^5I_8 \rightarrow {}^5G_{6,}$ is regarded as hypersensitive transition. Proportionality constant (K) for the relation $P\ \alpha\ \bar{\nu}\ T_6$ or $P\ \alpha\ \bar{\nu}\ T_4$,as proposed by R.D.Peacock, should be constant for the present systems. The value of proportionality constant K_2 (= $P/\ \bar{\nu}\ T_6$), is approximately constant for Ho^{3+} doped systems and value of K_1 (= $P/\ \bar{\nu}\ T_4$), is also constant for certain ligands. In the present study of Ho^{3+} doped systems, constancy was found more in K_1 in comparison to K_2.

III. Materials And Method :-

All the chemicals and the solvent used were of analytical grade. $HoCI_3.6H_20$ 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 $DyCI_3.6H_20$ to get systems of having metal to ligand ratio I : 2 and I : I 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 Ho $^{3+}$ doped systems:-

SN	Levels	Ho^{3+}+solvent		Ho^{3+}+Aa		Ho^{3+}+Ab		Ho^{3+}+Ba		Ho^{3+}+Bb		Ho^{3+}+ Ca		Ho^{3+}+Cb	
		P obs	P Cal	P obs	P Cal	Pobs	P obs	P obs	P Cal	P obs	P Cal	Pobs	PCal	P obs	P Cal
1	(^{5}G,^{3}H)$_5$	9.16 X10^{-6}	8.6230 X10^{-7}	1.0853 X10^{-5}	1.0287 X10^{-6}	9.5300 X10^{-6}	8.3158 X10^{-7}	1.9878 X10^{-5}	6.6554 X10^{-7}	1.4521 X10^{-5}	8.2555 X10^{-7}	1.7737 X10^{-5}	8.2616 X10^{-7}	1.485 X10^{-5}	8.9297 X10^{-7}
2	^{5}G$_4$	1.4407 X10^{-6}	2.4522 X10^{-7}	1.9331 X10^{-6}	2.9780 X10^{-7}	1.7349 X10^{-6}	2.4669 X10^{-7}	2.2145 X10^{-6}	1.8190 X10^{-7}	1.7493 X10^{-6}	2.1710 X10^{-7}	2.1481 X10^{-6}	2.2110 X10^{-7}	1.7855 X10^{-6}	2.4429 X10^{-7}
3	(^{5}G,^{3}G)$_5$	4.2824 X10^{-6}	2.3265 X10^{-6}	5.5763 X10^{-6}	2.9737 X10^{-6}	5.1271 X10^{-6}	2.6154 X10^{-6}	5.0825 X10^{-6}	1.5331 X10^{-6}	3.9573 X10^{-6}	1.5971 X10^{-6}	4.7357 X10^{-6}	1.7202 X10^{-6}	4.5571 X10^{-6}	2.0469 X10^{-6}
4	^{5}G$_6$	1.6665 X10^{-5}	1.4822 X10^{-5}	1.7062 X10^{-5}	1.4667 X10^{-5}	1.5799 X10^{-5}	1.3580 X10^{-5}	1.6484 X10^{-5}	1.2600 X10^{-5}	1.2700 X10^{-5}	1.0061 X10^{-5}	1.5300 X10^{-5}	1.2008 X10^{-5}	1.3589 X10^{-5}	1.0931 X10^{-5}
5	^{3}K$_8$	7.4953 X10^{-7}	6.2512 X10^{-7}	1.1115 X10^{-6}	6.8824 X10^{-7}	1.0330 X10^{-6}	5.5999 X10^{-7}	1.0422 X10^{-6}	5.2235 X10^{-7}	8.1634 X10^{-7}	6.0301 X10^{-7}	8.1978 X10^{-7}	6.1716 X10^{-7}	6.3371 X10^{-7}	6.2377 X10^{-7}
6	^{5}F$_2$	8.5604 X10^{-7}	4.2152 X10^{-7}	1.2016 X10^{-6}	4.7229 X10^{-7}	1.0963 X10^{-6}	3.4923 X10^{-7}	1.1287 X10^{-6}	3.6706 X10^{-7}	8.5045 X10^{-7}	5.0302 X10^{-7}	9.0355 X10^{-7}	4.8336 X10^{-7}	7.9236 X10^{-7}	4.9405 X10^{-7}
7	^{5}F$_3$	1.6722 X10^{-6}	7.4140 X10^{-7}	2.2603 X10^{-6}	8.3064 X10^{-7}	2.0773 X10^{-6}	6.1425 X10^{-7}	1.9290 X10^{-6}	6.4514 X10^{-7}	1.3923 X10^{-6}	8.8417 X10^{-7}	1.6970 X10^{-6}	8.5017 X10^{-7}	1.4441 X10^{-6}	8.6903 X10^{-7}
8	^{5}F$_4$	4.7384 X10^{-6}	2.1782 X10^{-6}	5.7529 X10^{-6}	2.5677 X10^{-6}	5.0136 X10^{-6}	2.0441 X10^{-6}	6.6613 X10^{-6}	1.7232 X10^{-6}	5.8173 X10^{-6}	2.1873 X10^{-6}	6.6541 X10^{-6}	2.1636 X10^{-6}	6.1655 X10^{-6}	2.3127 X10^{-6}
9	^{5}F$_5$	2.3848 X10^{-6}	2.1253 X10^{-6}	3.0181 X10^{-6}	2.5693 X10^{-6}	2.4849 X10^{-6}	2.1165 X10^{-6}	2.6577 X10^{-6}	1.5939 X10^{-6}	2.4878 X10^{-6}	1.9253 X10^{-6}	2.6252 X10^{-6}	1.9428 X10^{-6}	2.4874 X10^{-6}	2.1369 X10^{-6}
12	rms→ deviations	σ =± 3.0770X10^{-6}		σ =± 3.7237X10^{-6}		σ = ± 3.3497X10^{-6}		σ = ± 6.9037X10^{-6}		σ = ± 4.9033X10^{-6}		σ =± 6.0663X10^{-6}		σ =± 5.0117X10^{-6}	

Table-2: Computed values of oscillator strength for Ho $^{3+}$ doped systems:-

SN	Levels	Ho^{3+}+Da		Ho^{3+}+Db		Ho^{3+}+Ea		Ho^{3+}+Eb		Ho^{3+}+Fa		Ho^{3+}+Fb	
		Pobs	P Cal	Pobs	P Cal	P Cal	P obs	P Cal	P Cal	P obs	P Cal	P obs	P Cal
1	(^{5}G, ^{3}H)$_5$	1.6862X10^{-5}	6.1882X10^{-7}	1.4292X10^{-5}	7.686X10^{-7}	1.036X10^{-5}	1.147X10^{-6}	9.8188X10^{-6}	1.0340X10^{-6}	9.4178X10^{-6}	1.2788X10^{-6}	8.6728X10^{-6}	1.1597X10^{-6}
2	^{5}G$_4$	2.1272X10^{-6}	1.7052X10^{-7}	2.0109X10^{-6}	2.088X10^{-7}	2.039X10^{-6}	3.036X10^{-7}	1.9103X10^{-6}	2.8008X10^{-7}	1.8010X10^{-6}	3.3564X10^{-7}	1.6598X10^{-6}	3.0883X10^{-7}
3	(^{5}G, ^{3}G)$_5$	4.3782X10^{-6}	1.4773X10^{-6}	4.0565X10^{-6}	1.730X10^{-6}	4.613X10^{-6}	2.270X10^{-6}	4.4790X10^{-6}	2.2712X10^{-6}	4.4759X10^{-6}	2.4267X10^{-6}	4.1854X10^{-6}	2.3583X10^{-6}
4	^{5}G$_6$	1.5214X10^{-5}	1.1964X10^{-5}	1.4335X10^{-5}	1.173X10^{-5}	1.700X10^{-5}	1.494X10^{-5}	1.6380X10^{-5}	1.4425X10^{-5}	1.5412X10^{-5}	1.3684X10^{-5}	1.3575X10^{-5}	1.1989X10^{-5}
5	^{3}K$_8$	7.5366X10^{-7}	4.8447X10^{-7}	6.2232X10^{-7}	5.704X10^{-7}	9.734X10^{-7}	8.449X10^{-7}	9.1025X10^{-7}	7.5436X10^{-7}	8.8928X10^{-7}	9.1332X10^{-7}	8.1692X10^{-7}	8.0873X10^{-7}
6	^{5}F$_2$	8.6429X10^{-7}	3.3229X10^{-7}	7.0458X10^{-7}	4.294X10^{-7}	1.072X10^{-6}	6.893X10^{-7}	9.7124X10^{-7}	5.8731X10^{-7}	9.8059X10^{-7}	7.8578X10^{-7}	9.6977X10^{-7}	6.8852X10^{-7}
7	^{5}F$_3$	1.6858X10^{-6}	5.8472X10^{-7}	1.2598X10^{-6}	7.556X10^{-7}	1.846X10^{-6}	1.212X10^{-6}	1.5571X10^{-6}	1.0331X10^{-6}	1.7679X10^{-6}	1.3822X10^{-6}	1.3931X10^{-6}	1.2112X10^{-6}
8	^{5}F$_4$	5.7023X10^{-6}	1.5928X10^{-6}	5.6641X10^{-6}	1.996X10^{-6}	6.482X10^{-6}	3.022X10^{-6}	5.9967X10^{-6}	2.6928X10^{-6}	6.5857X10^{-6}	3.3908X10^{-6}	5.9904X10^{-6}	3.0523X10^{-6}
9	^{5}F$_5$	2.4543X10^{-6}	1.4912X10^{-6}	2.3656X10^{-6}	1.835X10^{-6}	2.545X10^{-6}	2.676X10^{-6}	2.3299X10^{-6}	2.4561X10^{-6}	2.6119X10^{-6}	2.9707X10^{-6}	2.4305X10^{-6}	2.7241X10^{-6}
12	rms→ deviations	σ = ± 5.8312X10^{-6}		σ =± 4.8578X10^{-6}		σ = ± 3.4990X10^{-6}		σ = ± 3.3317X10^{-6}		σ = ± 3.0930X10^{-6}		σ = ± 2.8472X10^{-6}	

Table 3:- Computed values of Judd-Ofelt parameters for Ho^{3+} doped systems.

SN	**Systems**	**M:L**	**T_2**	**T_4**	**T_6**	**T_4/T_6**	**T_4/T_2**
1	Ho $^{3+}$ + Solvent	-	3.28X10^{-10}	1.82X10^{-10}	1.04X10^{-10}	1.74603	5.54X10^{-1}
2	Ho^{3+} +Aa	1:2	2.94X10^{-10}	2.32X10^{-10}	1.17X10^{-10}	1.99198	7.89X10^{-1}
3	Ho^{3+} +Ab	1:1	2.80X10^{-10}	2.04X10^{-10}	8.62X10^{-11}	2.36995	7.29X10^{-1}
4	Ho^{3+} +Ba	1:2	2.98X10^{-10}	1.20X10^{-10}	9.07X10^{-11}	1.32066	4.02X10^{-1}
5	Ho^{3+} +Bb	1:1	2.17X10^{-10}	1.25X10^{-10}	1.24X10^{-10}	1.00406	5.75X10^{-1}
6	Ho^{3+} +Ca	1:2	2.70X10^{-10}	1.34X10^{-10}	1.20X10^{-10}	1.123703	4.98X10^{-1}
7	Ho^{3+} +Cb	1:1	2.24X10^{-10}	1.60X10^{-10}	1.22X10^{-10}	1.308852	7.15X10^{-1}

8	Ho^{3+} +Da	1:2	$2.84X10^{-10}$	$1.15X10^{-10}$	$8.21X10^{-11}$	1.406072	$4.07X10^{-1}$
9	Ho^{3+} +Db	1:1	$2.64X10^{-10}$	$1.35X10^{-10}$	$1.06X10^{-10}$	1.274167	$5.13X10^{-1}$
10	Ho^{3+} +Ea	1:2	$3.28X10^{-10}$	$1.77X10^{-10}$	$1.71X10^{-10}$	1.040143	$5.40X10^{-1}$
11	Ho^{3+} +Eb	1:1	$3.15X10^{-10}$	$1.77X10^{-10}$	$1.45X10^{-10}$	1.221542	$5.63X10^{-1}$
12	Ho^{3+} +Fa	1:2	$2.82X10^{-10}$	$1.89X10^{-10}$	$1.94X10^{-10}$	0.975489	$6.72X10^{-1}$
13	Ho^{3+} +Fb	1:1	$2.37X10^{-10}$	$1.84X10^{-10}$	$1.70X10^{-10}$	1.082181	$7.77X10^{-1}$

Table-4 :- Computed values of Judd-Ofelt parameters for Ho^{3+} doped systems.

SN	**Systems**	**M:L**	**T_2**	**T_4**	**T_6**	**T_4/T_6**	**T_4/T_2**
1	Ho^{3+} + Solvent	-	$3.28X10^{-10}$	$1.82X10^{-10}$	$1.04X10^{-10}$	1.74603	$5.54X10^{-1}$
2	Ho^{3+} +Aa	1:2	$2.94X10^{-10}$	$2.32X10^{-10}$	$1.17X10^{-10}$	1.99198	$7.89X10^{-1}$
3	Ho^{3+} +Ab	1:1	$2.80X10^{-10}$	$2.04X10^{-10}$	$8.62X10^{-11}$	2.36995	$7.29X10^{-1}$
4	Ho^{3+} +Ba	1:2	$2.98X10^{-10}$	$1.20X10^{-10}$	$9.07X10^{-11}$	1.32066	$4.02X10^{-1}$
5	Ho^{3+} +Bb	1:1	$2.17X10^{-10}$	$1.25X10^{-10}$	$1.24X10^{-10}$	1.00406	$5.75X10^{-1}$
6	Ho^{3+} +Ca	1:2	$2.70X10^{-10}$	$1.34X10^{-10}$	$1.20X10^{-10}$	1.123703	$4.98X10^{-1}$
7	Ho^{3+} +Cb	1:1	$2.24X10^{-10}$	$1.60X10^{-10}$	$1.22X10^{-10}$	1.308852	$7.15X10^{-1}$
8	Ho^{3+} +Da	1:2	$2.84X10^{-10}$	$1.15X10^{-10}$	$8.21X10^{-11}$	1.406072	$4.07X10^{-1}$
9	Ho^{3+} +Db	1:1	$2.64X10^{-10}$	$1.35X10^{-10}$	$1.06X10^{-10}$	1.274167	$5.13X10^{-1}$
10	Ho^{3+} +Ea	1:2	$3.28X10^{-10}$	$1.77X10^{-10}$	$1.71X10^{-10}$	1.040143	$5.40X10^{-1}$
11	Ho^{3+} +Eb	1:1	$3.15X10^{-10}$	$1.77X10^{-10}$	$1.45X10^{-10}$	1.221542	$5.63X10^{-1}$
12	Ho^{3+} +Fa	1:2	$2.82X10^{-10}$	$1.89X10^{-10}$	$1.94X10^{-10}$	0.975489	$6.72X10^{-1}$
13	Ho^{3+} +Fb	1:1	$2.37X10^{-10}$	$1.84X10^{-10}$	$1.70X10^{-10}$	1.082181	$7.77X10^{-1}$

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