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1991 J. Phys.: Condens. Matter 3 6069

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## Absorption spectra and electron correlation of the trivalent holmium ion in $\text{LiYF}_4$

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Received 12 April 1991, in final form 21 May 1991

**Abstract.** The optical absorption spectrum of  $\text{Ho}^{3+}$  in  $\text{LiYF}_4$  and its Zeeman effect in magnetic fields along the crystal  $c$  axis were studied in the range from 15 400 to 28 500  $\text{cm}^{-1}$ . The quantum numbers  $\bar{\mu}$  of all crystal field components of  $\text{Ho}^{3+}$  were determined using linearly and circularly polarized transitions from the ground state. The correlated and spin-correlated crystal fields were used for a fitting procedure of the energy levels.

### 1. Introduction

The energy levels of  $\text{Ho}^{3+}$  in  $\text{LiYF}_4$  in the spectral range from 0 to 21 300  $\text{cm}^{-1}$  were first determined by Karayianis *et al* (1976). Christensen (1979) extended this work and recorded the absorption spectrum of  $\text{Ho}^{3+}$  in  $\text{LiHoF}_4$  up to 26 070  $\text{cm}^{-1}$ . He did not, however, assign the energy levels of the multiplets  $^3\text{K}$  and  $^5\text{G}$ . Thus it was only possible to obtain crystal field parameters for the terms of the multiplets  $^5\text{I}$  and  $^5\text{F}$ . Consequently the effects of electron correlation for the  $\text{Ho}^{3+}$  ion could not be studied, since its influence on the crystal field splittings only becomes pronounced for the terms at higher energies.

Therefore, in continuing our previous investigations on  $\text{Ho}^{3+}$  in  $\text{YVO}_4$ ,  $\text{YAsO}_4$ ,  $\text{HoPO}_4$ ,  $\text{LaCl}_3$  and  $\text{Y}(\text{OH})_3$  (Enderle *et al* 1990b,c,a, Pilawa 1990, 1991a,b,c),  $\text{LiHoF}_4$  and  $\text{Ho}^{3+}$  in  $\text{LiYF}_4$  appeared to be appropriate candidates for the further study of the influence of electron correlation on the crystal field splittings of the  $\text{Ho}^{3+}$  ion.

In the first part of this paper, the results of an experimental reinvestigation of the polarized absorption spectrum of  $\text{Ho}^{3+}$  in  $\text{LiYF}_4$  in the energy range from 15 400 to 28 500  $\text{cm}^{-1}$  and its Zeeman effect in magnetic fields up to 4 T are described. These provide the energies of all the crystal field components of  $\text{Ho}^{3+}$  in this range and their crystal quantum numbers. Subsequently, the measured splittings are handled theoretically by using a crystal field operator which includes the effects of electron correlation.

### 2. Experimental details

Single crystals of  $\text{Li}(\text{Y}_{1-x}\text{Ho}_x)\text{F}_4$  with  $x = 0.03, 0.1$  and  $1.0$  were grown by Dr Ward at University of Oxford (Gabbe and Harmer 1968). They crystallize in the scheelite

structure  $I4_1/a$ . The point symmetry of the  $\text{Ho}^{3+}$  sites is  $\bar{4}$ . The crystals were cut and polished so that they could be irradiated parallel and perpendicular to the tetragonal inversion axis ( $c$  axis). The experimental equipment used was the same as described by Enderle *et al* (1990b,c).

The eigenstates of the  $\text{Ho}^{3+}$  ion in  $\text{LiYF}_4$  are characterized by their transformation behaviour with respect to the symmetry operation  $\bar{4}$ . With regard to the  $\bar{4}$ -axis all crystal field states are symmetry singlets. They can be labelled by the crystal quantum number  $\bar{\mu}$

$$\bar{4}|j\bar{\mu}\rangle = e^{i(2\pi/4)\bar{\mu}}|j\bar{\mu}\rangle$$

where  $j$  is a running index. The crystal field states  $|j\bar{\mu}\rangle$  are linear combinations of the free-ion states  $|\gamma JM\rangle$  (Enderle *et al* 1990b).  $J$  and  $M$  are the quantum numbers of the total angular momentum and  $\gamma$  distinguishes between states with the same  $J$ .  $\bar{\mu}$  can take the values 0,  $\pm 1$  and 2. The states  $\bar{\mu} = \pm 1$  are Kramers degenerate doublets. Since the symmetry transformation  $\bar{4}$  is part of the site symmetry  $42m$  of  $\text{Ho}^{3+}$  in host lattices of the tetragonal zircon structure,  $\text{YVO}_4$ ,  $\text{YAsO}_4$  and  $\text{HoPO}_4$ , the selection rules in  $\bar{\mu}$  can be found in one of our preceding papers (Enderle *et al* 1990b). The absorption spectra of  $\text{Ho}^{3+}$  in  $\text{LiYF}_4$  show that the symmetry quantum numbers  $\nu$  and  $S$ , which are valid for the zircon structure, are really not defined for this structure.

### 3. Experimental results

#### 3.1. Analysis of the spectra

In order to determine the energy levels of  $\text{Ho}^{3+}$  in  $\text{LiYF}_4$  the measurements of the absorption spectra were mainly carried out at low temperatures between 1.4 and 5 K. At these temperatures only the lowest crystal field component of the ground term  $^5I_8$  of  $\text{Ho}^{3+}$  in  $\text{LiYF}_4$  is thermally populated. Both linearly and circularly polarized spectra were taken. If transitions from the lower split component of the ground state ( $\bar{\mu} = +1$ ) are observed, the symmetry quantum number of an excited crystal field component can be determined directly from the polarization of the respective absorption line. If the direction of the incident light is perpendicular to the  $c$  axis, electric dipole transitions to excited doublets with  $\bar{\mu} = -1$  are  $\pi$ -polarized and transitions to the excited singlets are  $\sigma$ -polarized. Magnetic dipole transitions which can be observed for excited terms with  $J = 7, 8$  and  $9$  are  $\sigma$ -polarized for transitions to excited doublets with  $\bar{\mu} = +1$  and  $\pi$ -polarized for transitions to excited singlets. If the direction of the incident light is parallel to the  $c$  axis, circularly polarized transitions to the excited singlets can be observed. Electric dipole transitions to the singlets  $\bar{\mu} = 0$  and  $2$  are  $\sigma^+$ - and  $\sigma^-$ -polarized, respectively. Magnetic dipole transitions to the singlets  $\bar{\mu} = 0$  and  $2$  are  $\sigma^-$ - and  $\sigma^+$ -polarized, respectively. The Zeeman effect of all absorption lines was recorded for magnetic fields along the  $c$  axis only.

#### 3.2. Discussion of the individual terms

The energies and the splitting factors of the determined crystal field components are given in table 1. The energies are related to the lowest component of the ground

term  ${}^5\text{I}_8$ . The absolute errors of the energies are  $\pm 0.4 \text{ cm}^{-1}$ ; the relative errors of the distances between the components of one term are usually smaller. The splitting factors are determined in the limit of low magnetic fields (for a definition including sign conventions see Enderle *et al* 1990b). Their errors amount to  $\pm 0.2$ , as a rule. Larger errors caused by broad absorption lines or extremely non-linear magnetic shifts are given in the table. Most of the data were obtained from  $\text{LiYF}_4$  samples containing 3%  $\text{Ho}^{3+}$ . For some terms ( ${}^5\text{S}_2$ ,  ${}^3\text{H}_{5,6}$  and  ${}^5\text{G}_2$ ) samples containing 10%  $\text{Ho}^{3+}$  had to be used to render possible a complete analysis. Pure  $\text{LiHoF}_4$  was only necessary for the term  ${}^5\text{F}_2$ .

Table 1. Energies, crystal quantum numbers and Zeeman splitting factors.

$J$ -term	Centre of gravity ( $\text{cm}^{-1}$ )	Energy of the component ( $\text{cm}^{-1}$ )	Crystal quantum numbers $\bar{\mu}$	Splitting factor	
${}^5\text{I}_8$		0.0	$\pm 1$	-6.87	
		7.0	2		
${}^5\text{F}_5^a$	15 582.1	15 489.2	2	-2.0	
		15 495.2	$\pm 1$		
		15 512.6	0		
		15 558.7	0		
		15 622.8	$\pm 1$		0.6
		15 632.0	2		
		15 639.8	0		
15 667.3	$\pm 1$	5.2			
${}^5\text{S}_2^b$	18 511.0	18 486.8	2	-1.8	
		18 490.7	0		
		18 522.0	-1		+1.8
		18 522.1	+1		
		18 533.5	2		
${}^5\text{F}_4^a$	18 656.9	18 604.9	0	0.6	
		18 607.6	2		
		18 614.5	+1		-0.6
		18 614.6	-1		-3.5
		18 685.3	$\pm 1$		
		18 685.4	0		
		18 700.7	2		
		18 713.9	0		
${}^5\text{F}_3^a$	20 707.0	20 642.0	+1	-0.6	
		20 642.2	-1		+0.4
		20 663.1	0		
		20 711.4	2		
		20 754.8	2		
		20 767.7	+1		-2.1
		20 768.0	-1		+2.1
${}^5\text{F}_2^b$	21 265.1	21 118.3 <sup>c</sup>	2	1.1	
		21 132.5	0		
		21 173.7	$\pm 1$		
		21 227.1	2		

Table 1 *Continued.*

<i>J</i> -term	Centre of gravity (cm <sup>-1</sup> )	Energy of the component (cm <sup>-1</sup> )	Crystal quantum numbers $\bar{\mu}$	Splitting factor
<sup>3</sup> K <sub>8</sub> <sup>a</sup>	21 428.9	21 370.0	2	
		21 376.5	0	
		21 378.4	-1	-3.4
		21 378.6	+1	2.1
		21 384.3	0	
		21 395.0	-1	-0.1
		21 395.0	+1	0.8
		21 399.5	2	
		21 410.0	0	
		21 455.1	-1	4.0
		21 455.2	+1	-4.0
		21 469.5	0	
		21 469.5	0	
		21 477.1	2	
		21 492.4	2	
		21 492.4	-1	-5.2
21 492.5	+1	5.4		
<sup>5</sup> G <sub>6</sub> <sup>a</sup>	22 269.1	22 130.6	2	
		22 137.6	±1	0.4
		22 150.9	0	
		22 226.6	2	-7.8
		22 226.6	2	7.8
		22 289.7	0	
		22 313.8	±1	5.0
		22 378.8	±1	-0.2
		22 390.7	0	
		22 423.1	2	
<sup>5</sup> F <sub>1</sub> <sup>a</sup>	22 440.1	22 424.0	0	
		22 448.1	±1	1.4
<sup>5</sup> G <sub>5</sub> <sup>a</sup>	24 078.7	23 980.9	0	
		23 990.3	-1	-1.8
		23 990.1	+1	1.8
		24 021.4	2	
		24 082.6	-1	-3.0
		24 082.6	+1	2.5
		24 117.0	0	
		24 120.9	2	
		24 150.5	-1	0.9
		24 150.4	+1	-0.1
24 178.7	0			
<sup>5</sup> G <sub>4</sub> <sup>a</sup>	25 974.0	25 903.1	2	
		25 905.7	0	
		25 957.4	-1	0.9
		25 957.7	+1	-1.4
		25 985.1	0	
		25 986.1	±1	-1.2
		26 019.0	2	
		26 066.2	0	

Table 1 Continued.

$J$ -term	Centre of gravity ( $\text{cm}^{-1}$ )	Energy of the component ( $\text{cm}^{-1}$ )	Crystal quantum numbers $\bar{\mu}$	Splitting factor
$^3\text{K}_7^a$	26 214.1	26 191.0	+1	-5.6
		26 191.3	-1	$5.9 \pm 0.3$
		26 194.3	2	
		26 198.9	2	
		26 202.4	-1	-0.4
		26 202.6	+1	$0.1 \pm 0.5$
		26 206.6	0	
		26 215.7	0	-3.6
		26 215.7	0	3.6
		26 225.2	-1	-2.6
		26 225.2	+1	$0.9 \pm 0.8$
		26 228.8	2	
		26 234.5	-1	3.5
		26 234.4	+1	$-0.8 \pm 0.3$
		26 244.4	2	
$^3\text{H}_5^b$	27 672.8	27 572.5	2	
		27 578.6	$\pm 1$	0.5
		27 592.2	0	
		27 679.5	-1	-3.9
		27 679.6	+1	3.9
		27 709.6	0	
		27 732.9	2	
		27 753.9	$\pm 1$	1.8
		27 769.5	0	
$^3\text{H}_6^b$	27 917.5	27 809.1	2	
		27 818.2	0	
		27 864.8	2	
		27 867.6	$\pm 1$	$1.1 \pm 0.4$
		27 887.1	2	
		27 891.4	$\pm 1$	$-2.5 \pm 1.0$
		27 951.0	0	
		27 990.6	0	
		28 023.7	$\pm 1$	$1.3 \pm 0.4$
28 041.7	2			
$^5\text{G}_2^b$	28 351.1	28 299.1	2	
		28 317.4	0	
		28 354.2	$\pm 1$	1.0
		28 430.8	2	

<sup>a</sup> Crystals containing 3%  $\text{Ho}^{3+}$  were used to analyse the terms.

<sup>b</sup> Crystals containing 10%  $\text{Ho}^{3+}$  were used to analyse the terms.

<sup>c</sup> Pure  $\text{LiHoF}_4$  was used.

3.2.1.  $^5\text{I}_8$ . Similar to  $\text{Ho}^{3+}$  in  $\text{YAsO}_4$  and  $\text{HoPO}_4$ , the lowest crystal field component is a doublet  $\bar{\mu} = \pm 1$ . The value of the splitting factor  $s_{\bar{\mu}}$  was determined to be  $-6.87 \pm 0.2$  for 10%  $\text{Ho}^{3+}$  in  $\text{LiYF}_4$  which is in good agreement with the value found for pure  $\text{LiHoF}_4$  ( $-6.9 \pm 0.25$ , Battison *et al* 1975). The large value of  $s_{\bar{\mu}}$  indicates that the ground doublet mainly consists of  $M = \pm 7$ . Therefore the lowest Zeeman

component has the symmetry quantum number  $\bar{\mu} = +1 (M = -7)$ .

*3.2.2.  ${}^5F_5$ .* All the transitions allowed at low temperatures were observed. Measurements of the Zeeman effect additionally showed one weak  $\sigma$ -polarized transition to the  $\bar{\mu} = +1$  component of the lowest doublet. If the site symmetry is strictly  $\bar{4}$ , such a transition would be allowed in magnetic dipole radiation only if the term has  $J=7, 8$  and  $9$ . The transition to the upper doublet was very strong, so that the spectrum was over-absorbed. The energy of this doublet could be fixed, however, with higher accuracy by the weak transition from the first excited singlet  $\bar{\mu} = 2$  of the ground term at  $20$  K.

*3.2.3.  ${}^5S_2$  and  ${}^5F_{4,3,2}$ .* All crystal field components were determined at low temperatures. Again forbidden transitions from the ground state to the  $\bar{\mu} = +1$  component of the doublets were observed. These were  $\sigma$ -polarized for the cases of transitions to the lower doublet of the term  ${}^5F_4$  and to all doublets of  ${}^5S_2$  and  ${}^5F_3$ . For  ${}^5F_3$  the forbidden transition also existed in the  $\sigma^+$ -spectrum. This suggests admixtures of the nearby  $\bar{\mu} = 0$ . Since the transition to the lower singlet  $\bar{\mu} = 2$  of  ${}^5F_2$  is very weak this singlet could only be recorded in the  $\sigma^-$ -spectrum of the pure  $\text{LiHoF}_4$  sample.

*3.2.4.  ${}^3K_3$ .* All crystal field components could be ascertained at low temperatures. Electric and magnetic dipole transitions were observed to all doublets of the term. The  $\bar{\mu} = +1$  component of the lowest doublet also appeared in the  $\sigma^+$ -spectrum indicating an interaction with the neighbouring singlet  $\bar{\mu} = 0$ . Electric dipole transitions were found to the lower three singlets  $\bar{\mu} = 0$ , whereas both electric and magnetic dipole transitions were found to the upper pair of singlets  $\bar{\mu} = 0$ . This pair of singlets is degenerate. It mainly contains  $M = \pm 8$  states, so that the Zeeman shift of the upper singlet cannot be observed because a transition from the ground state component  $\bar{\mu} = +1$  (mainly  $M = -7$ ) is forbidden, in agreement with the selection rules (Enderle et al 1990b). The symmetry quantum number of this singlet pair was confirmed by the transition from the first excited singlet  $\bar{\mu} = 2$  of the ground term which was observed at  $20$  K in  $\pi$ -polarization. Electric dipole transitions were found to the lower two singlets  $\bar{\mu} = 2$  and both electric and magnetic dipole transitions could be recorded to the upper two singlets  $\bar{\mu} = 2$ .

*3.2.5.  ${}^5G_6$  and  ${}^5F_1$ .* All crystal field components were observed at low temperatures. The middle pair of the singlets  $\bar{\mu} = 2$  of  ${}^5G_6$  is degenerate. It shows a linear Zeeman shift. The shift of its upper component could only be observed in magnetic fields above  $2$  T. The singlet  $\bar{\mu} = 0$  of  ${}^5F_1$  lies close to the upper singlet  $\bar{\mu} = 2$  of  ${}^5G_6$ . Its energy could be ascertained by backward extrapolation of the Zeeman shift of the absorption line observed in the  $\sigma^+$ -spectrum. A forbidden transition was seen to the  $\bar{\mu} = +1$  component of the lowest doublet of  ${}^5G_6$ .

*3.2.6.  ${}^5G_5$  and  ${}^5G_4$ .* All crystal field components could be determined at low temperatures. Forbidden transitions to the  $\bar{\mu} = +1$  components could be observed to all doublets of  ${}^5G_5$  and to the lower doublet of  ${}^5G_4$ .

*3.2.7.  ${}^3K_7$ .* All components could be ascertained at low temperatures. Magnetic dipole transitions occurred to all doublet components  $\bar{\mu} = +1$  and to the two upper singlets  $\bar{\mu} = 2$ . The upper singlet pair  $\bar{\mu} = 0$  turned out to be degenerate.

3.2.8.  $^3H_5$ . All crystal field components were determined at low temperatures. One forbidden transition to the  $\bar{\mu} = +1$  component of the middle doublet was found.

3.2.9.  $^3H_6$ . The absorption lines were relatively broad. Therefore the errors of the energies ( $\pm 2 \text{ cm}^{-1}$ ) and of the splitting factors are very large. The second singlet  $\bar{\mu} = 0$  remains uncertain. The corresponding absorption lines were weak but could be observed in  $\sigma^+$ - as well as in  $\sigma^-$ -polarization.

3.2.10.  $^5G_2$ . All crystal field components were determined at low temperatures.

The transitions between the ground state component  $\bar{\mu} = +1$  and the components  $\bar{\mu} = +1$  of the doublets in the excited terms, which were observed in the Zeeman spectrum of nearly all terms, are generally very weak and  $\sigma$ -polarized. A probable explanation may be that a component of the magnetic field perpendicular to the  $c$  axis produces admixtures of singlet states into the doublets. Indeed a dependence of the absorption intensity on the magnetic-field strength was observed for the terms  $^3K_8$  and  $^5G_6$ . But for all other terms there was no field dependence. A possible explanation using the assumption that the site symmetry for the  $\text{Ho}^{3+}$  ions may be reduced, e.g. by small local distortions, seems very unlikely since the reduction would even lead to triclinic symmetry. The consequence would be that transitions allowed in the  $\sigma$ -spectrum should also be seen in the  $\pi$ -spectrum and *vice versa*, which was not the case.

#### 4. Crystal-field calculation

For a comparison of the observed crystal field components with calculations the following crystal field Hamiltonian (Pilawa 1991c) was used

$$H_{\text{CF}} = \sum_{k=2,4,6} \sum_{q=0,\pm 4} e_{q,1}^{(k)} \left[ O_{q,1}^{(k)} + \sum_{\substack{t \\ t \neq 1}} c_{q,t}^{(k)} * O_{q,t}^{(k)} \right].$$

Here the  $O_{q,1}^{(k)}$  are one-electron operators which are proportional to the tensor operators  $\sum t_q^{(k)}(i)$  ( $i$  is the electron on which the operator acts). The  $O_{q,t}^{(k)}$  ( $t \neq 1$ ) are linear combinations of the  $\sum t_q^{(k)}(i)$  and operators of the correlated (Bishton and Newman 1970) or spin-correlated crystal field (Judd 1977). The site symmetry  $\bar{4}$  of the  $\text{Ho}^{3+}$  ions allows the parameters to be complex. Karayianis *et al* (1976) found, however, that the complex contribution to the crystal field is really small. Thus all crystal field parameters were supposed to be real. The calculation was restricted to those operators  $O_{q,t}^{(k)}$  which were found to be essential in describing the correlation effects within the investigated spectral range for  $\text{Ho}^{3+}$  in other compounds (Pilawa 1991a,b,c). These operators are given in table 5 of the Pilawa paper (1991a). The atomic intermediate coupling wavefunctions were calculated with the parameter values  $E^1 = 6698.4 \text{ cm}^{-1}$ ,  $E^2 = 28.79 \text{ cm}^{-1}$ ,  $E^3 = 630.2 \text{ cm}^{-1}$ ,  $\zeta = -2086.7 \text{ cm}^{-1}$ ,  $\alpha = 31.20 \text{ cm}^{-1}$ ,  $\beta = -992.6 \text{ cm}^{-1}$  and  $\gamma = 0$  (Domann 1974). The matrix elements of  $H_{\text{CF}}$  were calculated for all terms up to  $35\,000 \text{ cm}^{-1}$  and the full matrix was then



diagonalized. It includes 316 states which could be attached to the different symmetry types of  $\bar{4}$ . The measured crystal field components were compared with the calculated ones. All crystal field parameters were allowed to vary freely. The obtained parameter values are given in table 2. The parameters  $e_{q,i}^{(k)}$  are in good agreement with the values obtained by Christensen (1979), who also supposed the parameters to be real. The corresponding  $e_{q,i}^{(k)}$  values given in column (c) of table 2 were calculated from his  $B_q^{(k)}$  parameters using the following conversion factors:  $-0.77$  for  $k = 2$ ,  $1.07$  for  $k = 4$ ,  $-1.07$  for  $k = 6$ . The correlated crystal field parameters  $c_{q,i}^{(k)}$  are very similar to those found for zircon structures (Pilawa 1991c). Table 3 shows the root-mean-square (RMS) deviations between the measured and calculated crystal field components which can be obtained without and with inclusion of correlated crystal field operators. The introduction of correlated crystal field operators has a small influence on the RMS deviation of the terms  ${}^5I$  and  ${}^5F$  but significantly improves the fits of the terms  ${}^3K_{3,7}$ ,  ${}^5G_{5,4,2}$  and  ${}^3H_{5,6}$ . There is no improvement for  ${}^5F_1$  and even a worsening for  ${}^5S_2$  and  ${}^5F_2$  which may be attributed to the same causes as those given for  $\text{Ho}^{3+}$  in the zircon structures by Pilawa (1991c). The RMS deviations are generally larger than those obtained for the zircon structures. The reason may be that the complex parts of the crystal field parameters, which are not necessarily zero for the site symmetry  $\bar{4}$ , were not included in our calculation.

**Table 2.** Crystal-field parameters. Column (a), parameter values obtained by fitting the multiplets  ${}^5I$  and  ${}^5F$  to the crystal field operator without inclusion of correlation effects. The experimental splittings of the terms  ${}^5I_{7,6,5,4}$  were taken from Christensen (1979). Column (b), parameter values determined by fitting all measured crystal field components to the crystal field operator including correlation effects. Column (c), parameter values deduced from Christensen (1979). All figures have the units  $\text{cm}^{-1}$ .

	(a)	(b)	(c)
$e_{0,1}^{(2)}$	-300	-294	-292
$e_{0,1}^{(4)}$	-680	-715	-670
$e_{4,1}^{(4)}$	900	958	889
$e_{0,1}^{(6)}$	53	117	56
$e_{4,1}^{(6)}$	-706	-729	-651
$c_{,3}^{(2)}$	—	-0.10	—
$c_{,3}^{(4)}$	—	-0.05	—
$c_{,4}^{(4)}$	—	0.22	—
$c_{,5}^{(4)}$	—	0.21	—
$c_{,2}^{(6)}$	—	-0.06	—

## 5. Conclusions

By analysing the spectrum of  $\text{Ho}^{3+}$  in  $\text{LiYF}_4$  and its Zeeman effect in the range between  $15000$  and  $28500 \text{ cm}^{-1}$  it was possible to determine the energies and the crystal-quantum numbers of all crystal field components in this energy region with

Table 3. RMS deviations between the observed and the calculated crystal field components of the individual terms: column (a), RMS deviations for a calculation with the parameters of column (a) of table 2; column (b), RMS deviations for a calculation with the parameters of column (b) of table 2. All figures have the units  $\text{cm}^{-1}$ .

	(a)	(b)
$^5\text{I}_7$	1.2	1.5
$^5\text{I}_6$	1.7	1.7
$^5\text{I}_5$	4.6	4.5
$^5\text{I}_4$	1.9	3.4
$^5\text{F}_5$	2.7	3.3
$^5\text{S}_2$	3.2	7.0
$^5\text{F}_4$	3.2	3.0
$^5\text{F}_3$	2.2	3.0
$^5\text{F}_2$	3.6	4.9
$^3\text{K}_8$	7.8	2.6
$^5\text{G}_6$	11.5	8.0
$^5\text{F}_1$	10.7	10.5
$^5\text{G}_5$	30.4	4.4
$^5\text{G}_4$	15.3	5.7
$^3\text{K}_7$	14.3	6.4
$^3\text{H}_5$	18.2	5.5
$^3\text{H}_6$	26.6	11.4
$^5\text{G}_2$	24.4	9.6

great certainty. These experimental results were used for a comparison with crystal field calculations including the influence of electron correlation. It turned out that the correlation is clearly perceptible only in a few multipole contributions and that these are the same as found for  $\text{Ho}^{3+}$  in other host lattices.

### Acknowledgment

The authors would like to thank Dr R. C. C. Ward, Clarendon Laboratory, University of Oxford for growing the crystals.

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