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Doppler limited laser spectroscopy on hafnium lines. Part II: Hyperfine structure of odd-parity levels

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Abstract. The hyperfine structure of selected odd-parity levels of the configurations $5d6s\ ^2\!6p$ and $5d\ ^2\!6s6p$ of ^{177}Hf I was studied in 10 lines lying in the red spectral region. Hyperfine spectra were obtained by the method of laser induced fluorescence in the plasma of a liquid nitrogen cooled hollow cathode discharge. The observed hyperfine structure constants A and B, together with results from earlier studies were analyzed by means of a parametric method. The interpretation has been carried out based on a refined multiconfigurational fine structure calculation including the main Rydberg series configurations mutually interacting. The set of fine structure parameters as well as the leading eigenvector percentages of levels relevant for this paper are given. The following single electron hfs parameters were deduced for ^{177}Hf: $a^{01}_{5d} = 98.8(0.8)$ MHz, $a^{01}_{6p} = 204.6(6.4)$ MHz, $b^{02}_{5d} = 4129(133)$ MHz, $b^{02}_{6p} = 7847(266)$ MHz for the lowest configuration.

PACS. 31.30.Gs Hyperfine interactions and isotope effects, Jahn-Teller effect

1 Introduction

Measurements of isotope shift (IS) and hyperfine structure (hfs) by means of high-resolution spectroscopy have been intensively carried out over the last decade and yielded important information on nuclear properties and atomic structures; for example, a comparison between deformation and charge radii changes may provide an interesting and detailed insight into the collective and single particle nuclear features. However, there are few data available for refractory elements for which the conventional method for production of an atomic beam by an electrically heated oven cannot be applied.

It is very interesting to study the nuclear properties of refractory elements [1] with $Z = 71$–78 and particularly hafnium which has the longest known isotopic chain of 31 isotopes; the $_{72}$Hf isotopes with neutron numbers 100-110 are located close to the well-investigated rare earths in the very interesting mass region of strongly deformed nuclei (see for instance figures 12 and 13 of Ref. [2])

The only two stable odd-mass hafnium isotopes are ^{177}Hf and ^{179}Hf with nuclear spins $I = \frac{7}{2}$ and $\frac{9}{2}$, respectively. As is commonly the case with the $5d$-elements, the hafnium atom has many low-lying levels below $3 \times 10^4\,\mathrm{cm}^{-1}$ which are members of the even-parity configurations as well as the odd-parity ones. To study hfs of some levels of the latter different techniques have been successfully tried: Zimmermann *et al.* [3] reported hfs and IS measurements of 14 lines by means of laser spectroscopy using an electron bombardment technique; hfs for three optical transitions was measured using the laser ablation atomic beam [4] and the resonance gas cell methods [5].

We wanted to extend in turn these high-resolution hfs investigations to the red region using Doppler limited laser spectroscopy to get new experimental data in order to add them to those previously determined and to deduce one-electron hyperfine parameters thanks to more accurate intermediate coupling wave functions obtained from a new fine structure analysis as regards odd-parity levels.

2 The observations

High-resolution Doppler limited laser spectroscopy has been employed to investigate the hfs of selected lines in the red spectral region (Fig. 1 and Tab. 1).

Using laser induced fluorescence and optogalvanic detection methods, measurements were performed in the plasma of a liquid nitrogen cooled hollow cathode discharge in the atomic spectrum of hafnium.

The complete details of the experiments were already given in the first part of this work [6]. Nevertheless, let us mention once more that an actively stabilized cw titan-sapphire laser (Coherent, model 899-21) pumped by an

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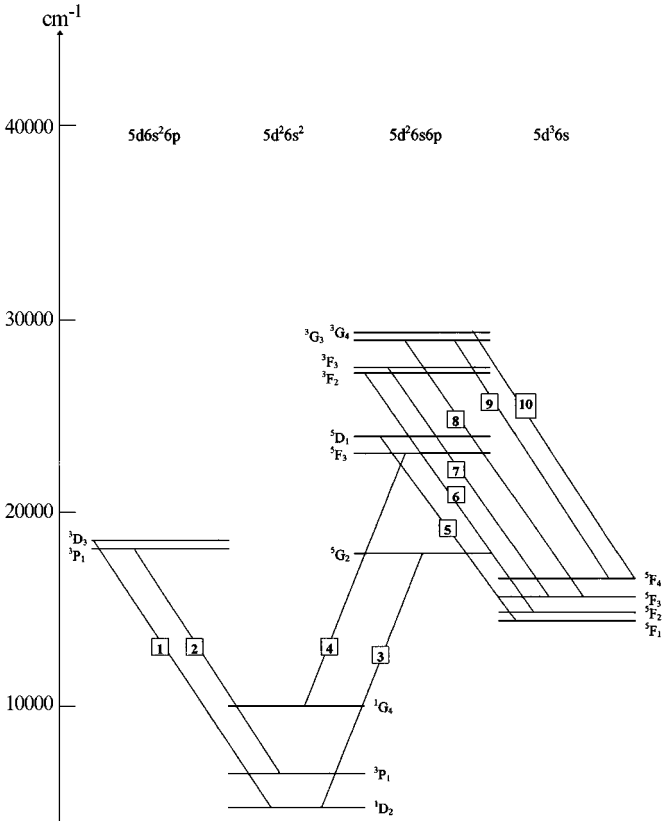

Fig. 1. Investigated lines in this work. The numbered wavelengths are given in Table 1.

Table 1. List of studied spectral lines. T_i is the energy value of the lower even level. T_u is the energy value of the upper level. N is the number assigned in Figure 1 to these lines.

$\lambda_{\rm air}$(nm)	lower level	T_i(cm^{-1})	upper level	T_u(cm^{-1})	N
784.537	$a\ ^1$D$_2$	5639	$z\ ^3$D$_3$	18382	1
864.004	$a\ ^3$P$_1$	6572	$z\ ^3$P$_1$	18143	2
808.027	$a\ ^1$D$_2$	5639	$z\ ^5$G$_2$	18011	3
774.017	$a\ ^1$G$_4$	10533	$z\ ^5$F$_3$	23449	4
900.474	$a\ ^5$F$_1$	14092	$z\ ^5$D$_1$	25194	5
805.647	$a\ ^5$F$_2$	14740	$y\ ^3$F$_2$	27150	6
834.425	$a\ ^5$F$_3$	15673	$y\ ^3$F$_3$	27654	7
774.017	$a\ ^5$F$_3$	15673	$z\ ^3$G$_3$	28584	8
846.000	$a\ ^5$F$_4$	16767	$z\ ^3$G$_3$	28584	9
801.059	$a\ ^5$F$_4$	16767	$z\ ^3$G$_4$	29247	10

argon-ion laser (15W) was used as laser source. Typical single mode output power was 1W in the red region (780-915 nm). The measurements were achieved with an enriched ^{177}Hf isotope probe. The abundances of the isotopes in the probe, as given by the supplier, were as follows: ^{174}Hf: 0.62%, ^{176}Hf: 0.87%, ^{177}Hf: 91.38%, ^{178}Hf: 4.92%, ^{179}Hf: 1.01%, and ^{180}Hf: 1.80%.

The schematic experimental set-up for laser induced fluorescence and optogalvanic detection is shown in Figure 2. Table 2 gives the experimental hfs data of seven $5d^26s6p$ levels and two $5d6s\ ^26p$ ones. For comparison we inserted experimental data achieved by Zimmermann *et al.* [3] who used lines lying in the visible spectral region, to reach the same levels that we studied.

3 Fine structure analysis

Twenty years ago Wyart [7] performed a systematic study of even configurations $5d\ ^N6s^2$, $5d\ ^{N+1}6s$ and $5d^{N+2}$ in neutral atoms of the platinum group resorting to a parametric method. In this work he also skimmed over the odd configuration fine structure of Hf I and Lu I (Tab. 8 in Ref. [7]). Unfortunately no calculated level energies nor eigenvector components were given. Furthermore he fitted 104 levels, observed by Meggers and Moore [8], by means of 35 parameters with a poor root mean squares deviation: 196 cm^{-1}.

Recently, Martin and Sugar [9] examined available data for the odd-parity energy level structures in Hf I [8] and gave the results of Hartree-Fock calculations for Hf $5d6s\ ^26p$ including configuration interactions within the $(d+s)^3p$ complex.

Aside from *ab initio* calculations achieved for Hf $5d6s^26p$ [9] and the parametric values obtained for the Hf $(5d+6s)^36p$ energy integrals [7], however, few details were given. So we performed here a more complete and reliable energy level analysis taking into account the upper terms of the $7p$ and $8p$ Rydberg series which can give rise to some perturbations.

The method used here was successfully tested for the system consisting of 33 configurations of Si I [10] and should find particular application for systems composed of many Rydberg configurations mutually interacting.

The configuration basis set-up used in this work consists of the following 7 configurations:

$$5d\ ^26s6p + 5d\ ^26s7p + 5d\ ^26s8p + 5d6s\ ^26p + 5d6s\ ^27p + 5d6s\ ^28p + 5d\ ^36p\,.$$

Although the total number of interaction integrals required for this basis is large the situation was made tractable by imposing physically realistic ratios of radial integrals as constraints [10].

The majority of the experimentally known odd-parity levels [8] located up to 5.5×10^4 cm^{-1} were fitted (some of them situated around 3.3×10^4 cm^{-1} were discarded).

All the parameters of the configurations $5d6s\ ^26p$, $5d^26s6p$ and $5d\ ^36p$ were adjusted. For these three configurations we defined only one spin-orbit parameter $\zeta(5d,5d)$. The relation between $\zeta(5d,5d)$ and the one-configuration spin-orbit parameters $\zeta_{5d}(5d^{N+M}6s^{2-M})$ is

$$\zeta_{5d}(5d^{N+M}6s^{2-M}) = \zeta(5d,5d) + [2/5][1-(N+M)]P_1 - [2(2-M)/\sqrt{5}]P_4 + [2/5]P_5\delta(M,0)\,,$$

where $N=1$ and $M=0,1,2$. We assumed $P_4=P_5=0$ in our fitting procedure.

In the configurations $5d6s\ ^27p$, $5d6s\ ^28p$, $5d\ ^26s7p$ and $5d\ ^26s8p$ the average energies only were fitted in view of

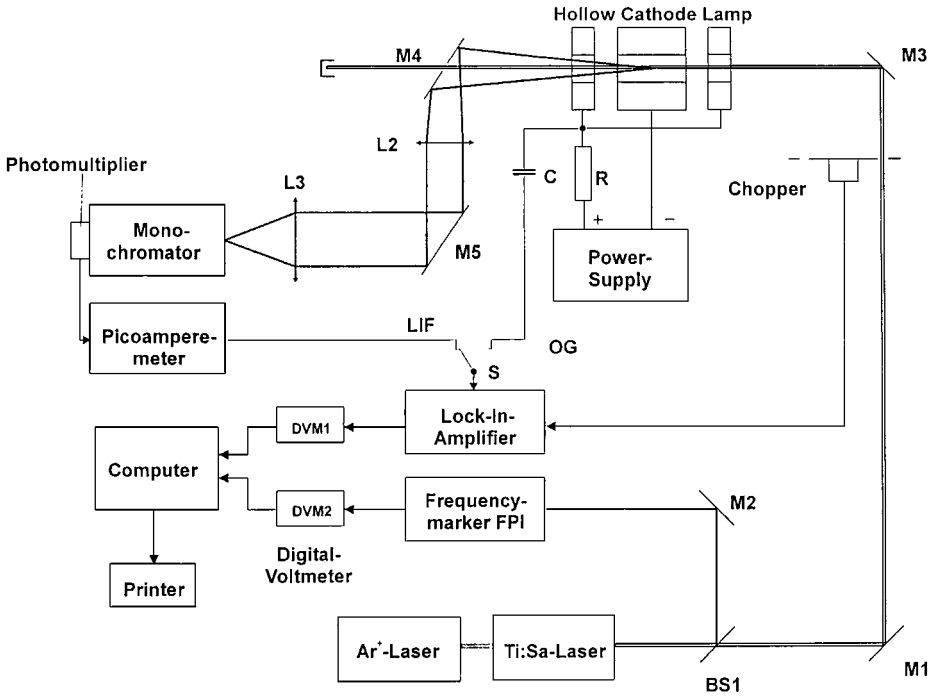

Fig. 2. Experimental set-up for laser induced fluorescence (LIF) and optogalvanic detection (OG).

Table 2. Hyperfine constants A and B in MHz determined from laser induced fluorescence measurements.

Level		Energy (cm^{-1})	A measured	B measured	
$5d^26s6p$	$z\ ^5G_2$	18011	-204.4 ± 0.4	2479 ± 4	
			-205.49 ± 30	2479.1 ± 3.3	Z
	$z\ ^5F_3$	23449	249.2 ± 0.5	1053 ± 4	
			250.08 ± 0.68	$1173.4^{(*)}$	Z
	$z\ ^5D_1$	25194	638.5 ± 1.5	-566 ± 10	
	$y\ ^3F_2$	27150	-150.2 ± 0.3	-746 ± 1	
	$y\ ^3F_3$	27654	279.3 ± 0.3	-845 ± 4	
	$z\ ^3G_3$	28584	-33.4 ± 0.3	923 ± 3	
	$z\ ^3G_4$	29247	207.8 ± 0.2	1292 ± 15	
$5d6s^26p$	$z\ ^3P_1$	18143	146.3 ± 0.3	-695 ± 3	
			146.28 ± 0.30	-694.7 ± 2.5	Z
	$z\ ^3D_3$	18382	151.4 ± 0.2	$-26.9^{(f)}$	
			151.40 ± 0.20	-26.9 ± 3.6	Z

$^{(*)}$ Deduced from B^{179} using the ratio of B values known from ABMR.
$^{(f)}$ A determined with B fixed to the value given in reference [3].
Z: Zimmermann *et al.* [3].

the limited number of available levels. So in order to reduce the number of free parameters, the relation between radial integrals differing only by the principal quantun number of the excited electrons has been used:

$$R^k(a,b) = [n^*(c)n^*(d)/n^*(a)n^*(b)]^{3/2} R^k(c,d)\,,$$

where a, b, c and d stand for configurations. n^* is the effective quantum number of the configuration and is determined from the following relation:

$$n^* = [Z^2 R_\infty/(E_\infty - E_{\rm av})]^{1/2}\,,$$

where E_∞ is the position of the centre of gravity of the ionisation limits and $E_{\rm av}$ is the average energy, to the second order of perturbation theory, of the considered configuration, R_∞ is the Rydberg constant and $Z = 1$ for neutral atom, $Z = 2$ for singly ionized atom, etc.

Table 3. Values (in cm^{-1}) of the fs spin-orbit parameters ζ used in the hfs analysis.

$\zeta(5d,5d) = 1319(10)$			
$P_1 = 78(6)$			
	Configurations: $5d6s^2np$		
	$n=6$	$n=7$	$n=8$
ζ_{5d}	1319(10)	1319[a]	1319[a]
ζ_{np}	2952(48)	736[b]	286[b]
	Configurations: $5d^26snp$		
	$n=6$	$n=7$	$n=8$
ζ_{5d}	1288(116)	1288[a]	1288[a]
ζ_{np}	2559(23)	637[b]	247[b]
	Configuration: $5d^36p$		
ζ_{5d}	1257(113)		
ζ_{6p}	1838(50)		

[a] Taken as equal to the fitted parameter of the lowest configuration ($n=6$) of the same Rydberg series.
[b] The spin-orbit integrals are related by means of configuration effective quantum numbers according to definition in reference [10].

In addition we assumed that in the same Rydberg-series configurations the spin-orbit integrals for $5d$-electrons are equal whereas those for p-electrons are related, like that for the radial integrals, by means of effective configuration quantum numbers.

Thus 158 odd levels were fitted, recurring to 41 variable parameters. The standard deviation of the fitting procedure was equal to 39 cm^{-1}. The results of this fit and the fine structure analysis including predictions of unknown fs levels will be published separately [11]. In Table 3 we give the values of the spin-orbit parameters needed for the hfs analysis and in Table 4 part of the fs fit limited to the levels under hfs studies.

4 Hyperfine structure analysis

According to Sandars and Beck [12] the hyperfine interaction can be described relativistically correct (though using non-relativistic wave functions) by effective Hamiltonians. The magnetic dipole constant A as well as the electric quadrupole constant B are functions of effective radial parameters $a^{\kappa k}$ ($\kappa k = 01, 12, 10$) and $b^{\kappa k}$ ($\kappa k = 02, 13, 11$). Using the intermediate coupling wave functions obtained from the fine structure analysis the experimental A and B hyperfine constants of 16 levels under study were expressed as linear combinations of $45a^{\kappa k}$ and $35b^{\kappa k}$ parameters, describing the hfs of $5d$-electrons, np-electrons ($n = 6, 7, 8$) and the $6s$-electron.

In order to reduce the number of free parameters (necessarily less than sixteen in each expansion of A and B) we made the following approximations:

1. a^{10}_{6s} remains the same for the 3 configurations with an open s-shell.
2. $a^{\kappa k}_{6p}(v) = (\zeta_v/\zeta_x)a^{\kappa k}_{6p}(x)$ and $b^{\kappa k}_{6p}(v) = (\zeta_v/\zeta_x)b^{\kappa k}_{6p}(x)$, where (v) and (x) stand for one of the 7 investigated configurations.
   $a^{\kappa k}_{5d}(y) = (\zeta_y/\zeta_z)a^{\kappa k}_{5d}(z)$ and $b^{\kappa k}_{5d}(y) = (\zeta_y/\zeta_z)b^{\kappa k}_{5d}(z)$, where (y) and (z) stand for one of the 3 investigated sets of Rydberg series configurations ($\kappa k = 01, 12, 02$). Furthermore b^{13}_{5d} and b^{11}_{5d} are imposed equal for all configurations whereas a^{10}_{5d} changes for core polarisation part according to the cases where $6s$-subshell is closed, opened or empty [13].
   For the spin-orbit parameter ζ the values found from the energy level fit (Tab. 3) were used.
3. $a^{12}_{5d} = 1.233a^{01}_{5d}$, $b^{13}_{5d} = 0.545\ b^{02}_{5d}$ and $b^{11}_{5d} = -0.182\ b^{02}_{5d}$. The ratios between these parameters were taken from relativistic Hartree-Fock calculations [14].
4. $a^{12}_{6p} = 1.46\ a^{01}_{6p}$ and $b^{02}_{6p} = -3.95\ b^{11}_{6p}$. Here we used Sandars and Becks method [12] which recurs to Casimir's factors [15], tabulated by Kopfermann [16].

Finally eight $a^{\kappa k}_{nl}$ and the two $b^{\kappa k}_{nl}$ were used as free parameters in the hfs fit. For these parameters the errors are quoted in Table 5 and Table 6.

Let us point out why we preferred to fit in each case the sum of the A and B hyperfine constants of the two close levels at 23449 and 23645 cm^{-1} instead of their individual values: one can notice that they have a quite similar eigenvector composition (Tab. 4) but opposite differences between E_{obs} and E_{cal} and also between g_{obs} and g_{cal}.

In the absence of *ab initio* calculations available in literature we were not able to compare our one-electron hfs parameters as regards p- and d-electrons. Fortunately for open s-shell we can determine the most important parameter for contribution to the A magnetic factor, a^{10}_{6s} which provides information on the electron density at the origin. Using isotope shift data and particularly the derived value of the transition electronic factor $E_{6s} = \pi a_0^3|\psi_{6s}(0)|^2/Z$ we can deduce this parameter: $a^{10}_{6s} = 8R_\infty\alpha^2 g'_I E_{6s} F_r(1-\delta)(1-\varepsilon)/3$ with α the fine structure constant, and g'_I the nuclear g factor referred to the Bohr magneton. F_r is a relativistic correction tabulated in reference [16]. δ and ε are small corrections taking into account the effect of the extended distrubution of nuclear charge and of nuclear magnetic moment, respectively, on a^{10}_{6s}. For the electronic factor E_{6s} different experimental values are given: $E_{6s} = 0.437(22)$ [3], 0.454(27) [4], 0.63(2) [17]. When taking the average value, *i.e.* $E_{6s} = 0.507$ one can derive $a^{10}_{6s} = 3568$ MHz, exactly the same value as given in Table 5.

To test the quality of our hfs analysis we calculated the value of the quadrupole moment Q_{5d} using the values a^{01}_{5d} and b^{02}_{5d} given, respectively, in Table 5 and Table 6 and the formula [12,14]:

$$Q_{\kappa k}\ \frac{2\mu_I}{e^2}g_I\frac{b^{\kappa k}}{a^{\kappa k}}\frac{F^{\kappa k}}{R^{\kappa k}} = 0.4061 \times 0.2267 \times \frac{4129}{98.8} \times 0.971 = 3.74(31)b\,.$$

This value is closer to the Q_{5d} value $= 3.365b$ determined from muonic M X-rays [18] than to the other values evaluated from optical spectra $Q = 4.699b$ [1] or $Q = 4.9b$ [19].

Finally Table 7 shows the comparison between experimental and calculated values of the A and B hyperfine constants. The agreement is very satisfactory regarding

Table 4. Leading eigenvector components for the investigated odd parity levels of Hf I.

$E_{\rm obs}$ [8] ($\rm cm^{-1}$)	$E_{\rm cal}$ ($\rm cm^{-1}$)	ΔE ($\rm cm^{-1}$)	$g_{\rm obs}$ [8]	$g_{\rm cal}$	J	largest components %	%
18011	17977	34	0.394	0.3884	2	86.07 E ^{5}G +	8.18 A ^{2}D
19293	19293	0	0.952	0.9507	3	83.83 E ^{5}G +	8.53 A ^{3}F
20960	20963	−3	1.157	1.1607	4	88.97 E ^{5}G +	5.99 A ^{3}F
22901	22919	−18	1.276	1.2690	5	94.28 E ^{5}G +	2.36 E ^{5}F
23449	23429	20	1.204	1.1449	3	38.55 E ^{5}F + 35.46 A ^{1}F	
25194	25199	−5	1.447	1.4490	1	58.87 E ^{5}D + 10.64 C ^{5}D	
27150	27126	24	0.993	1.0230	2	20.00 E ^{5}D + 18.15 E ^{3}F	
27654	27583	71	1.157	1.1976	3	15.34 E ^{5}D + 13.21 A ^{3}G	
28584	28642	−58	0.940	0.9296	3	33.92 E ^{3}G + 23.18 E ^{3}G	
29247	29223	24	1.071	1.0757	4	34.56 E ^{3}G + 28.34 E ^{3}G	
14018	14000	18	0.542	0.5629	1	63.23 A ^{3}D + 14.31 E ^{3}D	
16163	16192	−29	1.172	1.147	2	61.56 A ^{3}D + 13.82 E ^{3}D	
18143	18173	−30	1.428	1.4115	1	68.07 A ^{3}P +	7.25 E ^{3}P
18225	18193	32	1.246	1.2462	4	66.04 A ^{3}F +	8.90 E ^{3}F
18382	18365	17	1.287	1.2718	3	55.70 A ^{3}D + 12.32 E ^{3}D	
23645	23678	−33	1.074	1.1369	3	39.62 E ^{5}F + 31.73 A ^{1}F	
26464	26458	6	0.996	0.9806	1	65.72 A ^{1}P +	4.26 B ^{1}P

A: $5d6s^26p$ configuration;
B: $5d6s^27p$ configuration;
C: $5d^36p$ configuration;
E: $5d^26s6p$ configuration.

Table 5. Hfs parameters for the magnetic dipole interaction (in MHz). The uncertainties given in parentheses are the standard deviations.

Config.	a^{01}_{5d}	a^{12}_{5d}	a^{10}_{5d}	a^{01}_{np}	a^{12}_{np}	a^{10}_{np}	a^{10}_{6s}
$5d6s^26p$	98.8(0.8)	121.8	444(13)	204.6(6.4)	298.0	−157(11)	
$5d6s^27p$	98.8	121.8	444	50.9	74.4	−39	
$5d6s^28p$	98.8	121.8	444	19.8	28.9	−15	
$5d^26s6p$	96.4	118.9	−525(11)	177.2	230.5	591(70)	3568(65)
$5d^26s7p$	96.4	118.9	−525	44.2	64.2	127	3568
$5d^26s8p$	96.4	118.9	−525	17.2	24.9	50	3568
$5d^36p$	94.1	116.0	−525	127.5	185.6	−348(12)	

Table 6. Hfs parameters for the electric quadrupole interaction (in MHz). The uncertainties given in parentheses are the standard deviations.

Config.	b^{02}_{5d}	b^{13}_{5d}	b^{11}_{5d}	b^{02}_{np}	b^{11}_{np}
$5d6s^26p$	4129(133)	2250(49)	−751(89)	7847(266)	−1985
$5d6s^27p$	4129	2250	−751	1954	−494
$5d6s^28p$	4129	2250	−751	761	−193
$5d^26s6p$	4030	2250	−751	6803	−1985
$5d^26s7p$	4030	2250	−751	1695	−429
$5d^26s8p$	4030	2250	−751	659	−166
$5d^36p$	3906	2250	−751	4889	−1985

Table 7. Comparison between experimental and calculated A and B hyperfine constants.

Level		Energy (cm^{-1})	A_{ex} (MHz)	A_{cal} (MHz)	B_{ex} (MHz)	B_{cal}	reference
$5d^26s6p$							
	$z\ ^5G_2$	18011	−204.4	−208.5	2479	2096	PW
	$z\ ^5G_3$	19293	152.20	161.08	1821.6	1561.7	A
	$z\ ^5G_4$	20960	258.60	262.58	1838.0	1590	Z
	$z\ ^5G_5$	22901	321.10	314.18	2191.8	1916	Z
	$z\ ^5F_3$	23449	416.48(*)	415.96	4179.6(*)	4560.0	Z
	$z\ ^5D_1$	25194	638.5	640.4	−566	−608	PW
	$y\ ^3F_2$	27150	−150.2	−145.9	−746	−874	PW
	$y\ ^3F_3$	27654	279.3	272.2	−845	−1153	PW
	$z\ ^3G_3$	28584	−33.4	−33.0	923	1129	PW
	$z\ ^3G_4$	29247	207.8	204.4	1292	981	PW
$5d6s^26p$							
	$z\ ^3D_1$	14018	−32.93	−31.09	269.3	244.3	Z
	$z\ ^3D_2$	16163	75.60	76.1	685	775	C
	$z\ ^3P_1$	18143	146.3	144.2	−695	−703	PW
	$z\ ^3F_4$	18225	179.21	188.73	3276.8	3221.7	Z
	$z\ ^3D_3$	18382	151.4	148.5	−26.9	161	PW and Z
	$z\ ^1F_3$	23645	416.48(*)	415.96	4179.6(*)	4534	Z
	$z\ ^1P_1$	26464	−21.9	−23.3	687.7	782	B

PW: Present work;
Z: Zimmermann *et al.* [3];
A: Anastassov *et al.* [4];
C: Cajko [20];
B: Boss *et al.* [21];
(*) We considered the sum of the A's and the sum of the B's of these two levels (see paragraph 4 in the text).

the magnetic interaction constants and quite good for the quadrupole interaction constants; we emphasize that—due to the scarcity of experimental data—we used 8 instead of 45 parameters to describe the A's and 2 instead of 35 to describe the B's.

5 Conclusion

Doppler limited laser spectroscopic measurements using a cooled hollow cathode discharge were performed for hfs studies on the refractory element Hf.

Using for the first time lines lying in the red spectral region and then taking advantage of smaller handicap of Doppler width we perfectly verified four data given in the excellent experimental work of Zimmermann *et al.*

Including results of earlier data, the hfs of altogether 16 fine structure states was analysed. A more refined treatment, including intermediate coupling and configuration mixing, achieved a satisfactory interpretation of all observed hyperfine constants.

As mentioned above the differences between Q_{5d} values evaluated from optical spectra may find their origin in the core polarisation effects of the $5d$-shell—contributions from excitations $5d \rightarrow n'l'$, where $n'l'$ stands for open or empty s, d, g shells—appearing in configurations differing by the number of $5d$-electrons. In this way complex experimental and theoretical studies of the atomic structure including both odd- and even-parity electron level systems of the hafnium atom seem to be very interesting for the determination of all kinds of one- and two-body contributions to the observed hyperfine structure. Furthermore it can help to explain observed differences between Q_{5d} values obtained from optical spectra and/or from muonic M X-rays.

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