



## Fine- and hyperfine structure investigations of the even-parity configuration system of the atomic holmium



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### ABSTRACT

In this work new experimental results concerning the hyperfine structure (*hfs*) for the even-parity level system of the holmium atom (Ho I) were obtained; additionally, *hfs* data obtained recently as a by-product in investigations of the odd-parity level system were summarized. In the present work the values of the magnetic dipole and the electric quadrupole *hfs* constants *A* and *B* were determined for 24 even-parity levels, for 14 of them for the first time. On the basis of these results, as well as on available literature data, a parametric study of the fine structure and the hyperfine structure for the even-parity configurations of atomic holmium was performed. A multi-configuration fit of 7 configurations was carried out, taking into account second-order of the perturbation theory. For unknown electronic levels predicted values of the level energies and *hfs* constants are given, which can facilitate further experimental investigations.

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### 1. Introduction

Holmium belongs to the heavier elements of the lanthanides series. Its single stable isotope  $^{165}\text{Ho}$  is characterized by a relatively high nuclear spin value  $I = 7/2$ , which renders the hyperfine structure of its electronic levels rather complicated. The nuclear magnetic dipole moment amounts to  $\mu = 4.17(3)\mu_N$  [1], which is a very large value, yielding very broad hyperfine structure patterns in transitions between the electronic levels; also the nuclear electric quadrupole moment is relatively high (values ranging from 2.716 to 3.6 b, determined with various methods, are quoted in [1]), causing considerable deviations from the Lande interval rule.

Early knowledge of the spectrum and term system of the atomic holmium was based on the extensive studies of Blaise et al. in 1972 [2,3] and Wyart et al. in 1973 [4]. Several years later, the analysis of Ho I spectrum was significantly extended by the elaborate work carried out by Wyart et al. in 1977 and by Wyart and Camus in 1978 [5,6]. Wyart et al. [5] recorded 1462 emission lines of holmium by means of Fourier transform spectroscopy in the spectral range of 2493–12344  $\text{cm}^{-1}$ . 913 of these lines were classified as transitions between 144 odd and 116 even levels. The magnetic dipole constants *A* and the electric quadrupole constants

*B* were determined for 111 odd-parity and 66 even-parity levels. Wyart and Camus [6] interpreted the experimental data by means of parametric studies of the groups of ( $4f^{11}6s^2 + 4f^{10}6s^26p + 4f^{11}5d6s$ ) and ( $4f^{10}5d6s^2 + 4f^{11}6s6p$ ) configurations.

Precise, atomic-beam magnetic-resonance measurements of the hyperfine structure splittings of the ground multiplet  $4f^{11}6s^2\ ^4I$  were presented by Dankwort et al. in 1974 [7], Burghardt et al. in 1982 [8] and by Childs et al. in 1983 [9], who also measured the *hfs* constants of several excited levels. In 1992, Reddy et al. [10] re-measured the *hfs* constants for 29 levels by means of optogalvanic spectroscopy and extended these investigations to 9 higher energy levels, 5 of which were assigned to  $4f^{11}6s7p$  configuration, not included in earlier studies.

In 1997, Kröger et al. [11] presented a theoretical interpretation of the hyperfine structure for configurations  $4f^{10}5d6s6p$  and  $4f^{10}5d^26s$ . New energy levels were found, including an excited level of the ground configuration  $4f^{11}6s^2\ ^4F_{9/2}$  and some of the lowest-lying levels of  $4f^{11}6s6d$  and  $4f^{10}5d^26s$  configurations.

In recent years the holmium atom gained a special interest due to its very rich electronic levels structure. Because of the high nuclear spin value the electronic levels comprise a high number of the hyperfine sublevels, and these in turn are split into a number of Zeeman sublevels in the magnetic field; e.g. the ground state, with  $J = 15/2$ , is split into  $(2J + 1) \times (2I + 1) = 128$  Zeeman sublevels, which is the highest number for any stable isotope. This

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**Table 1**

Spectral lines of the holmium atom investigated for the purpose of determination of the hyperfine structure constants for the even-parity upper levels, and the values obtained. The *hfs* constants of the odd-parity lower levels involved in the spectral lines were fixed at the available literature values in the least-squares fit procedure (for details see text, Section 3); the respective citations are given in a separate column. The energies of the levels were taken from the NIST Atomic Spectra Database [22], with its primary source [28, and the references therein].

No	Line		Lower Level					Upper Level			
	$\lambda_{air}$ (nm)	$k_{vac}$ (cm <sup>-1</sup> )	E (cm <sup>-1</sup> )	<i>J</i>	<i>A</i> (MHz)	<i>B</i> (MHz)	ref.	E (cm <sup>-1</sup> )	<i>J</i>	<i>A</i> (MHz)	<i>B</i> (MHz)
1	2	3	4	5	6	7	8	9	10	11	12
1	487.408	20510.96	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	25930.66	13/2	1010.2(0.1)	3238.6(2.5)
2	487.797	20494.61	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	25914.31	11/2	697.7(0.1)	-872.0(2.9)
3	487.868	20491.61	8605.16	11/2	1035.1396(0.0009)	-1052.556(0.030)	[8]	29096.77	11/2	1039.1(0.6)	278.7(9.5)
4	488.512	20464.62	8605.16	11/2	1035.1396(0.0009)	-1052.556(0.030)	[8]	29069.78	11/2	967.1(0.4)	-1103.5(6.9)
5	493.488	20258.27	0.00	15/2	800.5832(0.0001)	-1668.079(0.003)	[7]	20258.27	13/2	1426.8(0.2)	1755.1(9.8)
6	493.901	20241.31	0.00	15/2	800.5832(0.0001)	-1668.079(0.003)	[7]	20241.31	13/2	996.2(0.4)	286(14)
7	496.104	20151.45	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	25571.15	13/2	1407.0(0.1)	-1360.7(2.9)
8	497.779	20083.63	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	25503.33	11/2	1227.1(0.1)	644.4(4.3)
9	497.996	20074.89	0.00	15/2	800.5832(0.0001)	-1668.079(0.003)	[7]	20074.89	15/2	824.5(0.4)	1600(23)
10	503.564	19852.93	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	25272.63	15/2	555.0(0.1)	-768.6(3.5)
11	503.845	19841.85	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	25261.55	11/2	1026.8(0.1)	980.4(1.7)
12	516.908	19340.43	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	24760.13	13/2	626.2(0.5)	445(18)
13	517.432	19320.82	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	24740.52	13/2	707.8(0.1)	-771.4(2.3)
14	519.576	19241.10	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	24660.80	15/2	753.7(0.2)	-2496(11)
15	527.687	18945.34	13094.42	9/2	884.5(2.0)	1752(22)	[15]	32039.76	9/2	818.6(2.1)	-94(28)
16	527.762	18942.66	10695.75	9/2	1137.700(0.001)	-494.482(0.010)	[9]	29638.41	9/2	702.4(0.1)	-388.5(4.8)
17	531.516	18808.86	13094.42	9/2	884.5(2.0)	1752(22)	[15]	31903.28	11/2	764.7(2.2)	-29(28)
18	569.367	17558.49	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	22978.19	13/2	933.0(0.1)	1150.3(6.0)
19	573.405	17434.83	8605.16	11/2	1035.1396(0.0009)	-1052.556(0.030)	[8]	26039.99	9/2	1244.9(0.2)	-198.0(5.4)
20	576.901	17329.18	13094.42	9/2	884.5(2.0)	1752(22)	[15]	30423.60	11/2	792.2(2.1)	-1441(23)
21	577.024	17325.50	8605.16	11/2	1035.1396(0.0009)	-1052.556(0.030)	[8]	25930.66	13/2	1010.3(0.1)	3205.5(7.4)
22	577.569	17309.15	8605.16	11/2	1035.1396(0.0009)	-1052.556(0.030)	[8]	25914.31	11/2	697.2(0.2)	-844.5(6.7)
23	585.286	17080.92	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	22500.62	15/2	747.4(0.2)	431.2(7.9)
24	588.268	16994.34	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	22414.04	13/2	818.9(0.1)	586.8(3.3)
25	589.251	16965.99	8605.16	11/2	1035.1396(0.0009)	-1052.556(0.030)	[8]	25571.15	13/2	1408.8(0.1)	-1377.5(3.5)
26	591.616	16898.17	8605.16	11/2	1035.1396(0.0009)	-1052.556(0.030)	[8]	25503.33	11/2	1226.9(0.1)	634.4(3.9)
27	594.803	16807.64	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	22227.34	15/2	1310.3(0.1)	-2080.5(2.2)
28	597.272	16738.16	5419.70	13/2	937.2088(0.0003)	-1438.065(0.025)	[8]	22157.86	13/2	739.3(0.1)	156.1(6.1)
29	600.204	16656.39	8605.16	11/2	1035.1396(0.0009)	-1052.556(0.030)	[8]	25261.55	11/2	1026.6(0.2)	958.6(5.9)

extremely rich structure was proposed for a possible application in quantum information - collective encoding of multiqubit quantum registers [12]. This induced the efforts towards laser cooling of the holmium atoms in MOTs with the use of an ingenious method developed for rare earths atoms [13] and demonstrated earlier on three elements in the upper half of the series (Er, Tm and Dy). Laser cooling schemes for Ho were proposed in [12], and in 2014 a holmium MOT was reported [14] at the first-stage cooling transition  $\lambda = 410.5$  nm.

Proposed second-stage cooling transitions in the holmium atom have not yet been demonstrated. The knowledge of the *hfs* for the upper levels of the cooling transitions is of course of great importance; for most of the levels in question the *A* and *B* constants evaluated from Fourier-spectroscopic measurements were available so far, and in the present work the authors determined more precise values for 5 such levels.

## 2. Experimental details

In the present investigations of the even-parity levels in Ho I the same experimental method and setup as in our work concerning the odd-parity levels [15] were applied, essentially not much different from those used also in our investigations of Tb I and Tb II [16–20]. The recent work [15] provided a description of the conditions specific for holmium, and the reader is referred there for details. In the following the authors restrict themselves to the general overview only.

The method applied was laser induced fluorescence (LIF) in a hollow cathode discharge with spectral selection of the fluorescence channels (decay transitions from the upper level); selective observation of fluorescence, apart from enabling the improvement

of signal-to-noise ratio, also facilitates the unequivocal identification of the upper levels, and thus the spectral lines classification.

As the source of exciting radiation, two cw tunable single mode lasers, used previously, were applied - both modified Coherent, model CR 699–21, operated on the dyes Rhodamine 6G (in the yellow-orange region - 12 lines) and Coumarin 498 (in the blue-green region, optically pumped by a diode laser - 17 lines), respectively; the former was used throughout all our investigations of the rare earths elements, and the latter was developed recently [21] and used in most works concerning terbium. The laser beam was intensity modulated and phase-sensitive detection of the resulting fluorescence was applied. Along with the LIF signal, also a transmission signal of a FSR  $\approx 1500$  MHz (wavelength-corrected) frequency marker was recorded, which facilitated the construction of the relative frequency scale for the spectra.

The wavenumbers of the spectral lines excited were determined by a wavemeter (Burleigh, model WA-1500) and the fluorescence channels were selected by a monochromator.

Typically for each transition investigated, 10–20 frequency scans of ca. 30–40 s duration were recorded, so that the signal-to-noise ratio could be somewhat improved by averaging over groups of subsequent scans. For very weak lines the number was increased up to ca. 50 scans and the scan time was increased to several minutes, if necessary.

In the *hfs* spectra sometimes some saturation effects were observed. When the exciting transitions were very weak, reduction of the applied laser beam intensity, sufficient to eliminate saturation, yielded considerable reduction of the signal to noise ratio. An attempt to increase the population of the lower level by raising the discharge current caused an increase of the Doppler broadening and thus the deterioration of the spectral resolution. Both effects were undesired. However, it was observed on numerous

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