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Fine- and hyperfine structure investigations of even configuration system of atomic terbium

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ABSTRACT

In this work a parametric study of the fine structure (*fs*) and the hyperfine structure (*hfs*) for the even-parity configurations of atomic terbium (Tb I) is presented, based in considerable part on the new experimental results. Measurements on 134 spectral lines were performed by laser induced fluorescence (LIF) in a hollow cathode discharge lamp; on this basis, the hyperfine structure constants *A* and *B* were determined for 52 even-parity levels belonging to the configurations $4f^85d6s^2$, $4f^85d^26s$ or $4f^96s6p$; in all the cases those levels were involved in the transitions investigated as the lower levels. For 40 levels the *hfs* was examined for the first time, and for the remaining 12 levels the new measurements supplement our earlier results. As a by-product, also preliminary values of the *hfs* constants for 84 odd-parity levels were determined (the investigations of the odd-parity levels system in the terbium atom are still in progress). This huge amount of new experimental data, supplemented by our earlier published results, were considered for the fine and hyperfine structure analysis. A multi-configuration fit of 7 configurations was performed, taking into account second-order of perturbation theory, including the effects of closed shell-open shell excitations. Predicted values of the level energies, as well as of magnetic dipole and electric quadrupole hyperfine structure constants *A* and *B*, are quoted in cases when no experimental values are available. By combining our experimental data with our own semi-empirical procedure it was possible to identify correctly the lower and upper level of the line 544.1440 nm measured by Childs with the use of the atomic-beam laser-rf double-resonance technique (Childs, J Opt Soc Am B 9;1992:191–6).

1. Introduction

The present paper is the third one in the series of our systematic investigations of the hyperfine structure in the terbium atom.

Terbium is positioned slightly above the middle of the lanthanides series. It belongs to the heavier elements with partially filled 4f electron shell, which normally have extremely complex optical spectra for low ionization stages. Terbium possesses only one stable isotope $^{159}_{65}$Tb, with a relatively low nuclear spin value $I = 3/2$, which limits the number of the hyperfine sublevels to at most 4, irrespective of the J quantum numbers of the electronic levels; this in turn poses a limitation on the number of the hyperfine components in the spectral lines – according to the selection rules, 9 or 10 *hfs* components only can occur. This situation seems favorable, since the structure of the spectral lines is by far less complex than in many other elements in the lanthanides series; in spite of that, the knowledge of the spectrum and structure of the terbium atom was still very scarce until recently. The electronic levels density in energy scale is extremely high, even compared with the remaining lanthanides, which is characteristic of mid-shell elements, and this constitutes a source of particular difficulties in both the experimental investigations and the theoretical interpretation of its electronic levels structure.

The pioneering works on the Tb I spectrum were performed by Davis [1] and Klinkenberg and coworkers, which culminated in the publication of a series of six papers with a common title [2–7]. It is perhaps worth mentioning that the ground state of the terbium atom was not identified until 1970 [8].

A brief survey of the spectroscopic investigations of the terbium atom described in the literature was presented in our earlier works [9,10].

Those works summarize more extensive investigations of the hyperfine structure of the terbium atom with the method of laser induced fluorescence (LIF) in a hollow cathode discharge, focused first of all on the even-parity configurations, which we undertook after more than two decades of stagnation in this topic. The first

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work [9] concerned predominantly the levels of the configuration $4f^85d6s^2$. Altogether, *hfs* data for 14 levels were obtained for the first time. In 2016, new experimental results concerning 8 levels belonging to the configuration $4f^85d^26s$ were published [10]. As a by-product, *hfs* constants for a number of odd-parity levels, involved as upper levels in the transitions investigated, were also determined in both works.

Experimental investigations performed within this work were focused on determination of the *hfs* constants of further electronic levels belonging to the even-parity configurations $4f^85d6s^2$, $4f^85d^26s$ and $4f^96s6p$, which could supplement the results of our earlier works; in particular we concentrated our efforts on the energy region ca. 11 000–15 000 cm^{-1} – in many of the levels occurring in this range assignment to particular configurations was problematic.

Another issue concerning the *hfs* of terbium electronic levels, which deserved further examination, was a relatively small value of one of the *hfs* one-electron radial parameters obtained in preliminary semi-empirical analysis, namely a_{6s}^{10} (and thus the radial integral $\left\langle r^{-3}\right\rangle_{6s}^{10}$), which did not seem to fit to the overall tendency exhibited along the lanthanides series; we addressed this problem in our earlier work [10]. The parameters mentioned, as well as their mutual relation, are described in Section 4.

The number of experimental results on the *hfs* of the terbium atom greatly increased in the course of this work, so that altogether *A* and *B* constants for 80 even-parity levels are available currently. This extension of experimental database prompted us to attempt a parametric analysis for even-parity configurations of Tb I, as a comprehensive multi-configuration fit including 7 configurations and taking into account second-order of perturbation theory with the effects of closed shell-open shell excitations included. The present work gives inspiration for experimental search for energetically low-lying unknown levels with the smallest *J*-values ($J=1/2$, 3/2 and 5/2).

2. Experimental details

The experimental procedure was the same as applied in our earlier works concerning the terbium atom [9,10], and very similar to the basic one we used throughout in the investigations of lanthanides series elements, e.g. [11–13]. The experimental setup is also essentially the same as previously used, with some minor modifications. A more detailed description of both the procedure and the setup is provided in the works cited; within this work a general overview is given, and the specific elements, characteristic of the present investigations, are presented in detail.

The measurements within this work were performed with the method of laser induced fluorescence (LIF) in a hollow cathode discharge with spectral selection of the decay transitions from the laser-excited levels (commonly referred to as fluorescence channels).

In general, the terbium atom spectrum does not contain particularly strong lines, in contrast to other lanthanide elements; many of the transitions studied within this work were rather weak and did not occur in any spectral lines databases, but were expected to be allowed by the selection rules. Due to very scarce knowledge of the odd-parity levels system, in most cases both the lower and the upper level involved in a particular transition had unknown hyperfine structures. Within this work, altogether 125 spectral lines involving the even-parity levels as the lower levels were investigated quantitatively, and many more were observed, but their signal quality was found insufficient for more than an estimation of the hyperfine structures of both levels involved. In order to facilitate determination of *hfs* for the upper odd-parity levels, auxiliary measurements on 9 spectral lines were also performed (a closer explanation follows in the next section, in Section 3.1).

In the measurements on more than a half of the total number spectral lines investigated, the source of exciting radiation was a single-mode cw ring dye laser (modified Coherent CR 699-21) operated on Rhodamine 6G dye – the same as applied in our earlier works on the terbium atom [9,10]. This laser covered most of the yellow-orange spectral region (ca. 565-615 nm) with relatively high output power available (exceeding 500 mW around the maximum of the tuning range).

In majority of the remaining cases another single-mode ring dye laser (also a modified Coherent CR 699-21) was applied; this one was operated on Coumarin 498 dye and optically pumped by an economy-class diode laser, which provided a few watts of radiation at 445 nm (Lasever Inc., China, model LSR445SD-4W); this dye laser covered a part of blue-green spectral region (ca. 485-525 nm). After initial power decrease, a sufficient power level (above 100 mW around the maximum of the tuning curve, and at least of the order of 10 mW at the edges), could be maintained over elongated periods of time.

Finally, measurements on several lines were performed with the use of a single-mode external-cavity diode laser (Toptica Photonics, model DL pro, with DLC pro controller) operated in the near IR region (ca. 765–805 nm).

In order to improve the signal-to-noise ratio, the laser beam was amplitude modulated by a mechanical chopper and the phase-sensitive detection of the resulting fluorescence signal was applied.

Individual fluorescence channels were spectrally resolved by a monochromator; in some cases additionally color glass filters were used for more rigorous suppression of undesired stray light. Along with the LIF spectra, transmission signal of a stable FP interferometer of 1500 MHz FSR (referred to as the frequency marker) was recorded; this signal facilitated the construction of the relative frequency scale for the LIF spectra. The absolute transitions vacuum wavenumbers were determined by a wavemeter (Burleigh WA-1500), however, with variable measurement precision. In some particularly unfavorable cases, for dye laser generated radiation in the regions close to the edges of the tuning curves, it was limited to ca. 0.1 cm^{-1}, which does not match the nominal precision of 50–100 MHz. On the other hand, the wavenumbers of all the transitions excited with a diode laser were determined with the nominal precision of the wavemeter. In general, the centers of gravity of the lines' structures were found to be in good agreement with the values expected from the differences between the levels' energies, and the occurrence of the expected fluorescence channels provided additional verification of the identification of the exciting transitions.

The specific experimental conditions were essentially similar to those used in our earlier investigations of the terbium atom, with some minor modifications.

The hollow cathode discharge lamp was run at a typical discharge current of 40 mA; this was adjusted according to the observed signal strength – for very weak transitions the current up to 100 mA was necessary, while for the strongest ones among those investigated within this series of measurements it could be lowered to ca. 10 mA. The resulting Doppler-broadened profiles with typical widths ranging from 400 MHz to 1000 MHz, dependent on the discharge current and the spectral region (typically ca. 700 MHz for the yellow-orange region) were obtained. Two kinds of buffer gas were used alternatively: argon and neon. Argon was used in our earlier investigations of terbium atom and proved to be advantageous, yielding a stable discharge at the optimum pressure of $5.5 \cdot 10^{-1}$ mbar, which could be applied directly at the ignition of the discharge. However, in some cases the argon