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Radiative lifetime measurements and semi-empirical transition parameter calculations for high-lying levels in Ba I

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ABSTRACT

Radiative lifetimes for 18 highly excited levels belonging to 5d4f, 5d6p, and 6s*n*p ($n = 10$, 12, 14–19) configurations of barium were measured by time-resolved laser-induced fluorescence (TR-LIF) technique. The uncertainties of the results are less than 10%. To our best knowledge, there are 16 lifetimes of Ba I reported for the first time. Two lifetimes were measured to compare with the previous results, and good agreements were achieved. From the combination of these experimental lifetimes and theoretical branching fractions calculated using a pseudo-relativistic Hartree–Fock model including core-polarization effects, a set of semi-empirical transition probabilities and oscillator strengths was derived for 46 Ba I spectral lines in the wavelength region from 240 to 3765 nm. For about half of these transitions, the uncertainties affecting the new decay parameters were found to be equal or better than 50%.

1. Introduction

Barium (Ba) spectra are abnormally enhanced for a distinct class of peculiar giant stars that are named Barium stars. The spectral data for the strong lines of s-process elements, such as Ba II at 455.4 nm, are very useful for determining the chemical abundances, which are very important for building and testing the theoretical stellar models [1–3]. The atomic radiative lifetimes, transition probabilities, and oscillator strengths are important data for discovery of the isotopes, atomic structure calculations, and element abundance analyses [4–6]. Thus, the transition probabilities and oscillator strengths of barium, which can be deduced by combining the measured radiative lifetimes with experimental or theoretical branching fractions, are of crucial importance.

The investigations on radiative parameters (such as radiative lifetimes, transition probabilities, and oscillator strengths) of Ba I excited levels has a long history. The lifetimes for Ba I have been measured by Hanle effect, delayed coincidence technique, and hook methods since 1964 [7–20]. Subsequently, many lifetimes for Ba I high-lying excited levels were obtained by TR-LIF technique, which is proved to be one method having simple experimental devices and good veracity and reliability. [21–23] Meanwhile, some theoretical calculations for Ba I lifetimes have been done by Kulaga et al. [24] and Dzuba and Ginges [25]. As for transition probabilities and oscillator strengths, Miles and Wiese compiled transition probabilities for Ba I and Ba II through critical evaluation [26]. Some data on the transition probabilities and oscillator strengths of 6s*n*p ($n = 16$–42) states were updated by Klose et al. [27]. Curry compiled some transition probabilities for Ba I and Ba II in 2004 [28]. Kalyar et al. measured oscillator strengths of Rydberg transitions of Ba I [29]. In one of our recent works [30], branching fractions for 108 Ba I lines were measured using a high-resolution grating spectrometer with a hollow-cathode lamp. These data were combined to natural radiative lifetimes published in the literature to deduce the corresponding transition probabilities and oscillator strengths.

The uncertainties of deduced oscillator strengths and transition probabilities depend on experimental lifetime data, or on adopted experimental methods. Some complicated experimental devices that are sensitive to temperature will involve uncertainties larger than 25% or even up to 50% [26]. Hence, obtaining accurate radiative lifetimes is very significant.

In this paper, we report experimental radiative lifetimes for 18 highly excited levels of Ba I by TR-LIF method, among which 16 are determined for the first time. The excited wavelengths used in the experiment are between 308 and 568 nm, and most of

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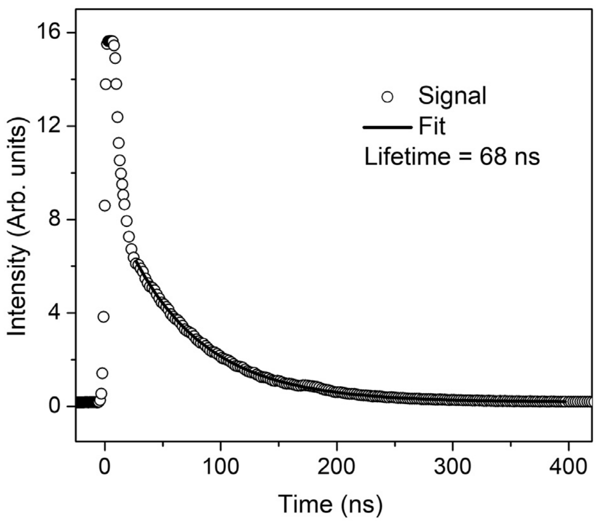

Fig. 1. A fluorescence decay curve of the 41,470.960 cm^{-1} level with an exponential fitting.

lifetime results in this paper correspond to high-lying levels located above 40,000 cm^{-1}. These new experimental data were then combined to theoretical branching fractions, which were calculated using the pseudo-relativistic Hartree–Fock method including core-polarization effects, to deduce semi-empirical transition probabilities and oscillator strengths for 46 Ba I spectral lines in the wavelength range from 240 to 3765 nm.

2. Lifetime measurements

Eighteen lifetimes of Ba I were determined by TR-LIF technique. Only a brief outline is presented here because the experimental setup in this paper is like the one described in our previous papers [30].

The free barium atoms were obtained through laser ablation by a 532 nm Q-switched Nd:YAG laser. Then the Ba I atoms at the ground or metastable states were selectively excited to an aim state by a beam of a dye laser (Sirah Cobra-stretch) operating with DCM or Rhodamine 6 G dye. The fluorescence emitted from an aim level was detected by a photomultiplier tube (PMT, Hamamatsu R3896), then were recorded and averaged by a 2.5 GHz digital oscilloscope (Tektronix DPO7254). The fluorescence signals were collected in the direction perpendicular to the dye laser and the atomic beams in order to escape the stray light. The second or third harmonics of the dye laser was produced by one or two type-I β-barium borate (BBO) crystals, and sometimes a hydrogen cell was also used to obtain first order of Stokes component as excitation light. A digital delay generator was used to adjust the delay time between the excitation and ablation pulses. The signals of measured level were recorded at different delay times, so some possible effects, such as collisional effect, radiation trapping, and flight-out-of-view effect could be found and minimized through changing experimental conditions. More than 1000 shots were averaged for all the fluorescence curves to improve the signal-to-noise ratio. Sometimes the filter was put in front of the entrance slit of monochromator to restrain the stray light of ablation and excitation pulses. The entrance and exit slits of monochromator were adjusted to obtain fluorescence curves with high signal-to-noise ratio.

According to our experience, the lifetimes longer than 40 ns can be obtained credibly by proper fitting recorded fluorescence curves to an exponential function directly. A fluorescence curve of the 41,470.960 cm^{-1} level with an exponential fit is shown in Fig. 1 for example. It is seen that the fluorescence curve has a good signal-to-noise ratio and is fitted well.

Table 1
Measured lifetimes of Ba I levels and comparison with previous results.

Upper level[a]				$\lambda_{Exc.}$ (nm)	Lifetime (ns)	
Config.	Term	*J*	*E* (cm^{-1})		This work	Previous
5d6p	$^1F^o$	3	26,816.266	568.157	46(4)	45[b], 44.5[c]
6s10p	$^1P^o$	1	39,311.950	358.210	107(5)	
5d4f	$(^5/_2,^7/_2)^o$	1	40,662.860	316.167	49(4)	48[d]
5d4f	$(^5/_2,^5/_2)^o$	1	40,736.810	315.429	107(8)	
6s12d	^3D	1	40,742.600	317.187	400(40)	
6s15p	$^3P^o$	1	41,159.830	313.045	628(60)	
6s16p	$^3P^o$	0	41,295.930	309.963	530(50)	
6s16p	$^3P^o$	1	41,296.960	311.707	248(20)	
6s16p	$^3P^o$	2	41,299.330	311.683	530(34)	
6s16p	$^1P^o$	1	41,307.880	309.848	284(28)	
6s17p	$^3P^o$	1	41,404.400	308.924	304(30)	
6s17p	$^3P^o$	2	41,406.530	333.209	411(40)	
6s17p	$^1P^o$	1	41,411.040	310.602	410(40)	
6s14g	^3G	3	41,470.960	357.702	68(5)	
6s18p	$^3P^o$	1	41,490.090	309.841	430(30)	
6s18p	$^1P^o$	1	41,494.390	309.800	552(22)	
6s19p	$^3P^o$	1	41,559.450	309.177	270(15)	
6s18g	^3G	3	41,693.910	354.872	48(4)	

[a] From Curry [28].
[b] From Matsuo et al. [22].
[c] From Brink et al. [15].
[d] From Zhang et al. [23].

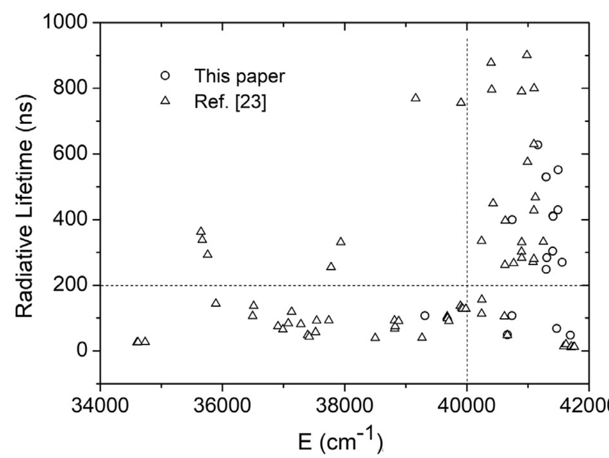

Fig. 2. Measured lifetimes of this paper and reference [23] as a function of energy level (cm^{-1}).

The lifetimes of 5d4f, 5d6p, and 6s*n*p ($n = 10$, 12, 14–19) levels of Ba I determined by TR-LIF technique together with previous results for comparison are listed in Table 1. One can see that the lifetimes between this paper and previous results for 26,816.266 and 40,662.860 cm^{-1} are approximately equal, which proves that our measurements are reliable. The uncertainties for the results measured in this paper are less than 10%. For 6s*n*p ($n = 10$, 12, 14–19) levels of Ba I, the lifetimes results are longer than 100 ns, which are relatively long. For further comparison, the lifetimes measured in this paper and in Ref. [23] are compiled as a function of energy level in Fig. 2. One can see that most of energy levels (more than 70%) with relatively long lifetimes (longer than 200 ns) are above 40,000 cm^{-1}.

3. Branching fraction calculations

The theoretical approach used in the present work for computing the branching fractions in Ba I was the pseudo-relativistic Hartree–Fock (HFR) method of Cowan [31] modified for taking

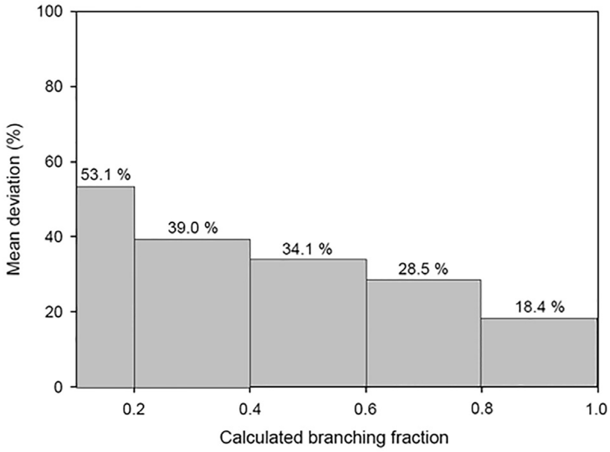

Fig. 3. Mean deviations between the branching fractions calculated in the present work and the experimental values measured by Wang et al. [30].

Table 2
Branching fractions, transition probabilities, and oscillator strengths obtained in the present work for highly excited levels of Ba I.

Upper level[a]		Lower level[a]		λ[b] (nm)	*BF*[c]	*gA*[d] (s^{-1})	log *gf*[d]	Unc[e]
E (cm^{-1})	*J*	*E* (cm^{-1})	*J*					
26,816.266	3	9596.533	3	580.568	0.133	2.02(7)	−0.99	E
τ = 46(4) ns		11,395.350	2	648.291	0.863	1.31(8)	−0.08	C
39,311.950	1	0.000	0	254.299	0.296	8.30(6)	−2.09	D+
τ = 107(5) ns		11,395.350	2	358.108	0.395	1.11(7)	−1.67	D+
		26,757.300	0	796.299	0.115	3.22(6)	−1.51	E
40,662.860	1	23,062.051	2	567.998	0.148	9.06(6)	−1.36	E
τ = 49(4) ns		23,479.976	1	581.813	0.476	2.91(7)	−0.83	D+
		23,918.915	2	597.065	0.122	7.47(6)	−1.40	E
40,736.810	1	23,918.915	2	594.440	0.119	3.34(6)	−1.75	E
τ = 107(8) ns		26,757.300	0	715.136	0.194	5.44(6)	−1.38	E
		28,230.231	0	799.359	0.262	7.35(6)	−1.15	D
40,742.600	1	12,266.024	0	351.065	0.326	2.45(6)	−2.34	D
τ = 400(40) ns		12,636.623	1	355.695	0.211	1.58(6)	−2.52	D
41,159.830	1	30,750.672	2	960.429	0.259	1.24(6)	−1.77	D
τ = 628(60) ns		32,943.774	2	1216.796	0.154	7.36(5)	−1.79	E
41,295.930	0	30,695.617	1	943.110	0.483	9.11(5)	−1.92	D+
τ = 530(50) ns		32,805.169	1	1177.428	0.270	5.09(5)	−1.97	D
41,296.960	1	30,695.617	1	943.018	0.107	1.29(6)	−1.76	E
τ = 248(20) ns		30,750.672	2	947.941	0.312	3.77(6)	−1.29	D
		32,943.774	2	1196.820	0.182	2.20(6)	−1.33	E
41,299.330	2	30,818.115	3	953.826	0.414	3.91(6)	−1.27	D+
τ = 530(34) ns		33,526.601	3	1286.198	0.247	2.33(6)	−1.24	D
41,307.880	1	0.000	0	242.011	0.335	3.54(6)	−2.51	D
τ = 284(28) ns		11,395.350	2	334.212	0.104	1.10(6)	−2.74	E
		28,230.231	0	764.453	0.107	1.13(6)	−2.00	E
41,404.400	1	30,695.617	1	933.557	0.124	1.22(6)	−1.80	E
τ = 304(30) ns		30,750.672	2	938.381	0.357	3.52(6)	−1.33	D
		32,943.774	2	1181.622	0.189	1.87(6)	−1.41	E
41,406.530	2	30,818.115	3	944.169	0.432	5.26(6)	−1.15	D+
τ = 411(40) ns		33,526.601	3	1268.700	0.247	3.00(6)	−1.14	D
41,411.040	1	9215.501	2	310.512	0.145	1.06(6)	−2.81	E
τ = 410(40) ns		11,395.350	2	333.063	0.100	7.32(5)	−2.91	E
		30,236.826	2	894.672	0.252	1.84(6)	−1.65	D
		30,750.672	2	937.797	0.200	1.46(6)	−1.71	D
41,470.960	3	34,602.765	2	1455.589	0.233	2.40(7)	−0.12	D
τ = 68(5) ns		37,394.868	2	2452.661	0.198	2.04(7)	0.26	E
		38,815.700	2	3765.083	0.127	1.31(7)	0.44	E
41,490.090	1	30,695.617	1	926.146	0.136	9.49(5)	−1.91	E
τ = 430(30) ns		30,750.672	2	930.894	0.326	2.27(6)	−1.53	D
		32,943.774	2	1169.775	0.200	1.40(6)	−1.54	D
41,494.390	1	0.000	0	240.923	0.112	6.09(5)	−3.28	E
τ = 552(22) ns		30,236.826	2	888.048	0.395	2.15(6)	−1.60	D+
		30,750.672	2	930.521	0.103	5.60(5)	−2.14	E
41,559.450	1	26,160.293	1	649.207	0.284	3.16(6)	−1.70	D+
τ = 270(15) ns		30,750.672	2	924.920	0.254	2.82(6)	−1.44	D+
		32,943.774	2	1160.357	0.133	1.48(6)	−1.53	E

[a] Level energies taken from Curry [28]. The experimental lifetimes, τ, measured in the present work are also given.
[b] Wavelengths deduced from experimental level energies
[c] Branching fractions computed using the HFR + CPOL method. Only *BF*-values larger than 0.1 are given.
[d] *gA*- and log *gf*-values obtained from the combination of HFR + CPOL branching fractions with experimental lifetimes. A(B) stands for $A \times 10^B$.
[e] Estimated uncertainties indicated by the same letter coding as the one used in the NIST database [35], i.e., C (≤25%), D+ (≤40%), D (≤50%), E (>50%) (see the text).

core-polarization effects into account (HFR + CPOL), as described by Quinet et al. [32,33]. The physical model considered was the same as the one used in our previous theoretical study of the barium atom [23]. More precisely, we assumed the Ba I atomic system as being composed of a xenon-like Ba III ionic core with 54 electrons surrounded by 2 valence electrons. The intravalence correlation was considered through configuration interaction (CI) by explicitly including in the calculations the following CI expansions: $6s^2 + 6sns + 6snd + 6sn'g + 5dn''s + 5dn''d + 6p^2 + 6p4f + 4f^2$ with $n \le 19$, $n' \le 15$ and $n'' < 9$ for the even parity, and $6snp + 6snf + 5dn'p + 5d4f$ with $n \le 19$ and $n' \le 9$ for the odd parity. The core-polarization contributions were evaluated using the dipole polarizability of the Ba III ionic core as calculated by Johnson et al. [34] using the relativistic random-phase approximation, i.e., $\alpha_d = 1.921$ a.u., while the cut-off radius was chosen to be the average radius of outermost core orbital (5p), as obtained in our HFR calculations, i.e. $r_c = 10.61$ a.u. In addition, a well-established least-squares fitting procedure was applied based upon the experimental energy levels compiled recently by Curry [28]. The standard deviations of the fits were 150 and 134 cm^{-1} for the even and the odd parities, respectively.

4. Semi-empirical transition probabilities and oscillator strengths

By combining the experimental lifetimes and the theoretical BFs obtained in the present work, we deduced the transition probabilities and oscillator strengths for all the lines depopulating the odd-parity levels of interest. The results obtained are reported in Table 2. These correspond to 46 Ba I spectral lines appearing in the wavelength range from 240 to 3765 nm. Note that concerning the BFs, only the values larger than 0.1 were retained in the table, most of the weaker decay channels being found to be affected by large cancellation effects [31] in our calculations.

The estimated uncertainties of the transition probabilities and oscillator strengths obtained in our work are also reported in Table 2, using the same letter coding as the one usually used in the NIST database [35]. They were evaluated as follows. First, an uncertainty was assigned to each of our calculated BF values by comparing the latter to those deduced from recent experimental measurements by Wang et al [30] for some transitions depopulating Ba I levels from 24,192 up to 41,622 cm^{-1}. Such a comparison allowed us to highlight some regular trends as far as the discrepancies between theoretical and experimental branching fractions are concerned. More precisely, as shown in Fig. 3, the mean deviations $(BF_{calc} - BF_{exp})/BF_{calc}$ were found to be equal to 18% for $0.8 < BF_{calc} < 1.0$, 28% for $0.6 < BF_{calc} < 0.8$, 34% for $0.4 < BF_{calc} < 0.6$, 39% for $0.2 < BF_{calc} < 0.4$, and 53% for $0.1 < BF_{calc} < 0.2$, respectively. We thus assumed uncertaintes affecting the branching fractions computed in the present work to be 20%, 30%, 35%, 40%, and 55% for BF = 0.8–1.0, 0.6–0.8, 0.4–0.6, 0.2–0.4, and 0.1–0.2, respectively. These uncertainties were then combined in quadrature with the experimental lifetime uncertainties derived from our measurements to yield the uncertainties of gA- and gf- values. As a final result, due to the fact that many of the highly excited levels considered in the present work are depopulated by quite a number of weak lines, among the 46 transitions listed in Table 2, about half of them have an estimated decay rate accuracy that is equal or better than 50%.

5. Conclusion

A new set of transition probabilities and oscillator strengths for 46 spectral lines of neutral barium has been obtained in the present work. They were semi-empirically deduced from the combination of accurate experimental radiative lifetimes measured for 18 highly excited levels located between 26,816 and 41,694 cm^{-1} with theoretical branching fractions calculated using a pseudo-relativistic Hartree–Fock model including core-polarization contributions.

Acknowledgments

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