Transition probabilities for infrared and visible lines in neutral barium

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From narrow-band and broadband spectra of barium hollow-cathode discharges obtained with the aid of a Fourier-transform spectrometer, we have determined the branching fractions of five moderately excited levels and the branching ratios between the infrared lines originating in the resonance level $6s6p\ ^1P^\circ_1$ of the neutral barium atom. By combining our emission measurements with lifetime and absorption data taken from the literature we were able to derive transition probabilities for 13 visible and 16 infrared transitions as well as upper limits for the A values of 15 additional infrared lines. The resulting transition probabilities cover five decades.

I. INTRODUCTION

Recent lifetime measurements[1–3] for moderately excited energy levels of the neutral barium atom make a determination of the corresponding branching fractions meaningful, since the latter can then be converted into spectroscopically useful transition probabilities. Given the energy-level structure of Ba I, a large number of transitions originating in levels with moderate excitation, i.e., with wave numbers $\sigma \gtrsim 25\,000\ \mathrm{cm}^{-1}$, have wavelengths within the infrared part of the electromagnetic spectrum, and only a minority of dipole-allowed and intercombination transitions lie in the visible part of the spectrum.

Literature values of transition probabilities in Ba I, that have been experimentally determined, concern almost exclusively visible lines,[3–7] namely, the transitions to the low-lying metastable levels $6s6p\ ^3P^\circ$, $6s5d\ ^{1,3}D$, to the resonance level $6s6p\ ^1P^\circ_1$, or to the ground level $6s^2\ ^1S_0$, respectively. A very similar situation exists for theoretical transition probabilities.[8–11]

For the lower-lying Ba I resonance level, on the other hand, three infrared decay channels (to the metastable levels $6s5d\ ^{1,3}D_{2,1}$ at wavelengths of 1.50, 1.13, and 1.11 μm, respectively) exist besides the green resonance line $6s^2\ ^1S_0-6s6p\ ^1P^\circ_1$ at 553.5 nm. As noted previously,[7] the absolute A values of the weak infrared lines originating in the resonance level depend sensitively on the intensity ratio $A(6s^2\ ^1S_0-6s6p\ ^1P^\circ_1)/\Sigma A(6s5d\ ^{1,3}D_{2,1}-6s6p\ ^1P^\circ_1)$, which at present has several proposed values that were measured by different techniques and that range from 280 to 550.[7,12–17] Furthermore, since the A values of the infrared transitions are approximately 3–5 orders of magnitude smaller than that of the resonance line, the infrared branching ratios have not yet been measured completely.

Based on radiometrically calibrated narrow-band spectra of Ba hollow-cathode discharges obtained by use of a Fourier-transform spectrometer (FTS),[18] we were able to determine the branching ratios for all the infrared lines originating in the Ba I resonance level $6s6p\ ^1P^\circ_1$, and by combining these measures with results from three independent laser excitation measurements,[13–16] we have derived the branching fractions of this level as well. By linking the results of the narrow-band spectra to data from our earlier broadband measurements obtained on the same instrument,[7] we could also determine the branching fractions of five levels of moderate excitation. The branching fractions belonging to three of these levels and those of the resonance level could be converted directly into transition probabilities, since lifetimes have been measured.[1,3,19] For the remaining two levels, lifetimes were calculated either by experimentally determined literature A values[4–6] or by the Ladenburg method.[20]

II. MEASUREMENTS AND DATA REDUCTION

A detailed description of the experimental as well as the data-reduction procedures is given in Ref. 7. Here, we restrict ourselves to a few explanatory notes and a description of the special and markedly different experimental arrangement used for some of the narrow-band measurements.

The branching fractions and ratios were obtained from five barium hollow-cathode spectra that have been recorded with the McMath Fourier transform spectrometer at the U.S. National Solar Observatory on Kitt Peak.[18] Two broadband spectra have been obtained by use of a symmetric hollow-cathode discharge (source I), based on a design developed by Danzmann and Kock.[21] Three narrow-band spectra were recorded through an asymmetric discharge (source II) that has been built according to a design by Engleman and Palmer.[22] This latter source consisted of a 32-mm-long cathode made out of pure Ba, with an 8-mm-diam bore. Both

TABLE I. Experimental conditions for the recordings of five spectra.

Source	Current (mA)	Ar pressure (Torr)	Range observed	Filters	$\delta\sigma$[a] (cm^{-1})
I[b]	2×400	0.35	320 nm–3.5 μm	None	0.041
	2×600	0.35	320 nm–3.5 μm	None	0.041
II[c]	40	0.40	830 nm–3.5 μm[d]	ir+vis	0.028
	78	0.40	830 nm–3.5 μm[d]	ir+vis	0.028
	234	1.0	830 nm–3.5 μm	ir	0.016

[a]Spectral resolution element.
[b]Symmetric discharge (Ref. 21).
[c]Asymmetric discharge (Ref. 22).
[d]One detector; the other detector received radiation in the wavelength band 553.5±10 nm.

discharges were run with Ar as carrier gas. The specific conditions used in recording the five spectra are given in Table I.

Since an FTS simultaneously records photons of all wave numbers (within the wave-number range seen by the detectors),[18] the signal-to-noise ratio (S/N) in a particular wave-number region can be increased considerably by reducing the photon flux in the instrument to that belonging only to the region of interest, i.e., by the use of narrow-band filters. During runs with source II, either an ir-interference filter ($\lambda=875$ nm to $\lambda=2.5$ μm) alone or combined with a bandpass filter ($\lambda=553.5$ nm, $\Delta\lambda=10$ nm) have been introduced into the light paths of the FTS (cf. Table I). The resulting S/N thus could be increased by at least one order of magnitude.

Most of the lines originating in moderately excited Ba I levels correspond to infrared transitions that are rather weak in comparison with the visible lines and could therefore not be detected in the broadband spectra, obtained with source I. Given the increased S/N within the narrow-band spectra, these lines could now be observed. By linking the broadband and narrow-band spectra through a suitable ir line of moderate intensity on a common radiometric scale, we were able to determine branching fractions for these levels.

The relative radiometric calibration of the narrow-band spectra, ranging up to 2.5 μm, was obtained in two independent ways: with an Optronics Standard lamp (operated at a current of 15 A); and using the Ar I and Ar II branching ratios from Adams and Whaling.[23] The latter method—which had already been used to calibrate the spectra stemming from source I (cf. Ref. 7)—could be applied for the infrared-wavelength region only, since adequate Ar-line groups[23] linking the radiometric scale of the infrared and visible range were lacking. Both procedures showed compatible results within the infrared spectral part, having uncertainties ranging from 5% to 10%. For wave-number calibration, line identification, and data reduction, we proceeded as described in Ref. 7.

III. RESULTS AND DISCUSSIONS

Our results are compiled and compared to other experimental[3–6,28] and theoretical[8–11] data in Tables II–IV. Transition probabilities of lines that originate in levels with measured lifetimes are listed in Table II, whereas in Tables III and IV, A values for lines stemming from levels with calculated lifetimes are given. For completeness, all possible $E1$ transitions (including intercombination lines) from a common upper level are listed in these tables, even if neither an experimental nor a theoretical A value is known. The wave numbers of the transitions and most of the level designations are taken from Moore.[29]

Transitions outside the calibrated range of our spectra had to be excluded from further analysis, because an extrapolation of the intensity calibration would have introduced too large an uncertainty. Strictly speaking, therefore, we list branching ratios rather than fractions. But, even if one assumes a dipole moment for the neglected lines that is as large as that of the strongest transition from a given upper level, the resulting branching fractions are so small—given the $1/\lambda^3$ dependence of A values—that their influence on the other transition probabilities remains within the experimental uncertainty. Therefore, these lines could safely be neglected.

For other lines we give upper limits of the branching fractions, since these lines were not observed in our spectra. The stated numbers correspond to the measured background intensities at the respective transition wave numbers and have an estimated uncertainty of ±30%. However, in computing the branching fractions it was assumed that there is no actual contribution from this transitions.

In Secs. III A and III B we describe the specific steps used to arrive at our results and discuss these by comparing them with literature values.

A. A values from measured lifetimes

The lifetimes used to convert our branching fractions to the transition probabilities listed in Table II have been obtained by use of different experimental methods. For the resonance level $6s6p\ ^1P^\circ_1$, we have chosen the value given by Kelly and Mathur,[19] which was determined by Hanle-effect measurements. Table V gives an overview of literature lifetime data for the moderately excited levels listed in Table II. The lifetimes of Hannaford and Lowe[1] (obtained from fluorescence measurements following stepwise or two-photon laser excitation) are in general longer than those reported by Jitschin and Meisel[2] (determined from linewidth measurements). Given the smaller uncertainty ranges (as compared to Ref. 2) and the inter-

TABLE II. Branching fractions and transition probabilities for upper levels with measured lifetimes.

Upper level	Lifetime (ns)	Lower level	Wave number (cm^{-1})	Wavelength[b] (nm)	Branching[c] fraction	Transition probabilities[a] (s^{-1}) Experimental Here	Experimental Literature[d,e]	Experimental Ref. 8	Theoretical Refs. 9 and 10	Theoretical Ref. 11
$6s6p\ ^1P_1^\circ$	8.37±0.08[f]	$6s^2\ ^1S_0$	18060.3	553.5	0.9966(0.2)[g]	1.19±0.01[8][g]	1.19[8]	1.24[8]	1.33[8]	1.22[8]
		$6s5d\ ^3D_1$	9026.3	1107.6	3×10^{-5}(14)[g]	3.6±0.5[3][g]		1.2[3]	0.41[3]	
		$6s5d\ ^3D_2$	8844.8	1130.3	0.0010(14)[g]	1.2±0.2[5][g]		1.2[5]	1.6[5]	
		$6s5d\ ^1D_2$	6664.9	1500.0	0.0023(14)[g]	2.8±0.4[5][g]	0.9±0.1[5][h]	0.46[5]	1.9[5]	2.8[5]
$6s8s\ ^1S_0$[i]	18.2±0.8[j]	$6s6p\ ^3P_1^\circ$	21734.2	460.0	0.74(4)	4.07±0.24[7]	4.5[7]	0.046[7]		
		$6s6p\ ^1P_1^\circ$	16310.5	612.9	0.11(17)	6.0±1.1[6]		7.4[6]	19.5[6]	
		$5d6p\ ^3D_1^\circ$	10178.7	982.2	0.10(27)	5.5±1.5[6]				
		$5d6p\ ^3P_1^\circ$	8666.6	1153.5	<0.04	<2.2±0.7[6]				
		$5d6p\ ^1P_1^\circ$	5816.5	1718.8	0.05(30)	2.7±0.8[6]				
		$6s7p\ ^3P_1^\circ$	3555.2	2812.0						
		$6s7p\ ^1P_1^\circ$	1823.7	5481.9						
$6p^2\ ^3P_0$	6.2±0.5[j,k]	$6s6p\ ^3P_1^\circ$	21857.3	457.4	0.75(4)	1.21±0.11[8]	1.3[8]	2.8[8]	2.0[8]	
		$6s6p\ ^1P_1^\circ$	16433.6	608.3	0.07(29)	1.1±0.3[7]	0.42±0.05[7][k]	0.26[7]		
		$5d6p\ ^3D_1^\circ$	10301.8	970.4	0.10(15)	1.6±0.3[7]				
		$5d6p\ ^3P_1^\circ$	8789.8	1137.4	0.08(19)	1.3±0.3[7]			1.3[7]	
		$5d6p\ ^1P_1^\circ$	5939.6	1683.2	<0.003	<4.8±1.4[5]				
		$6s7p\ ^3P_1^\circ$	3678.3	2717.9						
		$6s7p\ ^1P_1^\circ$	1946.8	5135.2						
$6s7d\ ^1D_2$[l]	7.0±0.5[j]	$6s6p\ ^3P_1^\circ$	22707.8	440.3	0.189(15)	2.7±0.5[7]				
		$6s6p\ ^3P_2^\circ$	21829.7	458.0	0.493(9)	7.0±0.8[7]				
		$6s6p\ ^1P_1^\circ$	17284.2	578.4	0.147(18)	2.1±0.4[7]			0.8[7]	
		$5d6p\ ^3F_2^\circ$	13279.8	752.8	0.019(50)	2.7±1.4[6]				
		$5d6p\ ^3F_3^\circ$	12397.0	806.4	<0.013	<1.9±0.6[6]				
		$5d6p\ ^1D_2^\circ$	12270.0	814.8	0.044(54)	6.3±3.4[6]			5.8[6]	
		$5d6p\ ^3D_1^\circ$	11152.4	896.4	<0.005	<7.1±2.1[5]				
		$5d6p\ ^3D_2^\circ$	10812.9	924.6	<0.006	<8.6±2.6[5]				
		$5d6p\ ^3D_3^\circ$	10364.6	964.6	0.074(17)	1.1±0.2[7]				
		$5d6p\ ^3P_1^\circ$	9640.3	1037.0	0.009(23)	1.3±0.3[6]				
		$5d6p\ ^3P_2^\circ$	9387.9	1064.9	0.019(19)	2.7±0.6[6]				
		$5d6p\ ^1F_3^\circ$	8528.1	1172.3	<0.002	<2.9±0.9[5]				
		$5d6p\ ^1P_1^\circ$	6790.2	1472.3	0.006(22)	8.6±2.0[5]				
		$6s7p\ ^3P_1^\circ$	4528.9	2207.4	<0.002	<2.9±0.9[5]				
		$6s7p\ ^3P_2^\circ$	4357.1	2294.5	<0.001	<1.4±0.4[5]				
		$6s7p\ ^1P_1^\circ$	2797.3	3573.9						

TABLE II. (*Continued*).

Upper level	Lifetime (ns)	Lower level	Wave number (cm^{-1})	Wavelength[b] (nm)	Branching[c] fraction	Transition probabilities[a] (s^{-1}) Experimental Here	Experimental Literature[d,e]	Experimental Ref. 8	Theoretical Refs. 9 and 10	Theoretical Ref. 11
		$6s4f\ ^3F_2^\circ$	741.6	13480						
		$6s4f\ ^3F_3^\circ$	727.7	13738						
		$6s4f\ ^1F_3^\circ$	608.0	16443						

[a]The values given are to be multiplied by powers of 10 with the exponents listed in brackets.
[b]Wavelength in air, according to Ref. 26.
[c]Values in parentheses indicate uncertainties in percent. All upper limits have an estimated uncertainty of ±30%.
[d]As recommended by Jahreiss and Huber (Ref. 5), all transition probabilities from Miles and Wiese (Ref. 4) with lower levels belonging to the term $6s6p\ ^3P^\circ$ should be multiplied by a factor of 0.45. The values given in this column are corrected accordingly.
[e]The A values given in Ref. 4 are estimated by their authors to be uncertain within ±50%, except for the resonance line, where the uncertainty lies within ±3%.
[f]Reference 19.
[g]Our measurements comprise the branching ratios between the infrared transitions only. The branching fractions listed here are based on a ratio of 290±40 (Refs. 13–16, see also Ref. 7) between the radiative decay rate of the resonance transition and that of the sum over all transitions to metastable levels. Gerke and Bushaw (Ref. 17) have determined a value of 440±40 for the same ratio. With this latter ratio, the transition probabilities to the metastable levels become $A(1.11\ \mu m)=(2.4\pm0.2)\times10^3\ s^{-1}$, $A(1.13\ \mu m)=(8.4\pm0.8)\times10^4\ s^{-1}$, and $A(1.50\ \mu m)=(1.9\pm0.2)\times10^5\ s^{-1}$. A(553.5 nm) does not increase noticeably.
[h]Reference 28.
[i]Designation according to Ref. 24.
[j]Reference 1.
[k]Reference 3.
[l]Designation according to Ref. 25.

nal consistency of their results, we have chosen the lifetimes of Ref. 1 for the levels $6s8s\ ^1S_0$ and $6s7d\ ^1D_2$. In the case of the level $6p^2\ ^3P_0$, we note a close agreement between the values from Ref. 1 and the one given by Fisk *et al.*[3] The latter datum was obtained from ac Stark effect measurements. For this level, therefore, we used a mean value of 6.2±0.5 ns, derived from Refs. 1 and 3.

We now turn to the discussion of the data presented in Table II and start with the resonance level $6s6p\ ^1P_1^\circ$. From our spectra the following branching ratios between the infrared lines originating in the resonance level have been obtained:

$$\frac{A(1.50\ \mu\text{m})}{A(1.13\ \mu\text{m})}=2.3\pm0.3,\quad \frac{A(1.50\ \mu\text{m})}{A(1.11\ \mu\text{m})}=80\pm10\ .$$

The value for the first ratio is a weighted mean value of 2.8±0.8 (cf. Ref. 7) and the more precise number of 2.2±0.2, that was obtained from measurements with source II. Listing a mean value is appropriate here, since our two results are based on different calibration procedures. The second branching ratio given is compatible with the lower limit of 30±13 reported in Ref. 7. The theoretical prediction of 40 for the same ratio, as given by Hafner and Schwarz,[8] may be considered as showing a fair agreement with our result in view of the weak transition at 1.11 μm. The values calculated by Trefftz,[10] on the other hand, imply a ratio of 460. No agreement between theory and experiment can be seen in case of the ratio $A(1.50\ \mu\text{m})/A(1.13\ \mu\text{m})$, the calculations giving either equal line strengths (Ref. 10) or even inverted values leading to 0.4 (Ref. 8) for that branching ratio.

In neither of the spectra obtained with source I nor source II could we obtain optically thin profiles of the resonance line (λ=553.5 nm). Therefore, we had to complete our measurements with a most probable value[7] of

$$\frac{A(553.5\ \text{nm})}{A(1.50\ \mu\text{m})+A(1.13\ \mu\text{m})+A(1.11\ \mu\text{m})}=290\pm40\ ,$$

based on three independent laser excitation measurements,[13–16] to be able to list branching fractions (and thus A values) for the lines originating in the resonance level. However, since the value of 440±40 measured by Gerke and Bushaw[17] for this ratio might be correct instead, we also list (in footnote g of Table II) the transition probabilities following from this latter ratio.

Note added in proof. The values given in Refs. 13–16 receive further support by a datum recently published by Kallenbach and Kock [J. Phys. B (to be published)]. Kallenbach and Kock have carried out extensive measurements and modeling on a Ba vapor which was excited with resonant and near-resonant laser radiation. They arrived at a value of 270±60 for the ratio in question.

The uncertainties of the branching fractions for the infrared lines originating in the resonance level are mainly determined by the uncertainty range of the chosen ratio of 290±40. Besides the resonance transition, only for the line $6s5d\ ^1D_2-6s6p\ ^1P_1^\circ$ is an experimentally determined A value published: $A(1.50\ \mu\text{m})=(9\pm1)\times10^4$, given by Bokhan,[28] is a factor of 3 below our result. We note, however, that this would imply an intensity ratio of 910 between the resonance line and the sum over the transitions to the metastable levels, and this is nearly twice the highest literature value, namely, 550.[7,12] Excellent agreement, on the other hand, is given between the calculated value by Bauschlicher *et al.*[11] and our experimentally derived datum for $A(1.50\ \mu\text{m})$.

For only a few of the lines originating in the moderately excited levels listed in Table II (i.e., $6s8s\ ^1S_0$, $6p^2\ ^3P_0$, and $6s7d\ ^1D_2$) theoretically or experimentally derived A values are available. As can be seen from Table II, our results for A(457.4 nm) and A(460.0 nm) are in good

TABLE III. Branching fractions and transition probabilities for an upper level with a lifetime calculated by use of the Ladenburg method.

Upper level	Lifetime (ns)	Lower level	Wave number (cm^{-1})	Wavelength[b] (nm)	Branching[c] fraction	Transition probabilities[a] (s^{-1}) Experimental Here	Experimental Literature[d,e]	Theoretical Ref. 8	Theoretical Refs. 9 and 10
$5d6p\ ^3P_2^\circ$	13±2	$6s5d\ ^3D_1$	16922.6	590.8	0.02(9)	1.5±0.3[6]	2.0[6]	2.6[6]	0.9[6]
		$6s5d\ ^3D_2$	16741.0	597.2	0.23(8)	1.8±0.3[7]	1.6[7]	2.2[7]	1.4[7]
		$6s5d\ ^3D_3$	16360.0	611.1	0.72(3)	5.5±0.9[7]	5.6[7]	5.5[7]	9.7[7]
		$6s5d\ ^1D_2$	14561.2	686.6	0.03(10)	2.3±0.4[6]			
		$5d^2\ ^3F_2$[f]	5022.5[f]	1990.5	<0.0007	<5.4±1.6[4][g]			
		$5d^2\ ^3F_3$[f]	4706.3[f]	2124.2	<0.001	<7.7±2.3[4][g]			
		$5d^2\ ^1D_2$	2894.5	3453.9					
		$5d^2\ ^3P_1$	2476.5	4036.9					
		$5d^2\ ^3P_2$	2037.6	4906.4					

[a]The values given are to be multiplied by powers of 10 with the exponents listed in brackets.
[b]Wavelength in air, according to Ref. 26.
[c]Values in parentheses indicate uncertainties in percent. The upper limits have an estimated uncertainty of ±30%.
[d]As recommended by Jahreiss and Huber (Ref. 5), all transition probabilities from Miles and Wiese (Ref. 4) with lower levels belonging to the term $6s5d\ ^3D$ should be multiplied by a factor of 0.56. The values given in this column are corrected accordingly.
[e]The A values given in Ref. 4 are estimated by their authors to be uncertain within 50%.
[f]Designation and level wave numbers according to Ref. 27.
[g]The branching ratio between these two infrared lines could be measured: $A(2.12\ \mu\text{m})/A(1.99\ \mu\text{m})=1.4\pm0.4$.

TABLE IV. Branching fractions and transition probabilities for an upper level with a lifetime calculated by use of two literature A values.

Upper level	Lifetime (ns)	Lower level	Wave number (cm^{-1})	Wavelength[b] (nm)	Branching[c] fraction	Transition probabilities[a] (s^{-1}) Experimental Here	Experimental Literature	Theoretical Ref. 8	Theoretical Refs. 9 and 10
$6s6d\ ^3D_3$	15.2±3.0	$6s6p\ ^3P^\circ_2$	17303.4	577.8	0.988(1)	6.5±1.3[7]	6.8±1.7[7][d] 6.2±0.9[7][e]	4.8[7]	4.7[7]
		$5d6p\ ^3F^\circ_2$	8753.4	1142.1	$<8\times10^{-4}$	<5.3±1.6[4]			
		$5d6p\ ^3F^\circ_3$	7870.7	1270.2	$<8\times10^{-4}$	<5.3±1.6[4]			
		$5d6p\ ^1D^\circ_2$	7743.7	1291.0	$<8\times10^{-4}$	<5.3±1.6[4]			
		$5d6p\ ^3F^\circ_4$	7061.0	1415.8	0.003(23)	2.0±0.6[5]			
		$5d6p\ ^3D^\circ_2$	6286.6	1590.3	$<3\times10^{-4}$	<2.0±0.6[4]			
		$5d6p\ ^3D^\circ_3$	5838.3	1712.4	0.005(21)	3.3±1.0[5]			
		$5d6p\ ^3P^\circ_2$	4861.6	2056.4	0.004(31)	2.6±1.0[5]			6.5[5]
		$5d6p\ ^1F^\circ_3$	4001.8	2498.2	$<8\times10^{-4}$	<5.3±1.6[4]			

[a]The values given are to be multiplied by powers of 10 with the exponents listed in brackets.
[b]Wavelength in air, according to Ref. 26.
[c]Values in parentheses indicate uncertainties in percent. The upper limits have an estimated uncertainty of ±30%.
[d]The original value from Miles and Wiese (Ref. 4) is multiplied by a factor of 0.45, as recommended by Jahreiss and Huber (Ref. 5).
[e]Reference 6.

agreement with the values given by Miles and Wiese,[4] the latter being corrected as recommended by Jahreiss and Huber.[5] In the case of A(608.3 nm), on the other hand, the result obtained by Fisk *et al.*[3] lies roughly a factor of 3 below our datum, and the two values are incompatible within their uncertainty ranges.

For the lines considered in this section and not originating in the resonance level, only nine calculated A values have been published.[8–10] Good agreement is seen for A(612.9 nm) (Ref. 8), A(814.8 nm), and A(1.14 μm) (Refs. 9 and 10, respectively). For the relatively strong transition $6s6p\ ^3P^\circ_1-6s8s\ ^1S_0$ at $\lambda=460.0$ nm, our result deviates by nearly a factor of 100 from the one calculated by Hafner and Schwarz.[8] The remaining five cases show deviations of factors 3–4.

B. A values from calculated lifetimes

To convert the branching fractions of the level $5d6p\ ^3P^\circ_2$ (cf. Table III) into transition probabilities, we calculated a lifetime by use of the Ladenburg method.[7,20] This method can be applied if lines originating in two different upper levels—one of them having a known lifetime, the lifetime of the other level being unknown—share a lower level, and if the relative intensity of these lines has been measured. In addition, the branching fractions of both of these levels must be known as well. Since the A values of the lines originating in the upper level with the known lifetime are then given, the absolute transition probability scale for the other upper level can be linked through the intensity ratio of the lines sharing the lower level.

Such a favorable pattern is present twice in our data set: atoms in the upper level $5d6p\ ^3P^\circ_2$ (with unknown lifetime) can undergo transitions to the lower levels $6s5d\ ^3D_{2,1}$, as is the case for atoms decaying from the level $5d6p\ ^3D^\circ_1$. The transition probabilities of the lines originating in the latter level—having wavelengths of 659.5 and 667.5 nm, respectively—are known from lifetime data and branching fraction measurements.[30] The resulting lifetimes for level $5d6p\ ^3P^\circ_2$ are then 11±1 and 15±2 ns, depending on whether the link is made through A(659.5 nm) or A(667.5 nm). These two values are just not compatible within their uncertainty ranges, but we note that their mean value, 13±2 ns, is well supported by the calculated lifetime from Ref. 8. The resulting A values for the two strongest visible lines agree with experimental literature data[4,5] as well as with theoretical values.[8] On the other hand, for A(590.8 nm), the theoretical[8–10] data and our experimental value deviate by at least 25%.

It is worth mentioning that the branching ratio between two infrared lines listed in Table III, namely,

$$\frac{A(2.12\ \mu\text{m})}{A(1.99\ \mu\text{m})}=1.4\pm0.4\ ,$$

could be determined from the spectra obtained with source II. To link the intensities of these two lines to the radiometric scale of source I was, however, not possible,

TABLE V. Comparison of literature lifetimes for three moderately excited Ba I levels.

Level	Lifetime (ns) Ref. 1[a]	Ref. 2	Ref. 3
$6s8s\ ^1S_0$[b]	18.2±0.8	18.2±0.8	8.4±0.6	
$6p^2\ ^3P_0$	6.1±0.5	6.2±0.4	5.1±0.5	6.4±0.5
$6s7d\ ^1D_2$[c]	7.0±0.5	6.9±0.5	5.9±0.4	

[a]Hannaford and Lowe (Ref. 1) determined their values by measuring the fluorescence signal after stepwise (second column) and two-photon (third column) laser excitation.
[b]Designation according to Ref. 24.
[c]Designation according to Ref. 25.

and we therefore are only able to give upper limits for the corresponding A values.

Finally, regarding the upper level $6s6d\ ^3D_3$ (cf. Table IV), we could detect three infrared lines besides the visible transition at $\lambda = 577.8$ nm. For the remaining five transitions, we give upper limits of the corresponding branching fractions. To arrive at a lifetime for this level we used the fact that two compatible A values for the intense orange line are available in the literature.[4-6] The resulting lifetime of 15.2 ± 3.0 ns is based on a mean value derived from $(6.8 \pm 1.7) \times 10^7\ s^{-1}$—the A value from Miles and Wiese[4] corrected according to Jahreiss and Huber[5]—and $(6.2 \pm 0.9) \times 10^7\ s^{-1}$, as measured by Eicke.[6] The lifetime we thus obtained is shorter by approximately 25% than the theoretical lifetimes resulting from the calculated values for A(577.8 nm).[8-10]

IV. CONCLUSIONS

From broadband and narrow-band FTS spectra of two different types of hollow-cathode discharges, we have measured the branching fractions of six upper levels in neutral barium, whereby the branching fractions of the resonance level were obtained by augmenting our emission measurements with results from laser excitation experiments.[13-16] By converting these branching fractions to A values with the aid of four literature lifetimes[1,3,19] and two lifetimes calculated by either the Ladenburg method[7,20] or from literature A values,[4-6] we were able to determine the transition probabilities of 13 visible and 16 infrared lines. In addition, we present upper limits for the A values of 15 infrared lines originating in five upper levels.

The resulting transition probabilities cover approximately five decades. Narrow-band FTS emission spectra are shown to be a powerful method for measuring infrared lines of low intensity. In particular, this method permitted the determination of the transition probability of an extremely weak infrared transition originating in the Ba I resonance level, namely, $6s5d\ ^3D_1 - 6s6p\ ^1P^\circ_1$ at a wavelength of 1.11 μm: $A = (3.6 \pm 0.5) \times 10^3\ s^{-1}$.

The comparison of our results with experimental data taken from the literature is satisfactory, in general. However, there are two instances where severe deviations occur, namely, for A(608.3 nm) (Ref. 3) and A(1.50 μm) (Ref. 28), originating in the upper levels $6p^2\ ^3P_0$ and $6s6p\ ^1P^\circ_1$, respectively. The agreement with theoretical transition probabilities,[8-11] on the other hand, is only partial. This fact and also the absence of theoretically derived A values for most of the infrared lines presented here indicate that further calculations are needed.

We also note here that the correction factors for some of the literature A values that are based on absorption measurements,[4] as proposed by Jahreiss and Huber[5] and later used by Niggli and Huber,[7] are again supported by the present results, albeit in four of the six possible test cases only.

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