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Ba oscillator strengths from a laser-excited vapor

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We describe a method for evaluating the consistency of oscillator strengths in the neutral and ionized species of an atom. The method is based on the constant number density of a dense vapor. In our procedure the vapor is excited and then ionized within less than 1 μs by resonant laser irradiation. The column densities $N_i l_i$ of the levels that are appreciably occupied during the excitation and ionization are sampled by absorption, emission, and/or anomalous-dispersion measurements in a set of suitable spectral lines. Given correct oscillator strengths f_i for these lines, the column density $\sum_i N_i l_i$ (which is actually determined from the measured quantities $N_i f_i l_i$) will remain constant while excitation and ionization take place. In barium—with diagnostics based on accepted f values in the literature—the column density appeared reduced by a factor of 2 during the initial portion of the excitation, i.e., while only the low-lying levels of neutral barium were predominantly excited. Upon complete ionization, the column density took again the value derived before the laser irradiation, when all atoms had been in the ground state. From this behavior we concluded that the literature oscillator strengths for absorption lines involving the neutral ground state and the ground state and low-lying levels of Ba$^+$ were consistent on a relative scale, but that the f values for absorption transitions from the low-lying metastable levels and from the resonance level of neutral barium were too large by a factor of 2. We recommend a set of correction factors to be applied to the existing data in the literature. Our conclusions are supported by the results of a critical analysis of recent oscillator strength and lifetime measurements for neutral barium.

I. INTRODUCTION

The measurement of oscillator strengths of selectively excited atomic species has long been contemplated.[1] Bernhardt *et al.*[2] have made a branching-ratio measurement from a laser-excited level and Carlsten, McIlrath, and Parkinson[3] have reported photoionization cross sections out of a laser-excited metastable level. In general however, laser excitation in quantitative spectroscopy has been restricted to lifetime measurements (cf. Ref. 4 for a recent example).

We report here the results of an experiment employing selective laser excitation on barium, where it was found that the literature f values[5] of the Ba I absorption lines originating from the low-lying metastable levels as well as from the resonance level[6] are too large by a factor of 2. A part of this investigation, namely the study of the excitation and ionization mechanisms in resonantly excited Ba vapor, has already been published.[7,8] In this paper, we present our results on oscillator strengths.[9] After a short description of the experimental arrangement (Sec. II), we shall discuss the principle of measurement and our results (Sec. III). A critical evaluation of recent lifetime and oscillator-strength measurements (Sec. IV) will then be followed by a discussion of the finally adopted corrections for Ba I f values (Sec. V).

II. EXPERIMENT

The experimental apparatus has been described in detail in Ref. 7. In short, a barium vapor (contained in a furnace) was excited by a flash-lamp pumped dye laser that was tuned to the 553.5-nm resonance line of Ba I. A second laser, an N_2-laser-pumped dye laser operated in a broad band mode, was used to measure, by means of the "hook" method,[7] the number densities of the predominantly populated Ba and Ba$^+$ levels. The continuum emitted by the second laser had a very short pulse length (a few ns) as compared to the duration (ca. 1 μs) of the flash-lamp driven pump laser.

Since we had placed the furnace in the test arm of a Mach-Zehnder interferometer and employed a stigmatic spectrograph, we obtained a spectrally dispersed interferogram—the hook spectrum—displaying the anomalous dispersion in the vicinity of the spectral lines occuring in the Ba vapor. With the broadband laser we were able to photograph a hook spectrum in a single exposure with a known delay from the start of the pump laser pulse. Thus we could sample the product $N_i f_i l_i$, of column density $N_i l_i$ (m^{-2}) and absorption oscillator strength f_i, for a number of lower levels and associated lines, in different stages of excitation and ionization, i.e., as a function of time. In addition, we also monitored the populations of highly excited levels by observing the temporally and spectrally resolved fluorescence of the laser-excited Ba vapor with photoelectric detection in a wavelength region extending from 380 nm to 1 μm. (Emission and hook-method measurements were put onto the same scale by comparing the population of a level measured by both techniques.)

III. PRINCIPLE OF MEASUREMENT AND RESULTS

In the absence of excitation by the pump laser, the atoms in the barium vapor were in the ground state.[10] The hook (i.e., the interferometric anomalous dispersion) measurement on the 553.5-nm resonance line, whose oscillator strength is accurately known from lifetime measurements,[11–14] then yielded the total column density of Ba in the vapor:

$$N_0 l_0 = \frac{4\pi^2 \epsilon_0 m c^2}{e^2} \frac{K\Delta^2}{\lambda^3 f} . \tag{3.1}$$

Here, Δ is the separation of the hooks formed by the interference fringes in the vicinity of the resonance line, K is the hook-method constant (i.e., approximately the fringe order used), and λ and f are, respectively, the wavelength and oscillator strength of the line in question.[7,15,16] The remaining symbols have their usual meaning of electric field constant (ϵ_0), electron mass and charge (m and e), and velocity of light (c).

When the intense (7 $\mathrm{GW\,m^{-2}}$), resonantly tuned pump laser was turned on, its radiation burnt through the 0.25-m-long barium-vapor column (with a density of $6.3\times10^{20}\ \mathrm{m^{-3}}$) in less than 50 ns. The excitation histories of twelve Ba and Ba^+ levels, which had been determined with a time resolution of 5 ns, have already been reported.[7,8] From these data and from the analysis of emission measurements (which indicated the populations of higher-lying levels), we concluded that, at a time of 50 ns, a uniformly excited vapor column containing predominantly atoms in the levels between the ground state and the resonance level (cf. Fig. 1) had been established;[17] almost complete ionization was achieved within 400 ns.

Under the assumption of a constant total barium density,[18] we may set the column densities before and during laser excitation equal to each other

$$N_0 l_0 = \sum_i N_i l_i , \tag{3.2}$$

where the sum is to be taken over all appreciably populated levels of neutral and ionized barium.

In the course of the progressive excitation and ionization, the sum can be restricted; at 50 ns, only the levels up to the laser-excited resonance level $6p\,{}^1P_1^o$, whose column densities $N_i l_i$ had been obtained from anomalous-dispersion measurements, must be included. Later, higher excited levels—whose populations were observed through emission—and then the low-lying Ba^+ levels—again observed by anomalous dispersion—must be included in the sum as well.[19]

Since the determination of each $N_i l_i$ involves an oscillator strength [cf. Eq. (3.1)], Eq. (3.2) can be fulfilled for the measured values of $N_i l_i$ only if the f values that are used in the data reduction are correct. When literature f values[5,6] were used, we found (cf. Fig. 2) that $\sum_i N_i l_i \approx 0.5 N_0 l_0$ at 50 ns, but that the proper equality [Eq. (3.2)] was fulfilled again after ca. 400 ns, when nearly complete ionization had been reached.

We conjectured, therefore, that the apparent loss of density was, in fact, due to incorrect oscillator strengths of the lines used for determining the populations of those levels that were predominantly occupied at 50 ns, namely those belonging to the $6p\,{}^1P^o$, $5d\,{}^1D$, and $5d\,{}^3D$ terms. If all these oscillator strengths are multiplied by the same, time-independent correction factor of 0.49 ± 0.06, and if the literature f values are used for the ion lines and the lines originating in the neutral ground state and in the levels of the term $6p\,{}^3P^o$ (cf. below), a nearly constant $\sum_i N_i l_i = N_0 l_0$ is obtained throughout the pump-laser pulse as seen in Fig. 2.[20]

A uniform correction factor of 0.49 for the f values of lines originating in the three terms $6p\,{}^1P^o$, $5d\,{}^1D$, and $5d\,{}^3D$ can be justified as follows. The two levels $5d\,{}^1D_2$ and $5d\,{}^3D_2$ are populated by decay of the resonance level $6p\,{}^1P_1^o$ through stimulated infrared emission (observed during the first 50 ns, cf. Ref. 7). At delays around 50 ns, the relative populations of the three levels ${}^3D_2$, ${}^1D_2$, and

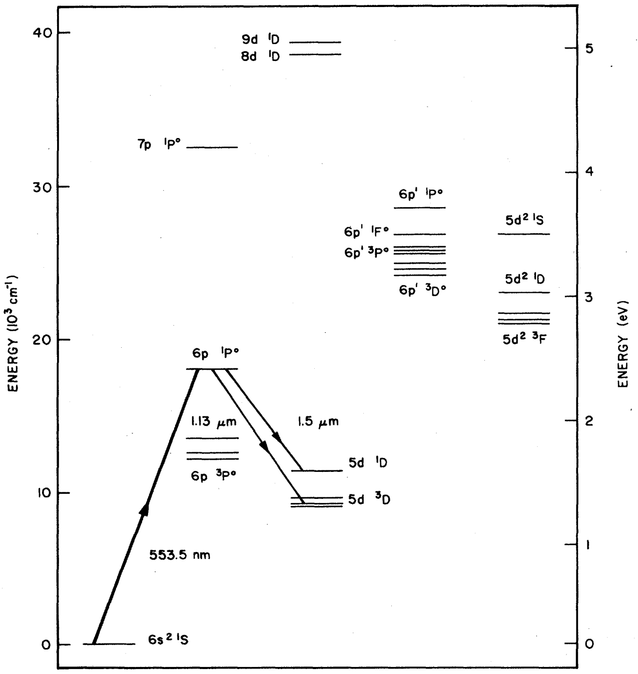

FIG. 1. Simplified term scheme of Ba containing only terms discussed in this paper. The transitions shown are relevant in connection with the initial excitation by resonant laser irradiation.

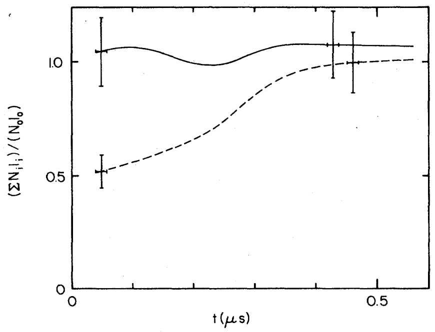

FIG. 2. The apparent ratio between the number of atoms during laser excitation and that before the excitation starts. The two curves shown represent ca. 30 data points. The dashed curve results if literature f values are used. The solid line is obtained if oscillator strengths of Ba I lines originating from levels in the $6p\,{}^1P^o$ and ${}^3P^o$ and in the $5d\,{}^1D$ and ${}^3D$ terms are decreased by a factor of approximately 2, as indicated in Table III.

TABLE I. Comparison of the literature f values with lifetime measurements.

Transition			Hook measurement			Lifetime measurements and corrected values				
Upper level	Lower level	Wavelength^a (nm)	f_{rel}^{b}	f_{lit}^{c}	$A_{lit}(10^8\ s^{-1})^c$	Lifetime (ns)	$A_{corr}(10^8\ s^{-1})^d$	f_{corr}^{d}	Correction factor based on this table^e	Recommended f values^f
$6s6p\,^1P_1^o$	$6s^2\,^1S_0$	553.548	1000	1.59	1.15	$(8.36\pm0.25)^g$	1.19	1.64	1.03	1.64 *A*
	$6s5d\,^1D_2$	1500.04		0.10	0.049	$(8.2\ \pm0.2)^h$	$\leq 0.0017^i$	$\leq 0.0034^i$	≤ 0.034	0.0034 *E*
	$6s5d\,^3D_2$	1130.304				8.37 ± 0.14^j				
						8.37 ± 0.08^k				
					$\sum A = 1.199$		$\sum A \leq 1.19$			
$6s7p\,^1P_1^o$	$6s^2\,^1S_0$	307.158	109^l	0.17	0.41	$13.2\ \pm0.4^m$	0.42	0.18	1.03	0.18 *D*
	$6s5d\,^1D_2$	472.644	87.6	0.14	0.69	$(13.5\ \pm0.6)^n$	≤ 0.34	≤ 0.068	$\leq 0.48\pm0.06$	0.062 *D*
	$6s5d\,^3D_2$	428.490								
	$5d^2\,^1D_2$	1054.010								
					$\sum A = 1.10$		$\sum A \leq 0.76$			
$6s8d\,^1D_2$	$6s6p\,^3P_2^o$	399.226				$20.1\ \pm0.8^o$				
	$6s6p\,^1P_1^o$	487.765		0.60^p	1.01^p		≤ 0.50	≤ 0.30	$\leq 0.49\pm0.02$	0.31 *D*
$6s9d\,^1D_2$	$6s6p\,^1P_1^o$	469.911		0.21^p	0.38^p	$25.5\ \pm1.4^o$	≤ 0.39	≤ 0.22	$\leq 1.03\pm0.06$	0.11 *D*
$5d6p'\,^1P_1^o$	$6s^2\,^1S_0$	350.111	102^l	0.16	0.29	12.17 ± 0.09^q	0.30	0.17	1.03	0.17 *D*
	$6s5d\,^1D_2$	582.628	187	0.30	0.97	$(12.4\ \pm0.9)^n$	≤ 0.32	≤ 0.092	$\leq 0.31\pm0.03$	0.13 *D*
	$6s5d\,^3D_2$	516.949								
	$5d^2\,^1D_2$	1820.41					0.20^i	0.61^i		
	$5d^2\,^1S_0$	5563.6^r								
					$\sum A = 1.26$		$\sum A \leq 0.822$			
$5d6p'\,^3P_1^o$	$6s^2\,^1S_0$	388.933	6.3^l	0.010	0.015	$11.7\ \pm0.9^n$	0.015	0.010	1.03	0.010 *D*
	$6s5d\,^3D_1$	599.709	157	0.25	0.46		≤ 0.30	≤ 0.14	$\leq 0.56\pm0.05$	0.14 *D*
	$6s5d\,^3D_2$	606.312	202	0.32	0.97		≤ 0.54	≤ 0.18	$\leq 0.56\pm0.05$	0.18 *D*
	$6s5d\,^1D_2$	698.679								
					$\sum A = 1.445$		$\sum A \leq 0.86$			
$5d6p'\,^3D_1^o$	$6s^2\,^1S_0$	413.243	6.22	0.0099	0.013	$17.0\ \pm0.5^n$	0.013	0.0102	1.03	0.0102 *D*
	$6s5d\,^3D_1$	659.533	295	0.47	0.72		≤ 0.39	≤ 0.25	$\leq 0.54\pm0.02$	0.26 *D*
	$6s5d\,^3D_2$	667.527	88.6	0.14	0.35		≤ 0.19	≤ 0.076	$\leq 0.54\pm0.02$	0.079 *D*
	$5d^2\,^3F_2$	3068.51^r								
					$\sum A = 1.083$		$\sum A \leq 0.59$			

TABLE I. (*Continued*).

Transition				Hook measurement		Lifetime measurements and corrected values				
Upper level	Lower level	Wavelength[a] (nm)	f_{rel}[b]	f_{lit}[c]	$A_{\mathrm{lit}}(10^8\ \mathrm{s}^{-1})$[c]	Lifetime (ns)	$A_{\mathrm{corr}}(10^8\ \mathrm{s}^{-1})$[d]	f_{corr}[d]	Correction factor based on this table[e]	Recommended f values[f]
$5d6p'\,{}^3D_3^o$	$6s5d\,{}^3D_2$	634.168	98.5	0.16	0.19	10.2 ±1.5[o]	≤ 0.17	≤ 0.15	$\leq 0.92 \pm 0.14$	0.089 *D*
	$6s5d\,{}^3D_3$	649.876	343	0.55	0.86		≤ 0.81	≤ 0.51	$\leq 0.92 \pm 0.14$	0.31 *D*
	$6s5d\,{}^1D_2$	735.929								
	$5d^2\,{}^3F_3$	2680.51[r]								
	$5d^2\,{}^3F_4$	2978.78[r]								
					$\sum A = 1.05$		$\sum A \leq 0.98$			
$5d6p'\,{}^1F_3^o$	$6s5d\,{}^1D_2$	648.291	245	0.39	0.44	44.5 ±1.8[q]	≤ 0.21	≤ 0.18	$\leq 0.47 \pm 0.02$	0.17 *D*
	$6s5d\,{}^3D_3$	580.569	12.7	0.020	0.040		≤ 0.019	≤ 0.0094	$\leq 0.47 \pm 0.02$	0.011 *D*
	$6s5d\,{}^3D_2$	568.000								
					$\sum A = 0.48$		$\sum A \leq 0.225$			

[a] Reference 33.

[b] Reference 24.

[c] Reference 5.

[d] Oscillator strengths for the resonance lines are accurately known. Upper limits (≤) are given for the other lines, because the branching ratios are not complete.

[e] Note that the correction factors given here are derived solely from the data presented in this table and do therefore not correspond to the correction factor of 0.49 from our measurements (cf. Sec. III).

[f] The recommended f values are converted from the literature data[5] by use of the modified correction factors of Table III (cf. Sec. V). The accuracies of the oscillator strengths follow those indicated in Ref. 5, except for the resonance line, where our correction now results in an uncertainty within 3% (indicated by the code letter *A*). The other code letters have the same meaning as in Ref. 5 (*D* and *E*: uncertainties estimated to be within 50% and larger than 50%, respectively).

[g] Reference 11.

[h] Reference 12.

[i] Reference 2.

[j] Reference 13.

[k] Reference 14.

[l] Reference 25.

[m] Reference 27.

[n] Reference 29.

[o] Reference 30.

[p] Reference 6.

[q] Reference 28.

[r] Reference 41.

${}^1P_1^o$ are those of their statistical weights, if literature f values are used to determine the populations.[21] This led us to conclude that the ratios of the literature f values in question were correct. (As already pointed out, the f values themselves should be shifted on an absolute scale, however.) Given the small fine-structure splitting of the $5d\,{}^3D$ term and the measurement technique used for the literature oscillator strengths[5] (hook method in a furnace), we may assume that the f values of lines originating in all levels of this term have the same relative scale.

Unfortunately, a similar argument cannot be given for the $6p\,{}^3P^o$ term, because its population mechanism is still unclear. However, we note that the fractional population of this term is relatively small (i.e., $<10\%$) at 50 ns. A correction of the corresponding f values by a factor of 0.5, for example, therefore has no perceptible influence on the total apparent Ba density and on the overall uniform correction proposed above.

In the following section we show that the hypothesis of the described correction is, in general, supported by a comparison with measured lifetimes of Ba levels. Somewhat more refined corrections and recommended oscillator strengths will be presented in Sec. V.

IV. LIFETIME AND RECENT OSCILLATOR-STRENGTH MEASUREMENTS

Table I (column 6) shows the lifetimes of several Ba levels, which have been measured in the past few years. If one knows the branching of the photon flux emitted by a given upper level k, i.e., if one knows the branching ratio R_{ki} (with $\sum_i R_{ki}=1$) for all the lines in the decay scheme of level k, the transition probabilities $A_{ki}=R_{ki}/\tau_k$ can be calculated from the lifetime τ_k of the level in question (cf. Refs. 22 and 23). Unfortunately, accurate and complete branching ratios are not yet available for Ba.

To derive transition probabilities—and thus also the oscillator strengths $f_{ik}=(g_k/g_i)(\epsilon_0 mc\lambda_{ik}^2/2\pi e^2)A_{ki}$ (where g_i and g_k are the statistical weights, respectively, of the lower and upper levels and λ_{ik} is the wavelength of the transition)—from the lifetime measurements, most of the authors cited in Table I have calculated branching ratios from oscillator strengths that had been measured with the hook method.[5] This procedure may lead to erroneous results, because f values belonging to lines with a common upper level, that were determined by the hook method, can be subject to scale shifts. The reason is that hook-method measurements require a knowledge of the populations of the lower levels of the transitions in question; an inaccurate knowledge of the thermodynamic state of the vapor will therefore introduce systematic errors, if the lines concerned have lower levels with different excitation energies.

We note that most of the levels that have measured lifetimes are of odd parity and therefore have an allowed transition to the ground state $6s^2\,{}^1S_0$. The probabilities of these transitions are expected to be accurate on an absolute scale: their relative values have been measured by the hook method and then normalized to the transition probability of the resonance line,[5,24–26] which is well known from lifetime measurements. (As discussed in Sec. IV B1 and shown in Tables I and III, however, we apply a 3% correction to the oscillator strengths of the resonance lines.)

To derive the required corrections for the transition probabilities involving excited lower levels, we proceeded as follows. For each upper level of odd parity with a known lifetime, we took the presumably correct, published A value for the transition to the ground state and scaled the remaining transition probabilities for lines with excited lower levels, so that $\sum_i A_{ki}=1/\tau_k$. (The additional, 3% correction of the transition probability of the resonance line—and of all the other transitions involving the ground state—will be discussed in connection with the lifetime of the $6p\,{}^1P_1^o$ level.)

Oscillator strengths that correspond to the transition probabilities corrected in this way are shown in Table I. The first two columns denote the levels and wavelengths belonging to the transitions out of the levels with known lifetimes. The relative oscillator strengths of column 3, which had originally been measured with the hook method,[24,25] have been adjusted by Miles and Wiese[5] to f and A values on the absolute scale (f_{lit} and A_{lit} of columns 4 and 5) given by the well-established oscillator strength of the resonance line (column 4 and 5). With the lifetime of the upper level (column 6) one then obtains, with the procedure described above, the corrected transition probabilities (column 7) and f values (column 8), as well as correction factors (column 9) for the original literature oscillator strengths of column 4. (The recommended f values of column 10 will be discussed in Sec. V.)

A. Accuracy of the lifetime measurements

The most reliable lifetime measurements are those of the resonance level. The four values shown in column 6 of Table I coincide within their mutual error limits. We also note that these lifetimes had been determined with different methods. Dickie and Kelly[13] and Kelly and Mathur[14] have obtained the most accurate results for the resonance level. We may assume, that the other levels measured by the same authors with the same method are reliable as well. The good agreement between the lifetimes for the level $6s7p\,{}^1P_1^o$ obtained by Dickie and Kelly[27,28] and by Brecht *et al.*[29] further suggests that the other measurements of Brecht *et al.* are also accurate. The identical lifetimes measured for the level $6s\,17d\,{}^1D_2$ (not shown in Table I) by Kaiser *et al.*[30] and by Aymar *et al.*[31] leads to a similar conclusion regarding the accuracy of the lifetimes of Ref. 30.

B. Remarks on some of the upper levels listed in Table I

1. Level $6s\,6p\,{}^1P_1^o$

Bernhardt *et al.*[2] have determined an (absorption) f value of ≤ 0.0034 for the transition $5d\,{}^1D_2$–$6p\,{}^1P_1^o$ at $\lambda=1500.04$ nm. This corresponds to a relative photon flux of $\leq 1/700$ of that in the resonance line; in contrast, the old value was $\approx 1/24$.[5] The relative photon flux of 1/600 calculated by McCavert and Trefftz[32] confirms the new experimental value. As Bernhardt *et al.* have not only measured the contribution of the line $5d\,^1D_2-6p\,^1P_1^o$ (as explicitly stated), but also that of $5d\,^3D_2-6p\,^1P_1^o$ at $\lambda=1130.04$ nm, their result actually represents the ratio between the sum of the transition probabilities for these two infrared lines and the transition probability of the resonance line.

There are no experimental data for the transition at 1.13 μm, so that we cannot exclude that this line—although being an intercombination line—is actually stronger than that at 1.5 μm. This follows from the intensity values given by Russell and Moore[33] and also from experiments by Bricks *et al.*[34] and by Whitkop and Wiesenfeld;[35] other experiments however seem to imply the opposite (cf. remarks to level $5d6p'\,^1P_1^o$ below).

Given the new branchings for the two infrared lines by Bernhardt *et al.*,[2] which now are negligible relative to the resonance line, the transition probability of the resonance line may directly be taken to be the inverse lifetime of its upper level. This results in a recommended absorption f value for the resonance line of $f(553.548\text{ nm})=1.64\pm0.02$, which is larger by 3% than the old value.

2. Level $6s\,7p\,^1P_1^o$

Theoretical calculations by Friedrich and Trefftz[36] for the line $5d^2\,^1D_2-6s7p\,^1P_1^o(\lambda=1054.01\text{ nm})$ yield f values ranging from 0.19 to 0.66, depending on the approximations used. If we included this result, we would obtain an upper bound of 0.030 for the f value for the $6s\,5d\,^1D_2-6p7p\,^1P_1^o(\lambda=472.644\text{ nm})$ line—corresponding to a correction factor of 0.21. In any case, $f(1054.01\text{ nm})\leq0.38$ must hold, or else, the listed lifetime would lead to a negative f value for the 472.644 nm line. (Friedrich and Trefftz[36] do not consider their values to be very reliable in the absence of a confirmation by further calculations or experiments.)

3. Level $5d6p'\,^1P_1^o$

Bernhardt *et al.*[2] have measured a ratio of 0.67 for the two transition probabilities of $5d^2\,^1D_2-5d6p'\,^1P_1^o(\lambda=1820.41\text{ nm})$ and $6s^2\,^1S_0-5d\,6p'\,^1P_1^o(\lambda=350.111\text{ nm})$. The resulting correction by a factor of ≤0.31 for the f value of the line at $6s\,5d\,^1D_2-5d6p'\,^1P_1^o(\lambda=582.628\text{ nm})$ is not inconsistent with the remarks made in the preceding discussion of level $6s7p\,^1P_1^o$, if there one were to consider the line $5d^2\,^1D_2-6s7p\,^1P_1^o(\lambda=1054.01\text{ nm})$ as well in establishing the correction.

As already pointed out in connection with level $6s\,6p\,^1P_1^o$, Bernhardt *et al.*[2] have neglected the line $6s\,5d\,^3D_2-6s\,6p\,^1P_1^o(\lambda=1.13\ \mu\text{m})$. This did not matter for the above calculation of the oscillator strength of the resonance line, but may be of importance here. It can be shown[37] that the above-mentioned intensity ratio of 0.67 may, in fact, be much smaller, if the influence of the 1.13-μm transition is considered. Without an exact knowledge of the contribution of this line, the correction factor for the oscillator strength of the 582.628-nm line can take any value between 0.31 and 0.54, whereby the lower and upper limits correspond to the A (18204.1 nm)$=0.20\times10^8\text{ s}^{-1}$ given in Table I and to $A=0$, respectively.

C. Corrections of f values for the absorption lines out of the $6s\,6p\,^3P^o$ term

As mentioned at the end of Sec. III, there are no straightforward arguments that could lead to a correction of the absorption lines out of the $6s\,6p\,^3P^o$ term, based on our own data. Furthermore, there are no lifetime measurements of even levels that have decay channels into the $^3P^o$ levels.

However, oscillator strengths for absorption lines out of the $^3P^o$ levels and other metastable levels are available from hook measurements on a shock tube.[38] These data are compared in Table II with the previously accepted values.[5]

The correction factor needed for the $6s5d\,^1D_2-5d6p'\,^1P_1^o$ ($\lambda=582.628$ nm) line is similar to that derived by us from the lifetime measurements (cf. Table I). This suggests that the f value[38] for the $6s\,6p\,^3P_2^o-6s6d\,^3D_3$ ($\lambda=577.762$ nm) line given in Table II is also more reliable than the earlier value.[5]

The ratio between the oscillator strengths of the lines at 580.023 and 577.762 nm, which have $6p\,^3P_2^o$ as common lower level, is available from our previous population measurements.[7] We prefer our ratio, 0.18 ± 0.03, which was obtained[39] with sizable populations and by use of the "hook vernier",[40] over those of 0.24 and 0.11 ($\pm60\%$) given in Refs. 5 and 38, respectively.

V. FINAL CORRECTION: A CRITICAL COMPARISON BETWEEN OUR AND LITERATURE DATA

Our final, recommended values represent a critical compromise between the data derived from the present

TABLE II. Comparison of literature data (Ref. 5) with hook-method measurements made on a shock tube (Ref. 38).

Transition	λ (nm)	f_{lit}[a]	f[b]	f/f_{lit}
$6s^2\,^1S_0-6s\,6p\,^1P_1^o$	553.548	1.59	1.6	
$6s\,6p\,^3P_1^o-6s\,6d\,^3D_2$	551.905		0.38 ± 0.13	
$6s\,6p\,^3P_2^o-6s\,6d\,^3D_3$	577.762	1.0	0.45 ± 0.09	0.45 ± 0.09
$6s\,6p\,^3P_2^o-6s\,6d\,^3D_2$	580.023	0.24	0.05 ± 0.02	0.21 ± 0.08
$6s\,5d\,^3D_3-5d6p'\,^1F_3^o$	580.569	0.020	0.0057 ± 0.0018	0.29 ± 0.09
$6s\,5d\,^1D_2-5d\,6p'\,^1P_1^o$	582.628	0.30	0.17 ± 0.06	0.57 ± 0.20

[a] Reference 5.

[b] Reference 38.

TABLE III. Recommended correction factors for literature oscillator strengths (Refs. 5, 6, and 26) belonging to lines with $\lambda \leq 1$ μm.

Lower term	Recommended correction factor
$6s^2\,{}^1S$	1.03
$6s5d\,{}^3D$	0.56
$6s5d\,{}^1D$	0.44
$6s6p\,{}^3P^o$	0.45
$6s6p\,{}^1P^o$	0.51

laser experiment, the correction factors from column 9 of Table I, and literature data obtained on a shock tube.[38] Although a blanket correction factor of 0.49 (cf. Sec. III) would, in principle, suffice, we advocate the somewhat modified set of correction factors given in Table III. These correction factors should be applied to most oscillator strengths of Refs. 5, 6, and 26 [for lines with wavelengths less than 1 μm (Ref. 9)]. For convenience we also gave in Table I (column 10) the recommended values for the lines listed there. The factors of Table III lead to a good agreement with the measured lifetimes and result in a total Ba density (cf. Fig. 1) that remains constant within the uncertainty of measurement at all times.

In modifying the blanket correction factor of 0.49, we took note of the uncertainties pointed out in connection with absorption lines out of the $6s5d\,{}^1D_2$ level in Secs. IV B2 and 3: the range of admissible correction factors suggests a slight lowering of the factor 0.49. In view of the factor of 0.31 or less for the $6s5d\,{}^1D_2-5d6p'\,{}^1P_1^o(\lambda=582.6$ nm) line and the other downward pointing indicators, we chose a value of 0.44. On the other hand we selected a correction factor of 0.56 for the absorption lines out of the ${}^3D$ levels, since—if any change is made here—a slight increase, which could be ascribed to a slightly reduced population through stimulated emission preferring the 1.5-μm over the 1.13-μm line,[21] was called for. The ratio of these two correction factors does not exceed 1.27, a value that would also be consistent within uncertainties with an equipartition of the populations in the levels ${}^1P_1^o$, ${}^1D_2$, and ${}^3D_2$ at times around 50 ns.

The correction factor of 0.45 for absorption lines out of the $6p\,{}^3P^o$ levels was derived from the data shown in Table II that had been obtained by hook-method measurements on a shock tube.[38] (An exception is the oscillator strength of the 580.023-nm line, which we related to that of the 577.762-nm line based on our own hook spectra that were taken on the laser-excited Ba vapor.[39])

VI. CONCLUSIONS

In an experiment investigating the excitation and ionization of a dense vapor under resonant laser irradiation,[7] we had noticed that diagnostic use of literature oscillator strengths belonging to absorption lines out of the excited levels up to (and including) the resonance level $6p\,{}^1P_1^o$ led to a spurious temporary reduction of Ba-vapor density.

The need for a correction of the f values used in the diagnostics and analysis of Ref. 7 is demonstrated here,[8] and the actual correction factors to be applied to the literature data are discussed in view of recent lifetime and oscillator-strength measurements.

We note that, with the method used, the total number of atoms during the experiment can be determined beforehand—in contrast to the conventional methods for measuring oscillator strengths from absorption or anomalous dispersion on furnaces or shock tubes, where the total number density can only be determined with additional assumptions on the thermodynamic state of the vapor or plasma.[23]

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[8] The results reported in Ref. 7 are already based on the corrected f values given in column 10 of Table I in the present paper.

[9] The correction of the literature data (Ref. 5) discussed here concerns only the data stemming originally from hook-method data taken on a furnace (Refs. 24 and 25). Our correction factors should also be applied to the more recently determined f values of Refs. 6 and 26, which had been scaled with the literature data of Ref. 5.

[10] Any thermal excitation was negligible. Given furnace temperatures below 1150 K, the total population of the low-lying metastable as well as resonance terms of Ba—$5d\,{}^3D$ and ${}^1D$ (excitation energy at ca. 1.2 and 1.4 eV) and $6p\,{}^3P^o$ as well as ${}^1P^o$ (1.6 and 2.2 eV)—was 10^4 times smaller than that of the ground state.

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[17] Except for fluorescence in the resonance line, no emission at wavelengths below 1 μm was observed at these early times. Emission in many lines could be well observed later, when the electron density was high enough to create a local thermal equilibrium. The estimate of the total population in all the levels lying above the resonance level under these conditions never exceeded 10%. For times, when no emission could be observed then, the population of the levels in question may safely be neglected.

[18] We could not find a mechanism that would temporarily change the density in the laser-pumped volume. In particular, no heating has occurred within the laser-excited volume at 50 ns, since at that time there is only a negligible number of electrons.

[19] From emission measurements we know that levels above the $6p\ ^1P^o$ resonance level have a negligible population at 50 ns. We concluded that, even between 200 and 400 ns, when the population of the high-lying levels peaked, the fraction of atoms in these levels never exceeded 10% of the total neutral Ba density.

[20] The upper curve of Fig. 2 is actually based on the slightly different final corrections to the literature f values given in Table III.

[21] We have observed that —at least within the uncertainty of measurement and averaged along the entire column length— the $5d\ ^1D_2$ and $^3D_2$ levels are being populated simultaneously.[7] A close inspection of the original hook spectra reveal that initially (i.e., for delay times of several ns), the $5d\ ^1D_2$ level is populated faster than $5d\ ^3D_2$. A somewhat more efficient population of $5d\ ^1D_2$ through the 1.5-μm transition might therefore not be excluded (and this has been considered in the choice of the correction factor for absorption transitions out of the $5d\ ^1D_2$ term in Sec. V). To calculate the relative column densities resulting from population through stimulated emission in the $5d\ ^1D_2$–$6p\ ^1P_1^o$ ($\lambda = 1.5\ \mu$m) and $5d\ ^3D_2$–$6p\ ^1P_1^o$ ($\lambda = 1.13\ \mu$m) lines, a model based on radiative transfer and taking into account the (currently unknown) relative strengths of these two lines would be required. Such a model could, in principle, result in column densities deviating from equipartition.

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[37] If $\chi = A(5d^2\ ^1D_2 - 5d6p'\ ^1P_1^o)/A(6s^2\ ^1S_0 - 5d6p'\ ^1P_1^o)$ is the ratio determined by Bernhardt *et al.*,[2] and $p = A(6s5d\ ^1D_2 - 6s6p\ ^1P_1^o)/A(6s5d\ ^3D_2 - 6s6p\ ^1P_1^o)$ is a ratio which had been set at infinity by these authors, because they had neglected the transition in the denominator, then the corrected ratio will be $\chi_{\text{corr}} = \chi - (1+\chi)/(1+p)$ and consequently $\chi_{\text{corr}} \leq \chi$. In order that $\chi_{\text{corr}} \geq 0$ and $\chi = 0.67$, $p \geq 1.5$ must hold, i.e., the line $6s5d\ ^3D_2 - 6s6p\ ^1P_1^o$ ($\lambda = 1.13\ \mu$m) must be weaker than $6s5d\ ^1D_2 - 6s6p\ ^1P_1^o$ ($\lambda = 1.5\ \mu$m), in contrast to what we noted above, when discussing the level $6s6p\ ^1P_1^o$.

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