Transition probabilities from the $6s6p\ ^1P^\circ_1$ resonance level of neutral barium

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From spectra of a very weak barium hollow-cathode discharge obtained with a Fourier-transform spectrometer, the branching ratio between the visible resonance line at 553.5 nm and the infrared line at 1500 nm of the barium atom was determined to be 485±40. Given the known lifetime and branching fractions between the infrared lines originating in the resonance level, it is now possible to present definitive branching fractions and transition probabilities for all transitions out of the $6s6p\ ^1P^\circ_1$ barium resonance level.

I. INTRODUCTION

The branching ratio between the resonance transition $6s^2\ ^1S_0-6s6p\ ^1P^\circ_1$ of Ba I at 553.5 nm and the infrared $6s5d\ ^1D_2, ^3D_2, ^3D_1-6s6p\ ^1P^\circ_1$ transitions at 1.50, 1.13, and 1.11 μm, respectively, determines the efficiency of optical pumping of the Ba I resonance line. The infrared decay channels, which lead to metastable levels, effectively remove the atoms from the pumping process, since they considerably delay the return of the atoms to the ground state. This branching ratio is therefore of specific interest for Ba-isotope separation by deflection through laser irradiation[1] and more generally for all applications involving optical pumping of barium via the resonance line.

Numerous attempts have been made to measure and calculate the relative photon flux between the spontaneous emission in the visible (535.5 nm) and the three infrared transitions (1.50, 1.13, and 1.11 μm). The ratio obtained from laser fluorescence experiments ranged between 50 and 550,[1-7] with a recommended weighted average of 290±40.[7] This value was also supported by data obtained from modeling the observed behavior of a resonantly pumped dense Ba vapor.[8] The ratios resulting from the two relevant calculations[9,10] are 380 and 740, respectively.

Recently, Niggli and Huber[11] have measured the relative photon flux between the three infrared lines, but because the resonance line was optically thick in their light source, they could not obtain a direct measurement of the branching ratio between resonance and infrared lines.

In this work we obtained a definitive experimental value for the relative photon flux between the resonance line at 553.5 nm and the infrared line at 1.5 μm, and consequently, by use of the data from Ref. 11, definitive and precise branching fractions. Given the lifetime of the $6s6p\ ^1P^\circ_1$ resonance level, definitive transition probabilities for all the lines originating from this level could now be determined.

II. EXPERIMENT

The branching ratio between the resonance line at 553.5 nm and the infrared line at 1.5 μm was obtained from two barium spectra recorded with the McMath Fourier-transform spectrometer (FTS) of the U.S. National Solar Observatory on Kitt Peak.[12] The light source was a hollow-cathode lamp, similar to the one described in Ref. 13, and was run with a flow of neon at a pressure of 2.7 mbar. In order to obtain an optically thin resonance line, the current through the lamp was set at 86 mA. Because of the widely differing wavelengths of the two lines of interest, reflective optics were used throughout and the two outputs of the FTS (Ref. 12) were monitored by different detectors: a midrange diode working in the visible region and an InSb detector (that was cooled with liquid nitrogen) in the infrared region.

The signal-to-noise ratio was kept high by restricting the photon flux to the spectral windows in the regions of interest by use of two 10-nm-bandwidth filters, Corion 5600 and Corion 1.5 for the visible and infrared regions, respectively. A scan with a path-length difference of ±30 cm was chosen to obtain a spectral resolution of $\sim 0.03\ \mathrm{cm}^{-1}$. The relative radiometric calibration of the spectrometer was obtained by replacing the hollow-cathode lamp by an Optronics Standard lamp operated at a current of 15 A.

III. RESULTS AND DISCUSSION

In Fig. 1 are shown the resonance line ($6s^2\ ^1S_0-6s6p\ ^1P^\circ_1$) at 18 060.266 cm^{-1} (553.5 nm) and the infrared line ($6s5d\ ^1D_2-6s6p\ ^1P^\circ_1$) at 6664.913 cm^{-1} (1499.98 nm).

The relative photon flux was obtained through a nonlinear least-squares fitting procedure that used Voigt profiles (DECOMP line-decomposition program, supplied by J. W. Brault of the National Solar Observatory, Tucson, AZ). The ringing seen in Fig. 1, which indicates that the resolution of the Fourier-transform spectrometer was not quite adequate to resolve the line profile, was removed by use of a (mathematical) filter.

The resulting branching ratio

$$\frac{A(553.5\ \mathrm{nm})}{A(1.50\ \mu\mathrm{m})}=485\pm 40$$

was found by averaging the values obtained in two independent measurements. The uncertainty value reflects

TABLE I. Branching fractions.

Upper level	Lower level	Wave number (cm^{-1})	Wavelength (nm)	Branching[a,b] fractions
$6s6p\ ^1P^\circ_1$	$6s^2\ ^1S_0$	18 060.27	553.5	0.9970 (0.03)
	$6s5d\ ^3D_1$	9026.3[c]	1107.6	2.6×10^{-5} (15)
	$6s5d\ ^3D_2$	8844.8[c]	1130.3	0.0009 (15)
	$6s5d\ ^1D_2$	6664.9	1500.0	0.002 06 (8)

[a]Based on our measurements and on the branching ratios between the infrared lines from Ref. 11.
[b]Values in parentheses indicate uncertainties in percent.
[c]Reference 11.

the uncertainty of the calibration of the standard lamp and the uncertainties in the determinations of the relative photon flux.

Strictly speaking, the value of 485 is only a lower limit, because we did not directly demonstrate that the resonance line was optically thin. However, a comparison with the radiance of the Optronics standard lamp yields a brightness temperature of 3100 K for the peak of the resonance line. An excitation temperature exceeding 4000 K would thus lead to optically thin conditions. Because the electron temperature of a hollow-cathode discharge at the pressure and current density of our source is of the order of 1 eV (11 400 K),[14] we may assume an excitation temperature which made the resonance line optically thin.

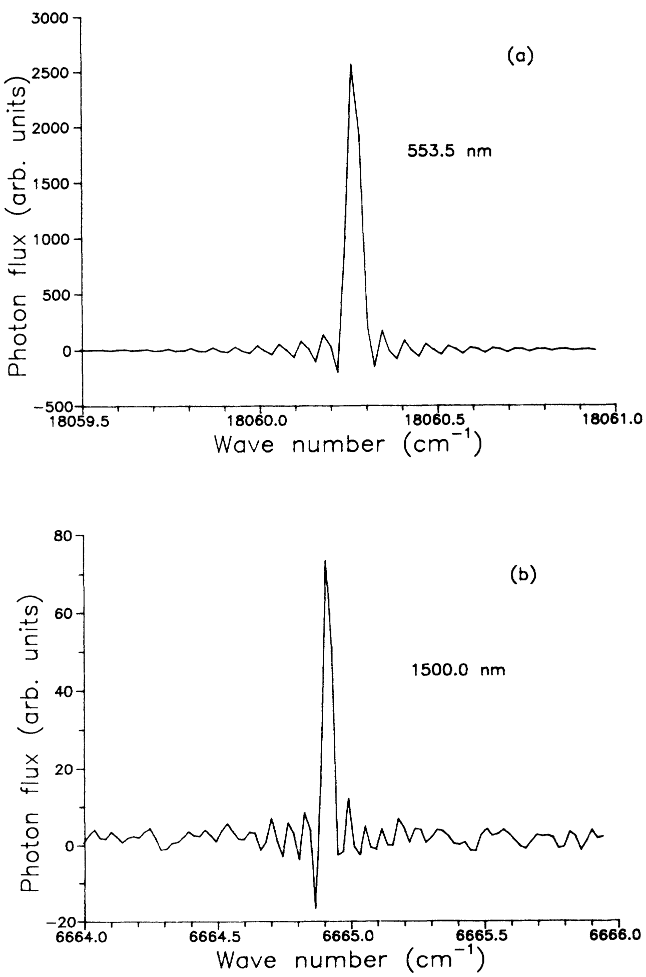

FIG. 1. (a) Line profiles of the resonance $6s^2\ ^1S_0$–$6s6p\ ^1P^\circ_1$ and (b) of the infrared $6s5d\ ^1D_2$–$6s6p\ ^1P^\circ_1$ transitions of Ba I, produced in a hollow-cathode lamp operated at a current of 86 mA with a neon carrier-gas pressure of 2.7 mbar.

Combining the above result with the branching ratios for the infrared lines obtained earlier by Niggli and Huber,[11]

$$\frac{A(1.50\ \mu m)}{A(1.13\ \mu m)}=2.3\pm0.3\ ,$$

$$\frac{A(1.50\ \mu m)}{A(1.11\ \mu m)}=80\pm10\ ,$$

we get

$$\frac{A(553.5\ nm)}{A(1.50\ \mu m)+A(1.13\ \mu m)+A(1.11\ \mu m)}=334\pm30\ .$$

This result is in good agreement with the value 300±45 that was obtained by Trajmar, Nickel, and Antoni[4] from a comparison between model calculations and scattering measurements of laser-excited Ba atoms. As expected, then, it is also in agreement with the "most probable value," 290±40, derived by Niggli and Huber[7] from the mean between Trajmar's result and the value 280±30 obtained by Lewis *et al.*[5] from measurements of the resonance fluorescence in an atomic beam. The value

TABLE II. Transition probabilities.

Wavelength (nm)	This work (s^{-1})[a]	Ref. 11 (s^{-1})[a]
553.5	1.19±0.01[8]	1.19±0.01[8]
1107.6	3.1±0.5[3]	3.6±0.5[3]
1130.3	1.1±0.2[5]	1.2±0.2[5]
1500.0	2.5±0.2[5]	2.8±0.4[5]

[a]The values listed are to be multiplied by powers of 10 with the exponents listed in brackets.

440±40, resulting from a pump-probe experiment,[6] may therefore now be excluded.

In Tables I and II we list the branching fractions and transition probabilities for all the lines originating from the $6s6p\ ^1P_1^\circ$ resonance level and compare these to one set of previous results. (An extensive listing of literature data was given in Ref. 11.) To convert branching fractions into transition probabilities (cf., e.g., Ref. 15), we used the lifetime of the resonance level, 8.37±0.08 ns, that had been determined by Kelly and Mathur from Hanle-effect measurements.[16]

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