

Atomic transition probability ratios between some Ar I 4s-4p and 4s-5p transitions

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All recent determinations of the atomic transition probabilities for the prominent Ar I 4s-4p lines in the red part of the visible spectrum have been carried out separately from determinations of the equally prominent 4s-5p lines in the blue part of the spectrum. A cross connection of these data is thus desirable to establish their mutual consistency. We have measured several transition probability ratios for 4s-4p to 4s-5p transitions and find very close consistency.

INTRODUCTION

The optical spectrum of neutral argon is dominated by two groups of 30 transitions each, the 4s-4p and 4s-5p transitions, which give rise to the prominent "red" and "blue" lines. A unified set of atomic transition probabilities of these and other argon transitions has been recently proposed¹ by combining the results of a new experimental approach¹ with critically evaluated recent literature data.²⁻¹⁰ The assembled set of transition probabilities was estimated to be accurate to $\pm 8\%$ for the strong lines and $\pm 15\%$ for the weak lines.

It is important to note that the data situation for the 4s-5p transitions differs appreciably from that for the 4s-4p transitions. For the 4s-4p transitions, three independent approaches have provided closely agreeing results:¹ (a) *absolute emission measurements*, carried out with wall-stabilized arcs; (b) *lifetime measurements* of the 4p states, in conjunction with branching-ratio measurements (electric dipole emission is possible into the 4s states only); and (c) *atomic structure theory*, specifically self-consistent-field (or Hartree-Fock) calculations.

On the other hand, for the 4s-5p transitions the situation is not as favorable because only one of the three approaches, the emission method, may be applied in the same straightforward manner. The theoretical methods suffer from appreciable cancellation of positive and negative contributions in the transition integrals,¹¹ which produce very uncertain results, and the lifetime measurements are difficult to interpret because the spontaneous radiative decay possibilities from the 5p levels are numerous because of various lower-lying 4s, 3d, and 5s levels. Up to now, only one lifetime result for a 5p level has been successfully combined with branching-ratio measurements for transition probability determinations.¹ To do this, a total of 15 branching ratios—involving mostly infrared lines—had to be determined.

Therefore, since the data for the 4s-4p transition array are on a much firmer and broader foundation than the 4s-5p data, it seems to be important to ascertain that the transition probability data that have been independently proposed for these two transition arrays are mutually consistent. For this purpose, we have measured the transition probability *ratios* between two 4s-4p and two 4s-5p transitions and compared them with the ratios derived

from the previously recommended data. To our knowledge, a study that specifically cross-connects the two arrays has not been carried out before.

EXPERIMENTAL METHOD

We have used the same laboratory setup that we had applied earlier in several emission spectroscopy experiments to determine the atomic transition probabilities of numerous C I, N I, and Ar I lines, and which we have described in considerable detail.^{8,12,13} We thus summarize only the instrumentation and techniques that are pertinent to the present experiment.

The emission source was a wall-stabilized arc with a central channel of 4 mm diameter and of about 80 mm length, and was operated at currents of 40 A for periods of several hours. The observations were made end on, but only the line radiation from the midsection of the arc was utilized. To accomplish this, the midsection was run in argon gas, while both end sections and electrode areas were operated in either nitrogen or helium. The separation of the gases was achieved by adjusting the gas flows and by appropriate positioning of the gas exits into the arc chamber.

The arc image was focused by a spherical mirror, enlarged by a factor of 2, on the entrance slit of a 2.25-m Czerny-Turner monochromator with a 1200-line/mm holographic grating, yielding a dispersion of 3.6 Å/mm. Both entrance and exit slits were set at 20 μm , yielding an instrument width of 0.007 nm. A photomultiplier tube with a broadband GaAs photocathode served as the detector. The photomultiplier was cooled to 243 K to reduce the dark current and improve the stability.

The Ar I lines were scanned in a stepwise manner, using typically about 50–60 data points across each line profile and sampling each point 500 times at 0.01-s intervals. A data-acquisition computer controlled the scanning sequence, recorded the photoelectric signals from the wall-stabilized arc and from a calibrated strip lamp, and monitored—and subsequently subtracted—the dark current.

The spectrometer-detector system was calibrated with a tungsten strip lamp radiation standard provided by the Radiometric Physics Division at the National Institute of Standards and Technology. A filter that completely blocked the radiation for wavelengths shorter than 590

TABLE I. Transition probability ratios between some Ar I 4s-4p and 4s-5p transitions.

Line Pair	Wavelengths (nm)	Transition probability ratios $A(4s-4p)/A(4s-5p)$	
		This experiment	Recommended data (Ref. 1)
4s[3/2] ₂ ⁰ -4p'[3/2] ₁ , 4s'[1/2] ₁ ⁻ -5p[1/2] ₀	714.7, 451.1	0.526	0.530
4s[3/2] ₂ ⁰ -4p'[3/2] ₁ , 4s'[3/2] ₂ ⁰ -5p'[5/2] ₂	714.7, 430.0	1.62	1.66
4s[3/2] ₁ ⁰ -4p'[1/2] ₀ , 4s'[1/2] ₁ ⁰ -5p[1/2] ₀	667.7, 451.1	0.200	0.200
4s[3/2] ₁ ⁰ -4p'[1/2] ₀ , 4s[3/2] ₂ ⁰ -5p'[5/2] ₂	667.7, 430.0	0.635	0.626

nm was employed to eliminate second-order radiation for the calibration of the long-wavelength lines.

Self-absorption for each line was checked by placing a concave mirror behind the arc source, which focused the arc radiation back onto itself. At all wavelength positions across the investigated line profiles, ratios were measured between (a) signals from the arc plus its image and (b) signals from the arc only, which was accomplished by blocking off the mirror with a mechanical shutter. For the 714.7-nm line these ratios decreased near the line center, indicating some self-absorption. By comparing the shapes of the profiles with and without the reflected radiation, an overall self-absorption correction of about 10% for this line was found for all experimental runs. No detectable self-absorption was found for the other lines.

Ratios of transition probabilities A between the 4s-4p (red) lines and the 4s-5p (blue) lines are obtained from the following relation, valid for a plasma in (partial or complete) local thermodynamic equilibrium (LTE):

$$\frac{A_r}{A_b} = \frac{I_r}{I_b} \frac{\lambda_r}{\lambda_b} \frac{g_b}{g_r} \exp \left[\frac{E_r - E_b}{kT} \right]. \quad (1)$$

$I_{r,b}$ are the total radiances of the red and blue lines, λ their wavelengths, and $g (=2J+1)$ the statistical weights of the respective upper levels (where J is the total angular momentum quantum number). E is the excitation energy of the upper atomic level and T the temperature.

Thus, for the determination of the transition probability ratios, the plasma temperature needs to be measured, in addition to the line radiance measurements. For the investigated line pairs, we obtain for our conditions $E_r - E_b \approx kT$, i.e., the exponent is close to unity. Therefore, the temperature measurement is not critically sensitive. In order to obtain the temperature, the plasma electron density is first determined from a measurement of the Stark width of the hydrogen H_β line. The Stark broadening theory of Vidal, Cooper, and Smith¹⁴—which, in the density range of interest, is closely confirmed by recent experiments—relates the Stark

width to the electron density. The temperature is then determined via the equilibrium and conservation equations for plasmas in local thermodynamic equilibrium,¹⁵ which are the Saha ionization equilibrium equation, the condition of local electrical charge neutrality, and the equation of state (Dalton's law). The equilibrium criteria as well as recent experimental studies indicate that, for the conditions of this experiment, complete LTE indeed exists,¹⁰ and these studies also show that for beginning deviations from LTE the "excitation" temperature hardly changes (while the "gas" temperature is more sensitively affected). Furthermore, since the excitation energies of the 4p and 5p levels only differ by about 1 eV ($E_r - E_b \approx kT$ for this experiment) even a systematic temperature uncertainty of a few percent—a very unlikely case—will only affect the transition probability ratios by the same amount.

RESULTS AND DISCUSSION

We have repeatedly measured the absolute radiances of two 4s-4p transitions at 667.7 and 714.7 nm as well as of two 4s-5p transitions at 430.0 and 451.1 nm. The wall-stabilized arc was always run at the same conditions, i.e., at a current of 40 A, resulting in an electron density of $N_e = 6.6 \times 10^{16} \text{ cm}^{-3}$ and a temperature of 12 010 K. We have thus determined four ratios of transition probabilities for these lines according to Eq. (1) and present them in Table I. Also listed are the ratios obtained from the recommended sets of transition probabilities of Ref. 1. It is observed that the agreement is extremely close—always within 2.5%—for all four ratios. This is well within our estimated uncertainty of $\pm 6\%$, which includes random measurement errors as well as systematic uncertainties in the radiance calibration and temperature measurement. Thus our cross-connecting line-ratio measurements fully confirm the consistency of the two independently established data sets for the 4s-4p and 4s-5p arrays of Ref. 1. The line pair at 714.7 and 430.0 nm has been measured by Nubbemeyer,¹⁶ and his result of 1.66 for this ratio is also in very close agreement with our data.

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