Atomic transition-probability measurements for prominent spectral lines of neutral nitrogen

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Relative transition probabilities of 29 lines belonging to the $3s$-$3p$, $3s$-$4p$, $3p$-$3s''$, and $2s2p^4$-$2s^22p^24p$ transition arrays of N I have been measured in emission with a wall-stabilized arc and were normalized to an absolute scale utilizing two recent lifetime results. Our experiment was undertaken to help resolve some long-standing discrepancies in the nitrogen data. Our results are usually slightly larger than two earlier emission measurements but are in closer agreement with recent calculations. The total uncertainties of our individual transition-probability values are estimated not to exceed a range from ±12 to ±15%, the larger uncertainties occurring for the very weak lines.

I. INTRODUCTION

The determination of atomic transition probabilities for lines in the optical spectrum of nitrogen, the most common element in the Earth's atmosphere, has been the objective of numerous experimental and theoretical studies published over the course of the past 30 years. Experimentally, the majority of the data were obtained from emission experiments with stabilized electric arcs.[1–4] The lifetimes of several pertinent energy levels have also been measured with various techniques.[5–9] Theoretical data are available from the Coulomb approximation[10] and approximate self-consistent-field calculations.[11] Two recent calculations include configuration-interaction treatments,[12,13] one of them on an extensive scale.[13] Our measurements were undertaken mainly as a contribution to help resolve some differences between previously published values. For this experiment, we have utilized lifetime data to establish our absolute scale and we estimate that our overall uncertainties for the stronger lines do not exceed ±12%. We have achieved this accuracy—aside from the just noted availability of good lifetime results—(a) by utilizing a well-defined emission source, (b) by using least-squares fitting techniques to obtain line profiles and to separate the profiles of partly overlapping lines, (c) by including line-wing intensity contributions, and (d) by applying an improved technique to diagnose the conditions of the emission source.

II. EXPERIMENTAL METHOD

Since the experimental setup and measurement techniques have been previously described in detail,[14] only a summary description of the major experimental components is given.

(a) Plasma source. The emission source used in these measurements was a wall-stabilized arc with a channel of 4-mm diameter and of 75-mm length, operating at currents between 35 and 50 A. For the end-on observation of N I lines, the midsection of the arc was operated in a flow of nitrogen gas with a small admixture of hydrogen (about 1–2 % by volume) for H_β Stark-width diagnostics. For the very strong $3s$-$3p$ quartet transitions in the red and near infrared, the nitrogen gas was strongly diluted with argon, up to a ratio of about 50:1, in order to eliminate significant self-absorption. The end sections of the arc, including the electrode areas, were operated in argon to minimize thermal gradients along the axial direction in the nitrogen plasma which would affect the N I line measurements.

(b) Spectrometer. A 2.25 m Czerny-Turner monochromator with a 1200-line/mm holographic grating was used that has a reciprocal linear dispersion of 3.6 Å/mm. The entrance and exit slits were set at 22.5 μm, while the entrance slit was masked to a height of 0.6 mm. A cooled photomultiplier tube with a GaAs photocathode was utilized as the detector. A filter, which completely blocked wavelengths shorter than 6000 Å, was used to eliminate second-order radiation for the long-wavelength lines. Spectra were scanned in a stepwise manner controlled by the data-acquisition computer.

(c) Data acquisition. Up to 60 data points were accumulated for each line profile by a computerized data-acquisition system. This system sampled the photoelectric signal 200 (or 500) times at 0.01 s intervals for each data point. The dark current was routinely monitored to correct for any variations during the stepwise scanning of a line profile.

(d) Spectral radiance calibration. The spectrometer-detector system was calibrated against a tungsten strip lamp standard calibrated by the Radiometric Physics Division at the National Institute of Standards and Technology. For this experiment, where the absolute scale is obtained from lifetime data, only the variation in spectral sensitivity over the investigated range of wavelengths had to be determined.

(e) Self-absorption correction. A concave mirror behind the arc source focused the arc radiation back on to itself. Ratios were obtained between (a) signals from the arc plus its image, and (b) signals from the arc only (utilizing a mechanical shutter in front of the mirror). These ratios were measured at all wavelengths across investigated line profiles. When the ratio became smaller near the line center, indicating self-absorption, a pointwise self-

absorption correction was carried out according to procedures discussed elsewhere.[14] This correction was only undertaken when the self-absorption corrections were fairly small. For cases of stronger self-absorption, the gas dilution procedure discussed in Sec. II A was applied until self-absorption became small.

(f) Plasma diagnostics. Measurements of the width of the hydrogen H_β line yielded the electron density via the Stark broadening theory of Vidal, Cooper, and Smith[15] which is in the density range of interest closely confirmed by recent experiments.[16,17] The temperature was then determined via the equilibrium and conservation equations for plasmas in local thermodynamic equilibrium (LTE).[18]

(g) Analysis procedure. Our objective was the determination of accurate transition probabilities for individual lines of N I multiplets which appear typically as partially overlapping lines at the electron density of the arc source. The method we employed for this purpose makes use of a least-squares fitting technique applied to the measured spectra to obtain analytical "synthetic" spectra.[19] The synthetic spectrum consists of Lorentzian profiles, including the complete line wings, for each line observed in the corresponding data scan and a cubic polynomial background function. The sum of these components provides an analytical composite curve for each spectrum which was made to match the actual data points of a spectral scan by least-squares fitting. Slight asymmetries of the line shapes due to ion broadening[20] were neglected.

(h) Relative transition probabilities. During each experimental run the composition and temperature of the plasma were held constant by precise current and gas flow controls while various multiplets of the neutral nitrogen spectrum were scanned. The spectral line at 746.831 nm was measured in every run and served as the reference line for the relative transition probabilities reported here. Relative transition-probability results for each run were then computed from the total radiance of each line from the relation[21]

$$\frac{A_x}{A_r}=\frac{I_x}{I_R}\frac{\lambda_x}{\lambda_R}\frac{g_R}{g_x}\exp\left[\frac{E_x-E_R}{kT}\right], \tag{1}$$

where the subscripts x and R denote the values associated with the line under consideration and the reference line, respectively. A represents the transition probability, I the total line radiance, λ the wavelength, E the excitation energy of the upper level, g its statistical weight, k the Boltzmann constant, and T the temperature. In separate runs with varying plasma conditions [electron density range, $(6.4-9.3)\times10^{16}$ cm^{-3}; temperature range, 11 840–12 650 K], four to six independent measurements of relative transition probabilities for each line were obtained and then combined in a final average. The uncertainties associated with the relative transition probabilities were estimated to be in the range from 6–12 %. The largest contribution to the uncertainty comes usually from the statistical scatter in the intensity ratio data between different scans within an experimental run and between different runs of the experiment and is typically ±4% for the strong lines and up to ±10% for very weak lines. (We define our measurement errors as two standard deviations of the average values.) Other contributions that we include are random and systematic uncertainties in the temperature determination (±2%), uncertainties in the spectral calibration procedure (±0.5% to 2%), and uncertainties in the self-absorption corrections (up to ±4%).

III. RESULTS AND DISCUSSION

A. Determination of an absolute scale via available lifetime data

The available lifetime measurements[5–9] for the relevant levels of N I are compiled in Table I. The most accurate experiments are based on the laser-induced fluorescence technique,[5,6,9] since it is state selective and very sensitive. For the $3p\ ^4D^\circ_{7/2}$ level, a lifetime of 27 ns was first measured by Bischel, Perry, and Crosley[8] and later revised to 34±12 ns by Copeland *et al.*[9] of the same group, who performed a new analysis of the results. Copeland *et al.* also undertook new measurements of this as well as the other $3p\ ^4D^\circ$ levels (with total angular momentum quantum numbers $J=\frac{1}{2}$, $\frac{3}{2}$, and $\frac{5}{2}$), using improved instrumentation. They found a common value of 43±3 ns, independent of the value of J. As indicated by their greatly improved accuracy, they consider the new results much more reliable than the older, revised value

TABLE I. Lifetime measurements for various $3p$ levels of N I.

Authors	Method	Reference number	Published year	Level	Lifetime τ
Desesquelles	Beam Foil	7	1970	$3p\ ^4P^\circ_{5/2}$	39
Bromander *et al.*	High-frequency deflection	6	1978	$3p\ ^4P^\circ_{5/2}$	55±5
Catherinot and Sy	Laser-induced fluorescence	5	1979	$3p\ ^4S^\circ_{3/2}$	23.3±2.3
Bischel *et al.*[a]	Laser-induced fluorescence	8	1981	$3p\ ^4D^\circ_{7/2}$	34±12
Copeland *et al.*[b]	Laser-induced fluorescence	9	1987	$3p\ ^4D^\circ$	43±3

[a]Lifetime as revised by Copeland *et al.*, Ref. 9.
[b]Same lifetime measured for all different levels of the $3p\ ^4D^\circ$ term.

TABLE II. Decay rates, in 10^8 s^{-1}, for the $3p\ ^4D°$ and $3p\ ^4S°$ levels of N I. The relative results of this experiment are normalized with a common normalization constant to achieve the best fit with the lifetime data.

Level (1)	Total decay rate (direct lifetime measurements) (2)	Partial decay rates into $3s\ ^4P$ This experiment best fit (3)	Partial decay rates into $2s2p^4\ ^4P$ [Theory (Ref. 13)] (4)	Total decay rate from this experiment and theory, sum of columns (3) and (4) (5)
$3p\ ^4D°$	0.233[a]	0.241[c]	0.010	0.251
$3p\ ^4S°$	0.429[b]	0.367	0.032	0.399

[a]Copeland *et al.*, Ref. 9.
[b]Catherinot and Sy, Ref. 5.
[c]For the $^3p\ ^4D_{1/2}$ level.

of Bischel, Perry, and Crosley.[8] We therefore apply only the new result by Copeland *et al.*[9] for establishing our absolute scale. In addition, we utilize another accurate lifetime measurement, carried out by Catherinot and Sy[5] for the $3p\ ^4S°$ level, again with the laser-induced fluorescence technique. On the other hand, we did not apply two lifetime results for the $3p\ ^4P°_{5/2}$ level, which were obtained with older techniques[6,7] and differed appreciably

TABLE III. N I absolute transition probabilities.

Transition	Wavelength (Å)	Transition probability A (10^8 s^{-1}) Emission experiments This work	Baronnet *et al.*[a]	Richter[b]	Theory Suskin and Weiss,[c] *LS* coupling
$3s\ ^4P$-$3p\ ^4D°$	8747.36	0.0107±13%	0.0072	0.0065	0.0134
	8728.91	0.040±13%	0.029	0.028	0.0449
	8718.84	0.066±12%	0.053	0.052	0.0811
	8711.71	0.126±12%	0.111	0.099	0.145
	8703.26	0.201±12%	0.200	0.181	0.227
$3s\ ^2P$-$3p\ ^2P°$	8655.87	0.110±12%	0.089	0.104	
	8629.24	0.269±12%	0.250	0.236	
	8594.01	0.231±12%	0.260	0.184	
	8567.74	0.052±13%	0.074	0.042	
$3s\ ^4P$-$3p\ ^4P°$	8242.37	0.135±12%	0.102	0.121	0.146
	8223.12	0.254±12%	0.214	0.249	0.273
	8216.32	0.204±12%	0.176	0.187	0.230
	8210.71	0.052±13%	0.048	0.050	0.0438
	8200.36	0.050±13%	0.047	0.040	0.0549
	8188.01	0.128±12%	0.125	0.102	0.138
	8184.85	0.085±12%	0.082	0.0670	0.0995
$3s\ ^4P$-$3p\ ^4S°$	7468.31	0.185±12%	0.159	0.157	0.203
	7442.30	0.120±12%	0.102	0.100	0.134
	7423.64	0.062±13%	0.050	0.048	0.0690
$3p\ ^2P°$-$3s''\ ^2S$	5411.88	0.0086±15%	0.0089	0.0075	
	5401.45	0.0043±15%	0.0045	0.0034	
$3s\ ^2P$-$4p\ ^2S°$	4935.03	0.0187±14%	0.0133	0.0161	
	4914.90	0.0086±14%	0.0060	0.00627	
$2s2p\ ^4\ ^4P$-$4p\ ^4D°$	5356.77	0.00187±15%			
	5328.70	0.00226±15%	0.0020		
$2s2p\ ^4\ ^4P$-$4p\ ^4P°$	5281.18	0.00245±15%	0.0023		
$3s\ ^4P$-$4p\ ^4S°$	4151.46	0.0101±14%	0.0091		
	4143.42	0.0061±15%	0.0051		
	4137.63	0.0028±15%	0.0024		

[a]Reference 22.
[b]Reference 2.
[c]Reference 13.

from each other (see also our comment at the end of Sec. III B).

In order to normalize our relative emission data, which are all on the same scale, we utilize the basic relation between atomic lifetimes τ and transition probabilities A:

$$\tau_k = \left[\sum_i A_{ki} \right]^{-1} , \tag{2}$$

where k denotes the atomic level of interest and i denotes all lower atomic levels to which radiative transitions occur. For the two levels of interest $3p\ ^4D^\circ$ and $3p\ ^4S^\circ$, the sums consist in each case of only two transitions ($i=1,2$). The principal (strong) transitions in each case $3s\ ^4P-3p\ ^4D^\circ$ and $3s\ ^4P-3p\ ^4S$ are those we have measured on the same relative scale. For the other transitions, $2s2p\ ^4\ ^4P-3p\ ^4D^\circ$ and $2s2p\ ^4\ ^4P-3p\ ^4S^\circ$, transition probabilities are available from theory on an absolute basis.[13] Thus we have two relations of the type

$$\tau_k^{-1} = c A_{k1}^{\text{rel}} + A_{k2} , \tag{3}$$

where c is the common normalization constant, A_{k1}^{rel} our relative emission A value and A_{k2} is the absolute A value available from theory. With the two known lifetimes we could determine c twice and have found the two normalization constants to agree within 14%. We have chosen the arithmetic mean for the conversion of all our relative values to the absolute scale.

In Table II we have assembled the relevant numerical data. We show the directly measured total decay rates (inverse lifetimes) for the two levels in column 1, and the partial decay rates resulting from our best-fit normalized measurements plus the theoretical contributions in columns 2 and 3. The two partial decay rates are then added up to total rates in column 4. It is seen that our measured transitions represent more than 90% of the total radiative decay rates. Since the normalization of our data to two independent lifetime results yielded close consistency, and since Copeland *et al.* estimate the uncertainty of their lifetime result to be ±7%, and Catherinot and Sy estimate ±10%, we cautiously estimate the

TABLE IV. Relative strengths of lines within multiplets. The strength of the weakest line in each multiplet is set equal to one.

Multiplet	Wavelength	J values	LS coupling	Relative line strength This work	Baronnet *et al.*[a]	Richter[b]
$3s\ ^4P$-$3p\ ^4D^\circ$	8747.36	$\frac{5}{2}$-$\frac{3}{2}$	1	1	1	1
	8728.91	$\frac{3}{2}$-$\frac{1}{2}$	1.67	1.87	1.99	2.1
	8718.84	$\frac{5}{2}$-$\frac{5}{2}$	9	9.2	10.9	11.9
	8711.71	$\frac{3}{2}$-$\frac{3}{2}$	10.7	11.6	15.2	15.4
	8703.26	$\frac{1}{2}$-$\frac{1}{2}$	8.3	9.3	13.6	13.9
$3s\ ^2P$-$3p\ ^2P^\circ$	8655.87	$\frac{3}{2}$-$\frac{1}{2}$	1	1	1	1
	8629.24	$\frac{3}{2}$-$\frac{3}{2}$	5	4.84	5.66	4.55
	8594.01	$\frac{1}{2}$-$\frac{1}{2}$	2	2.06	2.88	1.77
	8567.74	$\frac{1}{2}$-$\frac{3}{2}$	1	0.92	1.63	0.80
$3s\ ^4P$-$3p\ ^4P^\circ$	8242.37	$\frac{5}{2}$-$\frac{3}{2}$	5.4	5.48	4.42	6.08
	8223.12	$\frac{3}{2}$-$\frac{1}{2}$	5	5.11	4.60	6.24
	8216.32	$\frac{5}{2}$-$\frac{5}{2}$	12.6	12.3	11.3	14.0
	8210.71	$\frac{3}{2}$-$\frac{3}{2}$	1.6	2.09	2.05	2.52
	8200.36	$\frac{1}{2}$-$\frac{1}{2}$	1	1	1	1
	8188.01	$\frac{1}{2}$-$\frac{3}{2}$	5	5.11	5.31	5.12
	8184.85	$\frac{3}{2}$-$\frac{5}{2}$	5.4	5.07	5.24	5.04
$3s\ ^4P$-$3p\ ^4S^\circ$	7468.31	$\frac{5}{2}$-$\frac{3}{2}$	3	3.03	3.22	3.30
	7442.30	$\frac{3}{2}$-$\frac{3}{2}$	2	1.95	2.04	2.10
	7423.64	$\frac{1}{2}$-$\frac{3}{2}$	1	1	1	1
$3p\ ^2P^\circ$-$3s''\ ^2S$	5411.08	$\frac{3}{2}$-$\frac{1}{2}$	2	2.02	2.0	2.23
	5401.45	$\frac{1}{2}$-$\frac{1}{2}$	1	1	1	1
$3s\ ^2P$-$4p\ ^2S^\circ$	4935.03	$\frac{3}{2}$-$\frac{1}{2}$	2	2.20	2.25	2.57
	4914.90	$\frac{1}{2}$-$\frac{1}{2}$	1	1	1	1
$2s2p\ ^4\ ^4P$-$4p\ ^4D^\circ$	5356.77	$\frac{3}{2}$-$\frac{5}{2}$	1	1		
	5328.70	$\frac{5}{2}$-$\frac{7}{2}$	1.90	1.59		
$3s\ ^4P$-$4p\ ^4S^\circ$	4151.46	$\frac{5}{2}$-$\frac{3}{2}$	3	3.65	3.93	
	4143.42	$\frac{3}{2}$-$\frac{3}{2}$	2	2.19	2.13	
	4137.63	$\frac{1}{2}$-$\frac{3}{2}$	1	1	1	

[a]Reference 22.
[b]Reference 2.

TABLE V. Relative multiplet strengths of the $3s$-$3p$ quartets.

Multiplet	J file sum rule	This work	Baronnet *et al.*[a]	Richter[b]	Suskin[c] and Weiss
$3s\ ^4P$-$3p\ ^4S^\circ$	1	1	1	1	1
$3s\ ^4P$-$3p\ ^4P^\circ$	3	3.29	3.33	3.50	3.22
$3s\ ^4P$-$3p\ ^4D^\circ$	5	5.07	5.80	4.86	5.29

[a]Reference 22.
[b]Reference 2.
[c]Reference 13.

uncertainty of our thus established absolute scale to be no larger than ±10%.

B. Results and comparison of data

Table III contains all our results on an absolute basis, as well as comparisons with existing experimental and theoretical data. The other measurements for these lines were carried out with a wall-stabilized arc source similar to ours, by Richter,[2] and with a plasma jet, by Baronnet *et al.*[22] In these two experiments, the absolute scale was obtained from absolute line intensity and particle density measurements assuming local thermodynamic equilibrium—a method fundamentally different from our relative-emission approach, where the absolute scale is based on lifetime data. For the measured electron densities, the LTE assumption is valid according to an experimental study by Shumaker[4] and theoretical LTE validity criteria.[10] However, in these two earlier experiments no mention is made of the inclusion of the extended line wings in the line intensity measurements, so that it must be assumed that this problem was not addressed. The intensities of the extended line wings are of critical importance in their work, since neglecting then makes the transition probability data appear to be too small by about 15–20%. Indeed, the data of Richter[2] and Baronnet *et al.*[22] are often lower than ours by such amounts. (This problem is much less significant for measurements of line intensity *ratios*, as in our experiment, but we have nevertheless taken it into account by fitting the measured line shapes to Lorentzian profiles.)

Compared to the theoretical data of Suskin and Weiss,[13] our results are generally slightly smaller. This difference is entirely within the estimated experimental uncertainties, but similar uncertainties are also expected for the results of the multiconfiguration calculations performed by Suskin and Weiss.[13]

Our estimated overall errors given in Table III include both random measurement errors as well as estimates for random and systematic uncertainties resulting from the temperature measurement, the spectral radiance calibration, and self-absorption corrections. It is assumed that all errors are independent errors, so that the overall error is obtained as the square root of the sum of the squares of the individual errors.

In Table IV, we compare the *relative* line strengths for individual lines. The line strength S (in atomic units) is related to the atomic transition probability by $S=4.935\times10^{-19}g_k\lambda^3 A$, where $g_k=2J+1$ is the statistical weight of state k (J is the total angular momentum quantum number). We have tabulated the line strengths for the case of pure LS coupling[23] and compared them with the experimental data of Baronnet *et al.*,[22] Richter,[2] and the results of this work. The strength of the weakest line in each multiplet is set equal to 1. For most lines, our results adhere more closely to the LS-coupling ratios than the earlier measurements. A similar situation prevails for the multiplet line-strength data of the three $3s$-$3p$ quartets, which are given in Table V, together with the predictions of the J-film sum rule.

Using our data for the transition probabilities of the 8216.32 and 8184.54 Å lines (Table III), we can derive a lifetime τ for the $3p\ ^4P_{5/2}$ level. Our result $\tau=33$ ns includes the contribution of the rather weak $2s2p^4\ ^4P-3p\ ^4P_{5/2}$ transition, for which the theory[13] yields a decay rate of $0.010\times10^8\ s^{-1}$. This lifetime is in good agreement with the beam-foil lifetime measurement of Desesquelles,[7] who obtained $\tau=39$ ns, but differs appreciably from the lifetime result of Bromander *et al.*[6] of $\tau=55$ ns (see Table I).

Finally, we should note that the critical National Bureau of Standards (NBS) compilation[24] of nitrogen transition probabilities of 1966, with estimated uncertainties of ±25%, is fully consistent with our more accurate data. In the NBS compilation, mainly Richter's data were used; these values are often somewhat lower than ours, but the differences rarely exceed 20% (see Table III).

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