Transition probability ratios for selected multiplets of C I, N I, and O I, and comparisons with recent calculations

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With a wall-stabilized high-current arc, we have measured the transition probabilities of several multiplet pairs of C I, N I, and O I. Pairs have been selected for which two recent *ab initio* calculations have produced widely different results. All chosen multiplets are among the prominent, strong features of their respective spectra, and the pairing is in each case done for multiplets that hardly differ in their excitation energies. Our results do not favor either of the calculations, but are consistent with an earlier experiment.

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INTRODUCTION

During the last five years we have performed several emission spectroscopy experiments for a number of C I, N I, and O I transitions utilizing wall-stabilized high current arcs [1–3], and this paper describes some additional measurements. The principal purpose of these experiments was to determine atomic transition probability data for tests of recent multiconfiguration calculations, since these do not provide estimates of the accuracy of their result. The calculations were either carried out by members of the OPACITY Project (OP) team [4–6], who used an *R*-matrix code in conjunction with the close-coupling approximation, or by Hibbert *et al.* [7–9] who applied the CIV 3 configuration interaction code. The OP calculations yield multiplet values only, while the CIV 3 calculations provide transition probabilities for individual lines.

Comparisons between experiment and theory, as well as between the two types of calculations, revealed a surprising number of large differences in the data. To pinpoint their causes, it is desirable to check some of the earlier experimental results, using possibly a different approach. We again applied the standard emission spectroscopy technique with a wall-stabilized high-current arc, since this is the best light source to thermally excite these spectra, but we limited our measurements to the determination of transition probability ratios of selected multiplet pairs that obey special conditions discussed below.

EMISSION SOURCE

A high-current wall-stabilized arc with its central part operated in helium and with very small CO_2, N_2, or O_2 admixtures was used as the excitation source for the spectra of C I, N I, and O I. The arc was operated at currents from 35 to 70 A, but mainly at 40, 50, and 60 A. For these arc currents the effective plasma temperature was determined by the Boltzmann plot method based on O I line intensity measurements [3]. For side-on observations, effective temperature values of 11 300, 11 900, and 12 250 K were obtained at arc currents of 40, 50, and 60 A, respectively.

Measurements were mainly performed side on in order to avoid the continuum radiation from the part of the arc operated in argon, and to avoid interference with Ar I and Ar II lines. In the case of C I, end-on measurements were also carried out in order to avoid the background C_2 molecular radiation, which predominantly originates from the cooler boundary layers of the arc plasma. The intensities of spectral lines were varied by changing the amount of the admixture (N_2, O_2, or CO_2) to the helium gas flow, or by changing the arc current.

Self-absorption checks were performed by applying a technique described in Ref. [2]. During the time required for a set of relative intensity measurements, the intensity of one line was constantly monitored. If necessary, small corrections were applied to account for changes in admixture concentration as measured by variations in the monitored line intensity.

DATA ACQUISITION

The instrumentation used for measuring spectral line intensities, including the external optical system imaging the sources (arc, standard lamp) onto the entrance slit were previously described in detail [1,2]. The only difference is the use of a charge-coupled device (CCD) detector instead of the photomultiplier used in previous experiments. In order to check the linearity of response of the CCD detector, several spectra emitted from the tungsten strip lamp at various currents (light outputs) were taken with and without a gray filter (transmission 31%) in the light beam. These tests confirm the linearity of the CCD detector response from the signal level of the dark current (typically about 150–200 counts) to a level exceeding 40 000 counts.

SELECTION OF MULTIPLET PAIRS

As noted in the introduction, we have selected multiplet pairs fulfilling some special requirements.

(i) For one selected multiplet the agreement between two recent advanced configuration-interaction calculations is very good, with disagreements typically within 5%. These

TABLE I. Theoretical transition probability data A_{ki} for selected multiplet pairs. $\Delta E_k^{\mathrm{mult}}$ is the difference in excitation energy of the two multiplets.

Spectrum	Pair	Multiplet	λ_{Mult}	$\Delta E_k^{\mathrm{mult}}$ (cm^{-1})	A_{ki}^{mult} OP (in $10^8\ \mathrm{s}^{-1}$)	A_{ki}^{mult} CIV 3 (in $10^8\ \mathrm{s}^{-1}$)	Ratio of OP/ CIV 3
(1) Data for first member of a multiplet pair are in very good agreement:							
N I	(1*a*)	$3s'\ ^2D-3p'\ ^2F°$	9047.6		0.286	0.273	1.048
				−1597.95			
	(1*b*)	$3s'\ ^2D-3p'\ ^2P°$	7904.5		0.246	0.382	0.644
N I	(2*a*)	$3s'\ ^2D-3p'\ ^2F°$	9047.6		0.286	0.273	1.048
				177.93			
	(2*b*)	$3s'\ ^2D-3p'\ ^2D°$	9195.7		0.142	0.261	0.544
N I	(3*a*)	$3s\ ^2P-3p\ ^2D°$	9395.3		0.266	0.267	0.996
				82.66			
	(3*b*)	$3s\ ^4P-3p\ ^4S°$	7452.2		0.237	0.384	0.617
N I	(4*a*)	$3s\ ^2P-3p\ ^2P°$	8617.5		0.321	0.326	0.985
				2956.17			
	(4*b*)	$3s\ ^4P-3p\ ^4D°$	8691.6		0.183	0.259	0.707
O I	(1*a*)	$3p\ ^5P-4d\ ^5D°$	6157.3		0.0779	0.0744	1.047
				−42.86			
	(1*b*)	$3p\ ^3P-4d\ ^3D°$	7002.1		0.0332	0.0374	0.888
(2) Data disagree for both multiplets of a pair:							
C I	(1*a*)	$3s\ ^1P°-4p\ ^1S$	4932.05		0.0502	0.0602	0.834
				481.92			
	(1*b*)	$3s\ ^1P°-4p\ ^1D$	5052.17		0.0172	0.0260	0.662
N I	(5*a*)	$3s'\ ^2D-3p'\ ^2D°$	9195.7		0.142	0.261	0.544
				0.00			
	(5*b*)	$3s\ ^2P-3p'\ ^2D°$	4106.9		0.0232	0.0676	0.343

TABLE II. Ratios of multiplet transition probabilities.

Spectrum	Multiplet pair	This work	OP [4–6]	CIV 3 [7–9]	Expt. [2]
(1) Calculations agree for first multiplet of a pair:					
N I	$(3s'\ ^2D-3p'\ ^2F°)$ $/(3s'\ ^2D-3p'\ ^2P°)$	1.45±0.15	1.16	0.715	1.82±0.25
	$(3s'\ ^2D-3p'\ ^2F°)$ $/(3s'\ ^2D-3p'\ ^2D°)$	1.85±0.15	2.01	1.05	2.13±0.30
	$(3s\ ^2P-3p\ ^2D°)$ $/(3s\ ^4P-3p\ ^4S°)$	0.65±0.03	1.12	0.695	0.69±0.08
	$(3s\ ^2P-3p\ ^2P°)$ $/(3s\ ^4P-3p\ ^4D°)$	1.02±0.06	1.75	1.26	1.32±0.15
O I	$(3p\ ^5P-4d\ ^5D°)$ $/(3p\ ^3P-4d\ ^3D°)$	2.18±0.12	2.34	1.99	—
(2) Calculations disagree for both multiplets of a pair:					
C I	$(3s\ ^1P°-4p\ ^1S)$ $/(3s\ ^1P°-4p\ ^1D)$	2.38±0.10	2.92	2.32	—
N I	$(3s'\ ^2D-3p'\ ^2D°)$ $/(3s\ ^2P-3p'\ ^2D°)$	3.28±0.60	6.12	3.86	3.11±0.44

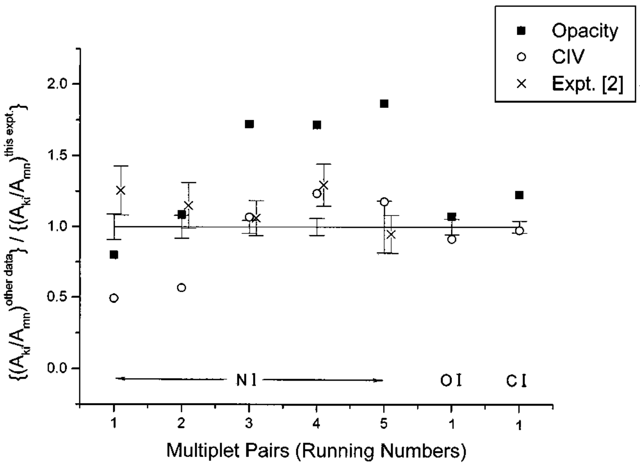

FIG. 1. Comparisons of measured and calculated multiplet *A*-value ratios with those determined in our experiment. The error bars along the line at unity represent the uncertainty estimates of this experiment.

calculations are either based on the CIV 3 configuration interaction code [7–9] or have been part of the OPACITY Project [4–6], in which essentially a multiconfiguration frozen-core approximation is used.

(ii) For the other multiplet the difference between the two theoretical results exceeds the typical uncertainties of multiplet ratio measurements which are based on relative line intensity (I_1, I_2) determinations $[\Delta(I_1/I_2) \leqslant 10\%]$.

(iii) Both multiplets of a pair originate from the same upper term E_k or from terms closely spaced in excitation energy, facilitating the conversion from line intensity ratios to transition probability ratios. Such intensity ratios are either independent of temperature or have only a weak temperature dependence.

(iv) Lines belonging to the multiplets are strong enough to be measured accurately and are isolated, i.e., they do not overlap with other spectral lines of the same element or lines of other plasma components.

(v) We also included a case in C I where the agreement between the calculations is only within 20% and a multiplet pair of N I where the two multiplet values show disagreements between the two calculations exceeding factors of 1.8.

Details about the selected multiplet pairs are listed in Table I.

RESULTS

We measured the multiplet ratios in at least 18 independent runs. Our uncertainties arise mainly from statistical and systematic errors in the line intensity measurements and the radiometric calibrations (for details, see Ref. [2]). The combined standard uncertainties are the root of the sum of the squares of the individual contributions. Table II lists our measured transition probability ratios, together with those from the above mentioned calculations [4–9], and a previous emission experiment by our group [2]. For consistency, we have converted the ''expanded'' uncertainties of Ref. [2] into standard uncertainties.

TABLE III. Transition probability data for reference multiplets (in $10^8\ s^{-1}$).

Multiplet	OP [4]	CIV 3[8]	Mean of [4, 8]	Expt. [2]
$3s'\ ^2D-3p'\ ^2F°$	0.286	0.273	0.280	—
$3s\ ^2P-3p\ ^2D°$	0.266	0.267	0.266	0.256
$3s\ ^2P-3p\ ^2P°$	0.321	0.326	0.323	0.297

In Fig. 1, we compare the *A*-value ratios obtained from OP, CIV 3, and another experiment (Ref. [2]) with those we have measured. Our data are not greatly different from the results of the earlier emission experiment for N I [2]. As Fig. 1 and Table II show, four of five multiplet ratios agree within the estimated uncertainties, and one is slightly outside. But both experiments show no clear pattern of preference for either one of the two calculations. For N I, where the discrepancies between the two calculations are especially large, each theoretical approach has only one good agreement with experiment, but four misses.

DATA ANALYSIS

A. Nitrogen

(I) From the measured set of eight transitions, we chose three A_{ki} multiplet values as reference data (see Table III), where the agreement between the OP and CIV 3 calculations is excellent, i.e., ±5% or better, the agreement with experiments (available for two of the multiplets) is also within 10%, and where in addition the ''scaling'' predicted by both calculations along the isoelectronic sequence (N I O II) agrees.

(II) Utilizing our measured transition probability ratios for the multiplets as given in Table II, and using the calculated mean values from Table III we have determined the A_{ki} values for the other N I multiplets and presented them in Table IV. For comparison, other results are listed, too.

Our measurements agree well with the OP value for the second multiplet, and with the CIV 3 value for the fourth multiplet. On the other hand, our measurements show large differences from the theoretical data for the other multiplets. But our data are in fair-to-good agreement with the earlier emission experiment [2]. All our N I transition probabilities are slightly higher than the other experiment, which is due to a different data normalization. In the 1995 experiment, the data were normalized against lifetime measurements, while

TABLE IV. Results for the transition probabilities (in $10^8\ s^{-1}$) for other N I multiplets from multiplet ratio measurements and comparisons with available data.

Multiplet	This work	OP [4]	CIV 3 [8]	Expt. [2]
$(3s'\ ^2D-3p'\ ^2P°)$	0.193	0.246	0.382	0.146
$(3s'\ ^2D-3p'\ ^2D°)$	0.151	0.142	0.261	0.124
$(3s\ ^2P-3p'\ ^2D°)$	0.046	0.0232	0.0676	0.040
$(3s\ ^4P-3p\ ^4S°)$	0.409	0.237	0.384	0.369
$(3s\ ^4P-3p\ ^4D°)$	0.317	0.183	0.259	0.225

our data are normalized against the most consistent theoretical results, given in Table III.

B. Carbon and oxygen

For each spectrum, we could find only one suitable pair of multiplets. For C I the numbers of Table II show that the CIV 3 results are in close agreement with our measurement, while the OP data are not. For O I our result is midway between the OP and CIV 3 data, but the differences with the calculational results are less than 10%.

SUMMARY

We have measured ratios of transition probabilities for seven multiplets of N I, C I, and O I, for which two recent calculations give divergent results. We placed considerable constraints on the selection of the multiplets by looking for pairs where both multiplets have large transition probabilities and either identical or nearly the same excitation energies. These restrictions permit accurate measurements, since only strong lines are involved, and the conditions of the emission source are not critical. Our results are generally consistent with an earlier experiment. The experimental data show no overall preference for either of the two advanced atomic structure codes.

In conclusion, it appears that the theoretical atomic structure codes are in need of further improvement, even for comparatively strong multiplet values. This finding is consistent with a recent experimental emission study for F I [10].

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