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TRANSITION PROBABILITIES FOR THE $3s^23p(^2P^o)$–$3s3p^2(^4P)$ INTERSYSTEM LINES OF Si II

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ABSTRACT

Intensity ratios of lines of the spin-changing "intersystem" multiplet of Si II ($^4P \rightarrow {^2P^o}$) at 234 nm have been used to determine electron densities and temperatures in a variety of astrophysical environments. However, the accuracy of these diagnostic calculations have been limited by uncertainties associated with the available atomic data. We report the first laboratory measurement, using an ion-trapping technique, of the radiative lifetimes of the three metastable levels of the $3s3p^2\,{^4P}$ term of Si II. Our results are 104 ± 16, 406 ± 33, and 811 ± 77 μs for lifetimes of the $J = 1/2$, 5/2, and 3/2 levels, respectively. A-values were derived from our lifetimes by use of measured branching fractions. Our A-values, which differ from calculated values by 30% or more, should give better agreement between modeled and observed Si II line ratios.

Subject heading: atomic data

1. INTRODUCTION

In diffuse plasmas, the rates for radiative decay—i.e., transition probabilities or A-values—of low-lying metastable atomic levels can be of the same order of magnitude as the rates for excitation and deexcitation by electron collisions. In such cases, intensity ratios involving emission lines from metastable levels are sensitive to electron densities and temperatures. Because of this sensitivity, such ratios are often the basis of diagnostic techniques (Jordan 1979; Doschek 1985; Dere & Mason 1981) used to determine these parameters in diffuse astrophysical plasmas. The accuracies of such diagnostic methods depend critically upon knowledge of the A-values of the transitions involved.

We report the first measured radiative lifetimes of the $J = 5/2$, 3/2, and 1/2 levels of the $3s3p^2\,{^4P}$ metastable term of Si II (see Martin & Zalubas 1983) and A-values determined from these lifetimes. This project (Calamai et al. 1992b) demonstrated for the first time that, under the right conditions, multiple-component decay curves obtained from observations of excited ions in a low-energy ion trap environment can be unambiguously analyzed to provide accurate radiative lifetimes for multiple metastable atomic levels.

The five spin-changing, "intersystem" lines of Si II multiplet 0.01 at ~234 nm, emitted as the fine-structure levels of the $3s3p^2\,{^4P}$ metastable term decay to the $3s^23p\,{^2P^o}$ ground term, are prominent features in the UV spectra of a variety of astrophysical objects: e.g., the solar chromosphere (Doschek et al. 1976), planetary nebulae (Köppen & Aller 1987), and the slow nova RR Tel (Penston et al. 1983). However, two recent attempts (Dufton et al. 1991; Judge, Carpenter, & Harper 1991) to use the Si II] line ratios as diagnostics were limited by uncertainties associated with the available atomic data, possibly the A-values. The results of our measurements should reduce these uncertainties.

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2. LIFETIME AND BRANCHING FRACTION MEASUREMENTS

The apparatus and procedures used for the lifetime measurements were similar to those of Calamai, Han, & Parkinson (1992a). Si^+ ions were produced and $3s3p^2\,{^4P}$ levels were excited inside an ion trap by electron bombardment (at 350 V for 2 ms) of research grade SiH_4 at four pressures between 1 and 13×10^{-8} torr. Cracking pattern data (O'Hanlon 1989) indicates that approximately 11% of the fragments were Si^+ ions. We estimated that our photon-counting efficiency was about 0.05% and, therefore, our observed signals indicate that more than 10^3 metastable ions were created and stored during each cycle of the trap. Most of our data were collected with spherical potential wells that ranged from 10 to 20 eV in depth.

Following ion creation, there was a 82 μs delay period during which the ion cloud stabilized and highly excited ions radiatively decayed to the lowest terms of each spin, i.e., to the ground and metastable terms. An f/2 CaF_2 lens focused light emitted by the decaying metastable ions onto a narrow-band interference filter (14% peak transmittance and 12 nm FWHM bandwidth at 233 nm) in front of an EMR 541Q photomultiplier tube (PMT) operated in photon counting mode. Detected photon counts were accumulated with a 40 Hz, two-phase, data accumulation technique that incorporated real-time background subtraction during alternate phases (Calamai et al. 1992a). This method produced 29 photon-decay curves, about seven at each pressure, with very good signal-to-noise ratios (see Fig. 1). The trap was cycled about 10^6 times, i.e., for about 12 hr, for each curve.

Several ancillary decay curves, for which experimental parameters such as data accumulation rate, channel bin width, electron bombardment energy, bombardment interval, and trap well depth and asymmetry were varied, were also obtained in order to explore for possible systematic effects. None were found.

Branching fractions (BFs) were needed in order to extract A-values from one of the measured lifetimes. The BF for the $^4P_{1/2}$ level was measured using the apparatus and method of Bergeson & Lawler (1993). A monolithic, B-doped, Si cylinder formed the cathode; the discharge was run in Ar. The lines, which were studied with a signal-to-noise ratio of 10–15, were completely resolved from blends with other lines of Si II, or

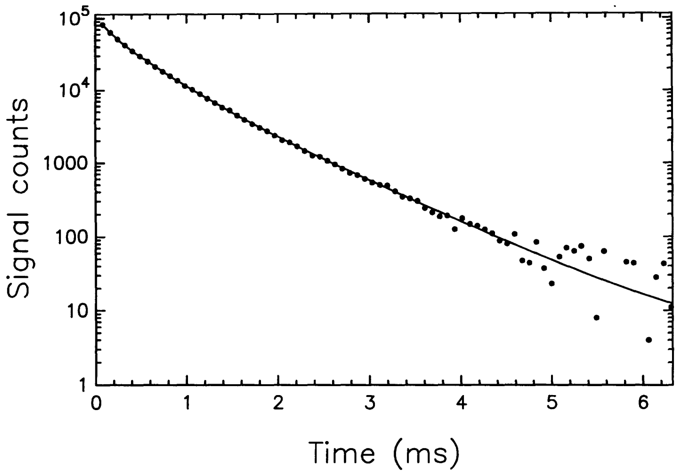

FIG. 1.—Logarithmic plot of the first 77 channels of a 128 channel decay curve for the $3s3p^2\ ^4P$ metastable level of Si^+. The solid curve is a fit of three independent exponentially decaying populations plus a constant background. Negative counts are not shown. Beyond 6 ms, the data are consistent with zero; residuals are shown in Fig. 3.

with lines of Si I, Ar I or II, and B I or II. The measured BFs showed no dependence upon discharge current or pressure.

3. DATA ANALYSIS

The logarithmic plots of the data indicated that, as expected, they corresponded to several populations exponentially decaying into a background. The data were best fitted to a model consisting of three exponentially decaying populations plus a constant background. Each decay curve was fitted using three different sets of input parameters that varied by more than factors of 2; consistent results, with reduced $\chi^2 \leq 1.0$ for nearly all cases, were obtained. The modeled constant background value was consistent with zero, and fits with six parameters gave results that were essentially the same as those from the seven-parameter fits when the data were fitted to an interval with sufficient signal intensity.

Several semiquantitative tests of the three-component model were performed. Fits using two or four initial populations produced large statistical uncertainties and values of reduced $-\chi^2 \geq 3.0$. Discrete integrals of the data were compared to the integrated form of the model, i.e., to $\sum_{i=1}^{3} N_i/\gamma_i$, where $\{N_i\}$ and $\{\gamma_i\}$ represent the best-fit values of the relative initial population and decay-rate parameters. Differences were less than 1%. Graphical measures of the accuracy of the fits were demonstrated by decompositions of the decay curves. In Figure 2*a*, calculated values of the signal counts associated with the "fast–" and "slow–decay rate" components of the data in Figure 1 have been subtracted from the full data set; in Figure 2*b*, the fast and medium components have been subtracted. The remaining signals clearly indicate that only single components remain. Moreover, as demonstrated in Figure 3, if all three components are subtracted from a data set, the residual counts are uniformly distributed about zero with an amplitude (about 30 counts) consistent with Poisson statistics. We noted that, if an incorrect set of initial population and decay-rate parameters are used to completely decompose our data, then the plots of residuals are modulated by a significant time-attenuated oscillation.

The decay rates measured at each pressure were averaged, plotted versus pressure, and extrapolated to zero. The mea-

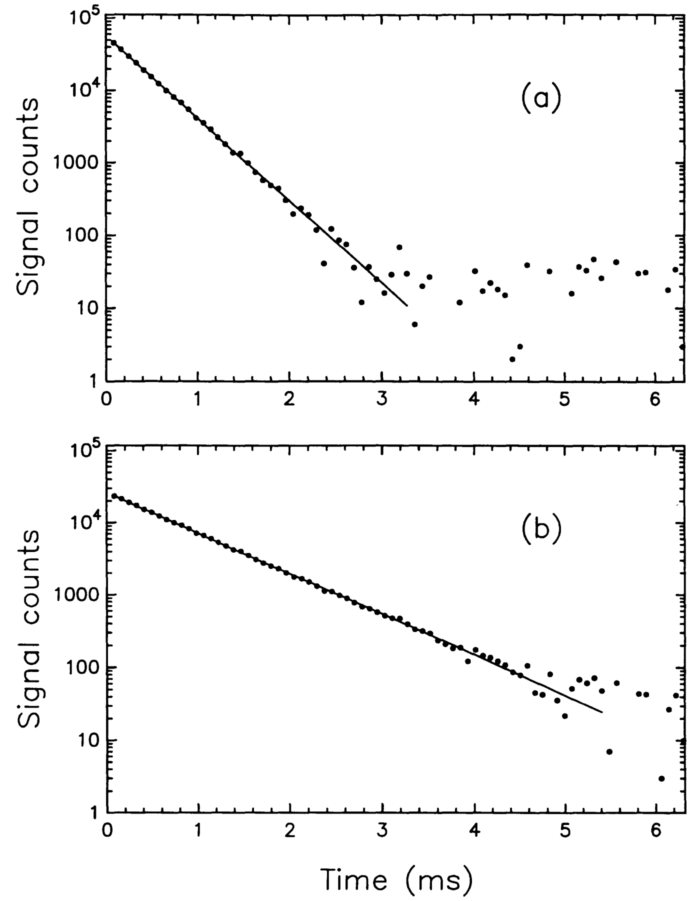

FIG. 2.—Modified logarithmic plots (negative counts not shown) of the decay curve shown in Fig. 1. (*a*) Residual signal counts in the data set after the results of the nonlinear least-squares fit were used to extract the fast- and slow-rate components from the data. (*b*) Residual signal counts in the data set after the fast- and medium-rate components were extracted from the data. In both cases, the straight-line (*solid curve*) behavior indicates that only one exponentially decaying population remains in either modified data set.

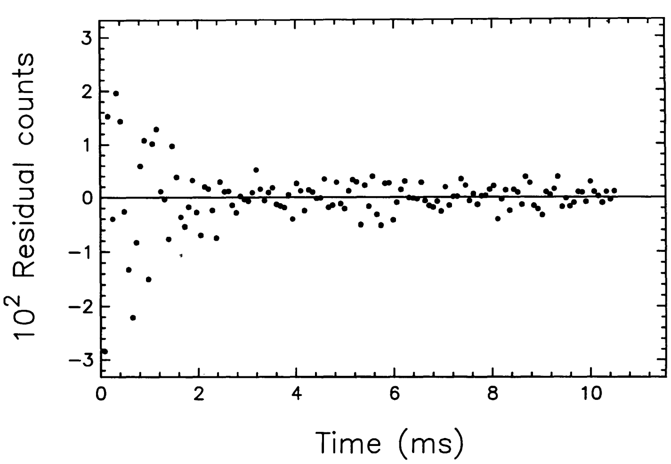

FIG. 3.—Linear plot of the residual counts remaining in a data set, shown in Fig. 1, after the results of the nonlinear least-squares fit were used to extract all three (fast-, medium-, and slow-rate) components from the data. The residual counts are uniformly distributed about zero with an amplitude consistent with Poisson statistics for the total counts (signal plus background).

TABLE 1

COMPARISON OF THEORETICAL AND EXPERIMENTAL RESULTS FOR THE RADIATIVE LIFETIME (μs) OF THE FINE-STRUCTURE LEVELS OF $3s3p^2\ ^4P$ METASTABLE TERM OF Si II

Authors	$^4P_{5/2}$	$^4P_{3/2}$	$^4P_{1/2}$	Type
This work	406 ± 33	811 ± 77	104 ± 16	Experiment
Dufton et al. 1991	370	581	89	Theory
Nussbaumer 1977	417	612	132	Theory

sured rates do not exhibit significance pressure dependence (Calamai et al. 1992b)—a finding expected if the collisional deexcitation coefficients for the Si^+ metastable levels are of the order of 10^{-9} cm^3 s^{-1} as found for other singly ionized species (Calamai et al. 1992a). The difference between the mean decay rate for each level and the extrapolated value was less than 2%.

Assignment of the appropriate ^4P level to the measured decay rates was based on (1) good correlation with calculated lifetimes (Nussbaumer 1977; Dufton et al. 1991) for the $J = 5/2$ and 1/2 levels, (2) agreement with the estimated relative photographic intensities from laboratory spectra (Shenstone 1961), and (3) agreement between our estimated relative upper level populations and values predicted by a model that started with a statistical population that was then distributed by allowed radiative decays.

Our measured results for the radiative lifetimes of the $J = 5/2$, 3/2, and 1/2 metastable levels of Si II are presented in Table 1. Statistical "errors" are the dominant source of uncertainty. We estimated that the sum of the possible systematic uncertainties is not more than ±3% of the measured radiative lifetimes, and we have added this systematic uncertainty for each level directly to the statistical uncertainties to arrive at total fractional uncertainties (at the 90% level of confidence) of 8.2, 9.5, and 15.4% for $J = 5/2$, 3/2, and 1/2 levels, respectively.

Thirty-four measurements of the $^4P_{3/2}$ branching fraction were made. The total uncertainty (systematic and statistical) was about 10% at the 90% level of confidence.

4. DISCUSSION

The possibility of detecting a spurious, time-dependent background luminescence signal is a major concern when making radiative lifetime measurements with our technique. Such a time-dependent background might occur if (1) another metastable state of a stored atomic or molecular ions could decay with the emission od radiation transmitted by the interference filter, (2) a population of stored ions with higher lying, long-lived levels could cascade down and repopulate the metastable levels, and/or (3) radiation could be produced by collisions between the stored ions and the neutral parent vapor.

Our signals were definitely from some Si ions or SiH_4 fragment; no photons were detected unless SiH_4 was present. We could not identify any excited state of a molecular ion that might have been produced from electron bombardment of SiH_4 and that would decay with emission within our detection bandpass. The parent SiH_4 molecules, which might have been excited by collisions, also do not exhibit emission features (Jacox 1988 and references therein) within our bandpass.

We searched for spurious effects by using combinations of interference filters and PMTs that allowed us to detect photons in the range 130–275 nm while blocking Si II radiation at 234 nm. No signals were seen. We believe, therefore, that the only identified possible cascade route, decays of the $3s3p3d\ ^4F^o$ term of Si II producing photons at about 140 nm, did not affect our measurements.

5. RESULTS AND CONCLUSIONS

Although, explicit quantitative comparisons are precluded by the lack of uncertainty information for the calculated data, our measured lifetimes for the $J = 5/2$ and 1/2 levels appear to be in moderately good agreement with the calculated values. However, a significant difference, about 30%, exists between our measured $^4P_{3/2}$ lifetime and both calculated results.

The A-value of the $^2P^o_{3/2}$–$^4P_{5/2}$ transition is simply the inverse of the radiative lifetime of the $^4P_{5/2}$ level. Calculations of Dufton et al. (1991) and Nussbaumer (1977) show that the $^4P_{3/2}$ branching ratio is about 99:1, with the line at 234:42 nm (in air) being stronger. The A-value for the weak branch is very uncertain. Our measured branching fractions were used to extract A-values from the $^4P_{1/2}$ lifetime.

Our results, and the values of Dufton et al. (1991) and Nussbaumer (1977) are presented in Table 2. There are some differences that are significantly larger than the uncertainties in our results. Using our A-values to calculate intensity ratios involving lines from the $3s3p^2\ ^4P$ metastable levels of Si II will bring the computed intensity ratios into better agreement with those observed by astronomers.

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TABLE 2

COMPARISON OF CALCULATED AND EXPERIMENTALLY DETERMINED A-VALUES (s^{-1}) FOR TRANSITIONS OF THE $3s3p^2\ ^4P$ METASTABLE LEVELS TO THE $3s^23p\ ^2P^o$ GROUND LEVELS OF Si II

Transition	λ_{air}(nm)	Branching Fraction	$A_{\text{this work}}$ (Measurement)	$A_{\text{Nussbaumer 1977}}$ (Calculation)	$A_{\text{Dufton et al. 1991}}$ (Calculation)
$^2P^o_{3/2}$–$^4P_{5/2}$	233.461	1.000	2460% ± 8%	2400	2700
$^2P^o_{3/2}$–$^4P_{3/2}$	234.420	0.992	1220 ± 10	1600	1700
$^2P^o_{1/2}$–$^4P_{3/2}$	232.851	0.008	10 ± 50	13	20
$^2P^o_{3/2}$–$^4P_{1/2}$	235.017	0.459	4410 ± 21	3000	4900
$^2P^o_{1/2}$–$^4P_{1/2}$	233.440	0.541	5200 ± 19	4600	6300

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