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Transition probability of the Si III 189.2-nm intersystem line

H. S. Kwong, B. Carol Johnson, Peter L. Smith, and W. H. Parkinson
Harvard-Smithsonian Center for Astrophysics, Cambridge, Massachusetts 02138

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A radio-frequency ion trap has been used to store Si^{2+} ions created by electron bombardment on SiH_4 or SiF_4. The lifetime of the metastable $3s\,3p\;{}^3P_1^o$ level, measured by direct observation of the ${}^1S_0$-${}^3P_1^o$ decay at 189.2 nm, was 59.9 ± 3.6 μs; $A=(1.67\pm0.10)\times10^4$ s^{-1}. The uncertainties indicate the 90%-confidence limit. Statistical uncertainty dominated. The measured value agrees with calculated values to within their quoted accuracies. Our result therefore confirms the calculated data which have been used heretofore in determinations of electron densities in astrophysical plasmas such as the solar transition zone.

I. INTRODUCTION

In many of the diffuse plasmas studied in astrophysics, the rates for collisional excitation and deexcitation are of the same order of magnitude as the radiative-decay rates for metastable levels in atomic ions, especially those levels which decay by spin-changing, electric dipole (i.e., intersystem) transitions. As a consequence, studies of intensity ratios involving lines connecting to such levels can be the most sensitive of electron-density diagnostic techniques.[1] These intersystem lines are also useful for astrophysical element abundance determinations in cases where allowed transitions are saturated.[2]

Interpretation of the observations requires accurate values of radiative transition probabilities (*A* values) for the intersystem lines. For low-*Z* ions, these *A* values are typically 4 orders of magnitude smaller than those for allowed transitions. None of the transition probabilities of ultraviolet lines used, for example, in solar transition zone electron-density determinations has been measured until now and, therefore, unconfirmed theoretical values have been used. Because the calculations are sensitive to configuration interaction, the resulting *A* values can differ by as much as a factor of 2.[2] In the cases where calculated data agree, their accuracy cannot be assessed in the absence of measured values.

This paper reports the measurement of the lifetime of the metastable $3s\,3p\;{}^3P_1^o$ level of Si^{2+} (Si III) which decays by emission of a photon at 189.2 nm to the $3s^2\;{}^1S_0$ ground state. Because there is only one decay channel, the lifetime is related directly to the transition probability.

II. MEASUREMENTS

The lifetime was determined by direct observation of the spontaneous emission from metastable Si^{2+} ions stored in a radiofrequency (rf) ion trap. The principles of such traps are discussed by Dehmelt.[3] Our apparatus was similar to those previously described.[4] Some improvements to the apparatus, procedure, and particular aspects of the Si^{2+} measurement are discussed below.

The cylindrical trap, with diameter and height of 3.3 cm, was machined from stainless steel. A 1.25-cm-diameter hole in the ring electrode permitted observation of the radiative decay photons. The rf frequency was 1.28 MHz; potential wells with depths from 10 to 20 eV were used.

The base pressure of the system, measured with an uncalibrated, nude ion gauge, was 10^{-9} Torr. Si^{2+} ions were created by electron bombardment of SiH_4 or SiF_4 at pressures between 10^{-8} and 10^{-7} Torr. Extensions of this range were not practical: At lower pressures the number of metastable ions and, therefore, the signal-to-noise level was very small; at higher pressures, the BaO dispenser cathode of the electron gun was poisoned. The uncertainty in the absolute values of the pressure is estimated to be less than a factor of 3.

The ions were created and excited during a 2-ms period. This was followed by a 60-μs delay during which allowed transitions could decay. Photons were then detected by an EMR 542-F, solar-blind photomultiplier (PMT) and analyzed by a gated, multichannel scaler with 256 channels at intervals of 10 μs. On alternate cycles, the trap was detuned, by applying an additional dc potential of about + 50 V to the ring electrode, so that no ions would be trapped. Background counts then subtracted from those accumulated. This procedure permitted the background *B* (e.g., PMT dark counts and possible photons from long-lived, metastable, neutral species), to be subtracted from the signal *S*. Thus the measurements were independent of any slow drift in the operating conditions of the apparatus.

The process for production of Si^{2+} ions in the $^3P_1^o$ level also created Si^{2+} ions in the metastable $^3P_2^o$ and $^3P_0^o$ levels and, in addition, Si^+ ions in the $3s3p^2\,^4P$ metastable term. For the doubly charged ions, the $^1S_0$-$^3P_2^o$ transition has $A \sim 10^{-2}\ s^{-1}$,[5] and the $^1S_0$-$^3P_0^o$ line is even weaker. Photons from these decays will not be significant. The ^4P Si^+ levels decay, with calculated lifetimes ranging from 132 to 612 μs,[6] by intersystem transitions at ~233 nm to the $3s^23p\,^2P^o$ ground term. Determination of the Si^+ metastable decay lifetimes is in progress.

In the Si^{2+} measurements, the photons from the Si^+ decays were reduced by using two interference filters—one with a peak transmission of 16% at 190 nm and a passband [FWHM (full width at half maximum)] of 20 nm and, the second, a blocking filter with about 70% transmission at 189 nm and 6% transmission at 233 nm. The combination had 12.5% transmission at 189 nm and less than 0.01% at 233 nm. The ion trap parameters were chosen, by employing the stability diagram (cf. Ref. 3), to optimize the number of trapped Si^{2+} ions. At these conditions, the measured ratio of Si^{2+} to Si^+ photons in the first 120 μs of our detection interval was about 4600. An additional measurement with the Si^+- to Si^{2+}-ion ratio maximized, but still employing the above-mentioned interference filters, gave a lifetime that was 21% longer than the mean of the other values. This result was not included in calculations of the Si^{2+} $^1S_0$-$^3P_1^o$ A value.

The selectivity of the trap was sufficient to permit us to change the Si^{2+}- to Si^+-ion ratio over several orders of magnitude. However, when the parameters were optimized for storage of Si^{2+}, species such as $SiH_n{}^{2+}$ ($n \leq 4$) could have been stored also. In order to confirm that we were observing Si^{2+}, one measurement was made with SiF_4 as the source gas. The lifetime was consistent with that obtained using SiH_4 (cf. Fig. 2).

III. DATA ANALYSIS, RESULTS, AND UNCERTAINTY

Eleven decay curves were analyzed; each comprised data from about 10^7 cycles, or 20 h of operation of the trap. Each channel of the scaler contained a value equal to $(S_i+B_i)-B_i' \equiv N_i$. S_i+B_i is the accumulated signal plus background added to the scaler in channel i when the signals from trapped ions were observed; B_i' is the accumulated background from the periods when the trap was detuned. Because the background subtraction was done in real time, only N_i and

$$T=\sum_i (S_i+B_i)+\sum_i B_i'$$

were available; the actual values of S_i+B_i and of B_i' were not.

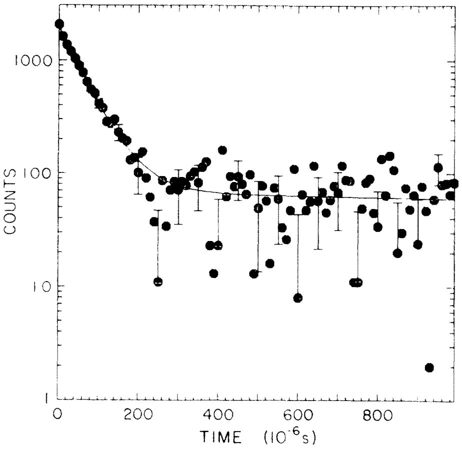

FIG. 1. Semilogarithmic plot of the decay of the $^3P_1^o$ level of Si III as a function of time. The data for all measurements were summed to produce the decay curve shown. Representative uncertainties, calculated in the manner described in the text, are given for each fifth datum. Error bars extending below 1 are not shown. The data and the computer analysis of them extended to 2.56 ms; the tail has been suppressed here to show detail of the decay. $2B$, the major contribution to the uncertainty of points in the tail (see text), was 1189. A two-exponential decay was fit to the data. The principal lifetime is 59.2 μs with an uncertainty at the 90%-confidence limit of 2.8 μs; i.e., $A_1=(1.69\pm0.08)\times10^4\ s^{-1}$. The other parameters were $A_2=(0.02\pm0.16)\times10^4\ s^{-1}$, $\alpha_1=1897\pm60$, and $\alpha_2+\beta'=68$. This fit had a $\chi^2/255=0.89$.

A nonlinear least-squares procedure, based on Bevington,[7] was used to fit the data in two ways. Firstly, each decay curve was fit to a single exponential plus background $I(t)=\beta+\alpha\exp(-tA)$. Secondly, all the data were summed and fit to $I'(t)=\beta'+\alpha_1\exp(-tA_1)+\alpha_2\exp(-tA_2)$.

The constant term β (or β') was small (cf. Fig. 1). It was attributed to a long-lived cascade contribution or to a slight asymmetry between the storage and nonstorage modes. The latter requires the addition of a dc potential to the ring electrode. The potential acts like an electrostatic lens which produces a different distribution in the electron beam and, thus a different number of background-producing species for the two modes of the trap.

In our case of Poisson statistics with a large expected value, the statistical uncertainty for each channel could be calculated: $\sigma_i=[(S_i+B_i)+B_i']^{1/2}$. Because S_i, B_i, and B_i' were not known, we assumed that $B_i \approx B_i'$ and noted, from ancillary measurements, that the values of B_i and B_i' were independent of the channel number. Thus

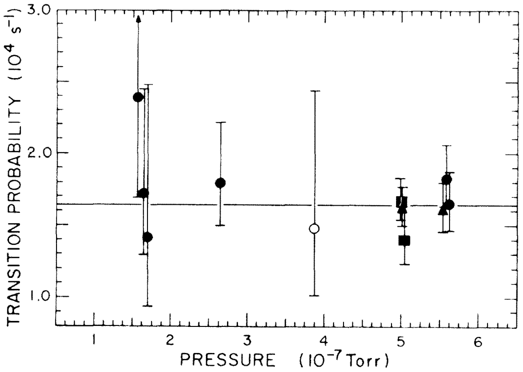

FIG. 2. Transition probabilities from single exponential fits to the 11 measured decay curves as a function of trap pressure. The square, circle, and triangle symbols are for estimated well depths of 10.5, 16, and 19 eV, respectively. The solid symbols are for data obtained using SiH_4; the open symbol, using SiF_4. The uncertainties shown are the 90%-confidence limits. The absolute scale of the pressure values is uncertain by a factor of 3. The weighted mean is shown.

$$\langle B_i \rangle = \langle B_i' \rangle = \left[T - \sum_i N_i \right] / 2 i_{\max} \equiv B \ ,$$

where $i_{\max} = 256$, the number of channels used. σ_i was approximated by

$$\sigma_i = [I^0(t_i) + 2B]^{1/2} \ ,$$

where $I^0(t_i)$ is the value of I (or I') at channel i using the initial values of the parameters, i.e., the values of σ_i were not varied as the fitting program searched for optimum values of the parameters.

The results from single exponential fits to the 11 decay curves are plotted as functions of trap pressure in Fig. 2. The uncertainties shown are those of the 90%-confidence limits, which were estimated by using the method of Lampton *et al.*,[8] who prescribe a general procedure for estimating joint confidence volumes for a correlated parameter fit. The most accurate values have uncertainties of ±8.6%. The weighted mean of the values plotted in Fig. 2 is $A = (1.64 \pm 0.07) \times 10^4$ s^{-1}. The uncertainty is the weighted mean 90%-confidence limit. $\chi^2/10 = 1.08$ for the calculation of the mean. No dependence on pressure, well depth, or source gas was observed.

The individual decay curves appeared to show a slight time dependence in the tail and, therefore, the data were fit to a two-exponential decay, $I'(t)$. Because no systematic differences among the measurements had been observed, all data were summed in order to improve the signal-to-noise ratio. The summed data and the fit to them are shown in Fig. 1. The result was $A_1 = (1.69 \pm 0.08) \times 10^4$ s^{-1}. Other parameters for this fit are given in the caption to Fig. 1.

Additional, single exponential fits were made to three subsets of the summed data: channels 1→20; channels 1→60; and channels 6→256. The results were consistent with A and A_1 above.

In addition to the statistical uncertainty discussed above, we also considered sources of systematic errors. A shortcoming of the method is the lack of wavelength selectivity. Because interference filters were used, the influence of Si^+ 233-nm photons was shown to be negligible in Sec. II; no other contaminating spectral features are known for species that may have been trapped. The inaccuracy resulting from that in the multichannel scaler time base was less than 1%. The presence of the source gas could have caused collisional redistribution of population among nearby energy levels in Si^{2+}, in particular to the fine structure $^3P^o_{0,2}$ levels. In our study, with the source gas pressure kept well below 10^{-6} Torr, it is unlikely that collisional transfer of population will affect our result because unnaturally large, mixing or quenching coefficients of about 2×10^{-8} cm^3 s^{-1} would be required to give a 1% effect on the measured lifetime. The absence of such effects was confirmed by the invariance of the measured lifetime with change of source gas pressure (cf. Fig. 2). The depth of the trap potential was varied in order to search for changes in measured lifetime caused by decrease of the ion cloud radius with time (cf. Ref. 4); none were seen (cf. Fig. 2). Another possible source of error is loss of ions from the trap. For our

TABLE I. Comparison of measured and calculated values of the transition probability for the $^1S_0$-$^3P^o_1$ line in Si III.

Method	A (10^4 s^{-1})	Uncertainty	Reference
Measurement	1.67	±0.10	this work
Semiempirical model potential	1.78	±0.36	11
Configuration interaction	1.46		12
Hartree-Fock	1.61	factor $\frac{3}{2}$	2
Distorted wave	2.25	factor 2	13

experimental parameters, a lower limit to the storage time of 0.1 s was measured by observing the ion signal as the ions were dumped from the trap following storage. The final source of possible systematic uncertainty considered was cascading from highly excited levels, a source of error in lifetime measurements which involves excitation by multiple-collision processes.[9] In our apparatus, the ratio of SiH_4 (or SiF_4) molecules to all other silicon-containing species is 10^3 or 10^4 to 1. We presume, therefore, that metastable Si^{2+} ions are created in a single dissociating collision of an electron on SiH_4 (or SiF_4). This process favors the production of Si^{2+} in low-lying levels.[10] Therefore, although the weak, second exponential decay could be a cascade contribution, such effects are unlikely to have influenced our results (see Fig. 1).

Because no significant systematic effects on our results were identifed, we conclude that the dominant uncertainty is statistical. We state our result as $A({}^1S_0\text{-}{}^3P_1^o)=(1.67\pm0.10)\times10^4\ \text{s}^{-1}$. This is the mean of the one- and two-exponential results and the uncertainty encompasses the 90%-confidence limit for both. The ${}^3P_1^o$ lifetime is, therefore, $59.9\pm3.6\ \mu\text{s}$.

IV. DISCUSSION

Our measured value for the Si III ${}^1S_0\text{-}{}^3P_1^o$ intersystem line transition probability is compared with calculated values in Table I. The calculated values agree with each other and with our result to within the accuracy quoted by the authors. Our result, which has an uncertainty of ±6%, is more precise than the calculated values.

This work demonstrates the ability to make straightforward measurements of the lifetimes of metastable levels of low-Z atomic ions of low charge. These lifetimes can readily be converted into transition probabilities in cases where there is a single decay channel.

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