Beam–foil lifetime measurements of low-lying levels in Si IV

S.T. Maniak, E. Träbert 1 and L.J. Curtis
Department of Physics and Astronomy, University of Toledo, Toledo, OH 43606, USA

Received 30 October 1992; revised manuscript received 10 December 1992; accepted for publication 18 December 1992
Communicated by B. Frick

New measurements of the radiative lifetimes of 3p, 3d, and 4s levels in Na-like Si IV have been made by fast ion beam excitation by a thin foil. Both curve fitting and ANDC joint decay curve analysis methods have been applied. The measured mean-life values (in ns) are: \(\tau(3p^3P_{1/2}) = 1.16(5)\); \(\tau(3p^3P_{3/2}) = 1.14(5)\); \(\tau(3d^3D_{3/2}) = 0.42(5)\); \(\tau(3d^3D_{5/2}) = 0.45(5)\); \(\tau(4s^2S_{1/2}) = 0.31(4)\). The results confirm isoelectronic trends established by cascade-free laser excited measurements in lower charge states and other ANDC measurements in higher charge states, and resolve discrepancies with earlier studies that used multieponential curve fitting reduction methods.

The brightness of the resonance lines of Na I played a central role in the historical development of both atomic spectroscopy and of laboratory astrophysics. Since these transitions are readily amenable to the absorption of dye laser light, they have also become preferred subjects for the study of the influence of light on the motion of atoms. Their isoelectronic counterparts in charged ions have important applications in areas such as fusion plasma diagnosis and interstellar elemental abundance determinations. These alkali-like systems, consisting essentially of one electron outside closed shells, have long been assumed to be theoretically calculable with high accuracy. However, small but puzzling discrepancies between theory and experiment exist for the Li I 2s–2p and Na I 3s–3p resonance transition probabilities.

A series of cascade-free beam-laser and pulsed-laser measurements [1–4] in Li I and Na I have yielded lifetimes that are consistently longer than those predicted by sophisticated ab initio calculations [5–7] by amounts just prescribed by the accuracies (typically 1% or better) of the measurements. Similar discrepancies with theory [8,9] exist for the Mg \(^+\) lifetime, measured in a beam-laser experiment [10], and for multiply charged ions in the Na sequence, measured by beam–foil spectroscopy [11–14]. The critical evaluation of the experimental data requires care, since early beam–foil work often relied solely on curve fitting techniques, and tended to overestimate these lifetimes. More recent beam–foil work has utilized the ANDC method [15], which fully accounts for cascade repopulation through the detailed joint analysis of all cascade-related decay curves. The results indicate that when cascade effects are removed, the discrepancy with theoretical predictions is reduced but not eliminated, and a small but systematic trend toward lifetimes longer than ab initio theory persists. However, semiempirical calculations by Theodosiou and Curtis [16] (using CAHS, the Coulomb approximation with a realistic potential with a Hartree–Slater core and spin–orbit and core polarization effects) agree with the experimental results. Agreement between ab initio calculations and experimental measurements improves for higher members of the isoelectronic sequence. Figure 1 presents an isoelectronic plot of the predictions of refs. [9] and [16] together with the experimental measurements.

There is a gap between the laser data for Na and Mg \(^+\) and the data for higher ionic charges which have been obtained by ANDC analyses. We report here new measurements for Na-like Si IV obtained using ANDC analysis. Two earlier studies of this ion have

1 Permanent address: Experimentalphysik III, Ruhr-Universität Bochum, Bochum, Germany.
been reported [17,18], both of which used only multiexponential curve fitting methods to determine the resonance transition. (Reference [17] employed the ANDC technique for the 3d, 4p, 4f, and 5g term lifetime, but their attempt to jointly ANDC analyze the 3s–3p and 3p–3d decay curves failed, and no attempt was made to separately measure the J dependence of the lifetimes.) We were able to improve upon the accuracy of the decay curve data, taking advantage of significant improvements in ion source, accelerator and spectroscopic detection technology in the two decades since the earlier studies.

The experiment was performed on the 330 keV Danfysik ion accelerator at the University of Toledo. A 0.5 μA ion beam of Si⁺ was produced by argon sputtering of solid silicon, which was isotopically separated in a 30 kV momentum analyser and post accelerated to an energy of 220 keV (chosen for optimum accelerator performance and light yield). The ion beam was directed into a target chamber through a 6 mm diameter aperture in front of a translatable foil wheel. The ions were excited and further ionized by passage through carbon foils of nominal areal density 2.2 μg/cm², with an accompanying mean energy loss of 6 keV [19]. Decay curves were obtained by collecting the ion-emitted light at 90° to the beam as a function of foil position with an Acton 1 m normal-incidence spectrometer. The spectrometer was equipped with two alternative externally indexable gratings and detection systems, one utilizing a channeltron and the other a solar blind or cooled Centronic photomultiplier tube. The beam current was measured by a Faraday cup and the light emitted a fixed distance from the foil was monitored with a photomultiplier tube with a fibre optic link. The experimental control and data collection were automated by an on-line computer system [20].

Decay curves were recorded for the 3s–3p, 3p–3d and 3p–4s transitions, and searches were made for other transitions that might contribute to the cascade repopulation of the 3p level. Evidence for repopulation by the 3p–4d transition was searched for because of the 1.9 ns predicted lifetime [21]. Only a weak intensity was observed for this transition.

Fig. 1. Charged scale line strength factors Z²S versus the reciprocal screened central charge 1/(Z−10). Experimental measurements in other ions are denoted by (O), the measurements for Si IV reported herein are denoted by (×), the solid lines trace the CAHS predictions of ref. [16], and dotted lines trace the MCDF calculations of ref. [9]. The theoretical prediction by Guet et al. [7] comes very close to the trend of the CAHS data shown in the graph (and thus to experiment).
(which occurs in second order near 1120 Å), supporting the prediction that 90% of the decay is branched via 4p–4d [21]. To test for blending, some of the decay curves were measured in both first and second diffraction orders. The decay curves were followed from a point just upstream of the foil to a point corresponding to a flight time of over 10 ns after excitation. This covers a time interval about ten times the 3p lifetime, and at the downstream extremum all transitions not repopulated by the yrast chain had fallen to the background level.

The decay curves of ns–np resonance lines in one-electron ions are well known to be functionally complicated. The Δn > 0 cascades from low-lying levels are shorter-lived than the Δn = 0 primary decay, causing a pronounced growing-in cascade contribution, but long-lived cascades from high-n yrast and Rydberg cascades also cause a significant growing-out tail. Any attempt at multiexponential curve fitting must employ a manageable small sum of exponential decay components to represent the infinite number of lifetimes present, and a system as complicated as this is likely to suffer from systematic errors. Nevertheless, multiexponential fitting analyses (using the standard nonlinear least squares program DISCRETE [22]) are included here for two reasons. One reason is to obtain a comparison with earlier multiexponential fitting analyses to verify that discrepancies that arise are attributable to cascade repopulation, and are not due to systematic errors in decay curve measurements. The second reason is to utilize the multiexponential representation of the decay curves (which does not necessarily reflect the lifetimes of identifiable levels) as a smoothing function for use with ANDC algorithms [15] such as CANDY [23]. After applying this multiexponential filter to reduce statistical fluctuations, these algorithms can then produce reliable lifetime values from the decay curves of the primary level and all levels that have significant cascades into it.

In the ANDC analysis, decay curves of various cascade-coupled levels are jointly evaluated. This method does not require preknowledge of the relative detection efficiencies at the various wavelengths of the observed decays, since these enter as scale factors that are then adjusted by the algorithm. However, it is essential that all curves are measured on a common time scale. This can be done by starting the decay curve upstream of the foil, and accurately noting the rise of the curve as the foil moves past the detection system (corrected for the detection efficiency and exponential content [24]). This t=0 alignment of the curves proved very important in this analysis. A small misalignment of the initial channels of the three decay curves could cause the ANDC process to reject the 3d and 4s cascade decay curves (i.e., assign a very small normalization factor to them) and a poor χ² probability. Correct t=0 alignment was characterized by a sharp increase in the normalization factors for the cascades and a sharp reduction in the chi-squared probability of the fit. This matching of the time base of each of the decay

Table 1
Lifetimes (ns) of low-lying Si IV levels.

<table>
<thead>
<tr>
<th>Level</th>
<th>τ (ns)</th>
<th>ANDC calculations</th>
<th>curve fitting calculations</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>this work a)</td>
<td>this work b)</td>
</tr>
<tr>
<td>3p 2P1/2</td>
<td>1.16(5)</td>
<td>1.39 c)</td>
<td>1.34(8) d)</td>
</tr>
<tr>
<td>3p 2P3/2</td>
<td>1.14(5)</td>
<td>1.41 c)</td>
<td>1.42(8) d)</td>
</tr>
<tr>
<td>3d 2D5/2</td>
<td>0.42(5)</td>
<td>0.44(6) d)</td>
<td>1.2(4) e)</td>
</tr>
<tr>
<td>3d 2D3/2</td>
<td>0.45(5)</td>
<td>0.52(5) d)</td>
<td>0.46(5) e)</td>
</tr>
<tr>
<td>4s 2S1/2</td>
<td>0.31(4)</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

a) Obtained using program CANDY [23]. b) Obtained using program DISCRETE [22].
c) Effective fitting parameters, not identifiable with a specific level lifetime.
d) Ref. [18]. e) Ref. [17]. f) CAHS [16].
g) Coulomb approximation [21]. h) Ref. [7].
curves is so important, because the relative amplitudes of the components of each of the multieponential curves enter the analysis, and the relative amplitudes of a sum of exponentials, of course, vary strongly with time after excitation. On a scale of foil displacement (as the curves are recorded) this is a matter of a few micrometers; this corresponds to a time scale of the decays of order 20 ps in the present case.

The results of our analysis are presented in table 1, together with earlier curve fitted measurements, as well as CAHS [16] and multiplet Coulomb approximation [21]. Notice that the parameters extracted from our multieponential curve fits agree very well with earlier reported lifetimes of the 3p levels, indicating that the measured decay curves themselves are consistent. However, when the 3p–3d and 3p–4s cascade decay curves are incorporated into the analysis via the ANDC method, then the 3p lifetimes decrease significantly, agreeing very well with the semi-empirical CAHS calculations of Theodosiou and Curtis [16]. In fig. 1 our results are combined with other measurements for the Na sequence, and compared with CAHS and MCDF predictions on an isoelectronic plot.

We gratefully acknowledge valuable suggestions from Drs. D.G. Ellis, R.R. Haar, C.E. Theodosiou, and R.M. Schectman, and technical assistance from D.J. Beideck. The hospitality experienced by E.T. at Toledo is greatly appreciated. E.T. enjoyed financial support by the Max Kade Foundation (New York) and the Feodor Lynen Program of the Alexander von Humboldt Foundation (Bonn). L.J.C. and S.T.M. were supported by the US Department of Energy, Office of Basic Energy Sciences, Division of Chemical Sciences, under grant DE-FG05-88ER13958.

References