Observation of a Discrepancy between Experimentally Determined Atomic Lifetimes and Relativistic Predictions for Highly Ionized Members of the Na I Isoelectronic Sequence

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(Received 29 February 1988)

The beam-foil technique has been used to record the intensity decay of the $3s$-$3p$ and $3p$-$3d$ transitions in Na-like Ti, Fe, Ni, and Cu. Subsequent analysis using the ANDC-CANDY cascade correction method results in lifetime data for the $3p\ ^2P_{3/2}$ level which show a divergence from relativistic calculations as the nuclear charge increases. For the $3p\ ^2P_{1/2}$ level, theory and experiment are found to be in better agreement.

PACS numbers: 34.50.Fa, 32.30.Jc, 32.70.Fw

Transitions between the low-lying levels in sequences with only one active electron are very intense and well isolated spectral features. Hence, these transitions are used in many applications of atomic spectroscopy, e.g., the diagnostics of astrophysical and laboratory plasmas. Experimental studies of these transitions also provide a natural first testing ground for theoretical atomic-structure calculations.

In Na I experimental lifetimes for the $3p\ ^2P$ levels have been reported by Gaupp, Kuske, and Andrä[1] with an estimated uncertainty as low as 0.3%. This was accomplished by our performing careful, cascade-free beam-laser measurements. Beyond Mg II all experimental lifetime data originate from measurements by the beam-foil method. For the $3s\ ^2S$-$3p\ ^2P$ resonance transitions, the nonselective nature of the beam-foil excitation process leads to complicated decay curves because the $3p$ levels are repopulated by cascades from higher-lying levels. However, a series of recent papers on Na-like S,[2] Cl,[3] Ar,[4] and Fe[5] has shown that accurate $3p\ ^2P$ lifetimes can be obtained despite the severe cascade situation by use of the so-called ANDC technique.[6] In this method the decays of the most important directly cascading levels are also measured and the primary $3p\ ^2P$ lifetimes are then obtained in a joint analysis of all the measured decay curves.[7]

In S VI, Cl VII, and Ar VIII the multiplet oscillator strengths derived from the cascade-corrected $3p\ ^2P$ lifetimes were found to be in very good agreement with the nonrelativistic multiconfiguration Hartree-Fock (MCHF) calculation by Froese Fischer.[8] The single-configuration relativistic Hartree-Fock (RHF) calculation by Kim and Cheng,[9] on the other hand, consistently overestimated the f values. This is not surprising since the latter calculation neglects correlation effects which are known to be important, particularly in the low-Z end of the sequence. What is more surprising is that the results in Cl,[3] Ar,[4] and Fe[5] seemed to indicate that the discrepancies in the oscillator strength for the $3s\ ^2S_{1/2}$-$3p\ ^2P_{3/2}$ transition between the RHF calculation and experiment increased with Z. However, the observed discrepancy in any one ion is not very large (only about two standard deviations) and hence further experiments were needed to establish an isoelectronic trend.

In this Letter we present a remeasurement of the $3p\ ^2P$ lifetimes in Fe XVI with better time resolution than in the previous study,[5] together with new data for Ti XII, Ni XVIII, and Cu XIX. In all cases the ANDC technique has been used to correct for all cascading reaching $3p$ via $3d$. An isoelectronic comparison of theory and experiment for the $3s\ ^2S_{1/2}$-$3p\ ^2P_{1/2,3/2}$ oscillator strengths up to $Z=29$ is also given. The $3p$ lifetimes in Fe, Ni, and Cu have been measured in previous beam-foil studies,[10–13] but only in the case of the $3p\ ^2P_{3/2}$ lifetime in Fe XVI (Ref. 10) was a detailed cascade correction performed.

The measurements reported here were done at the 4-MV Dynamitron tandem accelerator at the Ruhr University in Bochum, with a standard beam-foil setup. Beam energies between 20 and 36 MeV were used, with beam currents ranging from 0.5 μA for Ti and Fe up to several microamperes for Ni and Cu. The ion beam was further ionized and excited by passage through a 20-μg/cm^2 carbon foil, and the emitted light was analyzed and detected with a 2.2-m McPherson grazing-incidence spectrometer (model 247), equipped with a 600-line/mm gold-coated grating and a Channeltron detector. Further details on the experimental arrangement can be found in Ref. 5.

Decay curves for the $3s\ ^2S$-$3p\ ^2P$ resonance transitions and the $3p\ ^2P$-$3D\ ^2D$ direct cascade transitions were measured for Ti XII, Fe XVI, Ni XVIII, and Cu XIX. In Fe

TABLE I. Experimental lifetimes (in nanoseconds) for the $3p\,^2P$ and $3d\,^2D$ levels in Na-like Ti, Fe, Ni, and Cu.

Ion	Level	This experiment ANDC	This experiment Multiexponential fit[a]	Other
Ti XII	$3p\,^2P_{1/2}$	0.241 ± 0.015	0.275 ± 0.020 (−0.093, 2.12, 15)	
	$3p\,^2P_{3/2}$	0.204 ± 0.013	0.237 ± 0.017 (−0.090, 1.04, 11)	
	$3d\,^2D_{3/2}$		0.102 ± 0.003 (0.532, 5.32)	
	$3d\,^2D_{5/2}$		0.110 ± 0.004 (0.766, 5.88)	
Fe XVI	$3p\,^2P_{1/2}$	0.170 ± 0.011	0.180 ± 0.006 (−0.038, 1.9)	0.167 ± 0.011,[b] 0.157 ± 0.010[c]
	$3p\,^2P_{3/2}$	0.138 ± 0.009	0.154 ± 0.008 (−0.061, 1.9)	0.135 ± 0.008,[b] 0.138[d]
	$3d\,^2D_{3/2}$		0.064 ± 0.010 (0.24, 2.2)	0.051 ± 0.011,[b] 0.067 ± 0.004[c]
	$3d\,^2D_{5/2}$		0.072 ± 0.005 (0.22, 1.6)	0.069 ± 0.010,[b] 0.083 ± 0.005[c]
Ni XVIII	$3p\,^2P_{1/2}$	0.141 ± 0.009	0.153 ± 0.004 (−0.029, 2.8)	0.137 ± 0.014[e]
	$3p\,^2P_{3/2}$	0.113 ± 0.008	0.112 ± 0.031 (−0.063, 0.32, 2.7)	0.108 ± 0.011[e]
	$3d\,^2D_{3/2}$		0.054 ± 0.002 (0.28, 2.8)	0.066 ± 0.007[e]
	$3d\,^2D_{5/2}$		0.062 ± 0.002 (0.46, 4.4)	
Cu XIX	$3p\,^2P_{1/2}$	0.123 ± 0.008	0.131 ± 0.011 (−0.033, 0.94)	0.085 ± 0.018,[f] 0.123 ± 0.004[g]
	$3d\,^2D_{3/2}$		0.031 ± 0.003 (0.15, 3.1)	0.054 ± 0.006,[f] 0.041 ± 0.002[g]

[a]Cascade lifetimes in parentheses. A negative sign denotes a growing-in cascade.
[b]Hutton, Engström, and Träbert (Ref. 5), ANDC.
[c]Buchet *et al.* (Ref. 10), multiexponential fit.
[d]Buchet *et al.* (Ref. 10), ANDC.
[e]Pegg *et al.* (Ref. 11).
[f]Pegg *et al.* (Ref. 12).
[g]Buchet-Poulizac and Buchet (Ref. 13).

and Cu the measurements were done with a masked grating to increase the time resolution from around 50 to 25 ps by narrowing of the length of beam viewed by the spectrometer. As the increased time resolution did not appear to influence the analysis of the Fe XVI data (see below), the measurements in Ti XII and Ni XVIII were done with an unmasked grating and consequently higher statistics. In Cu XIX, the $3s\,^2S_{1/2}$-$3p\,^2P_{3/2}$ transition is blended by a transition in Al-like Cu ($3s^2 3d\,^2D_{5/2}$-$3s3p3d\,^2F_{7/2}$) which can be seen as a strong line in a

TABLE II. Experimental and theoretical oscillator strengths for the $3s\,^2S$-$3p\,^2P$ transitions in Na-like Ti, Fe, Ni, and Cu.

Ion	Upper level	This work ANDC	Other experiment	Theory
Ti XII	$3p\,^2P_{1/2}$	0.143 ± 0.009		0.156[a]
	$3p\,^2P_{3/2}$	0.312 ± 0.019		0.322[a]
	Multiplet	0.455 ± 0.021		0.479,[a] 0.45,[b] 0.428[c]
Fe XVI	$3p\,^2P_{1/2}$	0.115 ± 0.007	0.124[d]	0.125,[a] 0.125[e]
	$3p\,^2P_{3/2}$	0.244 ± 0.015	0.244[f]	0.272,[a] 0.271[e]
	Multiplet	0.359 ± 0.017	0.368[d,f]	0.397,[a] 0.383,[b] 0.332[c]
Ni XVIII	$3p\,^2P_{1/2}$	0.109 ± 0.007	0.114[g]	0.115[a]
	$3p\,^2P_{3/2}$	0.226 ± 0.014	0.237[g]	0.255[a]
	Multiplet	0.335 ± 0.016	0.351[g]	0.376[a]
Cu XIX	$3p\,^2P_{1/2}$	0.112 ± 0.007	0.112[h]	0.111[a]
			0.16[i]	

[a]Kim and Cheng (Ref. 9), relativistic Hartree-Fock. Values for Ti, Ni, and Cu are interpolated.
[b]Froese Fischer (Ref. 8), MCHF. The theoretical oscillator strengths are recalculated with the experimental energies. The value for Ti XII is interpolated.
[c]Biémont (Ref. 18), Hartree-Fock with experimental energies.
[d]Buchet *et al.* (Ref. 10), multiexponential fit.
[e]Karwowski and Szulkin (Ref. 19), relativistic Hartree-Fock.
[f]Buchet *et al.* (Ref. 10), ANDC.
[g]Pegg *et al.* (Ref. 11).
[h]Pegg *et al.* (Ref. 12).
[i]Buchet-Poulizac and Buchet (Ref. 13).

spectrum taken 1 ns after the initial foil excitation; see Fig. 2 in Hutton *et al.*[14] Hence the decay time of the blending transition is quite long and an accurate measurement of the $3p\,^2P_{3/2}$ lifetime could not be made at the low beam energies employed here. Consequently, the cascade transition from $3d\,^2D_{5/2}$ was not measured either. Wavelength data for the transitions investigated in this work are given by Reader *et al.*[15]

The analysis of the measured decay curves was done in three steps. First, the multiexponential fitting routine DISCRETE[16] was used outside the region closest to the foil (which is affected by the finite width of the detection window). The decay constants and relative initial intensities obtained in these fits were then used as start values for the program FITH,[17] which deconvolutes the effect of the detection window and performs a constrained multiexponential fit to the data. Both methods of analysis led to essentially the same values for the $3p\,^2P$ lifetimes. A more detailed discussion of the data analysis with reference to the finite detection window can be found in Ref. 5. The $3p\,^2P$ and $3d\,^2D$ lifetimes obtained from the curve-fitting analysis are given in Table I. It can be noticed that for all the ions the $3d\,^2D$ lifetimes show up as growing-in cascades in the $3s$-$3p$ decay curves, indicating good internal consistency in the analyses.

From the results of the S VI work,[2] where the influence of different cascades ($3d\,^2D$, $4d\,^2D$, and $4s\,^2S$) in the ANDC analysis was investigated, we conclude that the $n \geq 4$ levels should have an insignificant effect on the ANDC analysis in the highly ionized atoms studied in this work. Thus, to obtain the final, accurate $3p$ lifetimes only the repopulation reaching the $3p$ levels via $3d$ was included in the ANDC analysis, which was performed with the computer program CANDY.[7] Table I shows that the detailed cascade correction provided by this technique results in an average reduction of the experimental lifetimes by around 10%. We also note that the new results for Fe XVI confirm our earlier findings,[5] and that the $3p\,^2P_{3/2}$ lifetime is in perfect agreement with the value obtained by Buchet *et al.*[10] In Table II, these lifetimes are then converted to oscillator strengths for the $3s\,^2S$-$3p\,^2P$ transitions with use of the observed transitions energies from Ref. 15, and compared with previous experimental and theoretical results.

In Fig. 1 we compare the most recent experimental oscillator strengths for the $3s\,^2S$-$3p\,^2P$ transitions in the Na I isoelectronic sequence with the MCHF calculations by Froese Fischer[8] (multiplet values), and the RHF results from Kim and Cheng.[9] To obtain a consistent comparison, Fig. 1 includes only experimental results where the perturbing influence of cascades has been minimized, either by performance of ANDC analysis or by cascade-free measurements (Na I). References to many other measurements in the sequence can be found in a recent review article.[20]

For all ions up to Ti XII there is excellent agreement

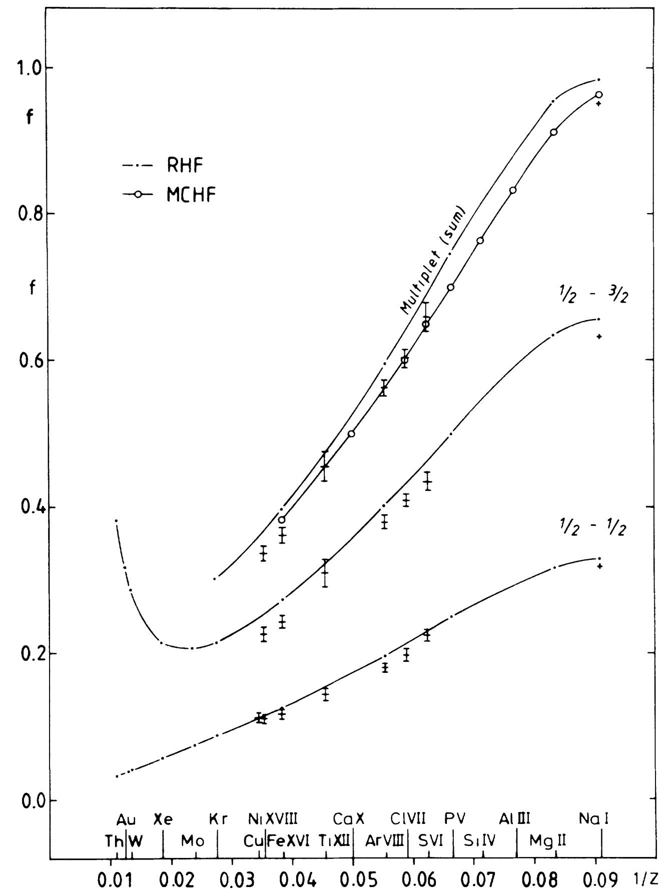

FIG. 1. Oscillator strength vs Z^{-1} for the $3s$-$3p$ transitions in the Na I isoelectronic sequence. The theoretical results are from Froese Fischer (Ref. 8) (nonrelativistic MCHF with experimental energy differences) and Kim and Cheng (Ref. 9) (single-configuration RHF). The experimental values are from the following references: Na, Ref. 1; S, Ref. 2; Cl, Ref. 3; Ar, Ref. 4; Ti, Fe, Ni, and Cu, this work. The ANDC method for cascade correction has been used in all except Na I. For a review of other experimental work in the Na I sequence see Ref. 20.

between the experimental values obtained with the ANDC procedure and those calculated with the MCHF technique, which treats correlation effects in detail. However, it must be stressed that the theoretical MCHF oscillator strengths shown have been recalculated with the experimental wavelengths.[15] This correction, which, e.g., amounts to about 7% in Fe XVI, compensates for the neglected relativistic effects, which mainly influence the energy separation of the $3s$ and $3p$ terms.

In the RHF calculation, on the other hand, correlation effects are largely neglected, but the calculated transition energies are in very good agreement with observations for the higher members of the sequence. If the f values in the beginning of the sequence are corrected by the experimental wavelengths the observed discrepancies are actually increased by a few percent. For the higher-Z ions, where correlation effects decrease in importance compared to the relativistic effects, one would expect the RHF results and experiment to converge. Instead, Fig. 1 clearly shows a small but significant diverging trend in the oscillator strength for the $3s\ ^2S_{1/2}$-$3p\ ^2P_{3/2}$ transition, whereas the agreement between theory and experiment for the $\frac{1}{2}$-$\frac{1}{2}$ transition is better. Since the calculated wavelengths are correct to better than one percent, also for the $\frac{1}{2}$-$\frac{3}{2}$ transition which should be most sensitive to the relativistic effects, the observed discrepancies must be related to the evaluation of the transition matrix elements.

From Fig. 1 it is clear that measurements for still higher ionization stages would be very interesting as a means to probe deeper into the relativistic domain. At the same time we hope that the results presented here will stimulate further theoretical studies of these systems which should be simple enough to be of fundamental interest.

The authors wish to express their gratitude to the skillful operators of the Bochum accelerator. Two of us (R.H. and L.E.) also thank the Bochum group for the friendly collaboration. This work was supported by the Swedish Natural Science Research Council (NFR).

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