# Mean-life measurements of ionized Ar and Cl excited states at grazing incidence wavelengths*

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We have measured Cl VI, Cl VII, and Ar VII, Ar VIII decay times at wavelengths in the range 50–550 Å using the beam-foil technique. The detection system was developed to obtain a spatial resolution of about 50 μm along the beam, allowing very short lifetimes of the order of a few picoseconds to be measured. Our results are compared with recent theoretical results and other experimental data in the Mg I and Na I isoelectronic sequences.

## I. INTRODUCTION

A primary need for accurate lifetime measurements of excited states in ionized atoms has occurred in investigations of material abundances in the sun and stars. However, more recently, highly stripped ions have been produced in hot laboratory plasmas. Analyses of these plasmas require knowledge of excited-state lifetimes for two main objectives, first, as part of diagnostic investigations through spectroscopic determination of the plasma densities and temperatures, and secondly, to determine the problems of plasma energy losses through radiative emissions of highly ionized impurity atoms. For the typical high plasma temperatures, highly stripped heavy-ion radiations provide most of the information in these techniques. In addition, as was pointed out in 1962 by Hinnov and Hofmann,[1] knowledge of transition probabilities may help to calibrate a diagnostic spectroscopic system *in situ*.

Beam-foil spectroscopy is a well-known technique for producing excited states of highly stripped ions.[2] However, the resonance lines of such ions occur principally in the ultraviolet, necessitating a grazing-incidence monochromator for detection of the emission spectrum. The resolving power needs to be sufficient to allow some spectral classification since these spectra are incompletely known. The most serious difficulty is the increasingly fast decay times for the higher stripped ions; thus typical electric dipole transition rates in the 50–500-Å range yield lifetimes of the order of 5–100 psec. Hence excellent time resolution is essential.

In this work we describe a specially modified beam-foil detection system, able to measure decay times of the order of a few picoseconds, which has been used to study decay times of highly ionized chlorine and argon in the wavelength range 50–550 Å. Previous lifetime work in these ions has been limited to wavelengths higher than about 500 Å, and to beam energies of below 2 MeV, except for the chlorine study of Bashkin *et al.*[3] In the Na I and Mg I isoelectronic sequences, some mean lives have been measured for most of the lighter ions—Cl, P, Si, and Al—since the compilation by Wiese *et al.*[4] Thus the transition probabilities of Ref. 4 in these sequences are based mainly on charge-expansion calculations of Crossley and Dalgarno[5] for the heavier ions. In addition to the recent experimental beam-foil results, Biémont[6] has made extensive calculations throughout the Na I isoelectronic sequence. It should be noted that his results show significant reductions in some of the transition probabilities compiled by Wiese *et al.*[4] Hence these measurements in Cl VII and Ar VIII should be useful in comparing the recent experiments with the more recent theory in this Na I sequence. In the Mg I sequence, on the other hand, the state of the theory is quite inadequate and the beam-foil experimental results give the only reliable lifetimes. Thus it is essential to compare the $f$ values in the isoelectronic sequence for a number of different ions to obtain reliable oscillator strengths.

## II. EXPERIMENT

The Argonne 4-MV dynamitron equipped with a physicon ion source was used to accelerate $^{35}Cl^+$ or $^{40}Ar^+$ to energies of 1.0 to 3.5 MeV. The beam was magnetically analyzed, directed through a thin (5 $\mu g\,cm^{-2}$) carbon foil, and collected in a Faraday cup whose integrated flux was used to normalize the measured photonemission to constant numbers of beam particles.

A McPherson 2.218-m grazing-incidence monochromator resolved the light emitted at approximately 90° to the foil-excited beam. A Bendix channeltron (dark count about 5 counts per minute) mounted behind the moving exit slit provided photoelectric detection at wavelengths between 40 and 600 Å. Both spectra and decay curves were stored

in an on-line ASI computer which normalized the photoelectron counts to beam charge and controlled the foil drive stepping motor. In this wavelength region pre-entrance-slit focusing optics is impractical, and the closest possible beam-to-slit distance with this commercial slit mounting is about 6 cm. Thus with the $f/40$ instrumental optics the length of beam observed is approximately 1.25 mm plus an entrance-slit width of 10–100 $\mu$m. Since the beam velocities used in the experiment are about 4.0 mm nsec$^{-1}$, this gives a time resolution of about 0.5 nsec, which is quite adequate for decay lifetimes of more than about 0.1 nsec. However, lifetimes for excited states in highly charged ions which decay by electric dipole emission are often much shorter, and reach a few picoseconds in the 10–500-Å wavelength range.

The spatial resolution of the detection system can clearly be improved, without changing the collection efficiency, by reducing the beam-to-entrance-slit distance. One possibility would be to redesign the entrance-slit flange to bring the fast ion beam very close to the entrance slit. However, the unique geometry of a grazing-incidence monochromator allows us to mount an additional entrance slit close to the beam, retaining essentially the same observation direction and also the focusing property of the Rowland circle instrument. Barrette[7] has applied this technique to lifetime measurements in ionized neon. In Fig. 1 we show the geometrical arrangement when the auxiliary slit is introduced. It should be noted that the grazing angles $\alpha$ and $\alpha'$ corresponding to the original and the auxiliary slit positions are reduced in the figure to clarify the changes involved. Thus in our geometry, where $L \sim 4$ cm, the change in $\alpha$ is small— for $\alpha = 86°$, $\alpha' \approx 85°$, and $x = L\cos\alpha' \approx 3$ mm.

The optical alignment of the auxiliary slit is relatively straightforward. Knowing the additional length $d$, we can calculate the position of the zero-order image to be a distance $t$ from the center of the grating $R$, where

$$t = 2r\cos\alpha' = [x^2 + (d + 2r\cos\alpha)^2]^{1/2} . \quad (1)$$

We set our exit slit at this position, and passing light in the reverse direction through the monochromator we can adjust the entrance slit to the correct position. Using the beam-foil source, we obtained the same spectral resolution with the auxiliary slits as with the original slits. Our auxiliary-slit widths were generally set at 50 $\mu$m, giving a resolution of about 0.3 Å at 300 Å. With this geometry, the center of the beam is about 5 mm from the spectrometer entrance slit, and the beam length viewed is essentially the slit width itself (50 $\mu$m).

With this improved spatial resolution, the measurements become sensitive to the foil surface unevenness. Only data taken using tightly stretched carbon foils (produced after a period of ion beam bombardment) gave reproducible decay curves for the fast lifetimes. The ion beam bombardment heats the foil, causing a shrinking effect which produces a macroscopically smooth and shiny surface at our beam energies. The foil finally ruptures.

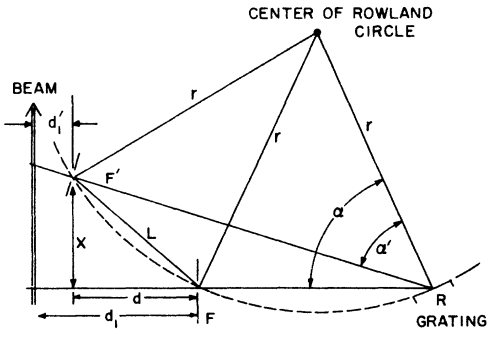


FIG. 1. Geometrical arrangment for the auxiliary spectrometer entrance slit. $R$ is the grating, $r$ the radius of Rowland circle, $F$ and $F'$ the old and new entrance slits, $\alpha$ and $\alpha'$ the old and new angles of incidence, and $d_1'$ the new beam–entrance-slit distance.

## III. RESULTS

### A. Spectra

In Fig. 2 we show a beam-foil spectrum of argon at a beam energy of 3.5 MeV. In the spectral region of 150 to 400 Å transitions of Ar VII and VIII predominate at this beam energy. A few transitions of Ar VI also occur, while Ar IV and Ar V are weakly excited. A complete list of transitions, most of which have previously been observed,[8] is available from the authors. The spectra of chlorine were similar, containing the same isoelectronic transitions but shifted to higher wavelengths. In Fig. 2 one can note that the same transitions (e.g., $3p$-$4d$ and $3p$-$4s$) can be observed in isonucleonic sequences for Ar VIII through Ar V.

Numerous strong unidentified transitions occur in the spectra, and we have identified some new transitions in Ar VIII, Ar VII, and Cl VI. These are listed in Table I. Identifications were made on the basis of quantum-defect comparisons or isoelectronic interpolation within the Mg I sequence.[9] The spectra were internally calibrated through known Ar VII, Ar VIII, and Cl VII transitions, the lines being fitted to Gaussian profiles. The wavelength accuracy of the new identifications is then dependent mostly on their proximity to previously classified lines and varies from ± 0.2 to 0.02 Å, two

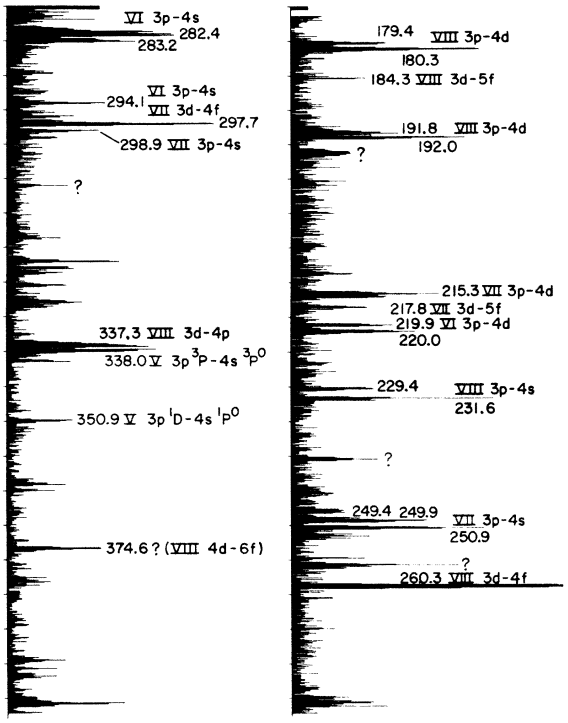


FIG. 2. Spectrum of foil-excited $Ar^+$ beam at 3.5 MeV between the wavelengths 170 and 400 Å, taken close to the foil. The wavelength scale is nonlinear, following the grating equation for the grazing-incidence monochromator.

TABLE I. New transitions in Ar VIII, Ar VII, and Cl VI.

| | Transition | Observed wavelength [a] (Å) | Energy of upper level [b] ($cm^{-1}$) |
|---|---|---|---|
| Ar VIII | $4f\ ^2F$-$6g\ ^2G$ | 407.87 | 961 918 |
| Ar VII | $3s3p\ ^3P^o_{1,0}$-$3s5s\ ^3S_1$ | 165.41 | 717 196 |
| | $^3P^o_2$-$^3S_1$ | 165.95 | 717 196 |
| | $3s3d\ ^3D$-$3s5f\ ^3F^o$ | 217.31 | 783 380 |
| | $3s3d\ ^3D$-$3s6f\ ^3F^o$ | 189.71 | 850 328 |
| | $3s3p\ ^3P^o$-$3p4p\ ^3P$ | 167.10 | 713 056 |
| | $3s3p\ ^3P^o$-$3p4p\ ^3D$ | 168.60 | 707 725 |
| | $3p3d\ ^3D^o$-$3p4p\ ^3D$ | 429.5 ± 0.2 | 707 725 |
| | $3p3d\ ^3P^o$-$3p4p\ ^3D$ | 422.4 ± 0.2 | 707 725 |
| | $3s3p\ ^1P^o$-$3s4d\ ^1D$ | 215.28 | 635 230 |
| | $3s3p\ ^1P^o$-$3s5s\ ^1S$ | 181.47 | 721 774 |
| Cl VI | $3s3d\ ^3D$-$3s6f\ ^3F^o$ | 256.5 ± 0.1 | 669 410 |
| | $3s2d\ ^1S$-$3s4p\ ^1P^o$ | 218.3 ± 0.1 | 457 792 |
| | $3s3d\ ^1D$-$3s4p\ ^1P^o$ | 437.5 ± 0.2 | 457 792 |
| | $3s3p\ ^3P^o$-$3s5s\ ^3S$ | 215.4 ± 0.1 | 564 220 |

[a] Accuracy estimated at ±0.05 Å, except where noted.
[b] Precision is determined by the accuracy of the observed wavelength, as the lower energy level is known more accurately. The quantum defects have been estimated from the energy levels given by C. E. Moore [*Atomic Energy Levels*, Natl. Bur. Stand. Circ. No. 467 (U. S. GPO, Washington, D. C., 1949)].

TABLE II. Mean lives in Cl VI and Cl VII.

| | Wavelength (Å) | Transition | Mean life (psec) This expt. | Other expts. | Theory | CA | Cascades | $R(0)$ [a] |
|---|---|---|---|---|---|---|---|---|
| Cl VII | 225.1 | $3p\ ^2P$-$4d\ ^2D$ | 114.0 ± 10 | | 112, [d] 113 [e] | 108.8 | 452 | 0.24 |
| | 240.5 | $3d\ ^2D$-$5f\ ^2F$ | 32.0 ± 5 | | | 50 | 390 | 0.40 |
| | 294.0 | $3p\ ^2P$-$4s\ ^2S$ | 54.0 ± 5 | | 43.5, [b] 58.1 [e] | 44.3 | 311 | 0.18 |
| | 340.3 | $3d\ ^2D$-$4f\ ^2F$ | 31.0 ± 5 | | 26.3 [b] | 27.3 | 367, 1050 | 0.55 |
| | 598.2 | $3p\ ^2P$-$3d\ ^2D$ | 173.0 ± 20 [g] | 180 ± 30 [c] | 164, [b] 151.4, [e] 159, [f] 187 [d] | 153 | 1004 | 0.88 |
| Cl VI | 243.9 | $3s3p\ ^3P^o$-$3s4d\ ^3D$ | 152.5 ± 15 | | | | 384 | 0.4 |
| | 325.2 | $3s3p\ ^3P^o$-$3s4s\ ^3S$ | 62.5 ± 5 | | 58.5 [b] | 68.2 | 571 | 0.27 |
| | 399.9 | $3s3d\ ^3D$-$3s4f\ ^3F^o$ | 59.0 ± 5 | | | 46.3 | 590 | 0.27 |
| | 555.5 | $3s3p\ ^3P^o$-$3s3d\ ^3D$ | 151.0 ± 5 | 220 [c] | 122 [b] | 124 | 797 | 0.27 |

[a] Replenishment ratio as defined by Curtis *et al.*, Ref. 10.
[b] Wiese *et al.*, Ref. 4.
[c] Bashkin and Martinson, Ref. 3.
[d] Laughlin *et al.*, Ref. 11.
[e] Biémont, Ref. 6.
[f] Crossley and Dalgarno, Ref. 5.
[g] Growing-in cascades from $4p$ and $4f$ reduce the precision of this measurement.

or three different recordings being used to verify the precision.

### B. Mean lives

For both chlorine and argon the strongest transitions observed in this wavelength region come from the $n=4$ and 5 levels of the Na I-like and Mg I-like ions, and we have restricted our mean-life measurements mainly to these two isoelectronic sequences.

In Table II we show the results of our analyses of the decay curves in Cl VII and Cl VI, and in Table III the analyses of the same transitions in isoelectronic Ar VIII and Ar VII. One measurement made of Ar VI is also included.

Each decay curve was measured several times, first with the original slit (spatial resolution ~0.8 mm) and then with the auxiliary-slit arrangement (spatial resolution ~0.06 mm). In Fig. 3 we show the typical improvement for data taken with the better spatial resolution. Thus the slow cascade is suppressed relative to the fast primary decay, and the effective replenishment ratio,[10] a measure of the impurity of the decay, is reduced from 0.40 to 0.26. For the faster decays the improvement is more marked, and the fast 18-psec decay of $3d\,^2D$-$4f\,^2F$ of Ar VIII was observable only with the auxiliary slit. The replenishment ratios in

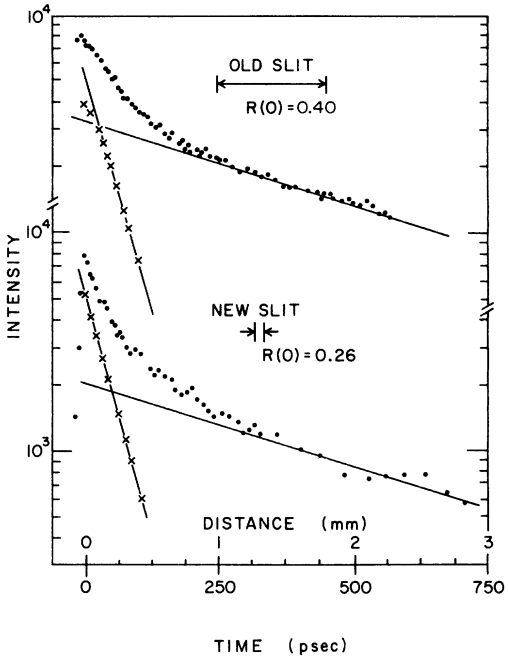


FIG. 3. Decay curves of the $3s3d\,^3D$-$3s4f\,^3F^o$ transitions of Cl VI (a) without and (b) with the auxiliary slit. The viewing length in each case is shown. The replenishment ratios at the foil, $R(x=0)$, are $R_a(0)=0.40$ and $R_b(0)=0.26$.

TABLE III. Mean lives in Ar VIII, Ar VII, and Ar VI.

| | Wavelength (Å) | Transition | Mean life (psec) This expt. | Theory | CA | Cascades | $R(0)$ [a] |
|---|---|---|---|---|---|---|---|
| Ar VIII | 159.0 | $3s\,^2S_{1/2}$-$4p\,^2P_{1/2,\,3/2}$ | 47.1±10 | 49.5,[g] 42.6,[b] 55.3,[c] 54.1[d] | 54.1 | 261 | 0.18 |
| | 179.4 | $3p\,^2P_{1/2}$-$4d\,^2D_{3/2}$ | 44.0±10 | 57.0,[g] 48.9,[e] 56.5,[c] 56.5[d] | 55.8 | 441 | 0.18 |
| | 180.3 | $3p\,^2P_{3/2}$-$4d\,^2D_{5/2}$ | 51.0±10 | 57.0,[g] 48.9,[e] 56.5,[c] 56.5[d] | 55.8 | 289 | 0.21 |
| | 184.3 | $3d\,^2D$-$5f\,^2F$ | 36.9±8 | 41.7,[b] 29.9[c] | | 222 | 0.42 |
| | 229.4 | $3p\,^2P_{1/2}$-$4s\,^2S_{1/2}$ | 24.0±5 | 28.3,[g] 28.6,[b] 32.6,[c] 36.9[d] | 29 | 283 | 0.40 |
| | 230.9 | $3p\,^2P_{3/2}$-$4s\,^2S_{1/2}$ | 25.5±5 | 28.3,[g] 28.6,[b] 32.6,[c] 36.9[d] | 29 | 125 | 0.33 |
| | 260.3 | $3d\,^2D$-$4f\,^2F$ | 18.8±4 | 23.5,[g] 15.4,[b] 16.1[c] | 16 | 158, 1200 | 0.33 |
| | 519.4 | $3p\,^2P$-$3d\,^2D$ | 153.8±10[h] | 154,[g] 137,[b] 132,[c] 127[d] | 130 | 596 | 0.11 |
| Ar VII | 176.6 | $3s^2\,^1S$-$3s4p\,^1P$ | 36.0±5 | | | 432 | 0.31 |
| | 192.6 | $3s3p\,^3P^o$-$3s4d\,^3D$ | 71.5±8 | | 139 | 366 | 0.21 |
| | 250.9 | $3s3p\,^3P^o$-$3s4s\,^3S$ | 30.4±5 | 36[b] | 41.8 | 398 | 0.30 |
| | 297.6 | $3s3d\,^3D$-$3s4f\,^3F^o$ | 48.7±5 | | 25.2 | 500 | 0.31 |
| | 479.4 | $3s3p\,^3P^o$-$3s3d\,^3D$ | 120.7±15[f] | 100.8[b] | 100 | 849 | 0.07 |
| Ar VI | 219.9 | $3p\,^2P$-$4d\,^2D$ | 282.0±10 | | 100 | | 0.00 |

[a] Replenishment ratio as defined by Berry *et al.*, Ref. 18.
[b] Wiese *et al.*, Ref. 4.
[c] Datla *et al.*, Ref. 12.
[d] Biémont, Ref. 6.
[e] Crossley and Dalgarno, Ref. 5.
[f] D. J. G. Irwin, A. E. Livingston, and J. A. Kernahan [Nucl. Instrum. Methods 110, 111 (1973)] have reported an experimental value of 126.
[g] Laughlin *et al.*, Ref. 11.
[h] Growing-in cascades from $4p$ and $4f$ reduce the precision of this measurement.

Tables II and III are those obtained with the auxiliary slit in position.

The error bars given in the tables are based on the reproducibility of mean lives from separate measurements. In the cases of resolved doublets, where we measured both decays we find consistency well within these error limits. The standard deviation for each separate decay-curve fit was less than half the given error bars.

We compare our mean-life results mainly with theory, as no other experimental data exist except for the transitions measured at the upper end of our wavelength range, where normal-incidence monochromators can also be used.

Oscillator strengths in the Na I isoelectronic sequence have been studied theoretically by Laughlin *et al.*[11] and Biémont[6] since the compilation of Wiese, Smith, and Miles, while many transition probabilities in Ar VIII have also been calculated by Datla *et al.*[12] For this sequence the compilation[4] was based essentially on a few experimental values in Na I and Mg II plus charge-expansion calculations of Crossley and Dalgarno[5] and frozen-core Hartree-Fock calculations of McEachran *et al.*,[13] with linear extrapolations to the hydrogenic limit at $1/Z=0$. Dalgarno[14] and Laughlin *et al.*[11] have pointed out that such linear extrapolations may frequently produce considerable error in estimated oscillator strengths. Laughlin *et al.*[11] introduced a method of obtaining both the oscillator strength and its derivative with respect to $1/Z$ at the hydrogenic limit. Thus they showed, in particular for the $3d$-$4p$ and $4p$-$4d$ transitions of the Na I isoelectronic sequence, that previous estimates of $f$ were appreciably too high for highly charged ions above on ionization stage of 7+. The subsequent works of Datla *et al.*[12] and Biémont[6] show the same trends for other Na I-like transition oscillator strengths.

We show an example in Fig. 4 of the oscillator strength of the $3p\,^2P$-$3d\,^2D$ transition as a function of $1/Z$. The calculations of Biémont here agree with the earlier work (contrary to the comparison shown in Ref. 6) and fall on the solid line taken from Smith and Wiese.[4] The other experimental values are taken from Refs. 15–20. Livingston[21] has also measured this Ar VIII mean life, obtaining the same value that we did.

It is clear that the experimental results for the high-$Z$ ions are in reasonable agreement with the calculations of Biémont, although at low $Z$ there remains considerable discrepancy. Only two other Na I-like transitions measured consist of the single allowed decays for which direct comparisons with theory can be made. The $3p$-$4s$ transition probability in Cl VII agrees with the calculation of Biémont, while in Ar VIII the experimental lifetime is slightly faster than all theories predict. For the $3d$-$4f$ transition the experimental lifetimes in Cl VII and Ar VIII are both about 20% longer than the compilation values.[4] Biémont has not calculated the $f$ values for this transition.

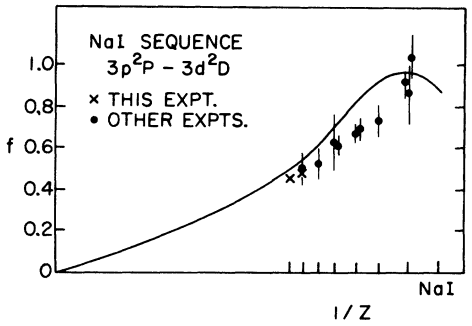


FIG. 4. Absorption oscillator strength for the $3p\,^2P$-$3d\,^2D$ transition of the Na I-like sequence as a function of $1/Z$. The solid curve is taken from Biémont (Ref. 6) (the same curve as Smith and Wiese, Ref. 4) and the experimental data are as follows: Mg II, Refs. 15–17; Al III, Ref. 15; Si IV, Ref. 18; P V, Ref. 19; S VI, Ref. 20; Cl VII, Ref. 3.

For the remaining Na I-like transitions measured, two or more allowed decays occur from the same upper term and the inverse of the sum of transition probabilities is listed as the theoretical mean life, and no direct comparisons of oscillator strengths could be made. The experimental and theoretical mean lives agree to within about 20%.

For the Mg I isoelectronic-sequence mean lives, there exist almost no previous experimental or theoretical results for comparison. Crossley and Dalgarno's charge-expansion calculations have been used primarily in the compilation of Smith and Wiese.[4]

We have calculated all of the transition probabilities from all of the observed terms using the Bates-Damgaard Coulomb approximation (CA).[22] In Tables II and III we list the results in the column labeled CA for both the Na I and Mg I sequences. As expected, in the Na I sequence the Coulomb approximation shows good agreement with the Hartree-Fock calculations and experiment. The comparison with experiment for the Mg I sequence shows that even for low-lying configurations of $n=3$ and 4, where the wave function of the excited electron overlaps considerably with the open core, the Coulomb-approximation result is within a factor of 2 of the experimental mean life.

The $3p\,^3P^o$-$3d\,^3D$ transition in Cl VI gives a $3d\,^3D$ lifetime which is in significantly better agreement with theory than the previous experimental result of Bashkin and Martinson.[3] The same 20% discrepancy is observed for this term

in Ar VII and for the $4s\,^3S$ term in both Cl VI and Ar VII.

No calculations or measurements of the lifetimes exist for the upper terms of the remaining transitions we have measured in the Mg I isoelectronic sequence.

## IV. CONCLUSIONS

We have adapted the beam-foil technique to measure very short lifetimes in the picosecond range at extreme vacuum-ultraviolet wavelengths. We have applied this technique to transitions in Na I-like and Mg I-like chlorine and argon. Reasonable agreement with Hartree-Fock calculations and $1/Z$ expansion and calculations is obtained in the Na I sequence. Almost no previous theoretical or experimental comparisons are available for the Mg I sequence measurements. Our Coulomb-approximation calculations which neglect configuration mixing with the displaced term system in this sequence are in surprisingly good accord with our experimental results.

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