

Experimental lifetimes of the $2p^5 3s^1 P_1$ and $^3 P_1$ levels in Ne-like sulfur and chlorine

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Lifetimes of the $2p^5 3s^1 P_1$ and $^3 P_1$ levels in S VII and Cl VIII have been measured using the beam-foil technique. The results are 17.4 ± 0.5 (13 ± 1) and 52 ± 2 (27 ± 1) ps for S VII (Cl VIII). Many theoretical results, obtained by a variety of methods, are available for these lifetimes. A summary of the theoretical lifetimes reveals a significant scatter of the results, and none of the calculations agrees with all the present experimental data.

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I. INTRODUCTION

The $2p^5 3l$ configurations in the Ne-isoelectronic sequence have attracted many investigators during recent years, largely because of the discovery of extreme ultraviolet (XUV) laser action between some $3s$ and $3p$ levels in Y and Se plasmas [1,2]. This interesting possibility motivated us to initiate an experimental investigation of atomic lifetimes in the lowest excited configurations in Ne-like ions. In two previous papers we have reported the results of beam-foil measurements of lifetimes for levels in the configurations $2p^5 3p$ and $3d$ in S VII, Cl VIII, and Ar IX [3,4]. In these analyses accurate $2p^5 3p$ lifetimes were obtained by using the ANDC/CANDY method for cascade correction [5,6]. In the present work we have extended our investigation of these systems to the $2p^5 3s^1 P_1$ and $^3 P_1$ levels in S VII and Cl VIII, i.e., to the lower levels involved in the laser scheme. In pure LS coupling all $3s$ levels except $^1 P_1$ should be stable against radiative ($E1$) decay since the only lower level is the $2p^6 1S_0$ ground state. However, the spin-orbit-induced mixing between the $J=1$ levels of $3s$ opens this decay channel also for $^3 P_1$. Furthermore, due to the large transition energy of these resonance lines the lifetimes of the $3s^1, ^3 P_1$ levels become much shorter than those of the $3p$ and $3d$ levels. An exception is the $J=1$ levels in $3d$ which have similar high probabilities for decay to the ground state.

The $3s^1, ^3 P_1$ lifetimes have previously been measured with the beam-foil method by Gardner *et al.* [7] in S VII and Cl VIII and by Berry *et al.* [8] in Cl VIII and Ar IX. Our values differ significantly from the earlier results, particularly for the $^1 P_1$ level in Cl VIII where our value is 60% longer than that reported in [8]. A number of theoretical calculations, using a variety of methods, have also been reported for the $3s^1, ^3 P_1$ lifetimes. The theoretical results are summarized in this paper and show significant scatter as well as substantial deviations from the present experimental data.

II. EXPERIMENTS

The measurements were performed with the beam-foil facility [9] at the 3-MV Pelletron accelerator at the Uni-

versity of Lund, Sweden. The design of the target chamber provides excellent time resolution, and, as discussed in Ref. [10], it is quite feasible to measure lifetimes down to a few ps at MeV energies. In the sulfur experiments the ion beam was accelerated to an energy of 3 or 5 MeV whereas for chlorine an energy of 6 MeV was used. After momentum analysis the beam was directed through a $5\text{-}\mu\text{g}/\text{cm}^2$ -thick carbon foil. The light emitted after the foil was analyzed with a Minuteman 310-G 1-m grazing-incidence monochromator with an angle of incidence of 85° and equipped with a 1200 lines/mm grating. Finally, the light intensity was registered with a channeltron detector.

The two transitions of interest in this work, $2p^6 1S_0 - 2p^5 3s^1 P_1$ and $2p^6 1S_0 - 2p^5 3s^3 P_1$, appear at 72.029 (58.673) Å and 72.663 (59.191) Å, respectively, in S VII [11] (Cl VIII [12]). This wavelength region has been extensively scanned in connection with an investigation of core-excited states in Na-like ions [13], and the Ne-like resonance lines are the strongest lines observed between

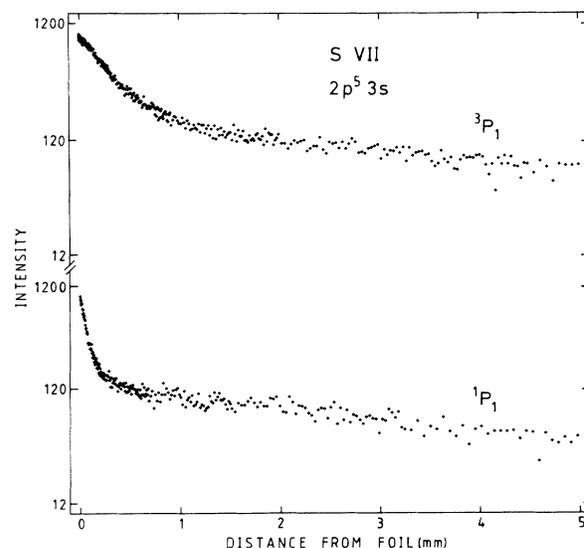


FIG. 1. The first part of the decay curves for $3s^1 P_1$ and $^3 P_1$ in S VII recorded at a beam energy of 5 MeV. The complete recordings extend over 60 mm.

40 and 200 Å. For a more detailed discussion of the spectral recordings we refer to [13] where we also show spectra of both S and Cl containing the Ne-like resonance lines. In both ions the two transitions are well isolated and no indications of any blends that could affect the decay measurements were observed. This conclusion was further confirmed by limited scans performed using a 2400 lines/mm grating. However, the possibility of a nearly perfect blend with Na-like core-excited transitions of the type $2p^6 nl - 2p^5 3l' nl$ ($l'=0$ or 2) with $n > 4$ cannot be excluded. But the intensities of such lines should be insignificant since, as discussed in [13], we have not found evidence of even the lines from $n=4$. Typical decay curves for the $3s \ ^1P_1$ and 3P_1 levels in S VII (recorded at a beam energy of 5 MeV) are shown in Fig. 1.

III. RESULTS

The decay curves were first analyzed by multiexponential curve-fitting using the program DISCRETE [14]. The decay curves were thus decomposed into three components for both the 1P_1 and 3P_1 levels, i.e., two cascades in addition to the component representing the decay of the primary level. Although the levels are repopulated by several transitions from the $2p^5 3p$ configuration only one cascade from this configuration is seen in the multiex-

ponential decompositions because all the $3p$ levels have about the same lifetime [3,4]. Furthermore, the decay time obtained for this component in both the 1P_1 and 3P_1 decay curves (Fig. 1), typically 0.7 ns in S VII and 0.6 ns in Cl VIII, is in perfect agreement with the direct measurements reported in [3,4]. The second cascade is long-lived, about 10 ns, and represents contributions from higher-lying states. A similar long-lived component was also present in the $3p$ and $3d$ decay curves [3,4].

In the next step we applied the ANDC/CANDY [5,6] method for cascade correction using our previously recorded decay curves of the $3p$ levels as the only cascades. In [3,4] we found that this treatment was essential when analyzing the decay curves for the $3p$ levels because the important $3d$ cascades had lifetimes quite close (within a factor of 3) to those of $3p$ and the lifetimes were lowered between 10% and 30% compared to the multiexponential curve-fitting results. However, the situation is very different for the $3s$ levels since these have lifetimes that are more than 20 times shorter than the cascading $3p$ levels, hence it is not surprising that the rigorous ANDC analysis gave the same results as the straightforward curve-fitting technique. Indirectly this consistency also shows that the influence of additional cascades from $n > 3$ is negligible. The resulting lifetimes are given in Table I.

Since there is only one possible decay channel from the

TABLE I. Experimental and theoretical lifetime (ps) for the $2p^5 3s \ ^1P_1$ and 3P_1 levels in S VII and Cl VIII.

		This work ^a	Other expts.	Theory
S VII	1P_1	17.4±0.5 (0.73±0.04, 10±5 ns)	12.0±3.2 ^b	9.3 ^c , 10.8 ^d , 16.9 ^e 11.4 ^e , 13.7 ^f , 26.6 ^g 17.2 ^h
	3P_1	52±2 (0.78±0.06, 13±18 ns)	48.7±13.3 ^b	42.4 ^c , 52.8 ^d , 78.4 ^e 54.4 ^e , 66.0 ^f , 124 ^g 77.3 ^h
Cl VIII	1P_1	13±1 (0.65±0.04, 9±7 ns)	9.9±1.9 ^b 8±2 ^k	6.7 ^c , 11.9 ^e , 8.2 ^e 10.3 ^f , 18.2 ^g , 12.9 ^h 12.7 ⁱ , 10.0 ^j , 10.8 ^k 8.5 ^k , 6.7 ^l , 10.9 ^m
	3P_1	27±1 (0.68 ±0.05, 11±7 ns)	34±12 ^b 30±5 ^k	22.5 ^c , 40.0 ^e , 28.1 ^e 35.7 ^f , 64.1 ^g , 42.7 ^h 41.3 ⁱ , 35.4 ^j , 33.4 ^k 25.2 ^k , 30.7 ^m

^a The first number is the final lifetime after ANDC analysis. Numbers in parentheses are the cascade lifetimes obtained from the multiexponential fits. Note that the lifetime of the cascades are given in ns.

^b Gardner *et al.* [7].

^c Fawcett and Hayes [19], SOC.

^d Fawcett and Hayes [20], SOC.

^e Shorer [27], the first value from DHF and the second from RRPA.

^f Kastner, Omidvar, and Underwood [23], HF.

^g Bureeva and Safronova [25], $1/Z$ expansion.

^h Curtis [17], semiempirical line strengths.

ⁱ Crance [26], model potential.

^j Loginov [22], HF.

^k Berry *et al.* [8], the first theoretical value from MCHF(r) without and the second with the $2p^5 3d$ configuration included.

^l Stewart [24], HF.

^m Fortner, Mathews, and Scofield [28], relativistic HF.

$3s\ ^{1,3}P_1$ levels the experimental lifetimes can be used to obtain accurate estimates of the degree of mixing between these levels if we assume that the only interaction is between these two levels and that the ground state is a pure LS state. The wave function for 1P_1 can then be expanded in the LS basis functions as

$$a|{}^1P\rangle + b|{}^3P\rangle$$

and for 3P as

$$b|{}^1P\rangle - a|{}^3P\rangle.$$

Consequently the expressions for the transition probabilities become (σ being the appropriate transition energy)

$$1/\tau({}^1P) = \text{const} \times \sigma_1^3 |\langle 2p^6\ ^1S | r | 2p^5 3s\ ^1P \rangle|^2 a^2, \quad (1)$$

$$1/\tau({}^3P) = \text{const} \times \sigma_2^3 |\langle 2p^6\ ^1S | r | 2p^5 3s\ ^1P \rangle|^2 b^2. \quad (2)$$

Thus, the ratio between (1) and (2) together with the normalization requirement, $a^2 + b^2 = 1$, give a^2 and b^2 .

For S VII we obtain $76 \pm 1\%$ and $24 \pm 1\%$ for a^2 and b^2 , respectively, which can be compared with the values of 81% and 19% obtained from fitting the Slater integrals to the observed level energies [15]. For Cl VIII the corresponding values are $67 \pm 2\%$ and $33 \pm 2\%$ from the lifetime measurements and 76% and 24% derived from the level energies [16]. An alternative formulation for the derivation of the degree of mixing (in terms of mixing angles) is given by Curtis in Ref. [17,18].

IV. DISCUSSION

In Table I we also give results of previous measurements of the $3s\ ^{1,3}P_1$ lifetimes. Gardner *et al.* [7] performed measurements in both S VII and Cl VIII. They recorded decay curves using the unresolved $2p^6\ ^1S_0 - 2p^5 3s\ ^{1,3}P_1$ multiplet, and derived the 1P_1 and 3P_1 lifetimes as the two shortest components in a multiexponential fit. This explains the rather large uncertainties quoted. Within the error estimates we obtain the same results for the 3P_1 lifetimes as they do while our results for 1P_1 are considerably longer both in S VII and Cl VIII. The Cl VIII lifetimes have also been measured by Berry *et al.* [8] using the spectrally resolved transitions, and in comparison with our data the 3P_1 lifetime agrees well (see Table I). But the discrepancy noted for the 1P_1 level when comparing with [7] is even more pronounced; our value of 13 ± 1 ps is 62% longer than the result of 8 ± 2 ps by Berry *et al.* [8]. The possible origin of this discrepancy is not understood; it should also be noted that the results of Berry *et al.* are consistent with the values they obtained in Ar IX. As arguments in favor of our results we note the following. (1) Careful tests using narrow slits and/or a 2400 lines/mm grating showed no sign of possible blends. (2) The consistency of many repeated measurements. (3) The decay time of the major cascade component, attributable to the $3p$ levels, is identical in the analysis of both the 1P and 3P decay curves, and in exact agreement with the values obtained in the direct measurements [3,4]. (4) The time resolution of our setup [9] in combination with the $1\text{-}\mu\text{m}$ -foil translation used in the recordings of the early part of the decay curves should

make a lifetime even shorter than 8 ps easily detected. As a further test we also performed fits that included the rising part of the decay measured curves by modeling the spatial efficiency function of the monochromator, as discussed in Ref. [10]. This analysis confirmed our quoted results.

In Table I we also present the available theoretical calculations of the 1,3P_1 lifetimes. The calculations by Fawcett and Hayes [19] and Fawcett [20] are both performed with the superposition-of-configurations (SOC) method of Cowan [21] and differ only in the number of configurations included and that the Slater integrals are fitted to experimental level energies in [20]. Loginov [22] also treated the Slater integrals as adjustable parameters and used Hartree-Fock wave functions for calculating the dipole transition matrix elements. The results by Kastner, Omidvar, and Underwood [23] and Stewart [24] are both from nonrelativistic Hartree-Fock calculations. Bureeva and Safronova [25] used the $1/Z$ expansion technique. Crance [26] employed the parametric potential method. Shorer [27] reports extensive calculations using both the Dirac-Hartree-Fock (DHF) and relativistic random-phase-approximation (RRPA) formalisms. Fortner, Mathews, and Scofield [28] performed their calculations using a relativistic Hartree-Fock procedure. In Ref. [8] Berry *et al.*, in addition to the measured lifetimes, also report theoretical results using a relativistic multiconfiguration Hartree-Fock technique [MCHF (r)], with and without including the $2p^5 3d$ configuration. Finally, we quote the semiempirical lifetimes obtained from scaled hydrogenic line strengths [17] and mixing coefficients and transition wavelengths derived from experimental level energies.

From the summary of the theoretical lifetimes in Table I a large scatter can be seen; thus the various methods differ in some cases by almost a factor of 3. Furthermore, none of the many reported calculations agree with all our measurements. For example, it is interesting to note that in both S and Cl the DHF result of [27] is in good agreement for the 1P_1 lifetimes but not for the 3P_1 value, whereas the RRPA result behaves just the opposite. Similarly, we note that the semiempirical results are also in perfect agreement for 1P_1 but deviates significantly for 3P_1 . Another way to compare the theoretical and experimental lifetimes is to study the ratio $\tau({}^3P_1)/\tau({}^1P_1)$ instead of the individual lifetimes. The first observation is that the scatter of the theoretical results becomes less pronounced. The mean value of the various calculations is 4.69 ± 0.14 and 3.26 ± 0.25 for S VII and Cl VIII, respectively. These values can be compared with our experimental results of 2.99 ± 0.15 (S VII) and 2.08 ± 0.18 (Cl VIII). The ratios obtained from the measurements by Gardner *et al.* [7] are 4.06 ± 1.55 (S VII) and 3.43 ± 1.38 (Cl VIII), whereas the investigation by Berry *et al.* [8] gives a ratio of 3.75 ± 1.13 for Cl VIII. Thus, the results of the previous experiments are generally closer to the calculated values.

In the case of the $2p^5 3p$ and $3d$ ($J \neq 1$) lifetimes we found generally a good agreement with theory [3,4] although the $3p$ and $3d$ configurations show a rather high degree of intermediate coupling. The good agreement

found for the $3s\text{-}3p$ transition probabilities indicates that the $3s$ orbital is well described theoretically. However, the calculation of the $3s$ lifetimes involves the $2p^6$ ground state, and a possible origin for the observed discrepancy between the theoretical and experimental results in the present case may be the handling of the different $2p$ orbitals.

In order to investigate if configuration-interaction effects could improve the agreement between calculated and observed lifetimes, we performed a series of calculations using the Cowan code [21]. According to these calculations there exists a small mixing between $2p^5 3p \ ^1S_0$ and the ground-state as well as between the $J = 1$ levels in $2p^5 3d$ and $2p^5 3s$, but including these effects actually increased the discrepancy with the experimental results compared to the single-configuration calculations. We also made LS -dependent single-configuration calculations with the Cowan code, i.e., optimized the wave functions on the $2p^5 3s \ ^1P$ and 3P terms individually. Using the dipole matrix elements calculated from these wave functions and the experimental wavelengths and mixing coefficients, we obtained a better agreement with our ex-

perimental values. The $3s \ ^1P_1$ lifetime became 16.6 (12.8) ps and the $3s \ ^3P_1$ lifetime 52.8 (26.9) ps in S VII (Cl VIII) compared to the values of 15.4 (11.8) and 48.9 (24.9) ps when using wave functions optimized on the center-of-gravity energy of the $2p^5 3s$ configuration. Although these values show good agreement with our experiments, they are semiempirical and it would be desirable with a careful and detailed theoretical study of the $2p^5 3s \ ^1,^3P_1$ lifetime to resolve the discrepancy that exists between the recent experimental and theoretical results. Additional experiments in other Ne-like ions are also needed in order to make meaningful and reliable isoelectronic comparisons.

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- [1] M. D. Rosen *et al.*, Phys. Rev. Lett. **54**, 106 (1985).
- [2] D. L. Matthews *et al.*, Phys. Rev. Lett. **54**, 110 (1985).
- [3] M. Westerlind, L. Engström, R. Hutton, N. Reistad, S. Bliman, M. Raphaelian, and H. G. Berry, Phys. Scr. **38**, 821 (1988).
- [4] M. Westerlind, N. Reistad, C. Jupén, and L. Engström, Nucl. Instrum. Methods B **31**, 300 (1988).
- [5] L. J. Curtis, H. G. Berry, and J. Bromander, Phys. Lett. **344**, 169 (1971).
- [6] L. Engström, Nucl. Instrum. Methods **202**, 369 (1982).
- [7] R. K. Gardner, C. L. Cocke, T. K. Saylor, and B. Curran, J. Opt. Soc. **68**, 830 (1978).
- [8] H. G. Berry, J. Desesquelles, K. T. Cheng, and R. M. Schectman, Phys. Rev. A **18**, 546 (1978).
- [9] L. Engström and K. Håkansson, Phys. Scr. **43**, 475 (1991).
- [10] L. Engström and P. Bengtsson, Phys. Scr. **43**, 480 (1991).
- [11] E. Ferner, Ark. Mat. Astron. Fys. **36A**, 65 (1948).
- [12] B. Edlén, Z. Phys. **100**, 726 (1936).
- [13] M. Westerlind, L. Engström, C. Jupén, and P. Bengtsson, Phys. Scr. (to be published).
- [14] S. W. Provencher, J. Chem. Phys. **64**, 2772, (1976).
- [15] E. Ya. Kononov, A. E. Kramida, L. I. Podobedova, E. N. Ragozin, and V. A. Chirkov, Phys. Scr. **28**, 496 (1983).
- [16] C. Jupén, Phys. Scr. **36**, 776 (1987).
- [17] L. J. Curtis, Phys. Rev. A **40**, 6958 (1989).
- [18] L. J. Curtis, Phys. Scr. **43**, 137, (1991).
- [19] B. C. Fawcett and R. W. Hayes, Phys. Scr. **36**, 80 (1987).
- [20] B. C. Fawcett, Phys. Scr. **30**, 326 (1984).
- [21] R. D. Cowan, *The Theory of Atomic Structure and Spectra* (University of California Press, Berkeley, 1981).
- [22] A. V. Loginov, Opt. Spektrosk. **61**, 11 (1986) [Opt. Spectrosc. (USSR) **61**, 6 (1986)].
- [23] S. O. Kastner, K. Omidvar, and J. H. Underwood, Astrophys. J. **148**, 269 (1967).
- [24] R. F. Stewart, Mol. Phys. **29**, 1576 (1975).
- [25] L. A. Bureeva and U. I. Safronova, Phys. Scr. **20**, 81 (1979). Data for Cl VIII are from U. I. Safronova (private communication).
- [26] M. Crance, At. Data **5**, 185 (1973).
- [27] P. Shorer, Phys. Rev. A **20**, 642 (1979).
- [28] R. J. Fortner, D. L. Mathews, and J. H. Scofield, Phys. Lett. **53A**, 336 (1975).