PHYSICAL REVIEW A VOLUME 11, NUMBER 2 FEBRUARY 1975

Lifetime of the $2\,^3S_1$ state in heliumlike sulphur and chlorine*

J. A. Bednar, C. L. Cocke, B. Curnutte, and R. Randall
Department of Physics, Kansas State University, Manhattan, Kansas 66506

(Received 4 September 1974)

Beam-foil measurements of the lifetimes of the $2\,^3S_1$ state in heliumlike sulphur and chlorine are reported. A Doppler-tuned x-ray spectrometer was used to detect the x rays emitted from the $1\,^1S_0 \leftarrow 2\,^3S_1$ transition. Time-of-flight techniques were used to trace the decay profile of the emitted x rays for 38–55-MeV sulphur and chlorine beams over flight paths of 56–861 cm. For foil-to-detector flight times greater than one half the theoretical lifetimes of 698 nsec (sulphur) and 374 nsec (chlorine), the respective experimental lifetimes of 706 ± 86 and 354 ± 24 nsec are in agreement with theory. For shorter flight times, the experimental decay profiles are not single exponentials. This experimental effect is the source of the previously reported discrepancy between experimental and theoretical lifetimes of the $2\,^3S_1$ state in heliumlike chlorine.

INTRODUCTION

The $2\,^3S_1$ state in heliumlike ions is expected to decay to the $1\,^1S_0$ ground state via a relativistically induced $M1$ transition. Measurements of the lifetimes of the $2\,^3S_1$ state for helium[1] ($Z=2$), chlorine[2] ($Z=17$), argon[3] ($Z=18$), titanium[3] ($Z=22$), vanadium[4] ($Z=23$), and iron[4] ($Z=26$) have recently been reported, and are presented in Table I along with calculations of these lifetimes by Drake[5] and Johnson and Lin.[6] For the cases of $Z=22$, 24, and 26, the respective experimental and theoretical lifetimes are in good agreement. However, for the cases of $Z=17$ and 18, the experimental lifetimes are shorter than the theoretical lifetimes, the discrepancy increasing with decreasing Z. Although the measurement for $Z=2$ is in agreement with theory, the uncertainties in this measurement are large.

Except for the case of $Z=2$, in all the experiments cited above the $2\,^3S_1$ state was produced by foil excitation and the lifetime was determined by tracing the decay profile of the emitted x rays as a function of foil-to-detector flight path. Because of the short span of flight paths used, for the cases of $Z=17$ and 18, it was necessary to assume a single-exponential decay profile.

The present experiment was conducted to reexamine closely the lifetime measurements for $Z=17$ (chlorine) and to extend them to $Z=16$ (sulphur). Particular importance was given to determining the shape of the decay profiles, since we consider a critical test of the assumption of single-exponential decay profiles important for establishing the validity of our results.

EXPERIMENT

The present set of experiments is similar in design to the beam-foil experiments discussed above. Sulphur and chlorine beams from the Kansas State University tandem Van de Graaff accelerator, ranging in energy from 38 to 55 MeV, were foil excited, and the decay profile of K x rays emitted from the excited beam was measured as a function of foil-to-detector flight path. A schematic diagram of the apparatus is presented in Fig. 1. Since the basic experimental techniques are completely described in Refs. 2 and 7, only a few pertinent details will be discussed here.

Two important improvements over previous experiments were made. First, the x rays emitted from the excited beam were detected by a Doppler-tuned x-ray spectrometer, DTS (Refs. 2, 7, and 8), consisting of the proportional counter, absorber, Soller slits, and divided circle shown in Fig. 1. This enabled us to isolate x rays from the $1\,^1S_0 - 2\,^3S_1$ transition at each foil position. Because of the low counting rates encountered in the lifetime experiments, Soller slits of $\pm5^\circ$ resolution were used in the DTS, yielding typical FWHM energy resolutions of 11 eV (sulphur) and 12 eV (chlorine) at the respective $1\,^1S_0 - 2\,^3S_1$ transition energies of 2431 and 2757 eV.[9] The second improvement involves the length of foil-to-detector flight paths used. In past experiments, decay profiles were examined over foil-to-detector flight paths of 270 cm or less. For the beam energies used, such flight paths correspond in flight time to only a small fraction of a mean life for the cases of $Z=17$ and 18, thus forcing the assumption of a single-exponential decay profile. In the present experiment, decay profiles were examined over foil-to-detector flight paths of 56–861 cm. For our average beam velocity (1.7×10^9 cm/sec), such flight paths correspond in flight time to 0.7 (sulphur) and 1.2 (chlorine) mean lives thus enabling us to examine the decay profiles more closely.

Angular collimation of the beam was achieved by a set of adjustable slits located 876 cm from the center of the detector chamber (collimator B, Fig. 1) and a circular aperture 0.32–1.27 cm in diameter located 17 cm from the center of the detector chamber (collimator A, Fig. 1). Collimator B was adjusted so that all the beam passed through the foil. Counting times were normalized to total charge collected in a Faraday cup located downstream of the detector chamber (Fig. 1). Because of the long foil-to-detector flight paths used, collimator A was needed to restrict the size of the excited beam passing through the DTS. The use of collimator A necessitates assuring that the spatial distribution of heliumlike ions in the 2 $^3S_1$ state in the central region of the excited beam and the spatial distribution of other charge states over the same angular region be the same, in order to avoid normalization difficulties due to the different angular admittance of collimator A for various foil positions. For this reason, we used a thick carbon foil (typically two layers of 40-μg/cm^2 carbon foil) 1.27 cm in diameter as an exciter. The use of two layers of foil also helped to ensure the uniform thickness of the exciter. Variation of the flight path was achieved by inserting foils at various positions between the collimators A and B. Usually three foil positions were used for a given run.

Various experimental tests of the apparatus, such as varying the beam energy, incident charge state, foil thickness, beamline pressure, detector chamber pressure, and the size of collimator A, were performed, with no statistically significant effects on the measured lifetimes. Also, since different foils were used at the various foil positions, several tests were conducted by cycling the foils through each position in an attempt to distinguish between foils. These tests exhibited no statistically significant differences between the excitation efficiencies of different foils.

An overall test of the apparatus was conducted by measuring the lifetime of the $2E1$ spontaneous decay of the $2s_{1/2}$ state in hydrogenlike oxygen and fluorine for foil-to-detector flight paths of 234–702 cm. The DTS was replaced by an electromagnet with a proportional counter viewing the magnet gap region. The remainder of the apparatus remained unchanged. The $2s_{1/2}$ state in oxygen and fluorine was produced by foil-exciting 15–35-MeV beams of these elements. The $2s_{1/2}$ component of the excited beam was monitored by mixing the $2s_{1/2}$ and $2p_{1/2}$ states in the magnetic field, and detecting the resulting monoenergetic x rays.[10] The experimental lifetimes from this test of 472 ±10 nsec (oxygen) and 242 ±20 nsec (fluorine) are in agreement with the respective theoretical lifetimes of 464 and 228 nsec.[11] We interpret this agreement as indicating that the apparatus used for our measurements of the 2 $^3S_1$ lifetimes for sulphur and chlorine is suitable for measuring lifetimes on the order of 10^2 nsec.

A detailed discussion of the identical $2s_{1/2}$ lifetime measurements with foil-to-detector flight paths of 56–305 cm is given in Ref. 10. In that experiment, we obtained agreement between experimental and theoretical lifetimes when a single exponential was fitted to the decay profile for foil-to-detector flight paths greater than 130 cm. All the decay profiles displayed a slight convex curvature for foil-to-detector flight paths of 56–130 cm. We noted that this curvature may result from moderately long-lived cascades (on the order of 10^2 nsec) through the $2s_{1/2}$ state with cascade lifetimes shorter than the $2s_{1/2}$ lifetimes. A cascade through the state under observation can only produce a convex curvature in the decay profile and thus lengthen the observed lifetime. This measurement indicated the possibility of moderately long-lived cascades in a foil-excited beam, a possibility which must receive consideration in beam-foil lifetime measurements for states with lifetimes on the order of 10^2 nsec.

In Fig. 2, we present a decay profile of x rays emitted from the $1\,^1S_0 \leftarrow 2\,^3S_1$ transition in 50-MeV

TABLE I. 2^3S_1 lifetimes in nsec.

Z	Theory [a]	Theory [b]	Experiment	Present work
2	7.9×10^{12}	8.0×10^{12}	$3.8\pm^{10}_{2}\times10^{12}$ [c]	⋯
16	710	698	⋯	706 ± 83 [g]
17	381	374	280 ± 25 [d]	354 ± 24 [g]
18	212	208	172 ± 12 [e]	
22	27.4	26.5	25.8 ± 1.3 [e]	
23	17.4	16.8	16.9 ± 0.7 [f]	
26	5.0	4.8	5.0 ± 0.5 [f]	

[a] Reference 5.
[b] Reference 6.
[c] Reference 1.
[d] Reference 2.
[e] Reference 3.
[f] Reference 4.
[g] $T/\tau_{\text{theor}} \geq \frac{1}{2}$.

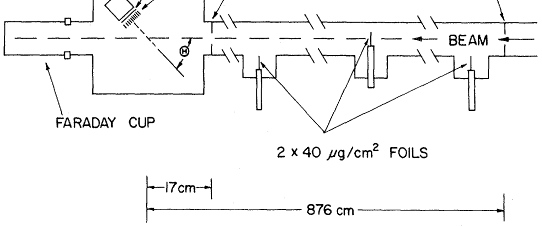

FIG. 1. Schematic diagram of the apparatus. The DTS consists of the proportional counter, absorber, Soller slits, and divided circle.

chlorine. Data from three separate runs, each with three foil positions, are presented in the decay profile, normalized to a foil-to-detector flight path of 500 cm. This decay profile gives a lifetime of 352 nsec when a single exponential is fitted to all the data points except for the point at 173 cm. The data point at 173 cm is high with respect to the other data points, and if included, it would decrease the value of the lifetime. As will be demonstrated shortly, this does not appear to be an error in the experiment, but rather a characteristic of the x-ray decay profiles of the systems being studied.

A typical integral spectrum of emitted x-ray energies is presented in the insert of Fig. 2. As is indicated, x rays were counted for energies both above and below the transition energy (points labeled "data point," inset, Fig. 2), and the difference between these two numbers was taken to be the number of x rays detected from the transition of interest. The number of detected x rays was corrected for dark count (0.1–0.2 per second) by monitoring the counting time and subtracting the appropriate number of counts. The beam velocity was corrected for energy loss in the foil by using the tables of Northcliffe and Schilling.[12]

RESULTS

Even with the advantage of examining decay profiles over a large range of foil-to-detector flight paths limited statistics and a limited number of foil positions create difficulties in establishing the shape of the decay profile for a single run. Thus we present in Fig. 3 a plot of lifetimes determined by fitting a single exponential to two-point decay profiles, as a function of mean foil-to-detector flight time T for the two foil positions. The average separation between foil positions is 198 cm. Both axes are presented in units of the theoretical lifetimes (τ_{theor}) given by Johnson and Lin (Table I). Such an analysis enables us to examine the shape of the decay profiles over the entire range of flight paths used, using data from many completely independent runs. The data presented in Fig. 3 represent the results of 56 two-point lifetime measurements, which were obtained over a total of 9 days of experiment time. Data points for which the flight times are nearly the same are averaged, the error bars representing the RMS deviation in lifetime values.

The results for both the sulphur and chlorine experiments are included in Fig. 3. We have used this format for presenting our results since the results for two ions examined in this experiment are similar when presented in universal units. We do *not* wish to suggest that the behavior exhibited in Fig. 3 is universal in Z.

The first characteristic of the results to be noted is that the curve in Fig. 3 is not a horizontal line, as one would expect for single-exponential decay profiles, thus suggesting the presence of more than one excited system. Effects due to the departure from single-exponential decay profiles are enhanced by the lifetime analysis presented in Fig. 3, in contrast with attempting to detect small departures from linearity in the natural logarithm of the decay profiles.

The second characteristic is that for $T/\tau_{\text{theor}} \geqslant \frac{1}{2}$ the decay profiles are approximately single exponentials. The mean lifetime values for $T/\tau_{\text{theor}} \geqslant \frac{1}{2}$ of 706 ± 86 nsec (sulphur) and 354 ± 24 nsec (chlorine) are in agreement with theory (Table I). These mean values are the average of 6 (sulphur) and 14

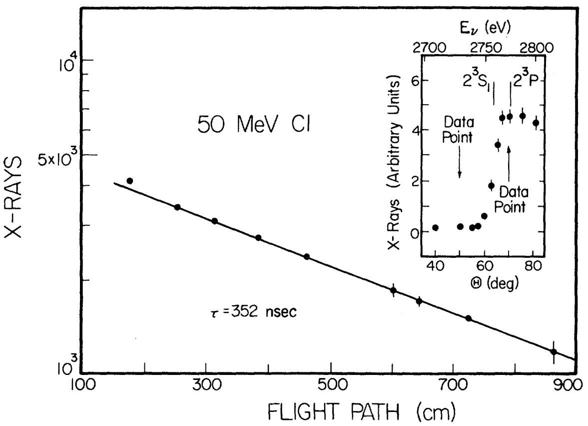

FIG. 2. X-ray decay profile obtained from three different runs using three different sets of foil positions. Exponential fits were used to normalize the data at a foil-to-detector flight path of 500 cm. The insert shows a typical energy spectrum obtained with the DTS.

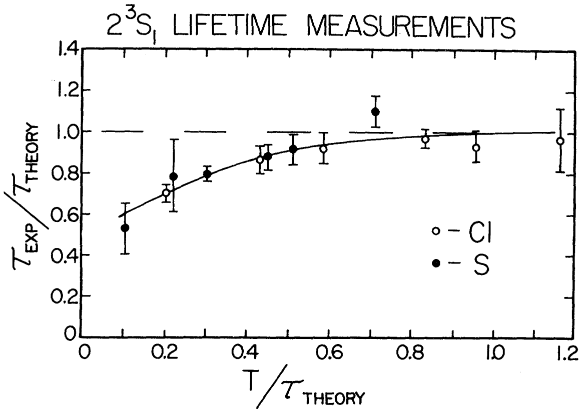

FIG. 3. $2\,^3S_1$ lifetimes from two-point decay profiles as a function of foil-to-detector flight time. Both axes are in units of the theoretical lifetime.

(chlorine) two-point lifetimes, and the uncertainties are RMS deviations.

Our present results are in agreement with the results reported by Cocke *et al.* (Table I) for the case of chlorine. In that experiment, the mean value of $T/\tau_{\rm theor}$ for all the reported measurements was 0.175, placing the reported 2 ^{3}S$_1$ lifetime value of 280 ±25 nsec ($\tau_{\rm expt}/\tau_{\rm theor} = 0.75 \pm 0.07$), in agreement with the results presented in Fig. 3.

We note that for the previous 2 ^{3}S$_1$ lifetime measurement for the case of argon (Table I), the mean value of $T/\tau_{\rm theor}$ for all the reported measurements was 0.29, making the reported 2 ^{3}S$_1$ lifetime value of 172 ±12 nsec ($\tau_{\rm expt}/\tau_{\rm theor} = 0.83 \pm 0.06$), consistent with the trend shown in Fig. 3. This suggests that the discrepancy between theory and experiment for the case of argon might be due to a departure from single-exponential decay profiles.

For the previous 2 ^{3}S$_1$ lifetime measurements for the cases of titanium, vanadium, and iron (Table I), the Z dependence of the 2 ^{3}S$_1$ lifetime is such that the mean values of $T/\tau_{\rm theor}$ are greater than unity.[13] Thus the reported values of $\tau_{\rm expt}/\tau_{\rm theor} \cong 1$ are consistent with the trend shown in Fig. 3.

DISCUSSION

Two possible sources of the effect exhibited in Fig. 3 have been examined. The first of these is the energy resolution of the DTS. Although the energy resolution used was adequate to show that the major source of x rays was the 1 ^{1}S$_0$ ← 2 ^{3}S$_1$ transition, we may have also been detecting a small number of x rays from the 1 ^{1}S$_0$ ← 2 $^3P_{2,1}$ transitions at the end of a moderately long-lived cascade. As noted in the discussion of the $2s_{1/2}$ lifetime measurements for oxygen and fluorine, there is evidence to suggest the presence of such moderately long-lived cascades in foil-excited beams. However, in contrast to cascading through the state under observation, cascading through an unresolved neighboring state can produce a concave curvature of the decay profile and thus a measured lifetime which is shorter than the 2 ^{3}S$_1$ lifetime, if both the mean lifetime of the cascade and the lifetime of the unresolved neighboring state are shorter than the 2 ^{3}S$_1$ lifetime.

We examined this mechanism by considering the following model. Heliumlike ions are created at the foil with an initial population $N(2\,^3{\rm S}_1)$ in the 2 ^{3}S$_1$ state and an initial population N_c in a highly excited state which cascades with a mean lifetime τ_c into the 2 ^{3}S$_1$ and 2 $^3P_{2,1,0}$ states. The 2 ^{3}S$_1$ and 2 $^3P_{2,1,0}$ states are populated by the cascade according to their respective statistical weights of 3, 5, 3, and 1. The initial populations of the 2 $^3P_{2,1,0}$ states at the foil are assumed to be fully depleted. The entire decay scheme of the 2 ^{3}S$_1$ and 2 $^3P_{2,1,0}$ states is analyzed to determine, as a function of T, the ratio $\tau_{\rm expt}/\tau_{\rm theor}$, which would result from a beam-foil lifetime measurement in which x rays from the 1 ^{1}S$_0$ ← 2 ^{3}S$_1$, 2 $^3P_{2,1,0}$ transitions were unresolved. The results of a least-squares fit of this model to the data in Fig. 3 is represented by the solid line in Fig. 3. This fit required $N_c = 0.05N(2\,^3{\rm S}_1)$ and $\tau_c = 0.15\tau(2\,^3{\rm S}_1)$. These values of the parameters N_c and τ_c in turn require decay rates R such that $R(1\,^1{\rm S}_0 \leftarrow 2\,^3P_{2,1})/R(1\,^1{\rm S}_0 \leftarrow 2\,^3{\rm S}_1) = 0.11$ for 44.2-MeV sulphur at a foil-to-detector flight path of 58 cm. Higher-resolution energy spectra of the emitted x rays for 44.2-MeV sulphur were taken at a foil-to-detector flight path of 58 cm. The FWHM energy resolution was 2.1 eV at the 1 ^{1}S$_0$ ← 2 ^{3}S$_1$ transition energy. A sum of three of these spectra averaged over five channels is presented in Fig. 4. The features appearing to the right of the 2 ^{3}S$_1$ line are believed to be due to structure in the sulphur K-absorption edge. These spectra indicate a maximum decay rate ratio of $R(1\,^1{\rm S}_0 \leftarrow 2\,^3P_{2,1})/R(1\,^1{\rm S}_0 \leftarrow 2\,^3{\rm S}_1) = 0.03$. Thus, based on the cascade model we considered, inclusion of 1 ^{1}S$_0$ ← 2 $^3P_{2,1}$ x rays does not appear to account for the observed effect.

A spin alignment of the excited beam provides a second possible source of the observed effect. If state with nonzero orbital angular momentum are created at the foil with an angular momentum alignment and decay into the 2 ^{3}S$_1$ state, spin-orbit coupling might transfer some alignment to the spin of the 2 ^{3}S$_1$ state. The difference in the net precession of such a spin alignment about Earth's magnetic field (taken to be $\frac{1}{2}$ G) for foil-to-detector flight paths of 56 and 861 cm would be 238° for a beam velocity of 1.7×10^9 cm/sec. Coupled with

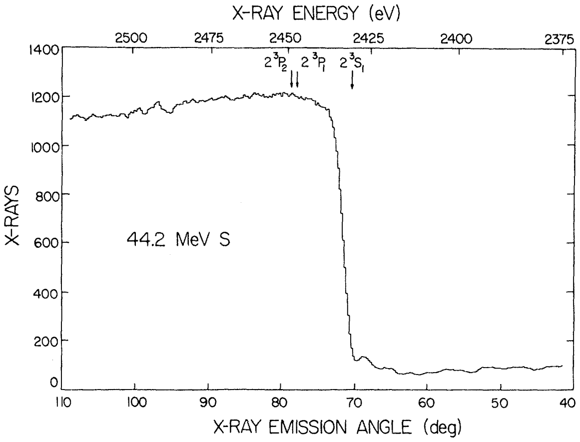

FIG. 4. High-resolution DTS energy spectrum of the x rays emitted from an excited 44.2-MeV S beam for a foil to detector separation of 56 cm.

anisotropic emission of x rays, such a spin alignment could cause the decay profile to depart from a single-exponential form. Since the net precession of a spin alignment for a given beam velocity is proportional to the magnetic field strength times the time during which the magnetic field acts, we simulated this possible precession by passing a 42-MeV foil-excited sulphur beam through the field region of an electromagnet which provided magnetic fields of up to 500 G over a flight path of 1.75 cm. X rays emitted from the excited beam were detected by a proportional counter viewing the beam after the magnetic field region, in a direction perpendicular to both the beam direction and the magnetic field direction. We looked for an oscillatory component in the number of x rays detected as a function of magnetic field strength. We can set an upper limit of $\frac{1}{2}\%$ of the total x-ray rate on the magnitude of any oscillatory component present. This is again too small an effect to account for the observed behavior.

The overall agreement between the results presented in Fig. 3 and the results of previous beam-foil measurements of the $2\,{}^3S_1$ lifetimes makes it clear that the non-single-exponential nature of the decay profiles for short flight times is responsible for the shorter lifetime reported previously for the case of chlorine.[2] However, we have not been able to find an entirely satisfactory explanation for this effect.

CONCLUSIONS

The results we have presented show that the assumption of a single-exponential x-ray decay profile from the $1\,{}^1S_0 \leftarrow 2\,{}^3S_1$ decay of heliumlike ions made in previous investigations of the lifetime of the $2\,{}^3S_1$ state is incorrect. We feel the previously reported discrepancies between experiment and theory on the value of the $2\,{}^3S_1$ lifetime in heliumlike chlorine[2] are due to this incorrect assumption. For foil-to-detector flight times $T \geq \frac{1}{2}\tau_{\text{theor}}$, the x-ray decay profiles are approximately a single exponential, and the experimental lifetimes of 706 ±86 nsec (sulphur) and 354 ±24 nsec (chlorine) are in agreement with theory.

---

*Supported in part by the Office of Naval Research and the Atomic Energy Commission.

[1] H. W. Moos and J. R. Woodworth, Phys. Rev. Lett. 30, 775 (1973).

[2] C. L. Cocke, B. Curnutte, J. R. Macdonald, and R. Randall, Phys. Rev. A 9, 57 (1974).

[3] H. Gould, R. Marrus, and R. W. Schmider, Phys. Rev. Lett. 31, 504 (1973).

[4] H. Gould and R. Marrus, Bull. Am. Phys. Soc. 19, 468 (1974).

[5] G. W. F. Drake, Phys. Rev. A 3, 908 (1971).

[6] W. R. Johnson and C. P. Lin, Phys. Rev. A 9, 1486 (1974).

[7] C. L. Cocke, B. Curnutte, and R. Randall, Phys. Rev. A 9, 1823 (1974).

[8] R. W. Schmieder, Rev. Sci. Instrum. 45, 687 (1974).

[9] H. T. Doyle, *Advances in Atomic and Molecular Physics*, edited by D. R. Bates (Academic, New York, 1969), Vol. 5, p. 337; R. E. Knight and C. W. Scherr, Rev. Mod. Phys. 35, 431 (1963).

[10] C. L. Cocke, B. Curnutte, J. R. Macdonald, J. A. Bednar, and R. Marrus, Phys. Rev. A 9, 2242 (1974).

[11] J. Shapiro and G. Breit, Phys. Rev. 113, 179 (1959); S. Klarsfeld, Phys. Lett. 30A, 382 (1969).

[12] L. C. Northcliffe and R. F. Schilling, Nucl. Data Tables 7, 233 (1970).

[13] R. Marrus (private communication).