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$2^2s_{1/2}$ STATE OF HYDROGEN-LIKE ARGON

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TWO-PHOTON DECAY AND LIFETIME OF THE $2^2s_{1/2}$ STATE OF HYDROGEN-LIKE ARGON[*]

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Abstract:

The lifetime of the $2^2s_{1/2}$ state of the hydrogen-like atom Ar XVIII has been measured by direct observation of spontaneous two-photon decay in a beam-foil time-of-flight experiment. Identification of the two-photon mode was made using photon counting techniques to observe the single-photon continuum, and the peak resulting from summing the energies of photon pairs detected in coincidence. The measured lifetime,

$$\tau(2^2s_{1/2}) = 3.54(25) \times 10^{-9} \text{ sec} \quad (95\% \text{ confidence})$$ is in excellent agreement with theoretical predictions.

The decay of the $2^2s_{1/2}$ state of hydrogen-like atoms has been a subject of long-standing theoretical interest. It was first shown by Breit and Teller[1] in 1940 that this state should decay to the $1^2s_{1/2}$ ground state primarily by the simultaneous emission of two photons. In this process, which we designate 2E1, the emission spectrum is a continuum, and the sum of the energies of the two photons equals the 1s-2s energy separation. Spitzer and Greenstein[2] invoked this mechanism to explain the continuous spectrum of planetary nebulae, and using non-relativistic theory, they accurately computed the predicted spectrum

and lifetime of metastable hydrogen. Later, Shapiro and Breit[3] obtained the

decay rate for a hydrogen-like atom of atomic number Z:

$$A_{2E1} \ (2^2 s_{1/2} - 1^2 s_{1/2}) = 8.226 \ Z^6 \ \text{sec}^{-1} \quad , \tag{1}$$

which agrees with Spitzer and Greenstein for $Z = 1$. Recently, a closed form

expression for the spectral distribution has been obtained[4] and several authors[5]

have theoretically treated the two-photon decay mode in the $2^1 S_0$ and $2^3 S_1$ states

of the helium isoelectronic sequence.

All of these calculations are non-relativistic, and consequently are

accurate only for small Z. Relativistic effects make it possible for

the $2^2 s_{1/2}$ state to decay to the $1^2 s_{1/2}$ state by magnetic dipole (M1) radiation,

a process that vanishes in the non-relativistic approximation. This mechanism

also was first noted by Breit and Teller, and recent calculations by Drake[6] and

by Schwartz[7] reveal that to relative accuracy $1/Z$, the M1 rate is

$$A_{M1} \ (2^2 s_{1/2} - 1^2 s_{1/2}) = 2.50 \times 10^{-6} \ Z^{10} \ \text{sec}^{-1} \quad , \tag{2}$$

and hence for $Z = 18$ contribute about 4% of the total decay probability of the

$2^2 s_{1/2}$ state.

Experimentally, the two photon decay mode was first observed in He II

by Lipeles, Novick, and Tolk,[8] who reported detection of coincidences and angu-

lar distribution measurements consistent with the $1 + \cos^2 \theta$ prediction. Rough

spectral measurements made with broad-band crystal filters also have been

reported.[9] A continuous spectrum observed in a plasma[10] has been attributed

to the two-photon decay of the $2^1 S_0$ state of helium-like atom Ne IX. The rela-

tivistic magnetic dipole mode has been observed recently in the solar corona and

in the laboratory.[12,13] Two recent measurements of the 2^1S_0 lifetime in He I,

which presumably decay by two-photon emission, have been reported,[15] but there

has so far been no measurement of the lifetime of the $2^2s_{1/2}$ state of any hydro-

gen-like atom.

In this letter we report the direct observation of the two-photon decay

mode in the hydrogen-like atom Ar XVIII using coincident photon counting techniques

and the measurement of the lifetime of the $2^2s_{1/2}$ state, using the beam-foil time-

of-flight method. The result is

$$\tau(2^2s_{1/2}) = 3.54(25) \times 10^{-9} \text{ sec} , \tag{3}$$

where the error indicates 95% confidence.

The apparatus used in this measurement has been described in previous

communications,[12-14] and only a brief summary is given here. Ions of ^{40}Ar in

the +14 charge state are accelerated in the Berkeley HILAC to an energy of

412 MeV (β = v/c = 0.148) and passed through a thin foil, from which they emerge

distributed among the +16 (helium-like), +17 (hydrogen-like), and +18 (fully

stripped) charge states. A significant fraction of the +16 and +17 ions may

emerge highly excited, but they undergo fast radiative or non-radiative de-exci-

tation to the ground or metastable states. The (forbidden) radiative decay of

the metastable states is detected in flight a few tens of cm downstream of the

foil by a pair of Si(Li) solid state x-ray detectors placed symmetrically per-

pendicular to the beam and about 2 cm from it. The photons were detected both

singly and in coincidence, using standard high-rate coincidence circuitry with

a resolving time $2\tau \sim 1$ μsec. In the singles mode, the energy   E1, E2 of every

detected photon in each detector was recorded. In the coincidence mode, the

detection of two photons in separate detectors within a time interval

$|T1-T2| \lesssim 5$ µsec was defined as a "coincidence," and resulted in the storage

of E1, E2, E1 + E2, and T1 - T2. With a typical beam current of 1 namp, single

counting rates were typically $10^2 - 10^3$ sec^{-1}, and coincidence rates were

$0.1 - 1$ sec^{-1}.

Since the lifetimes and spectra of the two-photon decays from 2^1S_0 and

$2^2s_{1/2}$ states are similar, it was necessary to discriminate against the helium-

like atoms in favor of the hydrogen-like atoms of interest here. To this end,

a two-foil technique was used. A thick (> 100 µg/cm^2) beryllium foil capable

of producing near charge equilibrium in the beam was placed ahead of the steering

magnet (at position A in Fig. 1 of Ref. 13). The steering magnet was then set

so that only fully-stripped ions were passed into our apparatus. The beam was

then passed through an extremely thin (< 10 µg/cm^2) carbon foil, much thinner than

is necessary to produce charge equilibrium. Since the capture of two or more

electrons in this thin foil is a less likely process than the capture of one

electron, the resulting beam has a substantially higher ratio of +17/+16 than

with the near charge-equilibrated beam.

A typical singles spectrum taken in this way, using a foil-detector

separation of 25 cm, is shown in Fig. 1. It consists of a broad continuum

between the detector threshold and E $\sim$ 3 keV, in general agreement with the

predictions.[2,4] The observed spectrum also includes the composite effects of

absolute detector efficiency (the step at 1.84 keV is due to absorption in the

detector at the K-edge of silicon), detector resolution ($\cong$ 0.2 keV), doppler

broadening due to the large acceptance angle of the detectors, low energy x-rays,

electronic noise, high energy background, and contributions from the decays of

any helium-like ions that might be present. Although the correction of the data

for all these effects is not yet complete, these data clearly verify the quali-

tative nature of the two-photon spectrum.

The purity of the beam as mostly hydrogen-like atoms can be assessed

in several ways: (1) by comparing spectra like Fig. 1 with similar spectra

taken with a nearly charge-equilibrated beam (measured to be 25% (+16), 50% (+17),

25% (+18); (2) by observing the spectrum at very large foil-detector separations

where only the magnetic dipole line[13] $2^3S_1 - 1^1S_0$ in Ar XVII is present; (3) by

measuring the sum energy of coincident photons (= 3.10 keV for Ar XVII, 3.30 keV

for Ar XVIII). Using these methods, we estimate that the contribution of helium-

like atoms to the two photon spectrum for the conditions of Fig. 1 is less than

a few percent.

To further verify the two-photon nature of the observed continuum, the

detectors were operated in the coincidence mode, so that only coincident events

(defined above) were stored. Figure 2a is a plot of the number of events versus

the time delay T1 - T2 between the two photons. The zero in this plot was

generated by introducing a fixed delay in one detector; it was calibrated using

a pulse generator to simulate a true coincidence. The peak in the time spectrum

of Fig. 2a is strong evidence that real coincident events are being observed.

The spectra observed as coincidences are shown in Figs. 2b and 2c. These

spectra represent only the true coincidences appearing under the peak of the time

spectrum in Fig. 2a. The contribution to that peak by accidental coincidences

was removed by subtracting the events occuring away from the peak (presumably

all accidentals), suitably normalized. The main difference between the singles

spectra in Figs. 1 and 2b is that the coincidence mode symmetrically discriminates

against the ends of the continuum, thus peaking it more strongly at the center.
The spectrum of the sum energy E1 + E2 observed as true coincidences is shown
in Fig. 2c. Except for residual noise due to accidentals which have been removed,
the spectrum is a single strong peak with a width roughly equal to the system
resolution, indicating that this peak represents a single line. That this line
falls at 3.3 keV is also strong evidence of the two-photon mode in Ar XVIII and
not Ar XVII. The fact that the singles spectrum observed in singles mode (Fig.
1) is the same as that observed in coincidence mode (Fig. 2b), and the associa-
tion of the latter (via Figs. 2a, 2c) with the 2E1 decay mode in Ar XVIII, per-
mits measuring the lifetime in the singles mode.

The decay of the $2^2s_{1/2}$ state was observed by varying the separation
between the foil and the detectors. A set of spectra like Fig. 1 was taken for
various separations (normalized to a fixed amount of integrated beam current),
and the total number of counts in the interval $0.75 \leqslant E \leqslant 2.5$ keV was obtained.
A plot of the normalized count rate versus distance is shown in Fig. 3. At large
distances the count rate levels off due to background and this is subtracted off
to leave the pure $2^2s_{1/2}$ decay. From the measured decay length and known beam
velocity, the mean (1/e) lifetime was determined. After a 1% correction for the
relativistic time dilation, the proper mean lifetime was found to be

$$\tau(2^2s_{1/2}) = 3.54(25) \times 10^{-9} \text{ sec.}$$

This result is in excellent agreement with the theoretical predictions.
The non-relativistic formula (1) predicts the value $\tau(2^2s_{1/2}) = 3.57 \times 10^{-9}$ sec.
If we combine this result with formula (2) for the relativistic magnetic dipole
decay using

$$\frac{1}{\tau} = A_{2E1} + A_{M1} \quad , \qquad\qquad\qquad\qquad (4)$$

the result $\tau(2^2s_{1/2}) = 3.47 \times 10^{-9}$ sec is obtained. Thus, our experimental results are in good agreement with the non-relativistic calculation.

FOOTNOTES AND REFERENCES

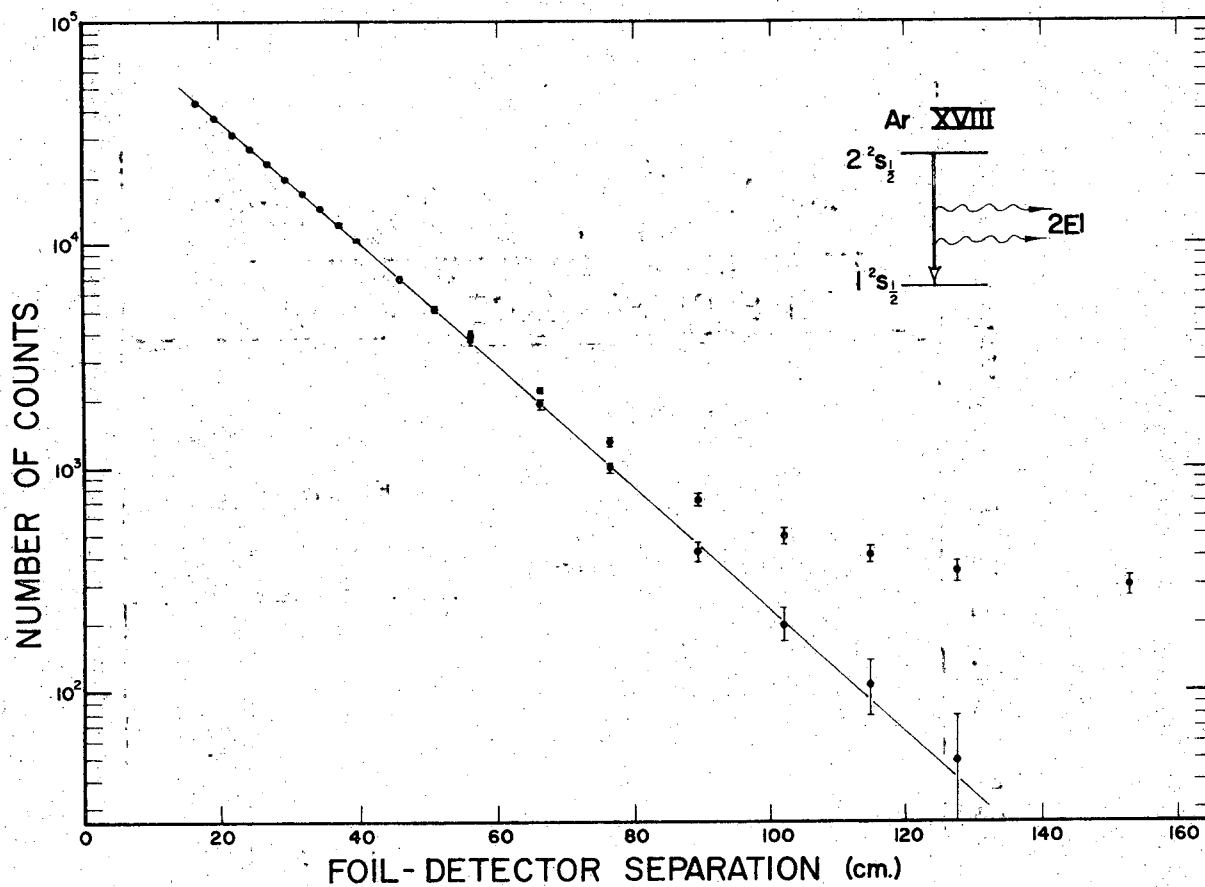* Work performed under the auspices of the U. S. Atomic Energy Commission.

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FIGURE CAPTIONS

Fig. 1. Typical energy spectrum observed in singles mode. This uncorrected

spectrum is predominantly the two-photon decay of metastable Ar XVIII. The

edge at 1.84 keV is an instrumental effect (see text). The detector reso-

lution is indicated.

Fig. 2. Spectra observed in coincidence mode: (a) Time difference between

coincident photon pairs. (b) Single photon energy spectrum, including only

events which participated in a true coincidence. (c) Sum of energies of

photons participating in a true coincidence.

Fig. 3. Decay curve obtained by recording the singles count rate versus foil-

detector separation. At large distances the observed counts level off to a

constant value, which was subtracted from each point to give the two-photon

counts. The error bars indicate the statistical error.

FOIL- DETECTOR SEPARATION (cm.)

XBL 709-6649

Fig. 3