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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,

$$\tau(2^2s_{1/2}) = 3.54(25) \times 10^{-9} \text{ sec (95% confidence)}$$

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 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 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:

\[
A_{2E1} \left( 2^2s_{1/2} - 1^2s_{1/2} \right) = 8.226 Z^6 \sec^{-1}, \tag{1}
\]

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 \(1^3S_0\) and \(2^3S_1\) 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 \(2^2s_{1/2}\) state to decay to the \(1^2s_{1/2}\) 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

\[
A_{M1} \left( 2^2s_{1/2} - 1^2s_{1/2} \right) = 2.50 \times 10^{-6} Z^{10} \sec^{-1}, \tag{2}
\]

and hence for Z = 18 contribute about 4% of the total decay probability of the \(2^2s_{1/2}\) 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 angular 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 \(1^3S_0\) state of helium-like atom Ne IX. The relativistic magnetic dipole mode has been observed recently in the solar corona and
in the laboratory. Two recent measurements of the $2^1S_0$ lifetime in He I, which presumably decay by two-photon emission, have been reported, but there has so far been no measurement of the lifetime of the $2^2s_{1/2}$ state of any hydrogen-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

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

where the error indicates 95% confidence.

The apparatus used in this measurement has been described in previous communications, 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 ($\beta = v/c = 0.148$) 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-excitation 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 perpendicular 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 $2\tau \sim 1 \mu\text{sec}$. In the singles mode, the energy $E_1, E_2$ 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 
\[|T_1 - T_2| < 5 \text{ usec}\] was defined as a "coincidence," and resulted in the storage of \(E_1, E_2, E_1 + E_2,\) and \(T_1 - T_2.\) With a typical beam current of 1 namp, single counting rates were typically \(10^2 - 10^3 \text{ sec}^{-1}\), and coincidence rates were \(0.1 - 1 \text{ sec}^{-1}\).

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 \(\mu\text{g/cm}^2\)) 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 \(\mu\text{g/cm}^2\)) 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 \(E \sim 3 \text{ keV}\), 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 (\(\approx 0.2 \text{ keV}\)), 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 qualitative 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 $^{13}S_{\perp} - ^{1}S_0$ 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 $T_1 - T_2$ 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 occurring 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 \( E_1 + E_2 \) 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 association of the latter (via Figs. 2a, 2c) with the 2E1 decay mode in Ar XVIII, permits 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 \( 0.75 < E < 2.5 \) 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

\[
\frac{1}{\tau} = A_{2E1} + A_{M1},
\]
the result $\tau(2^2s_{1/2}) = 3.47 \times 10^{-9} \text{ sec}$ is obtained. Thus, our experimental results are in good agreement with the non-relativistic calculation.

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FOOTNOTES AND REFERENCES

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6. G. W. F. Drake, University of Windsor (to be published).
7. C. Schwartz, University of California, (private communication).
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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 resolution 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.
Fig. 3
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