# Two-Phonon Decay and Lifetime of the $2^2s_{1/2}$ State of Hydrogenlike Argon*

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The lifetime of the $2^2s_{1/2}$ state of the hydrogenlike 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, $\tau(2^2s_{1/2}) = 3.54(25) \times 10^{-9}$ sec (95% confidence) is in excellent agreement with theoretical predictions based on a nonrelativistic calculation.

The decay of the $2^2s_{1/2}$ state of hydrogenlike 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 nonrelativistic theory, they accurately computed the predicted spectrum and lifetime of metastable hydrogen. Later, Shapiro and Breit[3] obtained the decay rate for a hydrogenlike atom of atomic number $Z$:

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

which agrees with the calculation by 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 $2^1S_0$ and $2^3S_1$ states of the helium isoelectronic sequence.

All of these calculations are nonrelativistic, 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 nonrelativistic 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

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

and hence, for $Z = 18$, contributes 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 broadband 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 $2^1S_0$ state of heliumlike atom Ne IX. The relativistic magnetic-dipole mode has been observed recently in the solar corona[11] and in the laboratory.[12] Two recent measurements of the lifetime of the $2^1S_0$ level in He I, which presumably decays by two-photon emission, have been reported,[13] but there has so far been no measurement of the lifetime of the $2^2s_{1/2}$ state of any hydrogenlike atom.

In this Letter we report the direct observation of the two-photon decay mode in the hydrogenlike 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

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

where the error indicates 95% confidence.

The apparatus used in this measurement has been described in previous communications,[12,14,15] and only a brief summary is given here. Ions of $^{40}$Ar in the +14 charge state are accelerated in the Berkeley heavy-ion linear accelerator 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 (heliumlike), +17 (hydrogenlike), 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 nonradiative de-excitation to the ground or metastable states. The (forbidden) radiative decay of the metastable states is detected in flight a few tens of centimeters downstream of the foil by a pair of Si(Li) solid-state x-ray detectors placed on opposite sides of 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 $2\tau \sim 1$ $\mu$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 $|T_1-T_2| \lesssim 5$ $\mu$sec 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 nA, single counting 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 heliumlike atoms in favor of the hydrogenlike atoms of interest here. To this end, a two-foil technique was used. A thick (>100 $\mu$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. 12). 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 $\mu$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.

The purity of the beam as mostly hydrogenlike 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, and 25% +18); (2) by observing the spectrum at very large foil-detector separations where only the magnetic dipole lines[12] $2^3S_1$-$1^1S_0$ 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 heliumlike atoms to the two-photon spectrum for the conditions of Fig. 1 is less than a few percent.

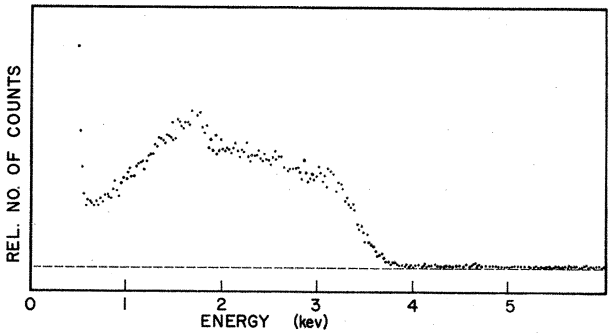


FIG. 1. Typical energy spectrum observed in singles mode. This uncorrected spectrum is predominantly the two-photon decay of metastable Ar XVIII. The edge at 1.84 keV is an instrumental effect (see text). The detector resolution is indicated.

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 heliumlike ions that might be present. Although the correction of the data for all these effects is not yet complete, these data clearly, if qualitatively, verify the two-photon nature of the spectrum.

To substantiate 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 2(a) 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. 2(a) is strong evidence that real coincident events were observed.

The spectra observed as coincidences are shown in Figs. 2(b) and 2(c). These spectra represent only the true coincidences appearing under the peak of the time spectrum in Fig. 2(a). 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 2(b) 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. 2(c). 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

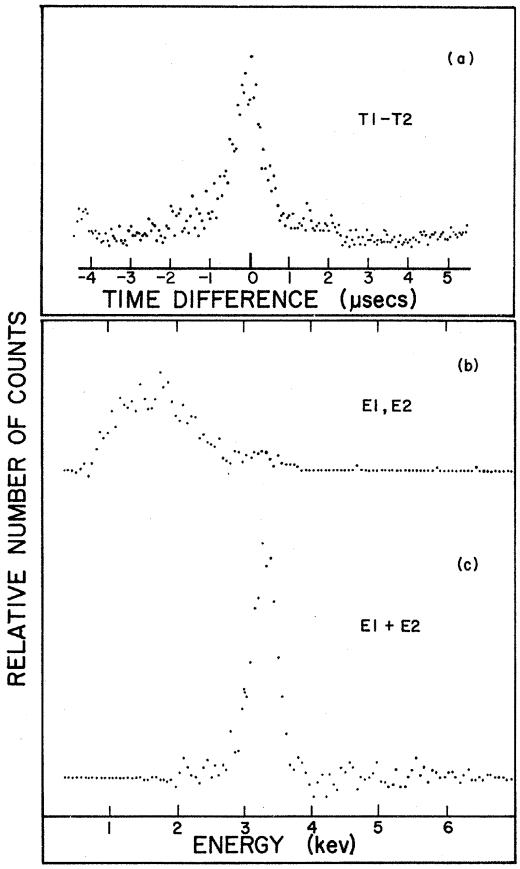


FIG. 2. Spectra observed in coincidence mode: (a) Time difference between coincident photon pairs. (b) Single-photon energy spectrum, including only events which participated in a true coincidence. (c) Sum of energies of photons participating in a true coincidence.

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. 2(b)], and the association of the latter [via Figs. 2(a) and 2(c)] 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 $0.75 \leq E \leq 2.5$ keV was obtained. A plot of the normalized counting rate versus distance is shown in Fig. 3. At large distances the counting rate levels off to that 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 dilatation, the proper mean lifetime was found to be $\tau(2^2s_{1/2}) = 3.54(25) \times 10^{-9}$ sec.

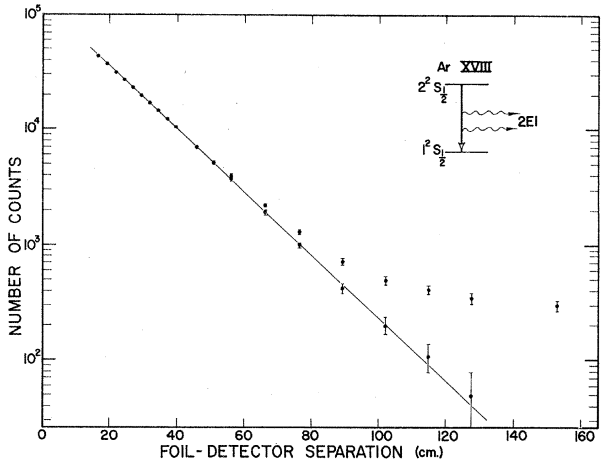


FIG. 3. Decay curve obtained by recording the singles counting rate versus foil-detector separation. At large distances the observed counts level off to a constant value, which was subtracted from each point to give the two-photon counts. The error bars indicate the statistical error.

This result is in excellent agreement with the theoretical predictions. The nonrelativistic 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

$$1/\tau = A_{2E1} + A_{M1}, \tag{4}$$

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 nonrelativistic calculation.

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# Many-Body Perturbation Calculation of the Indirect Spin-Spin Coupling Constant in HD Molecule*

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The linked-cluster many-body perturbation approach has been applied to the study of indirect nuclear spin-spin coupling constant $J_{HD}$ in HD molecule. The complete set of states used were the bound and continuum states of $H_2^+$ molecular ion with the internuclear separation appropriate to $H_2$ molecule. Our calculated value of $J_{HD}$ through the Fermi contact interaction mechanism is +42.57 Hz in good agreement with the most recent experimental value of $+42.7 \pm 0.7$ Hz.

Although the indirect spin-spin interaction between nuclei in molecules has been utilized extensively for qualitative understanding of electronic structures in molecules, its quantitative calculation even for the simple molecule HD has proved to be a formidable task, with the current situation far from conclusive.[1-8] The various theoretical approaches utilized so far can be divided broadly into two categories. In the first category is the conventional second-order perturbation as first proposed by Ramsey and Purcell.[1,2] The difficulty with such an approach is that one requires a knowledge of the complete set of ground and excited states of the molecule, which is not usually available.[3]

To obviate the knowledge of excited states, variation-perturbation procedures have been used by a number of authors.[4-8] In one class of such calculations, a diagonal-type perturbation procedure[4,5] was used, the second-order energy due to the hyperfine field of one nucleus being minimized to obtain the first-order perturbed wave function of the molecule. This function was then utilized to calculate the cross term in the second-order energy involving the other nucleus to obtain $J_{HD}$. The difficulty with this procedure was that the second-order nuclear self-coupling energy is infinite in nonrelativistic theory, no real minimum thus being attainable in a variational approach. A second class of variation perturbation calculation has attempted to extremize the cross terms in the second-order energy proportional to $J_{HD}$ using variational functions which describe the first-order perturbation due to both nuclei.[6-9] The difficulty with this procedure is that the cross term due to two perturbations has by itself no minimum and one does in fact get oscillatory behavior as the number of parameters increased.[9]

In this paper, we have revived the perturbation approach in a form that meets the major difficulty, namely, a knowledge of a complete set of states for the molecule. This is accomplished by using the linked-cluster many-body perturbation theory (LCMBPT),[10] where a neighboring Hamiltonian $\mathcal{H}_0$, for which the complete set of states can be obtained exactly, is used as the starting point for a perturbation treatment of $\Delta\mathcal{H} = \mathcal{H} - \mathcal{H}_0$. In our work here, a multiple perturbation approach is used in conjunction with the LCMBPT, using the sum of $\Delta\mathcal{H}$ and the two hyperfine interaction Hamiltonians $\mathcal{H}_H'$ and $\mathcal{H}_D'$ associated with the two nuclei. For the speed of convergence of the perturbation approach, it is nec-