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Lamb Shift in Hydrogenlike Argon

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The Lamb shift S in hydrogenlike argon (Z = 18) has been measured by a motional-electric-field-quenching time-of-flight experiment. We find $S = 38.0 \pm 0.6$ THz where the error includes 1-standard-deviation statistical error of 0.35 THz and an estimated systematic error of 0.48 THz.

The comparison between Lamb shift theory and experiment in hydrogenlike ions of high nuclear charge Z is a sensitive test of quantum electrodynamics in the region where higher-order corrections to the binding energy are large.¹ To data, Lamb-shift measurements on hydrogenlike ions through Z = 9 have been reported.² In this Letter we report a measurement of the $2S_{1/2}$ - $2P_{1/2}$ energy splitting (Lamb shift) of hydrogenlike argon (Z = 18). We find $S = 38.0 \pm 0.6$ THz which may be compared to values of S = 39.0 ± 0.16 THz calculated by Erickson³ and S = 38.25 ± 0.025 THz calculated by Mohr.⁴ This comparison constitutes the most rigorous test of quantum electrodynamics at high field strengths.¹

Our experiment is performed by measuring the electric-field-quenched lifetime of the metastable $2S_{1/2}$ state. At Z = 18 the unperturbed state decays predominantly by the simultaneous emission of two electric-dipole photons. A decay rate⁵ of $2.76 \times 10^8 \text{ sec}^{-1}$ is associated with this mode. There is also a relatively slow, single-photon magnetic-dipole channel with a rate⁵ $A = 9.08 \times 10^6 \text{ sec}^{-1}$. The frequency distribution of the emitted photons therefore consists of a continuous spectrum centered about 1659 eV falling rapidly to zero at the end points, plus a small peak at the $2S_{1/2}$ - $1S_{1/2}$ energy separation of 3318 eV.

The application of an external electric field mixes the wave functions of the $2S_{1/2}$ state with the *nP* states. Since *P* states decay by fully allowed electric-dipole radiation, the $2S_{1/2}$ lifetime is shortened. To lowest order the decay rate *R* of the $2S_{1/2}$ state as a function of electric field is given by⁶

 $R = R_{s} + R_{b} [V]^{2} / [\hbar^{2} (S^{2} + R_{b}^{2} / 4)],$

where R_s and R_p are the natural 2S and 2P decay

rates, S is the Lamb shift in radians per second, and V is the relativistic electric-dipole matrix element: $V=0.992\sqrt{3}eEa_0/Z$, where e is the electron charge, E the electric field in electrostatic units, and a_0 the Bohr radius. An electric field in the atom's rest frame is produced by passing atoms at a velocity of $v = 4 \times 10^9$ cm/sec through a homogeneous magnetic field. At field of B = 16kG, the 6.5×10^5 -V/cm electric field produces a quenching rate comparable to the natural $2S_{1/2}$ decay rate. The Lamb shift can thus be determined by measuring the velocity, magnetic field, and quenched lifetime.

The magnetic field is measured with an integrating flux meter calibrated against a nuclear magnetic resonance gaussmeter. The velocity is determined to a few parts per thousand by measuring the beam energy (approximately 340 MeV) with surface-barrier detectors which have been calibrated against a magnetically analyzed argon beam at 346 MeV. Errors from these velocity and magnetic field measurements make no significant contribution to the Lamb-shift error.

Lifetimes of the $2S_{1/2}$ state are measured by beam-foil time of flight. Bare argon nuclei, at velocities of 4×10^9 cm/sec, obtained from the Lawrence Berkely Laboratory SuperHILAC, are passed through an $8-\mu g/cm^2$ carbon foil located in a homogeneous magnetic field (Fig. 1). Among the approximately 10% of nuclei which capture electrons the ratio of hydrogenlike atoms to heliumlike atoms is 17:1. The lithiumlike fraction was too small to detect. From charge-exchange measurements⁷ of Ar⁺¹⁸ in N₂ we estimate an upper limit to the lithiumlike fraction of 1 part in 200 of the hydrogenlike fraction. No Ar⁺¹⁷ was observed in the absence of a foil. The cross section⁷ for Ar⁺¹⁸ \rightarrow Ar⁺¹⁷ in N₂ is $(1.1 \pm 0.3) \times 10^{-18}$



FIG. 1. Schematic diagram of the apparatus. Sideways-looking x-ray detectors are collimated to view approximately a 2-cm-long segment of beam.

 $cm^2/molecule$ at 8.5 MeV/amu. Thus in the 5mTorr pressure inside our apparatus the mean free path of Ar^{+18} exceeds 10^6 cm. X rays from decays in flight of the excited states are observed at a distance of approximately 0.6 m from the beam by a pair of Si(Li) x-ray detectors. A typical pulse-height spectrum is shown in Fig. 2. The raw data for the $2S_{1/2}$ decay curves are the pulse-height spectra measured as a function of x-ray detector position downstream of the foil. The intensity is normalized to the integrated current obtained by stopping the beam in a Faraday cup. The comparable intensities in the quench peak and two-photon spectra enable us to construct decay curves from either portion of the spectra.

The raw data must be corrected for deflection of the beam in the magnetic field. In addition to the change in solid angle and change in length of the beam viewed, there is a large change in brightness due to the motion of the deflected beam toward (or away from) the detectors.⁸ Representative correction factors for the change in counting rate due to deflection in a magnetic field of 20 kG are given in Table I. The change in the direction of travel of the beam allows both detectors to view a longer segment of the beam, increasing the apparent counting rate, and results in a lack of symmetry in the correction fac-



FIG. 2. Pulse-height spectrum from decays in flight of the $2S_{1/2}$ state of hydrogenlike Ar in a motional electric field of 7.8×10^5 V/cm at a foil-detector separation of 9 cm. The detector has a resolution (full width at half maximum) of 280 eV and the spectrum is Doppler shifted 4% as a result of the deflection of the beam in the magnetic field. Below 1500 eV the sensitivity of the detector drops rapidly to zero. In addition to the 3318-eV line from 1γ decay of hydrogenlike Ar there are weaker lines at 3126 and 3104 eV from decays of $2^{3}P_{2}$ and $2^{3}S_{1}$ states of heliumlike Ar. The hydrogenlike line can be separated from the heliumlike lines by computer fit.

tors for the two detectors as shown in Table I. Our model was tested by deflecting beams of heliumlike argon in the long-lived $2^{3}S_{1}$ state, so that changes in counting rate were primarily due to deflection effects rather than population depletion. At current levels of accuracy the uncertainties in the geometrical corrections make a negligible contribution to the experimental error.

Most of the hydrogenlike argon forms in the $1S_{1/2}$ ground state or low-lying excited states. Among the atoms formed in highly excited states, few cascade through the $2S_{1/2}$ state, because de-

TABLE I. Counting-rate and path-length corrections for a magnetic field of 20 kG. In column (a) the beam is deflected toward detector; in column (b) the beam is deflected away from detector.

Detector position (cm)	Path length (cm)	Relative counting rate	
		(a)	(b)
3.72	3.84	1.10	0.904
7.53	7.80	1.18	0.889
11.33	11.86	1.28	0.887
16.43	17.54	1.46	0.867
20.24	22.0	1.67	0.866

cay chains which populate the $2S_{1/2}$ state are far more likely to decay to the $1S_{1/2}$ state because of a much larger energy separation. Furthermore, atoms undergoing transitions in which the principal quantum number n decreases more rapidly than the orbital angular momentum l have a high probability of reaching states of maximum l = n - 1which are long lived and decay through the chain \dots , 4F, 3D, 2P, 1S. An upper limit to the cascading to the $2S_{1/2}$ state can be made from examination of the spectra (Fig. 2) as follows. Assume that all of the counts in the (240 eV Gaussian width) region 4000-4700 eV are from (Doppler shifted) decays $nP \rightarrow 1S$ (n > 2). The branching ra $tio^9 (nP \rightarrow 1S):(nP \rightarrow 2S) (n > 2)$ is about 7.5:1. By comparing the number of counts in the two-photon continuum with the $(n > 2)P \rightarrow 1S$ rate we find an upper limit of 0.2% of the observed two-photon counts originating from cascades to the $2S_{1/2}$ state, an insignificant contribution. At larger foil-detector separations the upper limit is much smaller. We emphasize that this upper limit is derived from purely experimental quantities and is model independent. The large energy gap between the series limit of the Balmer series at 1100 eV and the beginning of the Lyman series at 3318 eV assures the absence of interfering hydrogenlike lines in this region of the two-photon spectra.

In the observation of single-photon decay of the $2S_{\scriptscriptstyle 1/2}$ state, interference between the unresolved spectrum lines $2S_{1/2}$ + $1S_{1/2}$ and the cascade-fed $2P_{3/2,1/2} \rightarrow 1S_{1/2}$ results in a large systematic error in determining the $2S_{1/2}$ decay rate. In measurements of unquenched $2S_{1/2}$ lifetimes in hydrogenlike argon and hydrogenlike iron (Z = 26) we observe a significant number of counts arising from cascades through the 2P state. In the absence of information about the populations of the different nl states these cascade data are not directly applicable to quenching experiments as the electric field causes appreciable mixing of states of different parity. For the present, we base our Lamb-shift determination only upon $2S_{1/2}$ decay rates obtained by observing the two-photon decay channel.

Heliumlike argon present in the beam in small quantities has metastable states $2^{3}P_{2}$ and $2^{3}S_{1}$ whose decay produces x rays at 3126 and 3104 eV, respectively.¹⁰ (Counts from these lines can be separated from the 3318-eV line from hydrogenlike argon by a careful computer fit to the spectra using the measured x-ray detector response function.) However, the 2.3-nsec two-photon decay (2*E*1) of the $2^{1}S_{0}$ state in heliumlike $\operatorname{argon}^{10}$ is a continuum and cannot be distinguished from the two-photon decay spectrum of the $2S_{1/2}$ state of hydrogenlike argon. From observed population ratios $2S_{1/2}$: $2^{3}P_{2}$ in this experiment and population ratios $2^{2}P_{2}$: $2^{1}S_{0}$ in experiments on helium-like argon performed by Marrus and Schmieder,¹⁰ we find the initial population of $2^{1}S_{0}$ to be 0.03 \pm 0.01 of the initial $2S_{1/2}$ population. The uncertainty in the $2^{1}S_{0}$ fraction is the largest source of systematic error in this Lamb-shift measurement.

Lifetimes were measured over an average of 2.5 decay lengths in electric fields of \pm (5.93, 7.14, 8.06, 8.60) $\times 10^5$ V/cm with two detectors for a total of sixteen decay curves. Each decay curve contains at least 1.5×10^4 counts distributed among at least fourteen detector positions. Position-dependent background (which appears to be the result of gamma rays, Compton scattered in the detector) was measured as a function of detector position (see Fig. 1) and magnetic field with the foil 0.75 m upstream to allow the $2S_{1/2}$ state to depopulate before reaching the detectors. The background accounts for approximately 2%of the counting rate at a foil-detector separation of 5 cm and approximately 10% at 20 cm. The measured lifetimes corresponding to the electric fields are 1.87 ± 0.03 . 1.58 ± 0.06 . 1.40 ± 0.02 . and 1.29 ± 0.02 nsec, respectively. The data were sorted to yield Lamb-shift values as a function of field strength and polarity, direction of beam bending, direction of detector travel, and counting rate. No systematic differences were observed.

The result for the Lamb shift is 38.0 ± 0.6 THz. Our Lamb-shift value is an unweighted average of all sixteen measurements and the statistical error is the standard deviation of the mean. Contributions to the error are statistics; 0.35 THz which is mostly due to variation in the background; 0.45 THz, from uncertainty in the $2^{1}S_{0}$ fraction; and 0.15 THz, miscellaneous.

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Doppler-Tuned Hyperfine Spectroscopy of the Lithium Ion

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We have used the Doppler-tuned ion-beam, laser-spectroscopy method to measure the hyperfine intervals in the $2^{3}S$ and $2^{3}P^{\circ}$ states of ⁷Li⁺. Our resolution was 10 times that of the best previous measurement. In contrast to the earlier experimental work we find excellent agreement between our measurements and the theoretical values.

The fine and hyperfine structure of the $np^{3}P^{\circ}$ and $ns^{3}S$ states of the helium isoelectronic series are of considerable interest in atomic physics because their relative simplicity allows precise theoretical calculations. The fine structure of the $n^{3}P^{\circ}$ states (n = 2-5) has been calculated up to Z = 10 by Schiff and co-workers.¹ Their calculations included the contributions from the masspolarization correction, relativistic effects of order α^2 , and mixing of the singlet and triplet states, but not the Lamb-shift correction. For Li⁺, the predominance of the spin-spin magnetic interactions causes the three fine-structure levels of the $2^{3}P^{\circ}$ state to be partially inverted, so that a precision measurement makes possible a different test for the correctness of the two-electron, spin-spin Hamiltonian compared to helium where spin-spin and spin-orbit terms contribute about equally.² Jette, Lee, and Das³ have used the linked-cluster many-body perturbation method to calculate the contributions of the magnetic contact, dipolar, and orbital hyperfine interactions to the hyperfine structure of the $2^{3}P^{\circ}$ state of lithium ions. Their results agree to about 1 part in 10⁴ with the more recent numerical calculations by Aahamar and Hambro.⁴ Questions have been raised by the recent optical^{5,6} and beam-foil⁷ experimental results, deduced from incompletely resolved hyperfine structures for Li⁺, which showed significant discrepancies with

the accurate many-body calculations.⁸ Thus, it is important to make our independent high-resolution measurements.

It was first demonstrated by Wing *et al.*⁹ that the velocity-bunching effect, where the energy spread remains the same but the velocity spread is reduced with higher energies for accelerated ions, can be utilized in high-resolution laser spectroscopy. Linewidths can be significantly reduced, and the signal from the low-density, fast ion beam can thereby be increased by efficient collinear excitation with monochromatic laser light. This technique has been used in the spectroscopy of molecular HD⁺ ions, ⁹ Ba⁺ ions, ¹⁰ Xe⁺ ions, ¹¹ and neutral Na and Cs atoms.¹²

We have used this ion-beam, laser-spectroscopy method to measure the hyperfine intervals of the metastable 2^{3} S and the $2^{3}P^{\circ}$ states depicted in Fig. 1 for the lithium ion ⁷Li⁺. In Fig. 2, our observed linewidth is seen to be $\frac{1}{10}$ that of the most accurate previous optical measurement,⁶ where the linewidth was due to Doppler broadening from a liquid-nitrogen-cooled hollow-cathode lamp. Our narrow linewidth allowed us to resolve completely all the hyperfine transitions and to thereby obtain the hyperfine intervals with much improved accuracy. Our results, presented in Table I, show that in contrast to the previous work, we obtain excellent agreement with the theory of Jette, Lee, and Das² for all the