Collinear laser-beam–ion-beam measurement of the mean lifetime of the Ar II $4p'\,^2F^\circ_{7/2}$ level

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The mean lifetime τ of the $4p'\,^2F^\circ_{7/2}$ level of Ar II has been measured using a variant of the collinear laser-beam–fast-ion-beam spectroscopy technique. Our variant requires no mechanical motion or laser frequency tuning. The result is $\tau=8.414\pm0.025$ ns in agreement with an earlier, comparably precise, measurement based on a crossed laser–ion-beam technique. The result lies above all theoretical calculations.

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I. INTRODUCTION

The continued development of many-body perturbation-theory (MBPT) techniques now permits calculation of the mean lifetimes of excited states of certain atoms and ions, e.g., those having one electron outside closed shells, with accuracy and precision exceeding 1% [1]. Improved configuration-interaction calculations are also approaching the 1% level [2]. These calculations supercede those using earlier theoretical methods, which have uncertainties ranging from 5% to 25% or more. The MBPT calculations agree with the most precise and accurate data for electric dipole ($E1$) transitions [3] at the 1% level, although deviations between theory and experiment still occur at higher precisions [1]. Disturbingly, large discrepancies between theory and experiment have been found between magnetic dipole ($M1$) or electric quadrupole ($E2$) transitions in ions [4]. Since the calculational techniques are being extended to other isoelectronic sequences and to other configurations, improved mean lifetime data for both prompt and metastable transitions are needed to evaluate the efficacy of the approximations used in the perturbation-theory expansion.

The most precise mean lifetime measurements have been carried out on fast atoms by observing the optical decay in flight following excitation using a laser beam which crossed the ion beam [3]. This technique avoided the measurement complications associated with optical cascades from higher excited levels, since the excitation using the laser was selective. A variant of this technique employed the laser and ion beams in a collinear arrangement [5]. A further significant advantage of this latter method arises from the use of the Doppler effect to tune the ions into resonance with the laser, thus permitting the optical decays to be resolved from the exciting laser wavelength, reducing background effects. In the initial applications of this collinear method, the laser was tuned to excite the ions at different spatial positions, although the ion velocity could be swept by electrostatic potential variations. Unfortunately, in the initial tests of this method, two calibration measurements yielded conflicting results [5,6]. A measurement of the mean lifetime of the $6p\,^2P^\circ_{3/2}$ transition of Ba^+ was in excellent agreement with an earlier crossed-beam laser-ion measurement, but a similar comparison of the mean lifetime of the $6p\,^4P^\circ_{5/2}$ transition of Xe^+ by the two methods differed by about 5%, well outside expected error limits. No further measurements using the collinear "rapid Doppler switching" method of Gaillard and co-workers [5,6] were carried out. More recently, however, the calibration disagreement has been resolved in favor of the results of the rapid Doppler switching technique, by an independent crossed laser-ion beam measurement [7]. Consequently, the measurement scheme of Gaillard *et al.* [5] is capable of yielding both precise and accurate data, and its other advantages make it the method of choice for many applications. In particular, the use of ion velocity sweeping by variations in electrostatic potential along the ion-beam path is particularly useful. Thus the measurement of an ion excited-state mean lifetime can be carried out without any mechanical motion of the apparatus, and without changing the laser frequency, reducing potential problems with variations in background or signal due to changes in scattered light intensity and in beam direction associated with laser tuning.

For our inaugural measurements using this latter variation of the technique, we chose to measure the mean lifetime of the $4p'\,^2F^\circ_{7/2}$ level of Ar II. The mean lifetime of this level had very recently [8] been obtained using the fast-ion-beam–crossed-laser-beam technique with a precision of $\pm0.36\%$. This level was excited from either of the metastable $3d'\,^2G_{9/2,7/2}$ levels, which were populated in the ion source. The $^2F^\circ_{7/2}$ level has optical decays at several wavelengths, permitting a measurement free of background scattered light from the laser, while using only minimal resolution at the signal wavelength, provided either by a filter or a low-resolution monochromator. The result of the measurement is the mean lifetime $\tau=8.414\pm0.025$ ns, in excellent agreement with the earlier laser-based measurement, but with significant improvements in accuracy and precision compared to all other techniques. The closest theoretical result lies 3% below our experimental value.

II. EXPERIMENTAL OVERVIEW

Our experimental method is based on the technique developed by Gaillard *et al.* [5], which uses a collinear ion-beam–laser-beam geometry to allow maximal use of

the Doppler effect. The change in position along the joint beam axis between excitation and detection locations is obtained by varying the ion velocity, and so is restricted in its use to ions. As originally introduced by Gaillard *et al.* [5], the laser frequency was scanned, to change the spatial location of the resonance with the accelerated ions. Our implementation of the technique differs in that the laser wavelength is also fixed, and only the ion speed is changed by applying accelerating voltages over limited portions of the ion-beam path, an alternative mentioned by Gaillard *et al* [5], but not actually employed. The particular advantage of our wholly static method is that changes in laser intensity or overlap of the two beams associated with tuning are minimized.

The ions interact with the laser beam in a small portion of a region of uniform electric field produced by a fixed potential difference W across an array of plates, as discussed below, i.e., in a region of constant acceleration. Let z_0 specify the center of a narrow ($\cong 1$ mm long) spatial region between the plates as shown schematically in Fig. 1, from which optical decay photons are collected. For a laser tuned to a wavelength λ_L, ions which absorb at a wavelength centered at λ_0 can be Doppler shifted to resonance if $\lambda_0=\lambda_L(1\pm\beta)\gamma$ with $\beta=v_0/c$ and $\gamma=(1-\beta^2)^{-1/2}$, where v_0 is the ion speed at position z_0. The ion speed v_0 is determined by the potential U of the ion source relative to the entrance plate, and by the acceleration of the ions between the plates. If U is increased, the ions will be in resonance with the laser at a position $z_1<z_0$, where their speed is still v_0 due to the shorter time that the constant acceleration acts. Thus the position z_1 in space at which the ions are excited is determined by U. By sweeping U, the spatial interval z_1-z_0, and due to the ion speed also the time interval between excitation of the ions and detection of their subsequent optical emission, are varied. Since the ion speed is near 4×10^7 cm/s in these measurements, this interval is in the nanosecond range. A measure of resonantly scattered light intensity versus U provides a measure of the atomic mean lifetime.

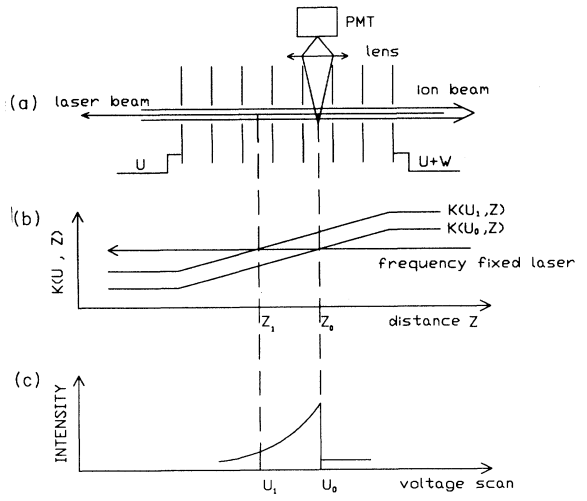

FIG. 1. Principle of the rapid Doppler switching technique. (a) Schematic of the apparatus. The potential difference W across the plate array produces a uniform electric field along the collinear ion-laser beam. (b) Ion kinetic energy as a function of position between the field plates and the potential U. (c) Expected fluorescence intensity change as the potential U is scanned.

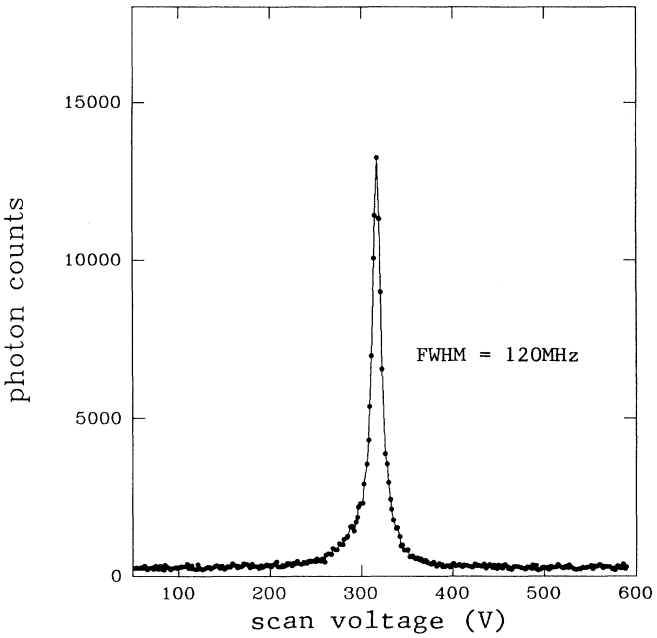

FIG. 2. Ar II $3d'^2G_{9/2}$-$4p'^2F^\circ_{7/2}$ resonance spectrum. The solid line is used to guide the eye.

If the potential difference between the plates W is reduced to zero and the potential U is scanned, the ions can be tuned through resonance to determine the linewidth of the excitation. In this instance, the resonance condition of the ion transition with the laser beam occurs over the whole region of plate separation. Again, the observed signal is photons from an optical decay transition at a wavelength different from that of excitation, observed as a function of voltage applied to the whole plate assembly. An example of a signal of this type appears in Fig. 2. The line shape is asymmetric, since ions excited upbeam from the location of light collection can decay into the detection system, while ions excited downbeam cannot, as the resonance is scanned. The asymmetry has been discussed in detail by Short *et al.* [9]. The width of the resonance is also determined by the energy spread of the ions in the ion source, by the spectral width of the laser line, and by the stability of the applied potentials.

III. APPARATUS AND TECHNIQUE

The Ar^+ ions were produced in a Danfysik 911A ion source at a potential near 30 kV. The ion source dc potential was measured to an accuracy better than 0.25% using a precision voltage divider, but there was additionally an ac ripple of 0.1%. After acceleration, the ions were charge-to-momentum selected using a small bending magnet, and focused into the collinear spectroscopy observation region through two collimating apertures, which had holes 0.635 cm in diameter and were located 50 and 100 cm upbeam from the entrance plate. Beam

currents near 4 μA were typically used. A small fraction of the ions were produced in the source in the metastable $3d'^2G_{9/2,7/2}$ states, for which the Ar^{2+} core is the highly metastable $3p^4\,^1D_2$ level. About 0.2% of these ions survived to the observation region, based on estimates of the efficiencies of ion excitation by the laser, and of detection of the optical decay photons. By using the measured laser excitation wavelength, obtained to $\pm 10^{-2}$ Å using a Burleigh WA-20VIS wavemeter, and by observing the dc potential U yielding the maximum intensity of excitation, calculations using the Doppler effect provided an accuracy of voltage determination comparable to the dc measurement precision.

To precisely obtain the electric field at the position of interaction of the laser and ion beams, the electric field plates were constructed according to the tested method specified by Gaillard *et al.* [5] to minimize edge effects, the principal source of error. They established that a similar plate assembly produced an electric field with accuracy and precision measured to better than 1%. A stack of eight plates, separated by insulators with a length of 9.276±0.01 mm, were used. The plates were 76 mm in diameter. The entrance and exit holes were 7.9 mm in diameter, while the interior guard ring plates had holes 8.8 mm in diameter. Precision dropping resistors which were matched to improve relative variations to 2×10^{-4} were placed between the plates. The effects of the apertures of the plates and the 0.38-mm thickness of each plate were determined analytically along the symmetry axis, and calculated numerically. If E_0 denotes the electric field $\Delta W/D$ between two infinite plates without apertures, separated by a distance D, then the variation of the field along the symmetry axis in the vicinity of an aperture is shown in Fig. 3. The net effect is a mean field near $0.9561E_0$ along the symmetry axis, with slightly higher values off axis. In practice, the potential difference W across the whole length of the set of plates was measured and divided by $7D$ to enable calculation of the value E_0, which was then corrected. With this arrangement, the value of the electric field was determined to an accuracy of 1.4×10^{-3}, assuming that the uncertainties of the parameters which go into the calculation of electric field add in quadrature. This is one of the main uncertainties of the measurement. A precision-programmed power supply stable to 5×10^{-4} provided the potential differences between the field plates. The whole plate assembly could be raised to a potential $\pm\Delta U$ (≤ 3000 V) relative to grounded plates, to accelerate or decelerate the ions just before entering the field-plate region.

The laser system consisted of a Coherent ring dye laser pumped by up to 6 W of 514-nm radiation from an argon-ion laser. The dye laser was operated below 200-mW power using R590 dye in a fixed single-frequency mode using étalons, but not actively stabilized, resulting in a linewidth near 30 MHz. The mode structure of the laser was monitored with an external étalon, and the wavelength was measured to be 6124.39 Å using the Burleigh WA-20VIS wavemeter. The laser beam was introduced to the observation vacuum chamber through an uncoated window at the end of the beamline, and an

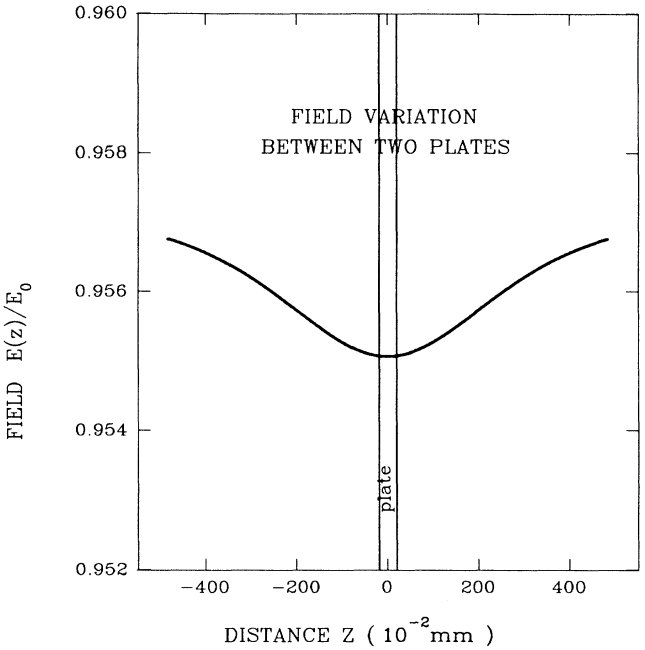

FIG. 3. The electric field variation between two field plates. One plate is located at $Z=0$ where the field has a minimum, while the maximum is the midpoint between two plates. The field is normalized to E_0, and has mean value $0.9561E_0$ (see text).

internal mask with an aperture 0.5 cm in diameter, used to reduce the effects of light scattering by the window. The ion beam was deflected electrostatically into a Faraday cup just before the window. The laser beam was guided through the apertures of the observation chamber, and finally through the ion beam collimator 1 m upbeam from the chamber. The laser beam was typically 0.4 cm in diameter in the interaction region. Since the ion beam was comparable in diameter to the 0.8 cm defining apertures, only a small fraction of the ions interacted with the laser light. The apertures defined the relative angle between the two beams to be ≤ 4 mrad.

The transitions $3d'^2G_{9/2}-4p'^2F^\circ_{7/2}$ at 6116.625 Å and $3d'^2G_{7/2}-4p'^2F^\circ_{7/2}$ at 6125.069 Å were separately excited by Doppler tuning, but longer wavelength transitions of the multiplet were not reached with the dye. The decay transitions $4s'^2D_{5/2}-4p'^2F^\circ_{7/2}$ near 4610 Å and $3d^2F_{7/2}-4p'^2F^\circ_{7/2}$ near 4906 Å were observed using a cooled 9862B photomultiplier tube. The photomultiplier had a typical dark rate of 1.5 s^{-1}. A small monochromator, with resolution reduced to 80 Å by opening the slits, was used to isolate these transitions. The light was collected using a lens system from a 1-mm region defined by optical baffles between two of the field plates. The prompt cascade transition $4s'^2D_{5/2}\uparrow 3p^5\,^2P^\circ_{3/2}$ near 672 Å which follows the 4610 Å decay was also observed, using an electron multiplier detector mounted opposite the photomultiplier.

To minimize background due to excitation of the beam ions by background gas, the beamline before the observation chamber was evacuated to typically 5×10^{-8} Torr during beam operation, by a cryopump. The collinear

beam chamber was evacuated by a 50 L/s ion pump, producing a residual pressure near 5×10^{-7} Torr. The background count rate near 4610 Å with laser and ion beams in operation was 40 s^{-1} per μA of beam current, due to nonresonant scattering.

IV. RESULTS AND DISCUSSION

Figure 2 shows photomultiplier counts due to the transition near 4610 Å, resulting from a voltage ramp through the Doppler-shifted ion absorption. The scan rate was equivalent to 500 ms per point, with a potential difference of 2 V between points. The full width at half maximum (FWHM) of the resonance is near 120 MHz. The Doppler compression of the random ion source velocities due to the acceleration of the ions through the potential difference U leads to a predicted resonance width $\Delta\nu=2\nu_0(\delta E_s+e\delta U)/(2mc^2eU)^{1/2}$, where δE_s is the range of kinetic energies of the ions in the source, and δU describes fluctuations in the accelerating voltage. The broadening effect due to the finite laser linewidth is assumed small. Based on the measured width, and the known stability of the power supplies, a spread of the ion energies in the source of about 5 eV is found, in accord with expectations based on ion source operation.

Each mean lifetime measurement cycle was initiated by a resonance sweep as described above, to verify ion-beam current and voltage stability. If the peak amplitude and voltage of the resonance were unchanged, the bias voltage of the electric field plate assembly was programmed to increase linearly with time to move the excitation point of the ions upbeam. The decay fluorescence counts were monitored as a function of this voltage sweep, which typically lasted for 150 s, with an observation time per point of 500 ms. This procedure was repeated several times, to yield one measurement. Thirty-eight separate measurements were carried out, under slightly different conditions. An example of a fluorescence decay curve is shown in Fig. 4(a), where the observed photomultiplier counts are plotted versus the applied voltage. The decay of the fluorescence will depart from a single exponential if photons from the excitation function are included. In practice, points were dropped from the high-intensity portion of the decay until no changes in the fitted time constant were observed. This corresponds to initiating the measurement at a voltage at which the excitation function intensity has dropped to zero. A plot of the logarithm of useful photon counts versus voltage appears in Fig. 4(b).

The relationship between elapsed time and voltage is given by $\Delta t=(2m/q)^{1/2}E^{-1}(U_A^{1/2}-U_B^{1/2})$ for ions with charge q, where E is the mean electric field at or near the observation region, and U_A and U_B are the electric potentials at spatial positions labeled z_A and z_B. Since the potential U of the whole plate assembly is varied, and since the ions must be in resonance with the laser at the two points A and B, U_A and U_B are just values of U. The relationship of time to voltage is clearly not linear in general, but since $U\approx30$ kV and $U_\Delta\cong(U_B-U_A)\lesssim300$ V, then

$$\Delta t\simeq(m/2qU)^{1/2}E^{-1}U_\Delta\ ,$$

with a systematic error no greater than 2.5×10^{-3}. The exact expression was used in the data analysis.

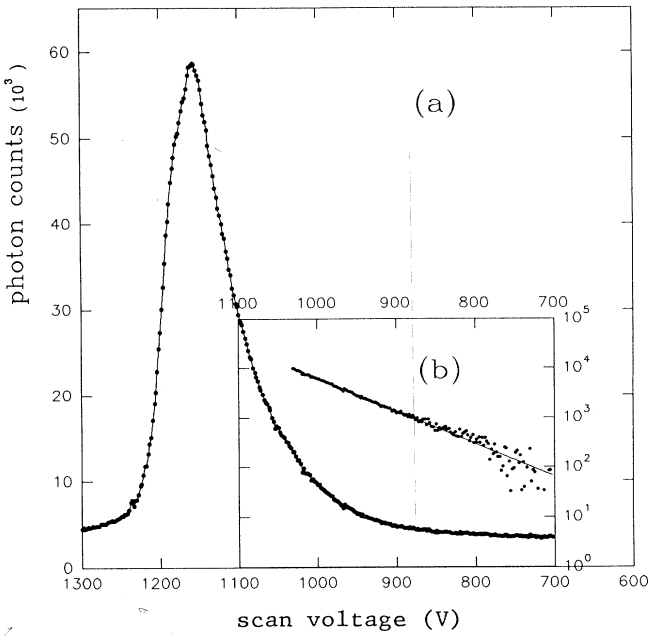

FIG. 4. (a) An example of data of the fluorescence decay curve of the Ar II $4p'^2F^\circ_{7/2}$ level. (b) The useful portion of the decay curve for extracting mean lifetime information. The plot is semilogarithmic with a constant background subtracted.

The optical decay intensity data were fitted with a function of the form $I(t)=C\exp(-t/\tau_m)+G$, where the fit determined the parameters C, G, and τ_m. The mean of the 38 measurements was $\tau=8.409\pm0.012$ ns (one standard deviation of the mean of the measurements) with extreme values of 8.61 and 8.28 ns. The results are plotted in Fig. 5. Only the upper value was more than two standard deviations of an individual measurement away from the mean. Some of the measurements of the set were carried out with particular measurement parameters varied (see below). Since the electric field in the beam interaction region did not exceed 250 V/cm, modifications of the mean lifetime due to the quadratic Stark effect were negligible. Measurements of the mean lifetime at different values of the electric field produced identical mean results.

A search was made for possible systematic effects which might affect the accuracy of our measurement. Since the local magnetic field was not shielded, and the laser light was partially polarized, an alignment or orientation of the excited state might be produced, which could precess in the residual magnetic field resulting in quantum beats, and a departure from a true single exponential decay. Twenty-one measurements with partial linear polarization resulted in a mean value $\tau_{LP}=8.396\pm0.015$ ns. The light polarization was converted to elliptical using a stressed-quartz quarter-wave plate [10] for 17 measurements, with the result $\tau_{EP}=8.425\pm0.018$ ns. The difference between the means of these two measurements slightly exceeds the larger standard deviation of the mean, indicating a possible small systematic effect. The magnitude of this effect was taken to be one-half the difference between the measured

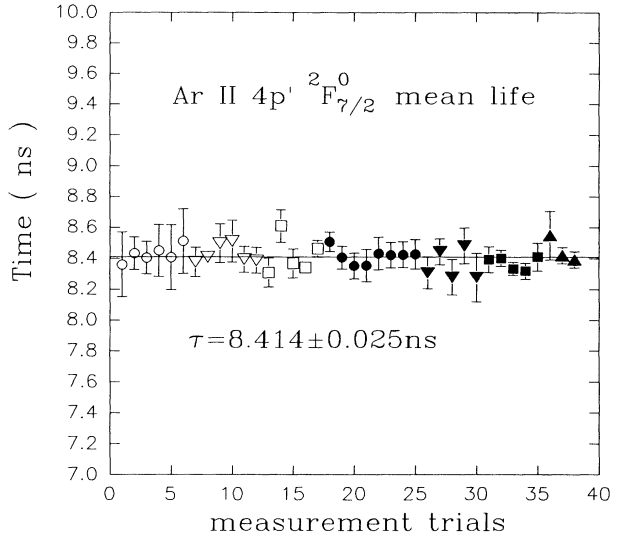

FIG. 5. Mean lifetime measurement trials. All solid symbols represent the measurements done using partially linearly polarized laser light, and open ones for elliptically polarized light. Similar symbols designate trials made during a continuous sequence of measurements. The raw measurement result was $\tau=8.409\pm0.012$ ns. After systematic corrections, the final result is $\tau=8.414\pm0.025$ ns.

means, or fractionally 2×10^{-3} of the mean lifetime.

The laser light intensity was also decreased by over a factor of 10 for several of the measurements in Fig. 5 without effect, except for a small decrease in signal-to-noise ratio.

Collisions of the beam ions with background gas molecules could possibly remove ions from the excited state, or excite ions to the excited state, leading to changes in the measured mean lifetime. When the ion beam and laser beam were present, but the laser beam was Doppler shifted off resonance, about 40 counts/s per μA beam current of background were observed. This is far less than one count during the mean lifetime of the level, so excitation effects were negligible. A collisional quenching cross section of 2×10^{-14} cm^2 was earlier measured for states of argon by Camhy-Val *et al.* [11]. At our beam velocity, this leads to a quenching rate of 7.6×10^{-7} cm^3 s^{-1}, resulting in a fractional increase to the final mean lifetime result of 1.7×10^{-4}. The corrections and systematic effects on the measurement result are compiled in Table I.

TABLE I. List of systematic errors.

Source	Correction (in units of 10^{-3})	Error (units of 10^{-3})
Collision with rest gas	+0.17	±0.17
E field		±1.4
Alignment of ion beam and E field	+0.44	±0.44
U determination		±1.1
Stark effect	negligible	negligible
Quantum beat		±2
Statistics		±1.2
Total	+0.61 (in sum)	±3.0 (in quadrature)

After the measured mean lifetime was corrected for the effects of quenching, the possible systematic uncertainty associated with quantum beats was added in quadrature to the relative uncertainty in the electric field determination and to the statistical uncertainty of the mean of the measurements, resulting in an overall limitation of the relative accuracy of the measurement of 3.0×10^{-3}, and a final result $\tau=8.414\pm0.025$ ns.

This measurement result can be compared to a measurement of the mean lifetime of the Ar II $4p'\,^2F^\circ_{7/2}$ state by the crossed-laser–fast-ion-beam method [8]. This earlier measurement was expected to be both accurate and precise, since the basic motivations were the same as for our measurement. The result was 8.41±0.03 ns, which agrees with the current measurement, and has basically the same precision. Improved precision in our measurements can be achieved by using a higher operating terminal potential on the ion accelerator, and magnetic shielding of the interaction region.

The relationship of the results of other measurement techniques, and of theoretical calculations, to the present results has been adequately presented in Ref. [8]. Calculations by Luyken [12] using the dipole-velocity form of the matrix element are closest to our data, falling about 3% lower.

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