Precision Measurement of the Metastable $6s\ [3/2]_2$ Lifetime in Xenon

M. Walhout,* A. Witte, and S. L. Rolston
National Institute of Standards and Technology, U.S. Department of Commerce, Building 221, Room A 167, Gaithersburg, Maryland 20899
(Received 13 January 1994)

Using a magneto-optical trap to isolate an isotopically pure sample of xenon, we determine the metastable $6s\ [3/2]_2$ state lifetime by measuring the rate of VUV emission due to magnetic quadrupole decay. We find lifetimes of 42.9(9) s and 42.4(13) s for ^{132}Xe and ^{136}Xe, respectively (1σ uncertainties). These values are less than half the theoretical predictions. We also find that nuclear-spin-induced decay produces a much shorter metastable lifetime in ^{129}Xe.

PACS numbers: 32.70.Fw, 32.80.Pj, 35.80.+s

Measurements of lifetimes of metastable atomic states provide sensitive tests of atomic theory, probing effects that do not strongly influence the positions of energy levels. Unfortunately, there have been few [1,2] experimental measurements of lifetimes > 1 s for neutral atoms, due to the difficulties of producing an unperturbed, state-selected atomic sample that can be observed for a long time. Laser cooling techniques offer an opportunity to surmount these limitations. The first metastable lifetime measurements using these techniques relied on separating the radiative and collisional rates of loss of Ar and Kr atoms in a magneto-optical trap [3]. We present a new, precision measurement involving direct detection of the VUV photons emitted from trapped xenon atoms due to the magnetic quadrupole ($M2$) decay of the $6s\ [3/2]_2$-($^3P_2$) metastable state. Our technique is insensitive to nonradiative deexcitation, which occurs primarily through Penning ionization in collisions between metastable atoms. For ^{132}Xe and ^{136}Xe, which have no nuclear spin, we determine the lifetime with a few-percent uncertainty, finding marked disagreement with theory [4,5]. In the case of ^{129}Xe, which has nuclear spin $I=1/2$, the mixing of fine-structure states by the hyperfine interaction is seen to shorten the lifetime dramatically. The precision of our measurements relies on the combined features of state selection, long observation times, and isotope separation that are afforded by laser cooling and trapping. Our results and those of Ref. [3] are prompting reevaluation of atomic structure calculations [5].

Our strategy is to record the rate of VUV emission from a trap and normalize that rate to the number of trapped atoms. In the normalization procedure, 823-nm light is used to quench the $6s\ [3/2]_2$ level, transferring population through the $6p\ [3/2]_2$ state and into the $6s\ [3/2]_1$, which undergoes prompt, electric dipole ($E1$) decay to the ground state and emits a VUV photon (see Fig. 1). The quench-induced VUV and the $M2$ radiation are detected with the same photomultiplier tube (PMT), which has nearly constant efficiency over the 2-nm wavelength range spanned by the $6s\ [3/2]_2$ and $6s\ [3/2]_1$ decays. Thus, the natural decay rate of the metastable level is approximately the $M2$ count rate divided by the number of counts from the quench, regardless of the solid angle of the detector. To make a precise measurement, we must be sure that the measured VUV rate is due only to the spontaneous decay of atoms in the $6s\ [3/2]_2$ state. We must also probe the number of trapped atoms accurately. Finally, we must also know the angular distribution of the radiation. These details are addressed below.

A diagram of our experiment appears in Fig. 2. The magneto-optical trap (MOT), which is described elsewhere [6], is composed of a magnetic quadrupole field and laser beams tuned a linewidth below the 882-nm cycling transition between the $6s\ [3/2]_2$ and $6p\ [5/2]_3$ states of a particular xenon isotope. The magnetic field gradient is typically 0.5 mT/cm, and the cloud of trapped atoms is less than 1 mm in radius. The trap lies in the path of a laser beam that decelerates metastable atoms emerging from a dc discharge source and travels along the z axis. This slowing beam is circularly polarized and tuned about 25 linewidths below the 882-nm resonance. All beams are chopped with a 60-ms period (43 ms on and 17 ms off) using acousto-optic modulators. Figure 3 shows the timing sequence. Measurement of the $M2$ decay rate takes place only when the laser beams are off and the atoms are in the metastable $6s\ [3/2]_2$ state.

$M2$ photons at 149 nm pass through a MgF_2 window and are detected using a Hamamatsu R1459 PMT [7] oriented at an angle of 90° with respect to $\hat{\mathbf{z}}$. If the magnetic sublevel (m_J) populations are known, and hence the spatial distribution of radiation, the total rate of $M2$ flouresence from the trap can be calculated from the

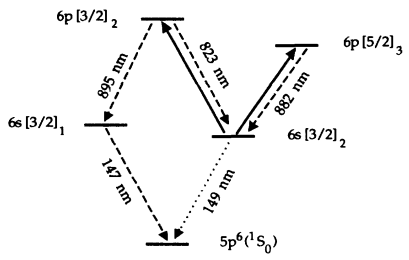

FIG. 1. Transitions used in the $6s\ [3/2]_2$ lifetime measurement. Solid lines represent laser excitations, dashed lines are electric dipole decays, and the dotted line corresponds to the weak $M2$ decay.

0031-9007/94/72(18)/2843(4)$06.00
© 1994 The American Physical Society

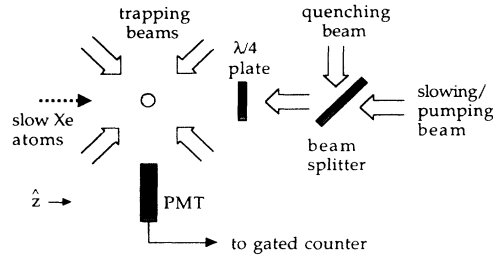

FIG. 2. Schematic diagram of the experiment. Trapping beams directed into and out of the page and coils for producing magnetic fields are not shown.

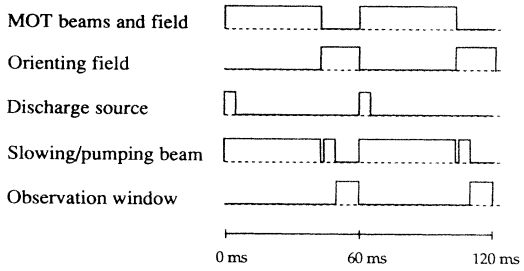

FIG. 3. Timing diagram. The dotted lines indicate "off" levels. Two periods of the trapping-pumping-detection cycle are shown. In order to probe the number of trapped atoms, the quenching laser is applied after 60 such periods.

VUV intensity at our single detector position. Though a MOT might be expected to produce a uniform m_J distribution, we create a known distribution by using the slowing laser to optically pump atoms into the $m_J=2$ state. Zeeman precession out of the state is avoided by replacing the quadrupole MOT field with an orienting field of 0.15 mT along $\hat{z}$. As the MOT and slowing beams are shut off every 60 ms, the MOT field is switched off and the orienting field is turned on. The slowing (optical pumping) beam is then applied for 5 ms. Such a long pumping time is necessary because of the large detuning of the slowing light. Photons from the oriented sample are counted in a 10-ms observation window during the remaining "MOT off" part of the chopping cycle. The $M2$ intensity at our detector is therefore related to the total intensity by the radiation pattern for $\Delta m=2$ quadrupole decay.

We measure the efficiency of optical pumping by comparing quenching rates for σ^+ and σ^- polarizations of the quenching light. This light is derived from a diode laser stabilized by grating feedback and locked to an absorption feature in a Xe discharge cell. A beam from the laser is directed along the slowing beam and switched with a mechanical shutter. The intensity and detuning are chosen so the quenching transition is driven well below saturation, effecting a slow transfer of population and eliminating detector pulse pileup. Since quenching acts on a $J=2\rightarrow J'=2$ transition for even isotopes, the σ^+ polarization should not be absorbed if atoms are pumped completely into $m_J=2$. By contrast, the σ^- polarization should quench about 75% of the atoms and pump 25% into the $m_J=-2$, which cannot absorb σ^- quenching light. For optical pumping pulses longer than 4 ms, the ratio of σ^- to σ^+ quenching rates is at least 10:1, indicating that more than 90% of the atoms are pumped to $m_J=2$. This degree of polarization persists for much longer than the 10-ms decay observation window. Equally satisfactory results are obtained for pumping into the $m_F=5/2$ state of ^{129}Xe, for which the pumping and quenching lasers couple to the $6p\ [5/2]_3(F=7/2)$ and $6p\ [3/2]_2(F=5/2)$ levels, respectively. The rate of quenching by σ^+ light is also used to minimize magnetic fields in directions orthogonal to $\hat{z}$. In the presence of transverse field components, Zeeman precession mixes m_J sublevels, so atoms that are initially optically pumped are rapidly enabled to absorb σ^+ quenching light. By applying compensating fields, we minimize the σ^+ quenching rate and null the unwanted components to $<1\ \mu$T.

Care is taken to eliminate background counts and the possibility of unwanted quenching of the metastable atoms during the observation phase. The MOT chamber is shielded with black material so that all external sources of quenching light are blocked. Quenching light from the beam source is avoided by pulsing the discharge for only 5 ms at the beginning of each 43-ms trapping interval. Using the pulsed source eliminates nearly all background VUV counts, since the only metastable atoms in the observation region after $\sim$40 ms are those confined in the MOT. Contributing only a few percent of the total rate, the remaining background is measured with the MOT beams either switched off or tuned blue of the 882-nm resonance, with both methods producing the same result.

After every 60 periods of the trapping-pumping-detection cycle, the number of atoms in the trap is probed by application of σ^--polarized quenching light and detection of the resulting 147-nm photons. To ensure that all atoms are accounted for, we apply quenching radiation in a sequence of two 10-ms pulses during two consecutive observation windows. The intervening exposure to trapping and pumping light restores the $m_J=2$ orientation to atoms that get pumped into the nonabsorbing $m_J=-2$ state during the first quenching pulse. Closing a shutter in the atomic beam eliminates reloading of the trap between pulses. Since the two pulses do not completely deplete the trap, we must still account for unquenched atoms. Assuming that each pulse quenches the same fraction of remaining atoms, the total number of trapped atoms that would produce VUV counts if quenched is

$$N=N_1/(1-N_2/N_1)\,, \tag{1}$$

where N_1 and N_2 are the counts accumulated during the first and second quenching pulses. As expected, the ratio N_2/N_1 depends on quenching polarization and is about 0.25 on average for a σ^- quench. This method of determining N gives consistent results that are independent of N_2/N_1. Furthermore, with the addition of a third pulse

we find $N_3/N_2 \approx N_2/N_1$, validating the series summation used to obtain Eq. (1).

If we detect $M2$ decay counts at rate R, then the decay rate of the metastable state is proportional to R/N. We determine this quantity by recording the sum of counts from 60 consecutive 10-ms observation windows and the number N found from the following pair of quenching pulses. Background is measured during 20 subsequent observation windows. The rate R is obtained by subtracting the weighted background from the number of decay counts and dividing the result by the total $M2$ observation time. The scatter in decay counts is well described by the expected Poisson distribution. We also find that $\overline{(R/N)}$ and $\overline{(\bar{R}/\bar{N})}$ are always equal within statistical uncertainty, affirming the stability of our experiment. In the case of ^{132}Xe we obtain $\overline{(R/N)}=0.0258(3)\ s^{-1}$, where we quote statistical uncertainties of 1 standard deviation. This result arises from more than 14000 measured decay counts in an accumulated observation time of 1620 s. Dedicating somewhat less observation time to other isotopes, we also find $\overline{(R/N)}=0.0261(6)\ s^{-1}$ for ^{136}Xe and $\overline{(R/N)}=0.0675(11)\ s^{-1}$ for ^{129}Xe.

A clear advantage of measuring the spontaneous decay rate by directly observing VUV fluorescence is the insensitivity of the method to nonradiative collisional processes that may remove metastable atoms from the trap. By contrast, the experiments of Ref. [3] measured the total trap decay rate and were limited primarily by uncertainties in collision rates and the necessary extrapolation to zero background pressure. For all the Xe isotopes that we study, $\overline{(R/N)}$ is independent of the Xe density in the MOT, which can be changed by orders of magnitude and monitored by detecting ions that are produced in collisions between trapped atoms. $\overline{(R/N)}$ also remains constant as we vary the background pressure of either air or Xe by a factor of 5. Within the $\sim 1\%$ statistical limitations of these tests, our results are unchanged by possible effects of collisionally induced VUV emission or trap loss.

To relate $\overline{(R/N)}$ to the metastable decay rate, we must know the spatial distributions of the $M2$ and quench-induced VUV photons. For the latter, the intensity radiated per unit solid angle is

$$\frac{dI}{d\Omega} = \Sigma_{m,m'} P(m) |C(m,m')|^2 X_{1|m-m'|}(\theta,\phi)|^2, \quad (2)$$

where $P(m)$ is the total population entering the magnetic sublevel m of the $6s\ [3/2]_1$ state, $C(m,m')$ is the Clebsch-Gordan coefficient governing the $E1$ decay of level m to the ground state level m', and $|X_{1|m-m'|}(\theta,\phi)|^2$ is the angular distribution of radiation for that decay [8]. The intensity striking our detector is obtained by integrating $|X_{1|m-m'|}(\theta,\phi)|^2$ over the appropriate solid angle, yielding a fixed coefficient in Eq. (2) for each $|m-m'|$. To obtain the populations $P(m)$, we solve the set of population rate equations that describe the optical pumping due to the quenching laser. The steady-state populations depend on the quenching laser polarization and saturation, the initial population distribution among the magnetic sublevels in the $6s\ [3/2]_2$ state, and the relative probabilities of the $6p\ [3/2]_2 \rightarrow 6s\ [3/2]_1$ and $6p\ [3/2]_2 \rightarrow 6s\ [3/2]_2$ decays. From the assumed initial population distribution we also compute the fraction of the $M2$ radiation that hits our detector, accounting for anisotropy with an equation analogous to Eq. (2), but for quadrupole decay.

TABLE I. Uncertainty budget.

Uncertain quantity	Uncertainty arising in τ_{132} and τ_{136} (%)
Statistics of $\overline{(R/N)}$	1.1(^{132}Xe)
	2.3(^{136}Xe)
Detector calibration	1.2
Optically pumped m_J distribution	0.8
Collisional quenching and trap loss	1.1
Branching ratio for $6p\ [3/2]_2$ decay	0.3
Quenching light polarization	<0.1
Direction of orienting field	<0.1
Stray quenching light	<0.1

These calculations yield a factor η by which $\overline{(R/N)}$ must be multiplied in order to account for anisotropic VUV emission and optical pumping during the quenching process. By varying the input parameters for the rate equations within the range of our experimental uncertainties, we obtain an uncertainty for η. Uncertainties in population distribution and polarization are constrained by the measured σ^-/σ^+ quenching ratio of $\geq 10{:}1$. Uncertainty in the branching ratio for the two $6p\ [3/2]_2$ decay channels is assessed using existing experimental data, which indicate that the $6p\ [3/2]_2$ branches with 70(4)% probability to the $6s\ [3/2]_2$ state [9,10]. We calculate $\eta=0.889(8)$ for the even isotopes and $\eta=0.769(19)$ for ^{129}Xe. The uncertainty in each value is the quadrature sum of independent uncertainty contributions.

Determining the metastable decay rate from our experiment also depends on the ratio of detection efficiencies at 147 and 149 nm. From the wavelength calibration of our PMT quantum efficiency and the transmission curve for our MgF_2 window, we obtain an additional correction factor of $\alpha=1.016(12)$. The uncertainty in this factor is evaluated by estimating the uncertainties in the slopes of published calibration curves.

For the even isotopes the natural decay rate is simply $\Gamma=\alpha\eta\overline{(R/N)}$. The result for ^{132}Xe is $\Gamma_{132}=0.0233(5)\ s^{-1}$, or a lifetime of $\tau_{132}=42.9(9)$ s. As expected, we obtain similar values for ^{136}Xe: $\Gamma_{136}=0.0236(7)\ s^{-1}$ and $\tau_{136}=42.4(13)$ s. The quoted uncertainties are quadrature sums of uncertainties listed in Table I. These sums are dominated by uncertainties in counting statistics, the wavelength calibration of detection efficiency, the purity of optical pumping, and the possibility of undetermined collisional effects. As an additional test of our knowledge of the angular radiation distribution, we compare the

^{132}Xe result with data acquired without any optical pumping. From measurements made with no pumping pulses or orienting field, but with all else the same as described above, we obtain $(\overline{R/N})=0.0218(5)$ s^{-1}. Assuming the unpumped atoms are distributed evenly over all m_J states, we find $\eta=1.04$, yielding a decay rate of 0.0230(6) s^{-1}, in agreement with the optically pumped case.

Our ^{129}Xe data reveal the effect of nuclear-spin-induced decay of the metastable state [11], though extracting a lifetime for this isotope is not possible. There are in fact two natural decay rates for ^{129}Xe corresponding to the two hyperfine levels ($F=5/2$ and $F=3/2$) in the metastable manifold. The $F=5/2$ state has only $J=2$ character, so that its lifetime should be the same as that of the even isotopes. In the $F=3/2$ state the hyperfine interaction mixes a small amount of the $6s$ $[3/2]_1$ state into the $6s$ $[3/2]_2$, leading to the breakdown of J as a good quantum number [12]. We can estimate the order of magnitude of the $6s$ $[3/2]_1$ population admixture to be the square of the ratio of the hyperfine structure constant ($\sim$0.03 cm^{-1}) to the fine structure splitting between the $6s$ $[3/2]_2$ and $6s$ $[3/2]_1$ levels ($\sim$1000 cm^{-1}). Such an argument leads to a $6s$ $[3/2]_1$ admixture of $\sim 10^{-9}$ in the $F=3/2$ metastable state. Since the $6s$ $[3/2]_1$ has a decay rate of $\sim 3\times 10^8$/s, the $F=3/2$ lifetime is expected to be only a few seconds.

In the ^{129}Xe experiment, we measure the total VUV decay rate and probe the number of $6s$ $[3/2]_2(F=5/2)$ atoms, finding $\alpha\eta(\overline{R/N}_{5/2})=0.0527(19)$ s^{-1}. This result cannot be regarded as the $F=5/2$ decay rate, since there may be counts arising from short-lived $F=3/2$ atoms. The characteristic time for off-resonant pumping of atoms into the $F=3/2$ state is known from trap loss measurements to be about 0.5 s. Once in this state, atoms remain in the PMT field of view for <0.1 s. We therefore expect 10%–20% of all atoms contributing to the signal to be in $F=3/2$. Assuming the $F=5/2$ lifetime is equal to the even-isotope lifetime, this degree of contamination can account for the additional VUV rate if the $F=3/2$ lifetime is $\sim$5 s. Such an estimate agrees with the calculation given above, suggesting that our measurement is indeed sensitive to nuclear-spin-induced decay. This result highlights the importance of isotope selection in our measurements.

The 43-s metastable lifetime in the even isotopes is more than a factor of 2 shorter than theoretically predicted. Moreover, the result is close to the $\sim$40-s lifetimes measured in Ref. [3] for both Ar and Kr. These findings have necessitated a reexamination of the relevant atomic structure theory. Early calculations for the metastable rare gases yielded lifetimes of 24, 56, 85, and 150 s for Ne, Ar, Kr, and Xe, respectively [4]. More recently, using relativistic wave functions that reproduce energy levels within 1%, Indelicato, Desclaux, and Kim have calculated 22, 51, 63, and 96 s for the same set of lifetimes [5]. In light of the experimental results, the trend of longer lifetimes for the heavier elements is clearly exaggerated by both of these predictions. Ongoing work by Indelicato, Desclaux, and Kim, however, demonstrates that the metastable lifetime is quite sensitive to electron correlation [5]. The sensitivity is especially pronounced for heavy elements, in which relativistic effects and electron correlation are large. For example, in preliminary calculations, valence correlation configurations that do not affect energy levels much are found to change the Xe $6s$ $[3/2]_2$ lifetime by as much as a factor of 2. Inclusion of such correlation configurations, which are neglected in Ref. [4], may resolve the discrepancy between previous theories and the measurements described here.

We thank T. J. McIlrath and W. D. Phillips for helpful discussions, C. Gerz and C. Orzel for technical assistance, and P. Indelicato, J. P. Desclaux, and Y.-K. Kim for sharing their results. This work was supported in part by the Office of Naval Research. A.W. acknowledges financial support from the Alexander-von-Humboldt foundation.

*Permanent address: Department of Physics, University of Maryland, College Park, MD 20742.

[1] J. R. Woodworth and H. W. Moos, Phys. Rev. A **12**, 2455 (1975).
[2] B. L. Wexler, B. E. Wilcomb, and N. Djeu, J. Opt. Soc. Am. **70**, 863 (1980).
[3] H. Katori and F. Shimizu, Phys. Rev. Lett. **70**, 3545 (1993).
[4] N. E. Small-Warren and L.-Y. C. Chiu, Phys. Rev. A **11**, 1777 (1975).
[5] P. Indelicato, J. P. Desclaux, and Y.-K. Kim (to be published).
[6] M. Walhout, H. J. L. Megens, A. Witte, and S. L. Rolston, Phys. Rev. A **48**, R879 (1993).
[7] Reference to a particular product or company appears only to ensure reproducibility of scientific results and does not represent endorsement by NIST.
[8] J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1975), 2nd ed., p. 753.
[9] J. A. Cabrera, M. Ortiz, and J. Campos, Physica (Amsterdam) **104C**, 416 (1981).
[10] H. Horiguchi, R. S. F. Chang, and D. W. Setser, J. Chem. Phys. **75**, 1207 (1981).
[11] R. Marrus and P. J. Mohr, Adv. At. Mol. Phys. **14**, 181 (1978).
[12] L. Armstrong, *Theory of Hyperfine Structure of Free Atoms* (Wiley, New York, 1971), p. 145.

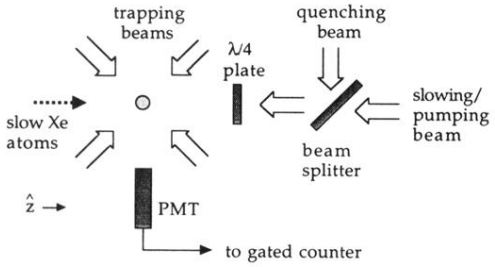

FIG. 2. Schematic diagram of the experiment. Trapping beams directed into and out of the page and coils for producing magnetic fields are not shown.