Lifetime measurements of individual hyperfine levels using localized Doppler tuning in laser excitation of fast ions

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(Received 7 December 1988)

Lifetimes of each hyperfine level of the $6p\ ^4P_{5/2}$ state of singly charged ^{129}Xe and ^{131}Xe have been measured using laser excitation of a fast-ion beam. The collinear laser- and ion-beam arrangement was used to achieve high spectral resolution, and transitions to each hyperfine level were locally Doppler tuned into resonance between a pair of post-accelerating electrodes. Decay to the $6s\ ^4P_{5/2}$ state was observed through a monochromator immediately after the post-acceleration field, and cascade-free lifetimes were then determined by mechanically varying the distance between the excitation and observation points. Depletion of close-lying hyperfine levels prior to the excitation region was necessary in ^{131}Xe to sufficiently isolate transitions for these measurements. These results form the first set of lifetime measurements for each hyperfine level within a particular J state of an atomic ion. Although no variation in lifetime with F quantum number was detected outside of the determined error bars, application to studies of hyperfine-induced mixing is discussed.

INTRODUCTION

Since the advent of fast-ion-beam laser spectroscopy,[1] a wealth of data has been collected regarding the structure and decay mechanisms of atomic ions.[2,3] Laser excitation eliminates the cascade problems often encountered with other nonselective means of excitation. This is important for accurate determination of oscillator strengths which are necessary for interpretation of astrophysical spectra, as well as for confirmation of theoretical approaches used in calculations of dynamical properties of complex atomic systems. High-resolution studies of ionic structure have also been made possible,[4] by exploiting the compressed longitudinal component of velocity spread in accelerated ion beams.[5] It is difficult, however, to simultaneously take full advantage of both the high spectral resolution afforded by a collinear ion- and laser-beam geometry, and the high accuracy achievable in lifetime measurements using selective laser excitation. Although rapid Doppler switching, by post acceleration of the ion beam, has been demonstrated as an effective means of localizing the excitation for collinear beams, previous experimental techniques[6–8] did not permit isolation of closely spaced spectral lines (as for instance, in hyperfine multiplets) for lifetime measurements of individual excited states. While Ceyzeriat *et al.*[8] performed a series of very accurate measurements on lifetimes of various levels in the $4p^5 5p$ configuration of singly ionized Rb, they were only able to resolve one hyperfine component sufficiently to measure its lifetime. Lifetimes of hyperfine components are generally not expected to vary within a certain J state; however, the hyperfine interaction can influence transition probabilities,[9,10] sometimes resulting in F-dependent lifetimes for the levels.[11] It therefore would be useful to have a technique available that provides the capability of measuring lifetimes of closely spaced levels (spacings on the order of the Doppler-broadened linewidth). We introduced such a technique in a recent paper.[12] This paper contains a detailed description of the essential features of this novel experimental method.

EXPERIMENTAL SETUP

A 400-kV electrostatic accelerator was used to provide a fast beam of singly charged Xe ions. Positive ions were produced in a universal low-voltage arc ion source, preaccelerated to 20 keV and injected into a 0.5-m-radius analyzing magnet, whereby isotopically pure singly charged ions were selected. Following a subsequent acceleration to 180 keV, a quadrupole triplet and horizontal and vertical deflection plates were used to focus and steer the beam through two 4-mm-diam apertures ($\sim$0.5 m apart) near the entrance and exit of the excitation chamber. Upon exiting the chamber, the beam was electrostatically deflected and collected in a Faraday cup. Transmitted ion-beam current varied between 0.5 and 1.0 μA. The beam line and excitation chamber were evacuated to $\leq 2\times10^{-6}$ Torr by diffusion pumps.

A single-mode frequency-stabilized Coherent 699-29 Autoscan ring dye laser produced a beam of $\leq$1-MHz-bandwidth laser light which was directed through a Brewster window at the end of the beam line, as shown in Fig. 1. The ring laser used Rhodamine-6G (R6G) dye pumped by an Innova 100-20 Ar-ion laser at 514.5 nm. A Spindler and Hoyer 03 8659 ($\times$7) beam expander was placed $\sim$30 cm after the laser-output coupler to focus the beam through the same 4-mm-diam apertures passed by the ion beam, thereby overlapping the ion beam in a collinear manner throughout the excitation chamber and beam line. The percentage of laser-output power passing both apertures was found to be 85–90% at full laser power, as measured prior to evacuation of the chamber and beam line.

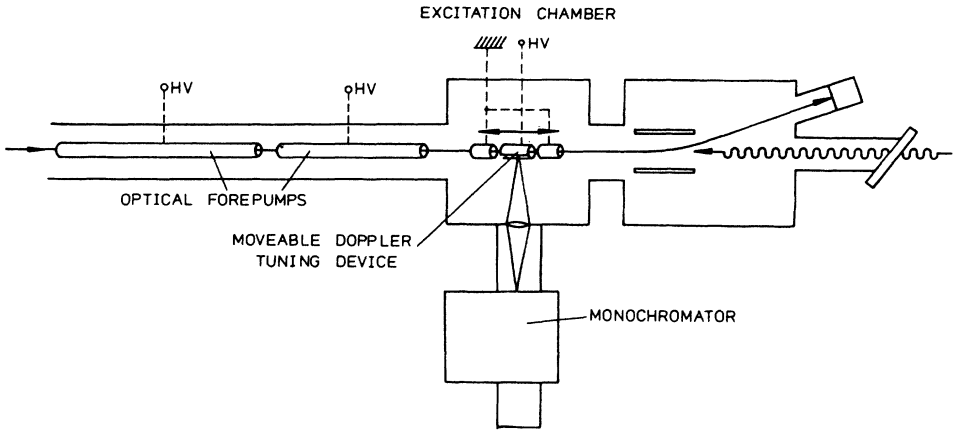

FIG. 1. Experimental arrangement. A single-mode laser beam is merged collinearly with an ion beam from a 400-kV electrostatic accelerator. Ions are postaccelerated into resonance in the first field region of the movable Doppler tuning device (DTD), and fluorescence from the decaying states is detected by a photomultiplier (PM) tube after passing a monochromator. Lifetime-decay curves are obtained by mechanically varying the distance between the excitation and observation points. Optical forepumps are used to isolate transitions in dense spectra by depletion of close-lying levels.

Inside the excitation chamber a set of three electrically isolated collinear cylinders (16 mm i.d.), denoted in the following as the Doppler tuning device (DTD), were attached to a movable carriage, which was mounted on steel rods and connected to a high-precision "ball-bearing"-type screw for positioning within the chamber. The two end cylinders were grounded and the center cylinder was connected to a Kepco BOP 1000M bipolar operational power supply and amplifier. Ions could thus be Doppler shifted into resonance with a fixed laser frequency by postacceleration (or deceleration). The center observation tube was slotted to allow fluorescence from the ion beam to be collected and focused onto the entrance slit of a Heath EUE 700 monochromator. In order to maximize the collection efficiency, the monochromator slits were fully opened to image a 2-mm-length section of the ion beam, giving a wavelength resolution of 4 nm for the decay channel. Photons passing through the monochromator were counted by an EMI 9789 QA photomultiplier (PM) tube which was cooled using a Hamamatsu C659A thermoelectric cooler.

A cross-sectional view of the DTD, along with the electrostatic potential variation on the common ion- and laser-beam axis, is shown for a typical observation-tube voltage of -800 V in Fig. 2. The ion-beam entrance aperture of the first grounded cylinder of the DTD was one of the two 4-mm-diam collimating apertures discussed above. In order to more sharply define the field region between this grounded tube and the high-voltage observation tube, without occluding the ion or laser beams, 5-mm- and 6-mm-diam apertures were placed at the other end of the grounded tube and the near end of the observation tube, respectively. An aperture of 10 mm diameter was placed at the downstream end of the observation tube in order to reduce field penetration into the cylinder at this end.

Post acceleration of the ion beam is often necessary to locally Doppler tune ions into resonance with the laser in the observation region. Otherwise, the lower level of interest may be optically pumped if ions are in resonance along the length of the beam line. This is especially true for limited metastable populations, but ground-state levels might also be significantly depleted by shelving into long-lived metastable levels. Spectral scans were thus obtained in the following manner. The DTD was fixed in a position such that ions excited in the electric field decayed before reaching the point inside the observation tube viewed by the monochromator. By scanning the laser frequency (or observation-tube voltage) prompt fluorescence from ions excited inside the observation tube was collected as a function of laser frequency (or postacceleration voltage).

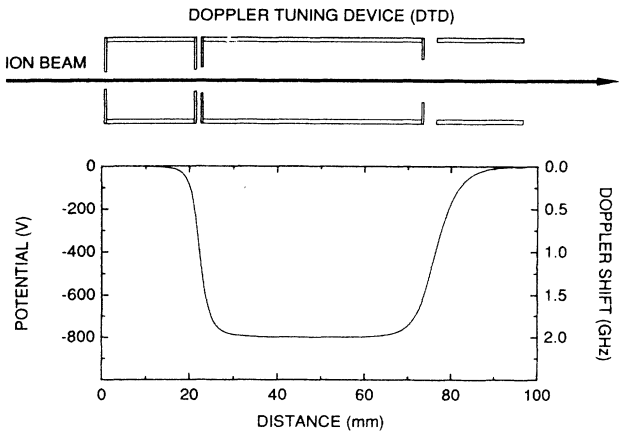

FIG. 2. Details of the Doppler tuning device. Schematic diagram of cross-sectional view of the DTD. A voltage is applied to the center cylinder and the two end cylinders are grounded. The potential variation along the ion-beam axis is depicted graphically for a typical center cylinder voltage of -800 V. The corresponding Doppler shift for an initial ion-beam energy of 180 keV is given on the right-hand y axis.

The special design of the DTD also gave the flexibility to collect lifetime-decay curves as well. The laser frequency and observation-tube voltage were chosen to Doppler shift a single transition through resonance in the first field gap, thus localizing the excitation along the beam axis. The fluorescence from the decaying excited state was then observed as a function of distance from the excitation point by mechanically varying the position of the DTD. The Doppler shift required for excitation of the ions with this collinear arrangement gave a rather direct, and inherently accurate, way to convert distance to a time scale for determining the lifetime of the excited state.

Two electrically isolated long cylinders, denoted "optical forepumps" in Fig. 1, were placed in tandem inside the beam line prior to the excitation chamber, coaxial with the ion and laser beams. The cylinders had an inner diameter of 25 mm, and a length of ~ 1000 and ~ 600 mm, respectively. Each was connected to a high-voltage power supply in order to allow Doppler tuning of the ion beam. In this manner, as introduced in our recent paper,[12] selective hyperfine levels could be depleted prior to the excitation chamber (cf. discussion of ^{131}Xe lifetime measurements below).

The central part of the data-acquisition system was a logical control unit originally designed for beam-foil–spectroscopy experiments.[13,14] The function of the unit in this application was to precisely control either the position of the DTD, or to control the observation-tube voltage. The dwell time per channel could be set in msec, or normalized to the digitized transmitted ion-beam current which was collected in the Faraday cup (Fig. 1). The laser frequency was controlled separately, using the "autoscan" program supplied with the laser. Spectra were accumulated in an Intertechnique BM 96 multichannel analyzer (MCA) and then transferred to a VAX 11/780 for analysis.

EXPERIMENTAL METHOD

Although the ultimate motivation for the design of the experimental apparatus described above was to measure lifetimes at high spectral resolution (e.g., lifetimes of individual hyperfine levels), a test experiment with the even isotope ^{132}Xe, which exhibits no hyperfine structure, was performed first. The lifetime of the excited $6p\ ^4P_{5/2}$ state was chosen as the first measurement, since this state is accesible by single-photon excitation ($\lambda_{air}=605.1$ nm in the ion rest frame) from the metastable $5d\ ^4D_{7/2}$ level populated in the ion source, and the decay to the quartet ground state $6s\ ^4P_{5/2}$ at 529.2 nm is sufficiently far from the laser-excitation wavelength to limit background problems from scattered laser light. Comparisons with earlier measurements also served as an indication of possible systematic errors inherent in this experimental technique.

It was first necessary to investigate contributions to the total spectral linewidth for this transition. High-resolution spectral scans were obtained by scanning the observation-tube voltage for a fixed laser frequency. Some examples are shown in Fig. 3. The laser frequency was set using the autoscan program of the ring-laser system, and read using an internal wavemeter. The setting is known to be reproducible to within 60 MHz and the absolute scale was verified to be accurate to within 200 MHz by comparison with a well-known fluorescence spectrum for I_2. The dwell time per channel for each scan was normalized to the Faraday cup current, to account for variation in ion-beam intensity. The laser intensity was monitored throughout the scans to verify stability of output power. Each channel in Fig. 3 corresponds to 1.2 V additional acceleration above the initial 180-keV energy. At this beam energy, the Doppler shift is ~ 2.5 MHz/V.

The full width at half maximum (FWHM) linewidth was observed to be slightly larger than 140 MHz for a laser power ≤ 30 mW [Fig. 3(a)]. The laser linewidth is known to be ≤ 1 MHz, and the natural linewidth of the transition can be estimated (from the known lifetime of ~ 8 ns) to be ~ 20 MHz. The dominant contribution to the total linewidth at low laser power, therefore, arises

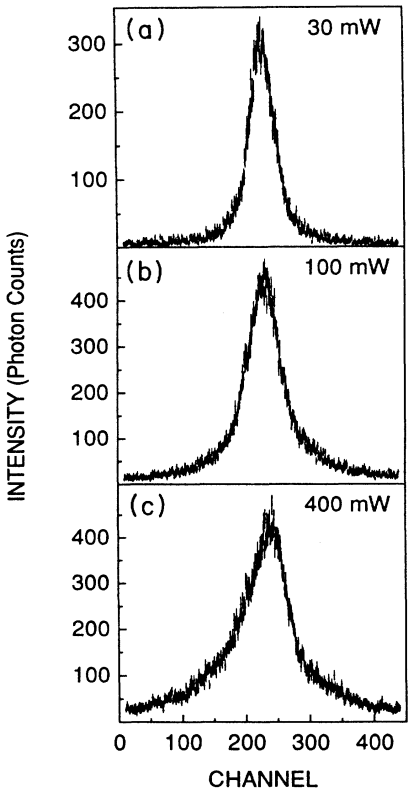

FIG. 3. Experimental line profile for the $5d\ ^4D_{7/2} \rightarrow 6p\ ^4P_{5/2}$ transition in singly charged ^{132}Xe (each channel corresponds to ~ 3-MHz effective laser frequency). (a) Negligible effects of power broadening are observed for 30-mW laser power. The width of the spectral line is primarily due to velocity distribution of ions. The natural linewidth is ~ 20 MHz. (b) The line profile was noticeably power broadened for 100-mW laser power. The peak height increased from ~ 320 to ~ 460 counts. (c) Line broadening increased as the laser power was raised to 400 mW (near maximum output power for this frequency). No further increase in peak height was observed, indicating a saturation of excitation at the peak of the velocity distribution.

from longitudinal velocity spread in the ion beam. With the assumption that the line profile obtained at 30 mW is negligibly influenced by power broadening, the velocity distribution was determined to have a FWHM of ~ 135 MHz. Although acceleration substantially compresses the velocity distribution along the ion-beam axis, the measured linewidth indicates an energy spread of ~ 55 eV, which is presumed to be primarily due to ripple on the accelerator high-voltage supply. Longitudinal velocity spreads from ion-beam divergence were estimated to give a nearly negligible contribution of ~ 1 MHz.

For higher laser powers, effects of power broadening were observed [Figs. 3(b) and 3(c)], which increased the FWHM to around 250 MHz near maximum laser power (400 mW), and changed the line profiles from an essentially Gaussian nature to include substantial Lorentzian-like tails. Each velocity class of ions has a natural line profile which is effectively broadened at high laser power, thereby increasing the contribution of fluorescence from neighboring velocity classes of ions at each point (i.e., each voltage). The asymmetry of the peak in Fig. 3(c) arises from the asymmetry of the excitation scheme. When a particular velocity class of ions is in resonance inside the observation tube, ions which lie on the faster side of the initial velocity distribution will be optically pumped at high laser power inside the field gap. This effect is naturally more dramatic on the right-hand side of the peak, since at these tube voltages the peak of the velocity distribution is excited in the field gap.

Spectral scans over a larger effective laser-frequency range were also possible. Figure 4 exhibits the hyperfine structure of the $5d\ ^4D_{7/2}$ and $6p\ ^4P_{5/2}$ levels in ^{129}Xe and ^{131}Xe. The structure of these hyperfine multiplets has been investigated in detail previously[15] and is not the subject of this paper. It is necessary, nonetheless, to be familiar with these multiplets to follow the discussion below of lifetime measurements of each upper hyperfine level, and for this reason these spectra are shown here.

Having established the experimental linewidth, it was then possible to proceed with the test measurement of the lifetime of the $6p\ ^4P_{5/2}$ state in ^{132}Xe. In order to locally confine the laser excitation inside the field gap, an observation-tube voltage of -800 V was chosen. The laser frequency was then set to excite the transition (at the peak of the velocity distribution) when the ions reach a potential of -370 V, roughly halfway between the first and second tubes of the DTD. From the maximum measured linewidth and calculations of the potential variation in the field gap, the length of the excitation (corresponding to the FWHM of the line profile) along the beam axis was determined to be ≤ 1 mm, giving an excitation time of ≤ 2 ns at the ~ 0.56-(mm/ns) ion-beam velocity. With this arrangement, ions "see" a laser frequency inside the observation tube that is Doppler shifted by ~ 1 GHz from the peak of the resonance profile, thus ensuring that only fluorescence from ions excited in the field gap is seen inside the observation tube.

Fluorescence-decay curves were obtained by stepping the DTD away from the monochromator slit and counting photons as a function of distance from the excitation point. The step size was 0.5 mm, and the dwell time per

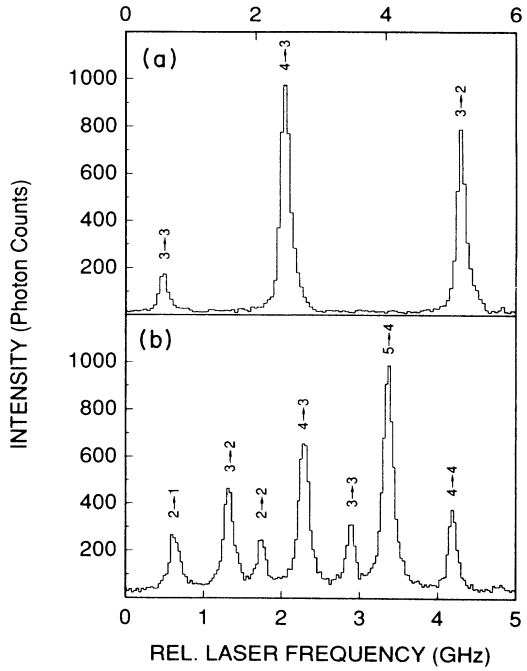

FIG. 4. Hyperfine structure in the odd isotopes of singly charged Xe. (a) Hyperfine multiplet of the $5d\ ^4D_{7/2} \rightarrow 6p\ ^4P_{5/2}$ transition in $^{129}Xe^+$. The nuclear spin of $\frac{1}{2}$ splits each J state into two hyperfine levels. (b) Hyperfine multiplet of the $5d\ ^4D_{7/2} \rightarrow 6p\ ^4P_{5/2}$ transition in $^{131}Xe^+$. The nuclear spin of $\frac{3}{2}$ splits each J state into four hyperfine levels.

channel was normalized to transmitted ion-beam current. Rapid scans in both directions were accumulated to better average small fluctuations in laser intensity. A typical dwell time was ~ 1 s/channel, giving a time of approximately 4 min for one forward and backward scan. The linearity and reproducibility of step size, as well as the collinearity of movement with the ion beam, have been thoroughly investigated in previous beam-foil experiments[14] which used the same translation apparatus. This system is capable of a spatial resolution that is better than $\frac{1}{10}$ mm; however, it is only 2 mm in this case due to the wide spectrometer slits.

Scattered laser light was essentially eliminated by viewing the decay through the monochromator, and by painting the interior and all other visible surfaces of the DTD with black conducting paint (Aquadag) to reduce reflections. Even though the spectral resolution of the monochromator was quite poor (~ 4 nm with a 2-mm entrance slit), the acceptance was sufficiently low so as to cut out most of the scattered laser light, since the laser wavelength was nearly 80 nm from the decay wavelength. Scans obtained without the ion beam showed that there was still some scattered light from the aperture plates at the position of the field gap, but there was none throughout the length of the observation tube where the lifetime measurements were made. Background from collision-induced fluorescence from the ion beam was also found to be negligible. Weak diffuse reflections of laser-induced ion-beam fluorescence from the black inner sur-

face of the observation tube may have given a small nonuniform background; however, no serious effect could be detected.

The localization of the excitation was checked periodically throughout the accumulation of each decay curve by checking the resonance voltage (i.e., the observation-tube voltage at which the peak of the velocity distribution is in resonance with the laser frequency) and then resetting the laser frequency to the appropriate value to bring the position of the resonance in the field gap to its original value. This was often necessary because of a long-term drift in the accelerator high voltage, but this drift could be efficiently tracked in this manner. Occasional mode hops in the laser frequency also occurred, which usually meant losing the data accumulated for a particular curve up to that point and starting again.

A typical decay curve obtained in this manner is shown in Fig. 5. The data were fitted using the DISCRETE computer program,[16] with parameters set to allow for up to two exponential components. The allowance for a second component served as a check on the validity of a data set, since the decay curves obtained in this manner should be cascade-free. Each curve was fitted between several beginning and end channels to check for consistency of the data. The close agreement between the fitted straight line and the logarithm of the data indicates a single-exponential decay, and thus suggests the absence of serious systematic errors. Nonetheless, possible sources of systematic error were investigated.

Stark shifts of the levels of concern are a potential problem with this technique. The excitation takes place inside an electric field which reaches a maximum value of ~ 250 V/mm along the center of the ion beam, becoming as high as 500 V/mm at the outer fringes of the ion beam, and the laser beam produces a much weaker ac field which has a maximum of ~ 5 V/mm at full laser power. The linear Stark effect does not exist for these states of well-defined parity; however, mixing from the quadratic Stark effect might shift energy levels, making selective excitation of closely spaced levels difficult. Contrary to the work of Gaillard *et al.*[7] and Pegg *et al.*,[17] the decay of the excited state is observed outside of the static electric field with this technique. Although the ac laser field is present throughout the observation region, it is very unlikely that this much weaker field could affect these measurements. Stark polarizabilities, which were determined from calculated oscillator strengths, resulted in energy level shifts of ≤ 1 MHz for the $6p\ ^4P_{5/2}$ level and ≤ 0.6 MHz for the lower $5d\ ^4D_{7/2}$ level at the maximum electric field value of 500 V/mm. Thus, Stark mixing has a negligible effect for this particular case.

Since the spectra were normalized to ion-beam current, and not to the total initial excitation intensity, we investigated the variation in excitation efficiency along the beam over the distance for which lifetime data were collected. This was accomplished by "viewing" a region near the upstream end (with respect to the ion-beam direction) of the observation tube through an optical fiber, one end of which remained in a fixed position on the DTD. There was no filtering of scattered laser light, so this source gave a large contribution to the total count rate in the PM tube at the other end of the optical fiber. By subtracting one scan of scattered laser light (without the ion beam) from a scan of combined beam fluorescence and scattered light, the excitation efficiency was verified to be constant to at least within 5% for the 5-cm scanning dis-

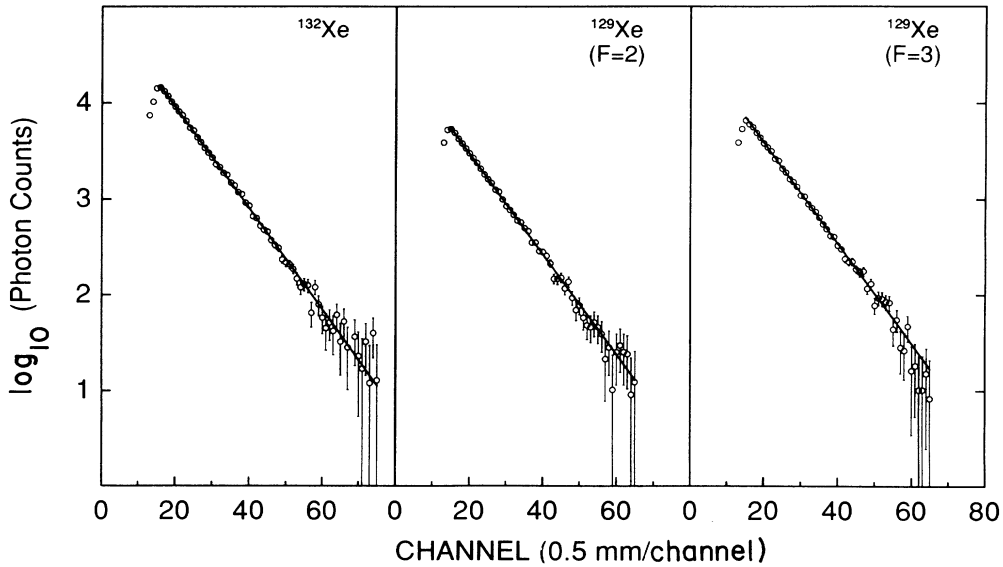

FIG. 5. Lifetime-decay curves for the $6p\ ^4P_{5/2}$ level in singly charged ^{132}Xe and ^{129}Xe. The number of photons counted (at 529.2 nm) is displayed as a function of distance from the point of laser excitation of the fast-ion beam. The isotope and hyperfine level are noted within each frame. Each curve was fitted to a single-exponential function which is shown as a solid line. Data and curves are plotted on a logarithmic scale with fitted background subtracted from each point. Error bars are derived from standard counting statistics.

tance. The possibility of ion-beam focusing effects, caused by lens action of the apertures on the DTD, was investigated in two ways. A ray-tracing computer program[18] showed that at the beam energy used there were no visible focusing effects. But in order to ensure that no systematic effects were introduced by the choice of polarity and value of the observation-tube voltage, decay curves were collected and analyzed for tube voltages in the ranges of -600 to -1000 V and $+600$ to $+800$ V, and no deviation of fitted decay constant outside statistical errors was found. In each case, the tube voltage must naturally still be sufficiently different from the resonance voltage in order not to have excitation inside the observation tube. Effects of power broadening were also of concern, so decay curves were collected for various laser intensities with no noticeable effect on the measured lifetime. Transit-time broadening is a potential problem, since the natural line profiles are Fourier broadened by short excitation times. This effect is expected to limit itself, however, at around 150 MHz (FWHM), because the excitation time in the field gap increases as the line profiles broaden.

RESULTS AND DISCUSSION

^{132}Xe

A set of 13 decay curves were collected in order to determine the lifetime of the $6p\ ^4P_{5/2}$ level in ^{132}Xe. Figure 6 displays the scatter in experimental lifetime values obtained from these curves. Distance along the ion-beam axis was converted to a time scale from a determination of the ion-beam velocity. The velocity was calculated from the measured Doppler shift of the well-known vacuum wavelength of the excitation transition.[19] The uncertainty associated with the laser wavelength meter, combined with changes in velocity during data acquisition (as a result of the accelerator high-voltage drift) produce a relative velocity error of $\leq 0.2\%$. The mean value for the lifetime of this level in ^{132}Xe was found to be 8.03 ± 0.10 ns, and is denoted in Fig. 6 by the solid line along with the error bars (one standard deviation) which are represented by dashed lines. The error bars include the statistical error derived from a calculation of the mean value of the decay constants for 13 curves, the uncertainty in the velocity calibration, as well as an estimate of the total contribution of the systematic errors discussed above. The present measurement agrees, at a comparable level of uncertainty, with the previous two most recent and reliable measurements. Ward *et al.*[20] obtained a value of 7.90 ± 0.15 (two standard deviations) using a crossed laser- and ion-beam arrangement, and Pegg *et al.*[17] arrived at a value of 7.95 ± 0.16 (one standard deviation) using a collinear geometry.

^{129}Xe

It was then possible to directly use this technique to measure the lifetime of each of the two hyperfine levels $F=2,3$ of the same state in ^{129}Xe. In Fig. 4(a) one can see that the transitions to these upper levels are separated by more than 2 GHz, which is equivalent to postacceleration voltages greater than 800 V (Fig. 2). It was therefore possible to set the laser frequency to excite only the $5d\ ^4D_{7/2}$ $(F=3)\rightarrow 6p\ ^4P_{5/2}$ $(F=2)$ transition in the center of the field gap (e.g., at -370 V for a tube voltage of -800 V) in order to measure the lifetime of the $F=2$ level. With this excitation scheme, neither the $4\rightarrow3$ or $3\rightarrow3$ transitions could be excited, since they lie 2.8 and 4.8 GHz, respectively, below the effective excitation frequency for the $3\rightarrow2$ transition. Eight decay curves were obtained in the same manner as for ^{132}Xe, one of which is shown in Fig. 5. The fitted results for each curve are displayed in Fig. 6, along with the mean value and error bars (8.02 ± 0.15 ns). Similarly, the laser frequency was next set to excite only the $4\rightarrow3$ transition in the field gap, and a set of eight decay curves were collected for the $6p\ ^4P_{5/2}$ $(F=3)$ level. A typical decay curve for this level is also shown in Fig. 5 and corresponding fitted values for each curve in Fig. 6. The mean value for the $F=3$ lifetime was 8.00 ± 0.15 ns. Again, the error bars represent one standard deviation and include statistical as well as estimated systematic errors. The increase in the uncertainty compared to the measurement for ^{132}Xe

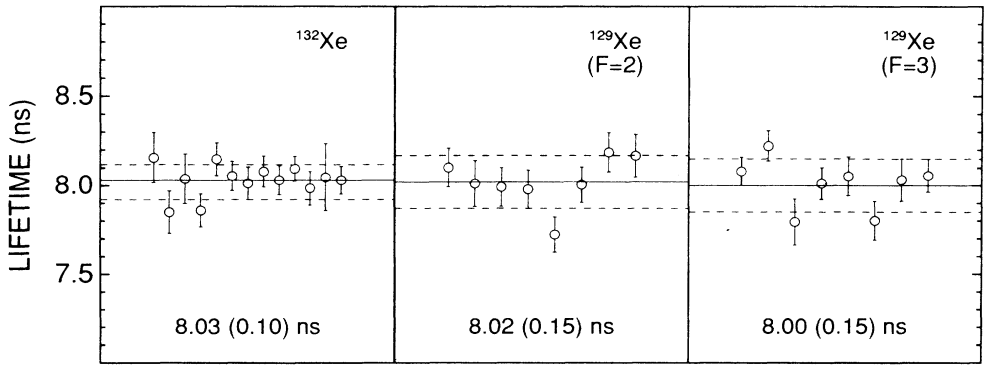

FIG. 6. Experimental values for the lifetime of the $6p\ ^4P_{5/2}$ level in singly charged ^{132}Xe and ^{129}Xe. Fitted results from individual decay curves are shown. The isotope and hyperfine level are noted within each frame. Error bars for each value represent the statistical uncertainty from fitted results. The arithmetic mean for each set of decay curves is given in the lower section of each frame. Error bars (in parentheses) include statistical errors as well as estimated uncertainty from systematic errors.

arises primarily from poorer signal-to-noise ratios, since the population of ions originally in the necessary metastable state for laser excitation is split in this case between the $F=3$ and 4 hyperfine levels in ^{129}Xe.

^{131}Xe

The other isotope that exhibits hyperfine structure (^{131}Xe) has a nuclear spin of $\frac{3}{2}$ which splits each J level into four F levels, giving a more dense and complicated spectrum of allowable transitions. Figure 4(b) shows the strongest seven transitions in this hyperfine multiplet—missing are two very weak allowable transitions ($2\rightarrow3$ and $3\rightarrow4$) which could be detected in spectra with better statistics, but are of no intrinsic interest for the lifetime measurements described in this paper. The close spacing of the spectral lines precluded direct use of the technique described above to determine the lifetime of each upper level, since in most cases, if one transition is excited in the electric field, an adjacent transition would be excited (at least with some probability) inside the observation tube. The fluorescence from the latter excited state would distort the decay curve for the intended level, making an accurate determination of the lifetime impossible.

In order to circumvent this potential problem, the technique of "optical forepumping"[12] was introduced prior to the DTD. A single transition to each upper level was isolated by depletion of close-lying hyperfine levels of the metastable $5d\ ^4D_{7/2}$ state, thus allowing the lifetime of each excited hyperfine level to be measured. Figure 7 (top row) shows portions of the ^{131}Xe hyperfine multiplet [cf. Fig. 4(b)] without optical pumping of selected hyperfine levels of the $5d\ ^4D_{7/2}$ state. The bottom row of Fig. 7 shows the pumping scheme for lifetime measurements of each of the four hyperfine levels in the upper $6p\ ^4P_{5/2}$ state. Each spectrum was obtained by scanning the observation-tube voltage while keeping a fixed laser frequency. The voltage was scanned in both directions, while normalizing to a preset value of integrated Faraday cup current, and each spectrum is the result of several complete scans.

Figure 7(a) displays the scheme used to measure the lifetime of the $F=1$ level of the $6p\ ^4P_{5/2}$ state. The $2\rightarrow1$ transition was isolated in the DTD field gap in the following manner. The laser frequency was chosen to be in resonance with the $3\rightarrow2$ transition at the velocity the ions had prior to reaching the DTD. Consequently, this transition was in resonance throughout the beam line and the population of the lower $F=3$ level was efficiently depleted prior to the lifetime measurement region. Ions were then excited from the lower level $F=2$ to the upper level $F=1$ using a positive observation-tube voltage, thus slowing the ions down to bring them into resonance with the fixed laser frequency in the field gap of the DTD. Since no transitions lie on the lower-frequency side (left-hand side in Fig. 7) of the $2\rightarrow1$ transition, depletion of other hyperfine levels was not necessary. For this particular case, the transition to the $F=1$ level was in resonance at $+260$ V, and the observation tube was held at $+500$ V, thereby placing the transition in approximately

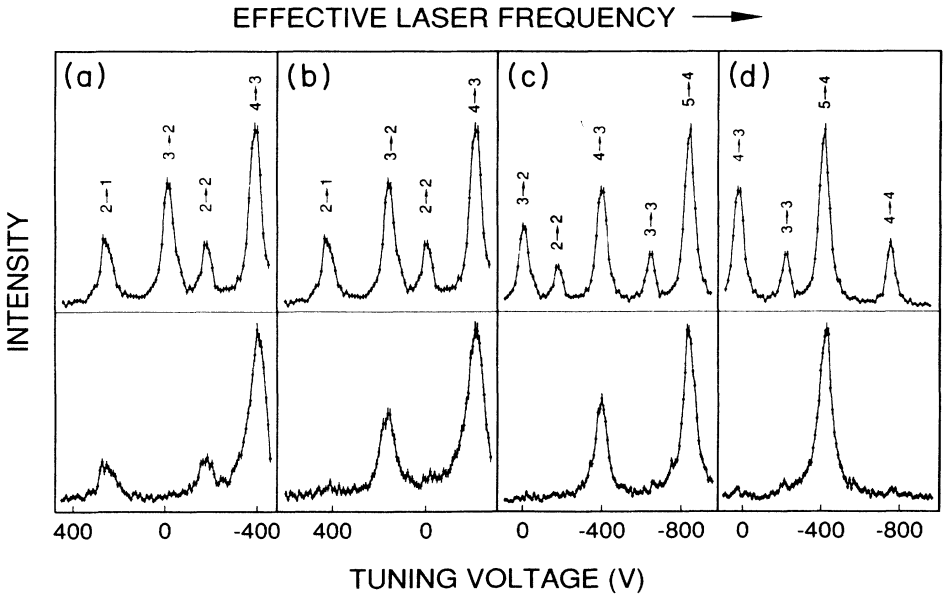

FIG. 7. "Optical forepumping" schemes for lifetime measurements of each hyperfine level of the $6p\ ^4P_{5/2}$ state in singly charged ^{131}Xe. The top spectra are unmodified portions of the $5d\ ^4D_{7/2}\rightarrow6p\ ^4P_{5/2}$ hyperfine multiplet. The lower spectra show the effect of the "optical forepumps." (a) The $2\rightarrow1$ transition has been isolated by selective depletion of the lower $F=3$ level. Measurement of the lifetime of the excited upper $F=1$ level was then possible. (b) The $3\rightarrow2$ transition has been isolated by selective depletion of the lower $F=2$ level. Measurement of the lifetime of the excited upper $F=2$ level was then possible. (c) The $4\rightarrow3$ transition has been isolated by selective depletion of the lower $F=3$ and $F=2$ levels. Measurement of the lifetime of the excited upper $F=3$ level was then possible. (d) The $5\rightarrow4$ transition has been isolated by selective depletion of the lower $F=3$ and $F=4$ levels. Measurement of the lifetime of the excited upper $F=4$ level was then possible.

the middle of the field gap, as in previous measurements. Three lifetime curves were obtained, one of which is shown in Fig. 8, giving a mean experimental value of 7.9±0.3 ns for the lifetime of the $6p\ ^4P_{5/2}$ $F=1$ level in ^{131}Xe.

Figure 7(b) demonstrates the pumping arrangement to isolate the 3→2 transition in order to determine the lifetime of the $F=2$ excited level. In this case, the laser frequency was chosen so as to be in resonance with the 2→2 transition along the beam line. Consequently both the 2→2 and 2→1 lines were effectively quenched, since they both arise from excitation from the $F=2$ lower level. The lower $F=2$ level may, in principle, be repopulated as the upper $F=2$ level decays. If repopulation of this "optically pumped" level were substantial, however, evidence for this would be directly seen in Fig. 7(b) (the same reasoning applies to discount possible repopulation of other F levels discussed below). With this arrangement, the 3→2 transition was excited in the field gap at approximately +170 V, and the observation tube was held at +500 V, as above, Two decay curves were then accumulated (see, e.g., Fig. 8), yielding a lifetime of 8.1±0.3 ns for the $F=2$ excited hyperfine level.

The "optical forepumping" scheme was more complicated for the remaining two hyperfine levels. The lifetime of the excited $F=3$ level was measured by isolating the 4→3 transition in the following way, as displayed in Fig. 7(c). The lower $F=3$ level was depleted by setting the laser frequency in resonance with the 3→2 transition along the beam line, and thus both the 3→2 and 3→3 lines were eliminated. The nearby 2→2 was quenched using the nearer (and shorter) of the two "optical forepumps", which are shown schematically in Fig. 1. The "forepump" tube was held at +250 V, thereby bringing the 2→1 transition into resonance along the 60-cm length of the tube, to optically pump the lower $F=2$ level. In this way, the 2→2 line was eliminated from the spectrum. This scheme placed the 4→3 transition of interest at resonance at −405 V inside the excitation field gap, using an observation-tube voltage of −600 V. A value of 8.4±0.4 ns was obtained for the lifetime of the $F=3$ level from the average of two decay curves, one of which is shown in Fig. 8. Figure 7(d) shows the pumping scheme for isolating the transition to the excited $F=4$ level. In this case, the 4→3 transition was in resonance along the beam line, thereby quenching both this line and the 4→4 line. The "forepump" tube was held at +405 V, which brought the 3→2 transition into resonance along the length of the tube, thus quenching the 3→3 line as well. The 5→4 transition was then excited at −420 V inside the excitation field gap, and the observation tube was held at −960 V. The experimental value for the lifetime of the $F=4$ upper level, derived from fits of two decay curves (Fig. 8), was determined to be 8.1±0.3 ns.

As with the other measurements, error bars represent the statistical and estimated possible systematic error for each set of measurements. The larger error bars for these measurements, as compared to those for the measurements for the other isotopes, are primarily due to the weaker intensity for these transitions. The lower count rate gave a poorer signal-to-noise ratio for each decay curve and greatly increased the amount of time required to accumulate each curve, which made it impractical to collect many curves for each excited hyperfine level. Another contributing factor, however, was the long-term drift in the accelerator high-voltage supply, which gradually changed the Doppler-shifted laser frequency that the ions experienced throughout the beam line and DTD. This gradual drift did not only shift the resonance position slightly in the field gap (which should have no pronounced effect on the decay curve), but also decreased the

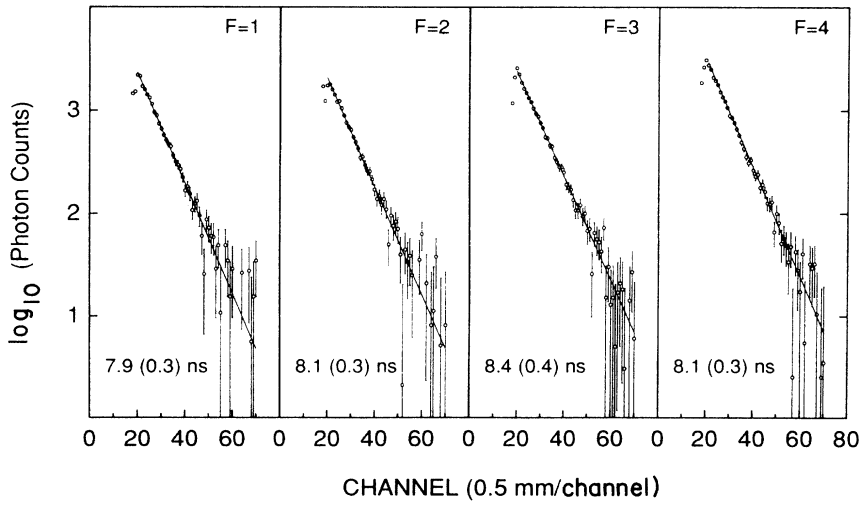

FIG. 8. Lifetime-decay curves for the $6p\ ^4P_{5/2}$ level in singly charged ^{131}Xe. The number of photons counted (at 529.2 nm) is displayed as a function of distance from the point of laser excitation of the fast-ion beam. The appropriate hyperfine level is noted within each frame. Each curve was fitted to a single-exponential function which is shown as a solid line. Data and curves are plotted on a logarithmic scale with fitted background subtracted from each point. Error bars for each data point are derived from standard counting statistics. The mean value for the lifetime, derived from a set of decay curves for each hyperfine level, is given in the lower section of each frame. Error bars (in parentheses) include statistical errors as well as estimated uncertainty from systematic errors.

optical pumping efficiency, since the effective laser frequency was shifted slightly off the peak of the velocity distribution. This effect was most pronounced in the decay curves for the $F=3$ excited level, since this measurement relied most heavily on the lower $F=3$ level being completely depleted. In this case, it was necessary to set the observation-tube voltage such that the $3\rightarrow 3$ transition could possibly be excited throughout the observation region, should there be some population of $5d\,^4D_{7/2}$ $F=3$ remaining in the ion beam when it reaches the DTD. The slightly higher value for this lifetime is believed to be due to this effect, and hence larger error bars were assigned for this measurement. The effect could not have been too severe, however, since the decay curves were still described well by single-exponential functions.

The longer optical forepump (Fig. 1) was not actually used in these measurements, but would provide additional pumping for even more complicated spectra. A limitation in the pumping effectiveness, however, is the divergence of the laser beam and ion beam. The focusing of both beams is chosen so as to optimize excitation in the field gap of the DTD, while keeping the overlap uniform through the scanning region for lifetime measurements. Consequently, the percentage of overlap, and therefore optical pumping efficiency, decreases with distance from the DTD. The net effect is to make the use of the first "optical forepump" less efficient than the second, despite the additional length. Improvement of the ion-beam quality (i.e., more parallel beam, and stabilization of the accelerator high voltage to eliminate the drift) and the fluorescence collection system should greatly increase the applicability of this technique to other systems and increase its inherent accuracy in the future.

CONCLUSIONS

The lifetimes of each hyperfine level of the $6p\,^4P_{5/2}$ state in the odd mass isotopes of Xe have been determined using a new technique designed to allow lifetime measurements at high spectral resolution, even for crowded spectra. No differences were found for the lifetimes of these levels at the present level of precision. To the best of our knowledge, these are the first lifetime measurements of each hyperfine level in an excited state of an atomic ion. Measurements of this type might play an important role in investigations of hyperfine-induced mixing in atomic ions, where mixing of hyperfine levels with other configurations results in a variation of lifetime with F quantum number. It has previously been shown[11] that lifetime measurements provide a sensitive means to extract mixing coefficients for individual hyperfine levels by comparing these results with the "unperturbed" lifetime of the same state in an isotope which has no hyperfine structure. The technique of "optical forepumping," however, might also have a wider range of applicability, since it provides a rather straightforward means of investigating unexplored multiplets or complicated spectra. Identification of all lines originating from a particular lower level would be immediately possible after this level has selectively been depleted using this technique. Troublesome line blends may also be reduced or eliminated with this method. In most cases, a blending line can be quenched by optical pumping of the lower level using another transition (at a substantially different laser frequency) originating from this level, thus not affecting the line of interest.

ACKNOWLEDGMENTS

This research was supported by the Swedish National Science Research Council (NFR). We also thank C.-G. Wahlström for his help in estimating the influence of Stark mixing.

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