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New technique for measuring excited-state lifetimes in ions using rapid Doppler switching

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A new and accurate method for measuring lifetimes of excited ionic states is described. The method employs selective laser excitation and rapid Doppler tuning to convert the spontaneous exponential decay of the excited state into a spatial variation of fluorescence intensity. The technique has been tested on the $6p\ ^2P^o_{3/2}$ Ba II level and our final result of 6.35 ± 0.09 nsec is in excellent agreement with previous high-precision measurements. The method should make it possible to routinely achieve 1 percent accuracy measurements of ionic lifetimes longer than a lower limit of a few nanoseconds, set by transit-time broadening and ion-beam velocity spread.

I. INTRODUCTION

Time-resolved experiments using fast-ion-beam laser spectroscopy (FIBLAS) are deceivingly simple in principle.[1] They have proven, however, to be experimentally rather complex and time consuming. In this paper, we introduce a new experimental method which should considerably simplify the data acquisition procedure while hopefully retaining the high-precision potential of previous selective laser excitation methods.

Starting with the pioneering work of Andrä *et al.*,[2] most previous FIBLAS lifetime measurements have been obtained by the crossed ion-laser-beam time-of-flight technique. In this arrangement, effectively a pulse of optical excitation is applied to the ions as they cross the laser beam in a time which can be made as short as one-tenth of a nanosecond. The resonance condition is achieved by either laser[3,4] or Doppler wavelength tuning.[2,5–8] Decay of the subsequent fluorescence is then monitored by varying the distance between a detector and the region of excitation. The ion-beam velocity, which is needed in this time-of-flight method to scale decay distances into time, must be measured independently using, for example, a calibrated electrostatic analyzer.[2]

Similar results have been obtained in the coaxial ion-laser-beam geometry introduced by Koch *et al.*[9] In this case, one can use a Doppler switching technique[10] whereby the ions are brought suddenly in and out of resonance with the applied laser light by in-flight changes of ion velocity. Winter and Gaillard[11] demonstrated that proper tailoring of a set of accelerating-decelerating electrodes can produce switching times in the nanosecond range. In the ion reference frame this is equivalent to the application of a laser pulse which is resonant whenever Doppler-tuning conditions are satisfied. In the early applications of the Doppler-switching concept, lifetime measurements were carried out by moving the set of electrodes upstream with respect to the observation point of a transverse detection system monitoring the beam fluorescence.[12] The ion-beam velocity was measured by determining the electric potential needed to Doppler shift a transition in the ion rest frame into resonance with the laser light.

Each of the beam-laser methods cited, however, requires accurate and necessarily slow mechanical motions under vacuum of either the target or the observation region. In the "rapid-Doppler-switching" technique, which we shall now introduce, the relative motion between the excitation and detection points is obtained by purely electrical means. This results, as we shall show, in a considerable simplification of the experiment. Further-

more, this new method circumvents the difficulties associated with ion-beam velocity measurements and requires instead accurate electric field and laser wavelength interval measurements which are somewhat easier to obtain.

After a brief exposition of the principles of the method, we shall describe the experimental arrangement which has been used in the initial experiment with encouraging results. From this set of measurements, we conclude that the present method should be applicable to atomic ions with levels which do not suffer appreciably from Stark quenching in electric fields of the order of a thousand volts per centimeter. The method is suitable for lifetimes in the range of a few to several hundred nanoseconds. As we shall demonstrate below, the lower limit is set principally by transit-time broadening effects present when the laser and ion beams interact for a very short time. As with other fast-beam methods, the upper limit arises from the prohibitive length of beam path which would have to be observed in order to collect the decay information on long-lived excited states. This laser excitation method is also clearly restricted in application to resonance levels or excited levels which are optically coupled to a metastable level that can be collisionially excited in the beam prior to the laser interaction. Finally, the method, being based on electric-field-induced velocity changes, applies only to ions.

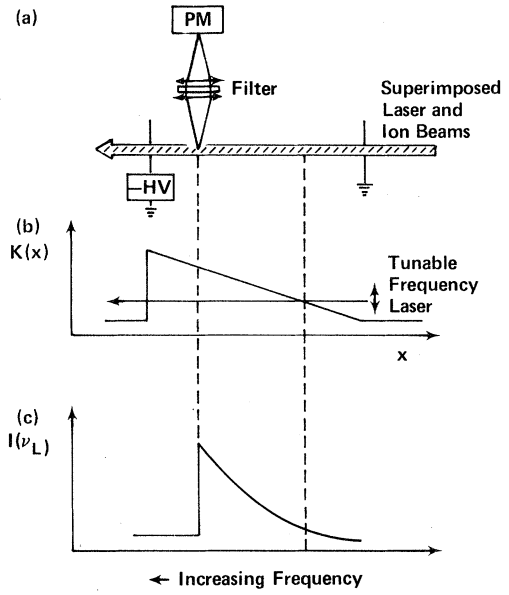

FIG. 1. Principle of the rapid Doppler switching method. (a) A schematic of the experimental arrangement showing the collinear beams passing through a parallel plate capacitor. (b) The ion kinetic energy $K(x)$ as a function of the distance along the common beam axis. (c) The expected variation of the recorded fluorescence as a function of laser frequency.

II. PRINCIPLE OF THE METHOD

The present technique makes full use of the narrow absorption line width which results from the velocity-bunching effect[13,14] obtained in coaxial fast ion-laser-beam arrangements. A number of recent FIBLAS experiments[15–19] based on this principle have achieved very high spectral resolution both with atomic and molecular ions.[20] With proper regard for the stability of the ion source and of the accelerating high-voltage power supply, it has been possible to observe resonances as narrow as 40 MHz full width at half maximum (FWHM). In addition, modern cw tunable dye lasers are able to deliver as much as 1 W in a single 1-MHz-wide laser mode. Doppler switching of such a laser line across the narrow beam absorption line can be achieved in a very short time in the ion rest frame by simply sending the superimposed ion and laser beams through a coaxial static homogeneous electric field zone operating either in an accelerating or decelerating mode as indicated in Fig. 1. Excitation occurs in front of the detection window whenever the laser wavelength is such that the Doppler-tuning condition is satisfied at the observation point in the accelerating (decelerating) field. For other wavelengths the excitation will occur at some other point in space along the common beam path. With a sufficiently narrow observation window, one can thus record a decay curve by simply monitoring the fluorescence at a given point along the ion path in the accelerating (decelerating) zone while tuning the laser wavelength. This is equivalent to moving the excitation point relative to the observation point. Alternatively, one could keep the laser wavelength fixed and change the incident-beam velocity, a solution which might be preferable under some circumstances. The main properties of the rapid-Doppler-switching method are readily established by considering the relationship between the spectral width of the laser Δ_L, the absorption width Δ_B of the ion beam, and the effective excitation time T. These variables are not entirely independent since transit-time broadening is likely to affect the absorption width for sufficiently short resonant interaction times.

Let us consider first that the ion beam has a well-defined initial velocity and is subjected to the

constant acceleration imparted by a coaxial electric field E. We shall further assume that power broadening has been made negligible by the use of sufficiently low laser power. Resonant excitation will occur over a beam length d, given by

$$d=\frac{W}{E(d\nu/dV)}=vT\ , \tag{1}$$

where W is the apparent laser width in the ion frame which moves with velocity v (assumed to be constant over the short distance d), $d\nu/dV$ is the dispersion of the Doppler-tuning process, and T is the width in the time domain of the equivalent resonant laser pulse applied to the ions. The dispersion can be written in the form

$$\frac{d\nu}{dV}=\frac{-2q}{Mc}\frac{\lambda_L}{\lambda_A^2-\lambda_L^2}\ , \tag{2}$$

where q is the ion charge, M is the ion mass, c is the velocity of light in a vacuum, ν is the frequency of the light, V is the accelerating voltage, λ_A is the wavelength of the transition in the ionic rest frame, and λ_L is the laser wavelength. It should, however, be quite clear that T and W are ultimately related to one another via a Fourier transformation between the time and frequency domains. Thus, adding quadratically the various contributions to the laser linewidth it can be seen that the following relation must also be satisfied:

$$W^2=\Delta_B^2+(2\pi T)^{-2}\ . \tag{3}$$

A combination of Eqs. (1) and (2) leads to a unique determination of both T and W. These quantities have been plotted in Fig. 2 for a case of practical interest.

In a real experiment, one must also take into account the initial spread of longitudinal-beam velocities, which tends to extend the resonant interaction region along the ion-beam path more than would be the case for a monoenergetic beam and thus enlarges the excitation time window as seen by the moving ions. Due to velocity bunching however, this effect is not nearly as detrimental using a fast ion beam as it would be in a low-energy-beam experiment. Assuming that the beam absorption linewidth Δ_B in zero applied electric field is no larger than 100 MHz, it is easy to show that the resonant interaction condition is satisfied over a maximum beam length of only a few 10^{-4} m in an applied field of the order of 40 kV/m. In the ion frame, this corresponds to an excitation time window in the nanosecond range; that is, rapid-Doppler-switching can be used to generate nanosecond-wide resonant light pulses in the ion reference frame.

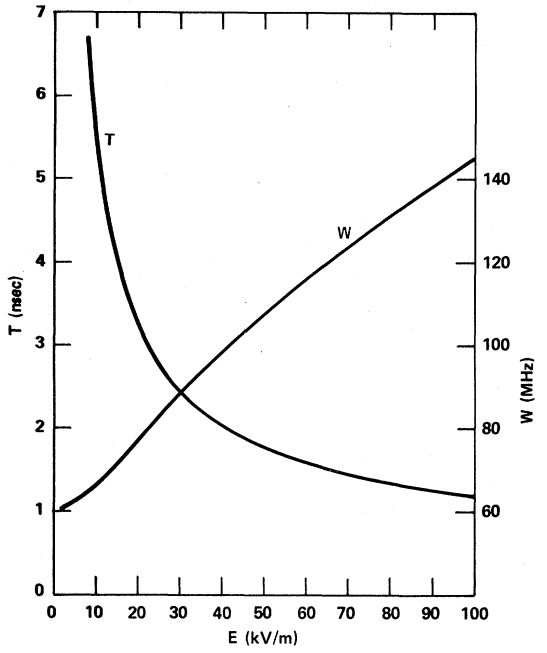

FIG. 2. Theoretical variation of the resonance transit time T and of the corresponding spectral width W as a function of the applied longitudinal electric field. The width W is a quadratic sum of contributions from the Doppler width and transit-time broadening. The theoretical curves shown as an example have been plotted for a ^{137}Ba II ion beam under the assumption that the Doppler broadening Δ_B is 60 MHz. This value was measured using an ion-beam velocity of 2.7×10^7 cm/s.

In order to extract lifetime information from the raw fluorescence data, it is necessary to determine a relationship involving time intervals between excitation and observation and the other experimental parameters. By monitoring the fluorescence in a region of space where the ions undergo a uniformly accelerated motion under an applied coaxial electric field E, the relationship between time intervals in the ion rest frame and the laser and ion wavelengths is

$$t-t'=\frac{2Mc}{qE}\frac{\lambda_A^2(\lambda_L-\lambda_L')(\lambda_L+\lambda_L')}{(\lambda_A^2+\lambda_L'^2)(\lambda_A^2+\lambda_L^2)}\ , \tag{4}$$

where primed and unprimed quantities correspond to values at different points along the beam path. The above equation reduces in first order to

$$t-t'\simeq\frac{M\lambda_A\Delta\nu_L}{qE}\ , \tag{5}$$

where $t-t'=\Delta t$ is the time interval between two excitation points and $\Delta\nu_L$ is the corresponding change in the laser frequency. The above equation shows that an approximate linear relationship exists between ion rest-frame time intervals and laser frequency intervals under the aforementioned experimental conditions. Equation (5) remains a good approximation at the 0.1% level for decay times corresponding to frequency intervals less than a few gigahertz. For longer sweeps and higher accuracy, the nonlinearity expressed by Eq. (4) in the frequency-to-time conversion should be taken into account. Another possible source of nonlinearity is evident in Eq. (4). Since the excitation at various points along the decay curve occurs for different values of the laser wavelength λ_L' there are slight systematic changes of the excitation time window, $\Delta t(\lambda_L')$, during a lifetime scan. This effect is again negligible for frequency excursions which do not exceed a few gigahertz.

Some important attributes of the technique are that the experimental time scale is established with one absolute wavelength measurement λ_A, one relative wavelength measurement $\Delta\lambda_L$, and one electric field measurement E, all quantities which can be accurately determined. In addition the technique is independent, to first order, of beam energy (velocity), is state excitation selective (and thus cascade free), and the measurement is done in high vacuum where collisional de-excitation effects are virtually eliminated.

III. EXPERIMENTAL ARRANGEMENT

A new FIBLAS apparatus has recently been constructed at the UNISOR mass separator at the Holifield Heavy-Ion Facility in Oak Ridge. Properties of the original UNISOR (University Isotope Separator—Oak Ridge) machine have been described in previous papers[21,22] and need not be repeated here. The new beam line is schematized in Fig. 3. It includes a four-plate electrostatic beam steerer which is used to make the ion beam overlap with the laser beam. The laser beam enters the beam line through a Brewster window and a hole in the last set of plates of the beam steerer. An Einzel lens (not shown) is used to optimize the ion optical properties of the beam before deflection. In order to avcid collisional quenching as well as collision-induced beam excitation, the background pressure in the beam line is maintained below 2×10^{-6} Torr by a 750 l/s turbomolecular pump.

The laser is a tunable, actively stabilized, ring dye laser system commercially available from Coherent Radiation. Two spectrum analyzers of free spectral ranges 30 and 1.5 GHz are used to monitor the laser scans. The laser wavelength is measured to a few parts in 10^7 with a traveling Michelson interferometer wave meter[23] which compares the unknown wavelength to that of a Lamb dip-stabilized HeNe laser at $\lambda=632.991\,45\pm0.000\,01$ nm.[24] The fluorescence spectrum from a room-temperature iodine vapor cell is recorded simultaneously with the ion-beam data as a wavelength reference which is based on the spectral tables of Gerstenkorn and Luc.[25] This spectrum is also used to monitor laser mode hops and correct for long-term frequency drifts of the laser.

Details of the laser-ion-beam interaction region are given in Fig. 3. Potentials applied to the accelerating (or decelerating) field plates are measured with a high-precision ($1/10^4$) voltage divider and a calibrated differential voltmeter.

Fluorescence photons from the interaction region are analyzed with a 30-cm Czerny-Turner spectrometer viewing perpendicular to the beam axis through two 20-cm focal length lenses which image the vertical spectrometer entrance slit in a 1:1 ratio on the ion-beam path. Great care has been taken to physically delineate the detection optics viewing angle and the laser path with carbon-black apertures and razor-sharp edges in order to reduce scattered laser light. The detection system includes a cooled photomultiplier and single-photon-counting electronics. Data acquisition via a multiple input ADC system interfaced to a PDP 11/34 minicomputer provides for both data storage and on-line data analysis.

The frequency intervals $\Delta\nu_L$ are determined by detecting the transmitted dye laser light through a commercially available spectrum analyzer of known free spectral range. This signal is used as the gate

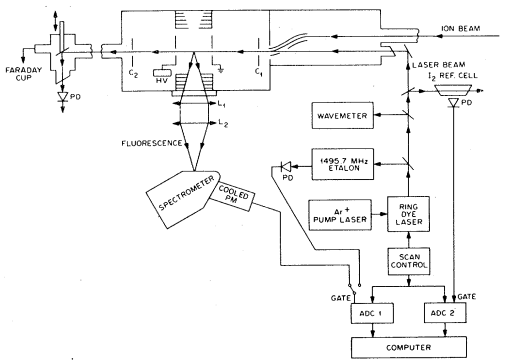

FIG. 3. Schematic diagram of the laser facility at the UNISOR isotope separator.

input to the ADC which causes the ramp from the dye laser control unit to be digitized and stored in the computer. In this way frequency interval markers could be generated quickly and used to calibrate the entire system from the dye laser through the ADC's. The spectrum analyzer itself was calibrated using the technique of exact fractions with an accurate wave meter as described by Goldsmith *et al.*[26] The result for the spectrum analyzer used here was 1495.7±0.1 MHz. This spectrum analyzer is not actively stabilized or evacuated, but at the level of accuracy required for this experiment and the manner in which it is used, such precautions are not needed. The rapidity of the scans relaxes the stabilization requirement and the dispersion changes less than $1/10^5$ for temperature changes of 5° C and a 3% change in atmospheric pressure. All other corrections are insignificant.

Experimentally, the potentially most difficult measurement in this technique is that of the electric field at the site of the interaction. The field plates were constructed in such a fashion as to minimize edge effects, the principal source of error. These plates are 12 cm in diameter and spaced 6.09 cm apart. The entrance and exit apertures for the beams are 0.8 cm in diameter. In order to obtain a uniform electric field throughout the interaction region five guard rings with precision dropping resistors were placed between the two end plates. To determine the uniformity of the electric field along the excitation region we combined rapid Doppler switching and in-flight optical pumping[27–29] to map the field in the region of interest. For this purpose, a second Doppler-tuning zone[27] had to be introduced upstream from the electric field plate region (Fig. 4). Conditions for efficient in-flight optical pumping of three-level systems have been described previously.[29] Velocity selective population changes can be made to produce dips or bumps in the fluorescence observed in the downstream interaction region. By adjusting the electrical potential applied to the upstream interaction region, it is possible to generate a narrow dip in the fluorescence (Fig. 5). Under such conditions the velocity of the ion beam at the position corresponding to the dip is the same as its velocity in the upstream interaction zone which, of course, can be determined by accurately measuring the voltage applied to the upstream zone. Thus we could verify, with a series of such measurements using different potentials, that the motion of the ions between the field plates was to a good approximation uniformly accelerated (i.e., a constant electric field existed in the plate region).

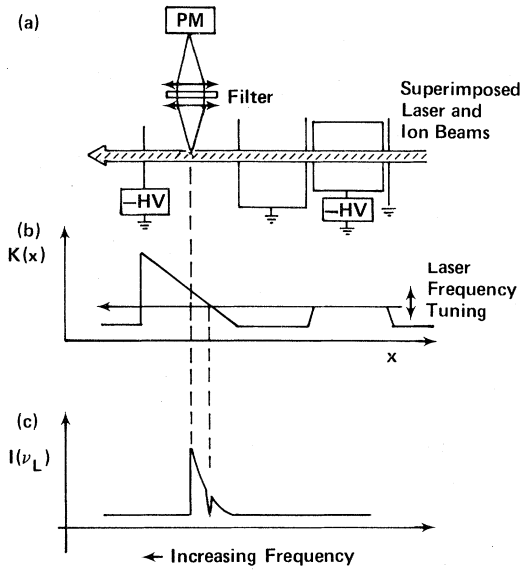

FIG. 4. In-flight velocity labeling by optical pumping in a three-level system. The apparatus shown is used to make velocity-dependent population changes and to monitor the subsequent dips in the fluoresence. The apparatus and description is the same as in Fig. 1 with the addition of a second upstream interaction region.

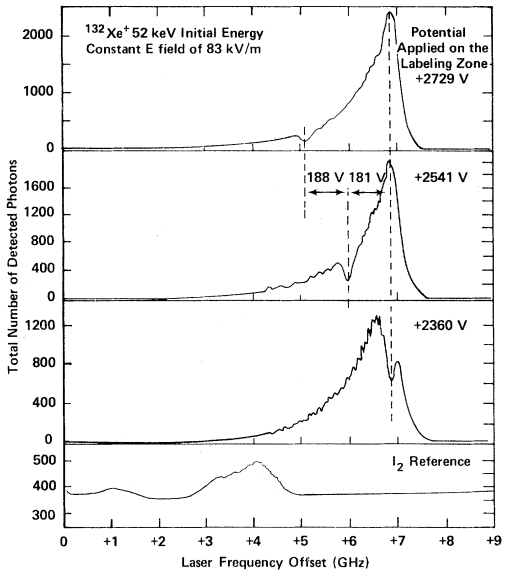

FIG. 5. Experimental results for in-flight velocity labeling obtained using a 52 keV, $^{132}Xe^+$ beam. The lower trace is a reference I_2 vapor spectrum recorded during a 10-GHz laser frequency sweep. Three $^{132}Xe^+$ fluorescent decay curves were recorded for three different values of the electric potential applied to the upstream labeling zone. The fluorescence dip in the downstream probe zone is observed to move across the fluorescence decay curve as a function of tuning the voltage applied to the labeling zone.

In a separate experiment, the absolute value of the electric field in the observation region was determined by observing the fluorescence decay curve under identical conditions except that the plates were moved relative to the point of observation a known amount between measurements. This means we have observed the decay at two different positions and the corresponding potentials, hence we can calculate the magnitude of the electric field. The result is 820 ± 7 V/cm, in excellent agreement with the expected value of 821 V/cm obtained by dropping a known potential over a known distance. We thus believe the value of the electric field used in Eq. (5) is known to better than 1% and is uniform. It may be worth pointing out that other field plate configurations were tried and did not satisfy the various consistency checks suggested.

IV. EXPERIMENTAL PROCEDURES AND RESULTS

The feasibility of our new lifetime measurement technique was tested on the well-known[8] lifetime of the $6p\ ^2P^o_{3/2}$ level in Ba II. The excitation and detection channels used in this work are shown in Fig. 6. The separator ion source was found to deliver a beam with sufficient metastable content for this experiment when operated in either the surface ionization or discharge mode.

The lifetime data were recorded using the apparatus described above and the following procedure. With the laser set for a scan range of 8 or 10 GHz encompassing the 585 nm absorption line, a series of many rapid scans was made while recording the fluorescent decay curve of the 455 nm line. Such rapid scans avoided normalization corrections due to fluctuations in ion- or laser-beam intensities. Simultaneously the I_2 spectra were recorded in another ADC in order to monitor small drifts over long periods and mode hops. Typically a series of several hundred scans was taken in an interval of five to ten minutes, recorded, and the procedure repeated. These groups of scans were adjusted for any slow drifts by monitoring the shifts in the centroids of the I_2 peak(s). The groups were then added together until sufficient counts were attained. Frequency markers were acquired for calibration purposes between the groups of scans by substituting the etalon gate signal (see Fig. 3) for the photomultiplier tube (PMT) gate signal, thus recording the frequency interval markers under the exact conditions used in obtaining the decay data. Fig. 7 shows a set of the Ba II $6p\ ^2P^o_{3/2}$ decay data taken in this manner. A major objection to scanning the laser rather than voltage applied to the observation region is the possibility of laser power

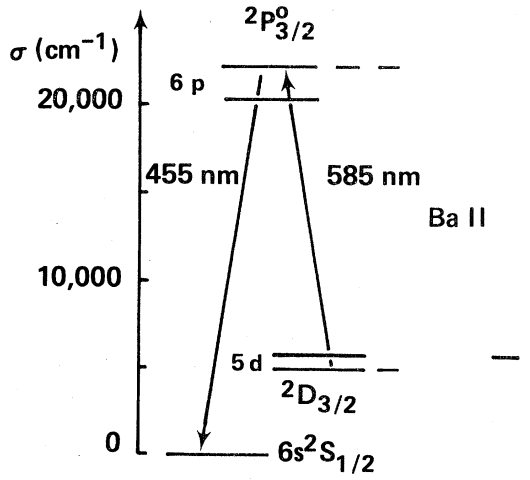

FIG. 6. Partial energy level diagram of Ba II showing the transitions used in the present investigation.

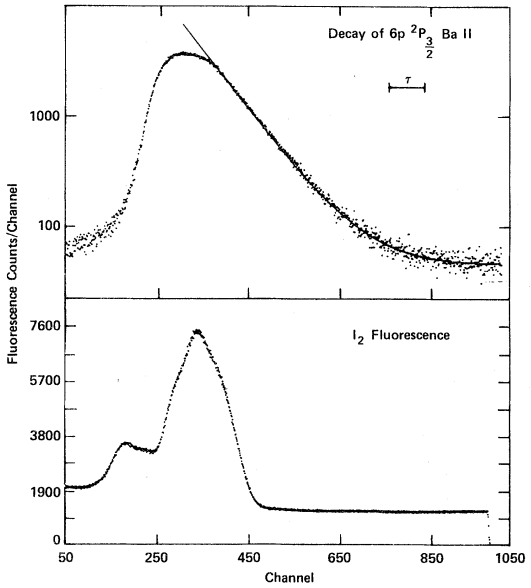

FIG. 7. Decay of the fluorescence at 455 nm in Ba II obtained in the present experiment. For channels close to the peak, the decay is influenced by the instrumental window function. At later channels, where the effect of the window function becomes negligible, the decay data fit well to a single exponential plus a constant (see solid curve). The lower part of the figure shows the I_2 vapor reference spectrum used in the present work to monitor laser frequency changes.

variation at the interaction site which can occur in some cases during dye laser scans. In order to take account of this effect, a number of scans were recorded observing the stray laser light at 585 nm. The variation in background count rate was found to be essentially flat with maximum excursions of only 5%. The background at the Ba II fluorescent wavelength due to collisional excitation of the ions by the residual gas also was examined and found to vary less than 1 percent across the scan range. As can be seen from Fig. 7, these small effects on the small background have negligible effect on the lifetime data. The data were acquired using standard ADC's in a multiscaling mode. As a consequence, the dead time is a function of the count rate and the channel in which the data is stored. A dead-time correction was applied to the data using standard techniques. Even at the highest count rates used in the experiment the dead-time correction to the lifetime was only $\sim 1\%$ and in most cases it was well below this. We thus conclude that this effect contributes much less than 0.5% to the overall uncertainty in the lifetime measurement.

An effect which must be considered carefully is the window function, that is, the effects of the finite sizes of the observation and excitation regions on the decay curve. The data were fit to a function of the form $N = N_0 \exp(-t/\tau) + B$, where N_0, τ, and B are constants determined from the method of least squares. To render negligible the effect of the window function on the decay data, the fitted parameters were determined as a function of the starting channel used in the fit. It can be shown that away from the peak the data approaches the form of a pure exponential plus a constant background. As one moves the starting channel of the fit farther from the peak of the decay curve, the fitted value of the lifetime initially decreases but eventually reaches an essentially constant value indicating that the window function no longer affects the data. We obtained a lifetime value for each decay curve by averaging the fitted values over this flat portion. Excursions from this average were found in most cases to be less than 1%. Figure 8 shows a decay curve with the constant background subtracted. It can be seen that the data are well fitted by a single exponential function.

Seven independent experiments were carried out and analyzed using the procedure described above. The unweighted average of these measurements with the corresponding standard deviation is 6.35 ± 0.05 ns and agrees with the accurate result obtained by Andrä[8] of 6.312 ± 0.016 ns to within 0.5%.

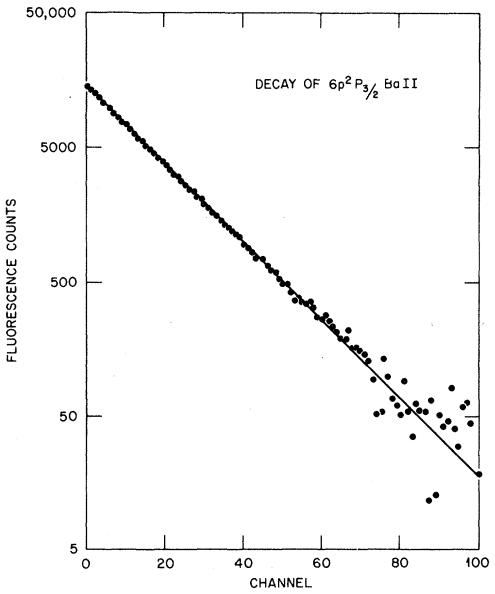

FIG. 8. A semilogarithmic plot of Ba decay data showing the least-squares fit after a constant background has been subtracted. It can be seen that the data are well fitted to a single exponential function from which the lifetime of the $6p\ ^2P^o_{3/2}$ level can be extracted.

V. ANALYSIS OF SYSTEMATIC ERRORS

A thorough inspection of systematic uncertainties is required in order to appraise the potential of any new measurement technique. Since in most of our work the errors associated with counting statistics were negligible, we shall focus our attention on the uncertainties associated with the use of Eq. (5). We have shown above that this approximation of Eq. (4) is good to better than 0.1% under the conditions used in the present work. In the application of Eq. (5) itself the constants M, q, and c are certainly known to better than 0.1%, which leaves the quantities $\Delta\nu_L$, λ_A, and E to be considered. The atomic transition wavelength λ_A is known to a few parts in 10^5 so this source of error is negligible. Our method of calibrating the entire optical plus electronic system with frequency interval markers from an etalon yields an accuracy better than 0.5% for the quantity $\Delta\nu_L$. The series of measurements with in-flight optical pumping indicates the electric field is uniform to better than 1% and we have measured its magnitude with an estimated error of less than 1%. There remain other less well-defined effects

which must be considered carefully before the final accuracy is quoted. These effects include the convergence or divergence of the laser and ion beams, the instrumental window function and the possible influence of the applied electric field on the excited levels.

The influence on the measured lifetime of the change in overlap of the laser and ion beams due to deviations from a perfect collinear geometry is an important consideration. If the beams are not exactly collinear or their relative overlap changes, the rate of excitation can vary for different points along the common beam axis. In the present measurement, however, the range of excitation points relative to the fixed detection point is only 7 mm. Geometrical considerations, including the relative spacings of beam apertures, indicate that there can be no more than a $\sim 5\%$ change in beam overlap over this range of measurements. This upper limit on the effect translates to a $\sim 1\%$ change in measured lifetime of the excited state. The beam overlap effect clearly becomes increasingly important in cases of longer-lived excited states. Stricter collimation of both the laser and ion beams, however, can greatly reduce the contribution to the uncertainty in the measured lifetime due to this effect.

Perhaps the least understood effect in the technique lies in determining the instrumental window function which should be used to deconvolute the fluorescence decay signal from the raw data. The effect of the window function can be minimized by using narrow spectrometer slits and low laser power. By delaying data analysis sufficiently far away from the peak fluorescence, as indicated above, we obtained good fits to the data using a single exponential plus a constant. We thus consider that the window function problem can be properly handled as long as the measured lifetime is larger than the excitation time. The theoretical discussion in Sec. II sets a lower limit in the nanosecond range on the excitation time window. Our experimental findings are in good agreement with the theoretical expectations. The reproducibility of our present result in Ba II, and its agreement with previous measurement shows that the lower limit of applicability of our method, as determined by the width of the window function, is somewhat below the measured 6.35 ns.

Some words of caution must be added concerning the possible influence of the applied electric field on excited level structure and lifetimes. In their recent review of the Stark effect, Kollath and Standage[30] recall the fundamental formula for a nonhydrogenic level:

$$\langle nJm \mid H_E^1 \mid nJm \rangle = \frac{-\alpha_0}{2}E^2 + \frac{-\alpha_2 E^2}{2}\left[\frac{3m^2-J(J+1)}{J(2J-1)}\right], \quad (6)$$

where α_0 is the scalar polarizability and α_2 is the quadrupolar (tensor) polarizability. Neither of these quantities has been measured in the state of interest in Ba II, but two different groups recently devoted their attention to the isoelectronic $6\,^2P^o_{3/2}$ state of Cs I.[31] The latest results indicate that in this case $\alpha_0=0.398(60)$ and $\alpha_2=0.065(10)$, both polarizabilities being expressed in units of MHz/(kV/cm)^2. Clearly in the fields used for the present lifetime experiment (<1 kV/cm), the Stark effect would give rise to very small spectral shifts and splittings in the case of Cs I. Since the isoelectronic variation of the polarizability is at least of the order of Z^{-4}, we can safely conclude that the influence of the applied electric field is also negligible in the Ba II case. In any other particular case this point would require special attention and our conclusion may well be altered for levels with a strong hydrogenlike character. Such states are known to be highly sensitive to electric fields, and Stark-induced state mixing is able to significantly alter the level lifetimes, thus making our method inapplicable in this case.

With the exception of the aforementioned hydrogenic state case, we have demonstrated a new technique for measuring lifetimes in ionic species. From a careful analysis of our systematic errors we estimate the total uncertainty in the lifetime result to be approximately 1.4%. This error limit is supported by the fact that the average of all of our measurements varies no more than 1.1% from the accurate value of Andrä.[8] We therefore quote an uncertainty of 1.4% on our present measurements giving a value of 6.35 ± 0.09 ns. While this is not the most accurate method developed to date, it does have a number of important attributes such as state selectivity and ion-beam velocity independence. It also has the advantage that the scans over the fluorescence curves can be made rapidly so that ion- and laser-beam intensity fluctuations are averaged out. By simultaneously recording a portion of a reference spectrum, one can add together a large number of scans in order to improve the statistics on weak transitions. Hence the overall sensitivity and accuracy of the method should permit significant new results to be obtained, even on weak beams.

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