PHYSICAL REVIEW
LETTERS

VOLUME 31 | 20 AUGUST 1973 | NUMBER 8

New Method for Precision Lifetime Measurements by Laser Excitation of Fast-Moving Atoms

H. J. Andrä, A. Gaupp, and W. Wittmann
I. Physikalisches Institut der Freien Universität Berlin, 1 Berlin 33, West Germany
(Received 21 May 1973)

A new method for precision measurements of atomic lifetimes by laser excitation of fast beams is described which is expected to allow accuracies of $\Delta\tau/\tau = 10^{-3}$. A first application to Ba^{+}-$6^2P_{3/2}$ yields as a preliminary result $\tau = 6.21 \pm 0.06$ nsec.

The present Letter introduces a new and probably the most accurate method to date for measuring radiative mean lives of excited atomic states. The method is expected to allow accuracies of the order of 1 part in 10^3 and is based on selective laser excitation of pure states of atoms or ions in fast monoenergetic beams. It can therefore be considered as a modern improvement of the early experiments by Dunoyer[1] (1913) and Koenig and Ellett[2] (1931), who first tried to measure atomic lifetimes after radiative excitation of thermal atomic beams. Or it can be considered equally well as a modification of the beam-foil spectroscopy[3] (BFS) technique after replacement of the foil by selective laser excitation. Thus the serious cascade problem in lifetime determinations with BFS[4] can be avoided, whereas all the other unprecedented properties of BFS[5] for lifetime measurements are conserved. Namely, one can observe the absolutely undisturbed emission of the excited atoms, completely unaffected by collisions, self-absorption, or induced emission in a vacuum of typically 10^{-6} Torr and at beam densities of typically 10^6 cm^{-3}. Moreover, the high and constant velocity of the beam guarantees an excellent time resolution and exceptional differential as well as integral linearity—in terms of delayed-coincidence techniques[6]—by detecting the photon emission from the beam as a function of distance downstream from the excitation region. Thus one has the cleanest conditions of all lifetime measurement methods[6,7] known, if the laser excitation of fast beams yields a well detectable reemission rate and if the beam velocity can be determined accurately enough for time-scale calibration.

The photoexcitation process is primarily governed by the probability per sec for excitation of an atom[8] in a photon flux $N(\nu)$ (cm^{-2} sec^{-1} Hz^{-1})

$$P = \int_0^\infty N(\nu) \cdot \sigma(\nu)\, d\nu, \tag{1}$$

with the cross section for absorption of a photon[9] of frequency ν

$$\sigma(\nu) = \frac{\lambda^2}{8\pi} \frac{2a+1}{2g+1} \frac{\Gamma \Gamma_\gamma}{(\nu - \nu_0)^2 + \Gamma^2/4}, \tag{2}$$

where Γ is the sum of the total widths of the upper and lower level, Γ_γ is the partial width for radiative transitions to the lower level, λ is the wavelength at resonance, a and g are the angular momenta of the upper and lower level, respectively, and ν_0 is the resonance frequency. Applied to the laser excitation of fast beams at some angle ϑ of intersection (see Fig. 1), one has to integrate P over the dwell time of the atoms in the interaction region in order to obtain the excitation probability per beam atom. In addition one has to account for effective Doppler broadening of $\sigma(\nu)$ due to the velocity straggling (proportional to $\cos\vartheta$), and due to the finite beam divergence

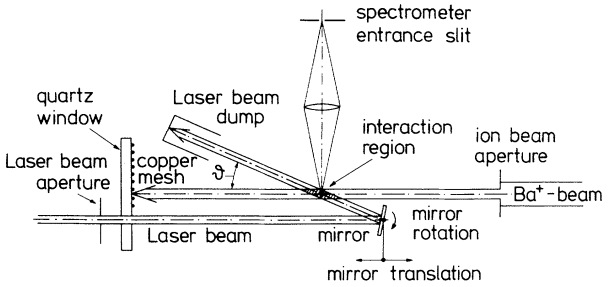

FIG. 1. Schematic experimental setup.

(proportional to $\sin\vartheta$), as well as for a frequency change $\nu(t)$ which each atom experiences when passing through laser radiation with some finite divergence (proportional to $\sin\vartheta$). These three effects cause a considerable reduction of excitation probability by factors of the order of 10^2–10^4, depending on each particular experiment, compared to absolutely nondivergent and monoenergetic beams. Although these effects can be minimized because of their different angular dependences by choosing an optimum angle ϑ, improvements will only be significant in exceptional cases where either the velocity straggling or the divergence become negligible. Furthermore, other experimental requirements like short excitation cutoff (proportional to $\cos\vartheta$) for high time resolution or the necessity to make a Doppler shift of a given laser line to an atomic resonance line allow only predetermined angles ϑ. Thus, in general, the feasibility of an experiment will mainly depend on the preparation of the best atomic beam quality possible, that is, a beam with the least velocity straggling, the least divergence, and the largest portion of atoms in the proper state.

If a suitable laser for an ionic resonance line is available, such conditions are most easily met by using a singly charged ion beam originating directly from an accelerator whose ion optics then solely determine the beam quality. Therefore, we performed the first experiment of this kind with a $^{138}Ba^+$ ion beam of 100 nA on 3 mm^2 with negligible relative velocity straggling ($<10^{-4}$) at a velocity of 0.631×10^8 cm sec^{-1}, corresponding to 285 keV with a divergence of 5 mrad. Ba^+ was chosen because its D_2-resonance line $6p\,^2P_{3/2} \rightarrow 6s\,^2S_{1/2}$ at 4554 Å closely coincides with a line at 4545 Å of an argon ion laser (Spectra-Physics Model 165) having 50-mW-cw output in multimode operation with a Gaussian envelope width of about 5 GHz at 0.5 mrad divergence. At the above velocity the wavelength gap between these two lines can be easily closed by performing a Doppler tuning of the angle ϑ to a fixed position of about 23° for which the experimental setup in Fig. 1 has been chosen. The Ba^+ beam transverses a typical BFS target chamber[3] with a transparent Faraday cup consisting of a quartz window with a copper mesh as the current collector. The laser radiation enters the chamber through this quartz window 10 mm underneath and exactly parallel to the Ba^+ beam, and hits a rotatable mirror which replaces the foil mount and can be translated stepwise along the beam axis like a foil with a spatial resolution of 0.4 mm. (For future precision experiments, fixed excitation is recommended in order to avoid uncertainties arising from not exactly parallel laser and atomic beams.) Thus with fixed mirror angle, the excitation region can be moved in front of a laterally viewing[10] spectrometer allowing a normal intensity decay measurement as in BFS.[5] After interacting with the Ba^+ beam the laser radiation is absorbed in a black dump in order to reduce background radiation at 4545 Å. Since in addition a Doppler separation of 9 Å exists between the exciting light at 23° and the re-emitted light at 90°, the 4545-Å laser radiation hitting the detector is completely eliminated by using 6-Å wavelength resolution at 4554 Å. This could be achieved by a spectrometer with 0.5-mm slit widths which also determined the time resolution by imaging the beam 1:1 on the entrance slit. As a result only detector noise is registered at full laser power without the Ba^+ beam, thus guaranteeing an absolutely flat background in the experiment, which is vital for the ultimate accuracy.

With the spectrometer viewing the interaction region and the mirror rotating by steps of 0.025°, the resonance was easily found with a count rate of 13 000 sec^{-1} for which an estimate from all experimental conditions had predicted 11 000 sec^{-1}. Typical BFS-type lifetime measurements[3,5] were then run with this resonance angle fixed by repeatedly scanning the mirror upstream and downstream, while keeping the optical axis of the detector in fixed position. Each step of 0.4 mm corresponds to one channel of a multiscaler into which counts were accumulated until a preset charge was collected in the Faraday cup at constant laser power.

The results are pure single exponentials with flat noise background convoluted with excitation and detection functions. Since the excitation function is unknown because of the fluctuating density distribution of the beam, the data analysis is al-

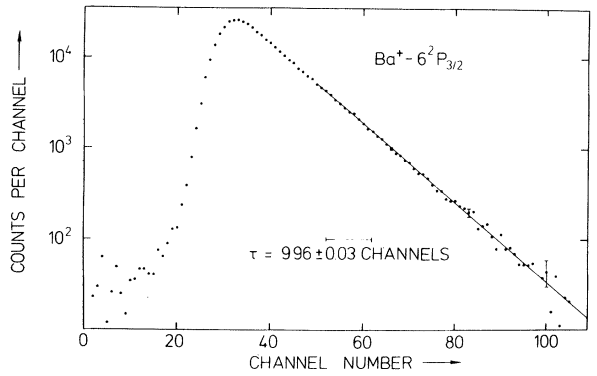

FIG. 2. Semilog plot of the result at 0.631×10^8 cm/sec and 0.4 mm per channel fitted by a single exponential (straight line) plus background, which is subtracted.

lowed only to start from channels where the excitation has stopped completely. Regarding this limitation the mean life τ was obtained by fitting $I(j) = Ae^{-j/\tau} + B$ to the data according to the weighted least-squares method.[11] Figure 2 shows a representative result after the fitted background of 186 counts per channel was subtracted and gives τ in number of channels (j) with an accuracy of 3 parts in 10^3. This result clearly demonstrates that our method can reach accuracies never obtained before as soon as the time-scale calibration can be achieved.

Dealing with ionic beams of all masses at energies of several hundreds of keV, it can be expected that an electrostatic velocity analyzer[12] will allow monitoring of the velocity profile during the whole experiment for obtaining an average value of the required accuracy. However, as to whether this averaged velocity coincides with the actual velocity of those ions excited by the laser (excitation depends on velocity!) represents an intrinsic problem of this procedure which has to be accounted for in each particular experiment.

Unfortunately, no such velocity analyzer was available for our first experiment. Therefore, we had to rely on a calibration of the accelerator voltage by nuclear reactions[13] to 1 part in 10^2, which finally yields a preliminary result out of several runs under different conditions of $\tau = 6.21 \pm 0.06$ nsec. This accuracy is already better than in earlier measurements[14-17] with a best value of $\tau = 6.27 \pm 0.25$ nsec,[15] and is expected to be further improved with a more advanced experiment.

Discussing now the applicability of this new method one notes that by the beam-gas interaction differently charged ground states (sometimes even metastable states) of all elements can be prepared under the described clean conditions with beam qualities as required for laser excitation. Thus one relies mainly on the available tunable cw lasers which nowadays cover only limited wavelength regions. However, all our considerations hold also for pulsed excitation at high repetition rates (≥ 500 sec^{-1}) with peak powers ≥ 1 kW. Since such laser operation conditions allow a much wider tunability and in addition frequency doubling, a wavelength region from 2500 to 7000 Å can be covered. Moreover, at ≥ 1 kW peak power the beam is saturated. Therefore, even stepwise excitation to higher nonresonant levels will become possible if a second pulsed laser is properly synchronized. Thus the laser excitation of hundreds of levels will be feasible and will therefore allow a rather universal application of our method for precision lifetime measurements. Aside from that it should be stressed that after polarized laser excitation even high-resolution studies can be performed on these levels under favorable alignment conditions with the level-crossing,[18] rf-resonance,[19] or quantum-beat techniques[20] in fast beams.

We wish to thank Professor E. Matthias for his interest in and for the continuous support of this work.

Note added.—After completion of this work Dr. L. Kay brought an unpublished memorandum to our attention in which he theoretically describes in some detail the problems of laser excitation of H beams.

[1]L. Dunoyer, Radium 10, 400 (1913).

[2]H. D. Koenig and A. Ellett, Phys. Rev. 39, 576 (1932).

[3]L. Kay, Phys. Lett. 5, 36 (1963); S. Bashkin, Nucl. Instrum. Methods 28, 88 (1964); *Beam-Foil Spectroscopy*, edited by S. Bashkin (Gordon and Breach, New York, 1968); Nucl. Instrum. Methods 90 (1970).

[4]L. J. Curtis, R. M. Schectman, J. L. Kohl, D. A. Chojnacki, and D. R. Shoffstall, Nucl. Instrum. Methods 90, 207 (1970); W. L. Wiese, Nucl. Instrum. Methods 90, 25 (1970).

[5]W. S. Bickel, Appl. Opt. 6, 1309 (1967), and 7, 2367 (1968).

[6]W. R. Bennett, Jr., P. J. Kindlmann, and G. N. Mercer, Appl. Opt. 1965, Suppl. 2: Chemical Lasers, p. 34, and references quoted there; E. W. Foster, Rep. Progr. Phys. 27, 420 (1964).

[7]W. L. Wiese, M. W. Smith, and B. M. Glennon, *Atomic Transition Probabilities*, U.S. National Bureau of Standards, National Standards Reference Data Series—4 (U.S. GPO, Washington, D.C., 1966), Vol. I; W. L. Wiese, M. W. Smith, and B. M. Miles, *ibid.*,

Series 22, Vol. II.
[8]A. C. G. Mitchell and M. W. Zemansky, *Resonance Radiation and Excited Atoms* (MacMillan, London, 1934).
[9]W. Heitler, *Quantum Theory of Radiation* (Clarendon Press, Oxford, England, 1949).
[10]M. Dufay, Nucl. Instrum. Methods 90, 15 (1970).
[11]D. C. Robinson, Nucl. Instrum. Methods 79, 65 (1970).
[12]R. G. Herb, S. C. Snowdon, and O. Sala, Phys. Rev. 75, 246 (1949).
[13]J. B. Marion, Rev. Mod. Phys. 33, 139 (1961).
[14]H. Bucka, J. Eichler, and G. V. Oppen, Z. Naturforsch. 21a, 654 (1966).
[15]A. Gallagher, Phys. Rev. 157, 24 (1967).
[16]M. Chenevier, R. Vialon, and J. C. Pebay-Peroula, Phys. Lett. 26A, 291 (1968).
[17]A. R. Schaefer, J. Quant. Spectrosc. Radiat. Transfer 11, 499 (1971).
[18]H. J. Andrä, Nucl. Instrum. Methods 90, 343 (1970); M. Carré, J. Desesquelles, M. Dufay, and M. L. Gaillard, Phys. Rev. Lett. 27, 1407 (1971).
[19]C. H. Liu, S. Bashkin, W. S. Bickel, and T. Hadeishi, Phys. Rev. Lett. 26, 222 (1971).
[20]K. Tillmann, H. J. Andrä, and W. Wittmann, Phys. Rev. Lett. 30, 155 (1973), and references quoted there.

Lifetime of the 2^3S_1 State of Heliumlike Argon ($Z = 18$) and Heliumlike Titanium ($Z = 22$)†

Harvey Gould, Richard Marrus,* and Robert W. Schmieder‡
Lawrence Berkeley Laboratory, University of California, Berkeley, California 94720
(Received 28 June 1973)

The $2^3S_1 \rightarrow 1^1S_0$ magnetic-dipole transition in the heliumlike atoms Ar^{+16} and Ti^{+20} has been studied by a time-of-flight technique. The lifetime of the 2^3S_1 state is found to be $172(12)\times10^{-9}$ sec in Ar^{+16} and $25.8(1.3)\times10^{-9}$ sec in Ti^{+20}, where the stated errors included statistical and systematic effects. These values can be compared with theoretical values of 212.4×10^{-9} sec for Ar^{+16} and 27.4×10^{-9} sec for Ti^{+20}.

The 2^3S_1 state of heliumlike ions decays to the 1^1S_0 ground state primarily by relativistically induced magnetic-dipole radiation. The existence of this single-photon process was first noted by Breit and Teller[1] in connection with the metastable state of hydrogen, and the theory for radiative decay of heliumlike ions in the 2^3S_1 state has now been considered by many authors.[2] The most detailed calculations of the lifetime of this state have been made by Drake[3] who finds that $\tau(2^3S_1) = 212.4\times10^{-9}$ sec for heliumlike argon and $\tau(2^3S_1) = 27.4\times10^{-9}$ sec for heliumlike titanium.

The single-photon decay of the 2^3S_1 state was first noted[4] in the x-ray spectra of heliumlike ions excited in the solar corona and has subsequently been observed in the laboratory in the spectra of Ar^{+16} [5] and He I.[6] A beam-foil measurement of this lifetime in Ar^{+16} using the 412-MeV argon beam from the old Berkeley heavy-ion linear accelerator (HILAC) yielded as a result $\tau(2^3S_1) = (172\pm30)\times10^{-9}$ sec, in rough agreement with the theory. In this Letter we report the results of a new series of measurements on this decay.

The systems studied were heliumlike Ar^{+16} and Ti^{+20} obtained from the new Berkeley heavy-ion accelerator (super HILAC). A schematic of the apparatus used is illustrated in Fig. 1. It is similar in basic idea to that employed in our previous work described in Ref. 5, but much of the hardware has been rebuilt and some of the dimensions changed. The beam emerging from the su-

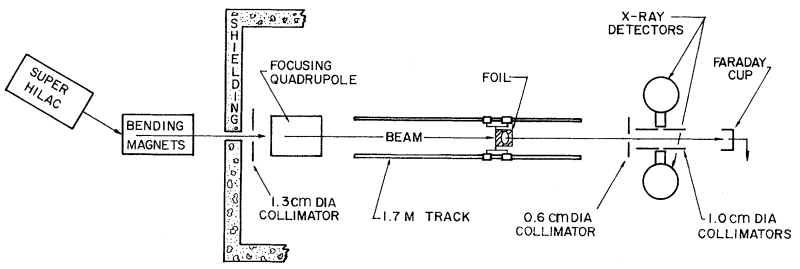

FIG. 1. Schematic of the apparatus.