

## Spectroscopy of the uranium ion using collinear fast-beam—cw-dye-laser modulation spectroscopy. Transition energies and excited-state lifetimes

O. Poulsen, T. Andersen, S. M. Bentzen, and U. Nielsen  
*Institute of Physics, University of Aarhus, DK-8000 Aarhus C, Denmark*  
(Received 26 May 1981)

Using collinear fast-beam—cw-dye-laser modulation spectroscopy, precise transition energies have been measured and excited-state lifetimes determined in the  $^{238}\text{U}^+$  ion. Wave numbers are measured using a real-time fringe-counting interferometer calibrated against the  $\text{I}_2$  absorption spectrum. The lifetimes are obtained by modulating the cw-dye-laser output by an opto-acoustic wave. A total of 25 excited-state lifetimes have been measured in  $^{238}\text{U}$  II, providing the first direct lifetime determinations in this ion.

### I. INTRODUCTION

From both a fundamental and applied-physics point of view, the study of heavy, complex atoms and ions is important. Experimentally, the optical spectra of most elements have been investigated thoroughly, and among those, the spectra of uranium represent one of the most prominent cases.<sup>1-4</sup> In recent years, some effort has also been put in the study of possible schemes for optical isotope separation. Böhm *et al.*<sup>5</sup> demonstrated isotope separation in neutral uranium, using pure optical methods, and Childs *et al.*<sup>6,7</sup> determined precise hyperfine-structure parameters for the two low odd levels 0 and  $620\text{ cm}^{-1}$  in  $^{235}\text{U}$  I, using rf-laser double-resonance spectroscopy. Carlson *et al.*<sup>8</sup> studied Rydberg series in  $^{238}\text{U}$  I by means of stepwise laser excitation and obtained a precise value of the first ionization limit.

Oscillator strengths provide additional spectroscopic information concerning an understanding of the dynamics of multistep excitation with a possible ionization as the final process. Oscillator strengths for U I have been measured recently by Miron *et al.*,<sup>9</sup> who used selective laser excitation, but no data at all have been available for the  $\text{U}^+$  ion. From an astrophysical point of view, the first ionized spectra are also of interest due to their appearance in the solar spectrum, and oscillator strengths are needed for ions such as  $\text{U}^+$  for testing the solar abundance of the heavy elements.

In the present work, the advantages of several known techniques are combined to achieve Doppler-free selective and isotopically pure excitation as well as the capability of identifying the in-

duced transitions uniquely. The output from an actively stabilized cw-ring-dye laser is superimposed on a magnetically separated ion beam from an isotope separator in a collinear geometry, ensuring Doppler-free excitation.<sup>10-12</sup> The transition energies are accurately determined, using a real-time fringe-counting interferometer calibrated against the  $\text{I}_2$  absorption spectrum. In the lifetime studies, the laser field is intensity modulated by an opto-acoustic wave. The scattered photons are energy analyzed and detected by means of standard delayed-fluorescence techniques. Several factors are of importance for these lifetime studies: (i) Vapor densities corresponding to  $10^6\text{ atom/cm}^3$ , which eliminate corrections due to pressure broadening and radiation trapping, (ii) a high duty cycle making data taking fast, and (iii) access to a wide variety of ions and atoms, the latter after charge exchange in metal vapors. The present technique has been used to study transition energies and excited-state lifetimes in  $^{238}\text{U}$  II after the applicability of the technique has been tested on  $^{90}\text{Zr}$  I and  $^{238}\text{U}$  I.

### II. EXPERIMENTAL

Isotopically pure beams of  $\text{Zr}^+$  and  $\text{U}^+$  were prepared using a 300-kV electrostatic accelerator equipped with a universal ion source. These fast beams were either used directly or charge exchanged in sodium vapor, thus producing a neutral beam containing metastable atoms. The charge-exchange process is very efficient in populating metastable levels ranging several eV in excitation en-

ergy. Also the discharge in the ion source populates many metastable levels in the ion. The existence of these metastables greatly enhances the applicability of this source to studies involving laser excitation.

A cw ring laser, actively stabilized on a reference cavity and with a resulting bandwidth below 1 MHz, was used to induce transitions between levels in the fast atomic absorbers. The collinear geometry was chosen in order to achieve Doppler-free excitation, the reason being the longitudinal cooling of the atoms in the beam.<sup>13</sup> The main source of spectral broadening stems from ripple on the high-voltage supply, limiting our resolution to 60 MHz at 75 kV of acceleration. When working in the complex spectra of the actinide elements, this resolution is useful for ensuring a clean selective excitation.

The transition energies are measured with a  $\lambda$  meter, which in real time displays the wavelength of the cw dye laser with an absolute precision of  $10^{-7}$ . With a  $100\times$  frequency extension, the intrinsic stability of the  $\lambda$  meter is below  $10^{-8}$ , the absolute accuracy being determined solely by the reference-laser wavelength, which is obtained from a calibration against the  $I_2$  absorption spectrum. As a reference laser we used a He-Ne, randomly polarized laser, stabilized in a polarization-temperature-sensitive servo loop. The relative sta-

bility of this laser is better than  $10^{-8}$ . The  $\lambda$  meter makes it easy to find and identify weak absorptions and also provides a convenient long-term control of the wavelength of the dye laser.

In order to measure lifetimes, the laser light was modulated by means of an opto-acoustic crystal. Pulse lengths down to 20 ns could be obtained by proper focusing of the laser light through the crystal and with repetition rates in the MHz range. The light pulses from the laser were detected with a photodiode and processed in standard fast-timing amplifiers and discriminators, as was also the fluorescent light after energy analysis in a grating spectrometer equipped with a cooled photomultiplier. A time-to-amplitude converter, started by the photon and stopped by the laser pulse, completed the delayed-fluorescence experiment. The selected start-stop procedure provides an effective use of the limited number of emitted photons. To ensure that the ion-neutral beam and the laser beam overlapped each other, the laser beam was adjusted to match the ion-beam profile using a telescope, and the astigmatism of the ion beam was reduced by means of an electrostatic quadrupole. With this equipment, which is shown in Fig. 1, lifetimes ranging from 10 to 5000 ns can be measured, the lower limit being set by the transit time of the acoustic wave through the laser beam and the higher limit by the flight time in the apparatus.

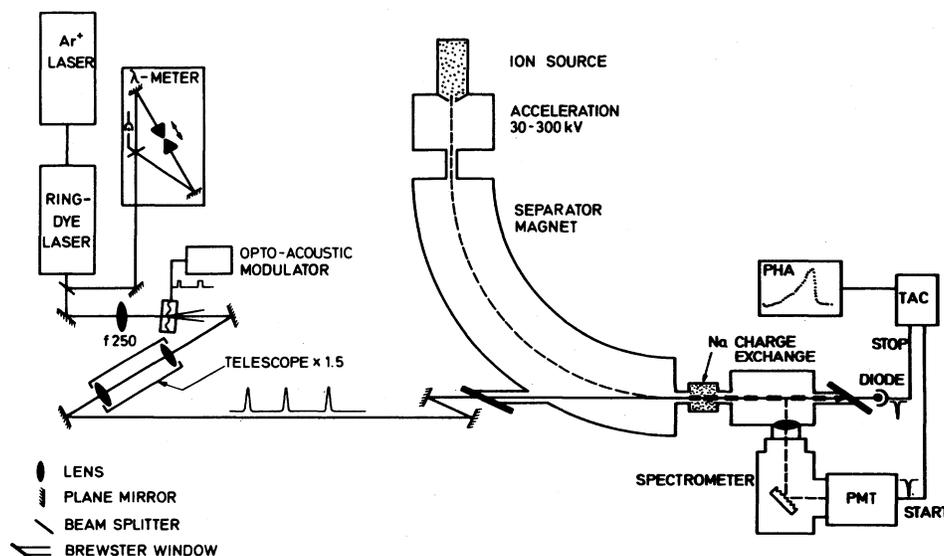


FIG. 1. The experimental apparatus, consisting of a 300-kV isotope separator, cw-ring-dye laser, and  $\lambda$  meter. Typical ion currents obtained are a few  $\mu\text{A}$ . Thus for  $^{238}\text{U}^+$ , we typically have a few nA of metastable ions. Also shown are the opto-acoustic modulator and the fast-timing electronics used recording the exponential decays.

To study possible systematic errors, decay curves have been obtained at beam energies ranging from 40 to 150 kV. The collimation of the ion beam and the associated mode matching of the laser have been carried out in several geometries to study effects associated with improper overlapping of the ion and laser beam. Beam parameters finally used were given by a waist  $\omega_0=2$  mm and a far-field diffraction angle  $\theta \simeq 10^{-4}$  rad for the laser field. The ion beam was collimated to match this laser-field distribution. Typical laser powers used were 10 mW/mm<sup>2</sup>.

The time resolution, which we have obtained by modulating the laser field, could also be performed by the "Doppler switching technique", as recently demonstrated by Ceyzeriat *et al.*<sup>14</sup>

### III. Results

When dealing with the very dense spectra of the actinide elements, it is important to be able to identify the particular transitions uniquely. Hence, before the lifetime results are presented, the transition energies found in the present work will be discussed and compared to previous work.

#### A. Transition energies

The frequency seen by the fast-moving atomic absorbers is given by

$$\nu_0 = \nu_L \frac{1 - \frac{v}{c} \cos \theta}{[1 - (v/c)^2]^{1/2}}, \quad (1)$$

where  $\nu_0$  is the transition frequency in the reference system moving with velocity  $v$  (the atoms),  $\nu_L$  is the frequency of the exciting laser in the laboratory system, and  $\theta$  is the angle between the wave vec-

tor  $\vec{k}$  and  $\vec{v}$ . By expanding Eq. (1) to second order, we find

$$\begin{aligned} \nu(\theta=0) &\simeq \nu_0 \left[ 1 + \frac{v}{c} + \frac{1}{2} \left( \frac{v}{c} \right)^2 \right], \\ \nu(\theta=180) &\simeq \nu_0 \left[ 1 - \frac{v}{c} + \frac{1}{2} \left( \frac{v}{c} \right)^2 \right] \end{aligned} \quad (2)$$

which displays the normal first-order Doppler effect ( $v/c$ ) and the second-order Doppler shift [ $-\frac{1}{2}(v/c)^2$ ] (Ref. 15). Thus, with known velocity  $v$  and  $\nu_L$  measured by the  $\lambda$  meter,  $\nu_0$  can be determined. The vacuum wavelength  $\lambda_\nu$  of the desired transition is related to a reference wavelength  $\lambda_{\text{ref}}$  as<sup>16</sup>

$$\lambda_\nu = \lambda_{\text{ref}} (C/N) (n_\lambda / n_{\text{ref}}), \quad (3)$$

where  $C$  is the counter reading,  $N$  is the counter preset, and  $n$  is the refractive index in air.

The total system has been calibrated against the I<sub>2</sub> atlas,<sup>17</sup> lines 5410–5415, and  $\lambda_{\text{ref}}$  is found to (6329.9201 ± 0.0010) Å. The uncertainty in  $\lambda_{\text{ref}}$  refers to the quoted absolute error in the I<sub>2</sub> atlas.

The second-order contributions in Eq. (2) can be evaluated using the known transition frequency  $\nu_0$  (Ref. 1) and the velocity found as  $v = (2 eV_0/m)^{1/2}$ , where  $V_0$  is the nominal acceleration voltage and  $m$  the mass of the ion. With 75 kV of the acceleration voltage, the second-order term is found to be of the order of 350 MHz for  $m = 238$ . Now, by measuring both  $\nu(\theta=0^\circ)$  and  $\nu(\theta=180^\circ)$ , an experimental value of  $\nu_0$  is determined as well as a corrected value of  $v$ . Corrections of the order of  $-3 \times 10^{-3}$  are found for  $\delta V/V_0$ , which is of no consequence for the second-order term evaluated from  $V_0$ .

Tables I and II show the experimentally determined transition energies of <sup>238</sup>U I and <sup>238</sup>U II, respectively, using Eqs. (1) and (3). Also shown are wave numbers determined at Los Alamos and

TABLE I. Measured transition energies in <sup>238</sup>U I (cm<sup>-1</sup>).

Transition	Present work <sup>a</sup>	Reference 1 <sup>b</sup>	Other references
0–16 900	16 900.387	16 900.378 (B)	16 900.387 (Ref. 3) 16 900.3875 (Ref. 4)
6249–23 572	17 323.056	17 323.048 (A)	17 323.057 (Ref. 3) 17 323.0544 (Ref. 4)

<sup>a</sup>Present work: ±0.002 cm<sup>-1</sup>.

<sup>b</sup>A – ±0.003, B – ±0.005 cm<sup>-1</sup>.

TABLE II. Transition energies in U II.

Transition	Present work <sup>a</sup>	Reference 1 <sup>b</sup>	Reference 2	$\Delta$ (MHz)
0—16 706	16 706.307	16 706.304 (A)		+ 90
0—17 392	17 392.211	17 392.212 (A)	17 392.212	— 30
289—17 434	17 145.323	17 145.323 (A)		0
914—18 200	17 285.330	17 285.329 (A)		+ 30
914—17 434	16 519.603	16 519.600 (A)		+ 90
1749—18 827	17 077.888	17 077.887 (A)		+ 30
2294—18 827	16 532.314	16 532.312 (A)		+ 60
4420—21 691	17 270.649	17 270.647 (A)		+ 60
4585—21 710	17 125.334	17 125.334 (A)		0
4585—21 975	17 390.154	17 390.154 (A)		0
4706—21 860	17 153.780	17 153.779 (A)		+ 30
5401—22 429	17 028.363	17 028.299 (E)	17 028.361	+ 1830
5401—22 642	17 240.975	17 240.974 (A)	17 240.975	+ 30
5401—22 868	17 466.531	17 466.529 (A)	17 466.530	+ 60
6445—22 917	16 472.418	16 472.416 (B)		+ 60
6445—23 241	16 796.332	16 796.331 (A)	16 796.330	+ 30
6445—23 241	16 796.000	16 796.000 (A)		0
6445—23 315	16 870.058	16 870.054 (B)		+ 120
6445—23 553	17 108.945	17 108.942 (A)	17 108.942	+ 90
6445—23 911	17 466.602	17 466.602 (A)		0
7166—24 288	17 121.373	17 121.372 (A)		+ 30
8276—25 492	17 216.186	17 216.184 (B)		+ 60
8394—25 163	16 769.540	16 769.538 (B)		+ 60
8510—25 714	17 203.184	17 203.183 (A)		+ 30
8755—26 191	17 435.672	17 435.671 (C)		+ 30
9075—26 713	16 637.902	16 637.902 (A)		0
9626—26 716	17 090.586	17 090.584 (B)		+ 60
10 740—28 154	17 413.490	17 413.488 (C)		+ 60
11 544—28 507	16 963.224	16 963.219 (B)		+ 150

<sup>a</sup>Present work:  $\pm 0.002 \text{ cm}^{-1}$ .

<sup>b</sup>Reference 1:  $A = \pm 0.003$ ,  $B = \pm 0.005$ ,  $C = \pm 0.10$ ,  $E = \pm 0.050 \text{ cm}^{-1}$ .

Aime-Cotton by means of classical spectroscopy and Fourier-transform spectroscopy.<sup>1-4</sup> Figure 2 gives a partial energy-level diagram for U II, showing the type of transitions induced in this work. They all have initial levels belonging to the odd configurations  $f^3s^2$ ,  $f^3ds$ , and  $f^3d^2$ .

It is seen that it is possible, with the combination of a  $\lambda$  meter and an electromagnetic isotope separator, to achieve an absolute precision within  $\pm 2 \text{ mK}$  ( $\pm 60 \text{ MHz}$ ), limited only by the precision of our reference laser linked to the  $\text{I}_2$  atlas<sup>17</sup>. Despite this, the agreement with the Los-Alamos line list<sup>1</sup> is well within our error limits. As a general trend, it is seen that our measurements are higher by  $1.5 \text{ mK}$  ( $45 \text{ MHz}$ ) compared to the LA-4501 line list,<sup>1</sup> which is also known to be  $0.6 \text{ mK}$  ( $20 \text{ MHz}$ ) below the wave numbers found in LA-8251-MS.<sup>2</sup>

The main feature displayed in Tables I and II is that laser spectroscopy performed on high-velocity atoms and/or ions can be carried out with an absolute accuracy comparable to or better than the best classical Doppler-limited techniques. With an improved high-voltage regulation and a  $^{22}\text{Ne}/^3\text{He}$   $^{129}\text{I}_2$  stabilized laser, which is being completed and installed in our  $\lambda$  meter, it will be possible to achieve absolute precisions around  $\pm 6 \text{ MHz}$  ( $\approx 10^{-8}$ ).

## B. Lifetimes

In order to test the applicability of collinear fast-beam—cw-dye-laser modulation spectroscopy for lifetime studies, a few test cases have been selected. The choice of Zr I is mainly dictated by

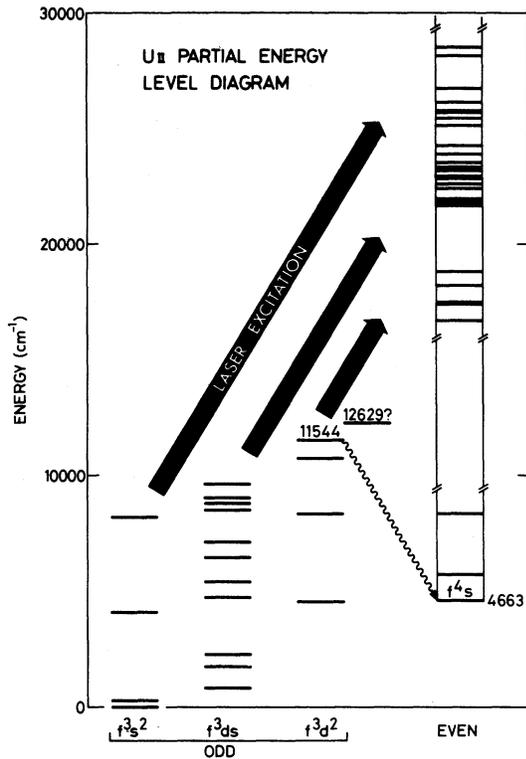


FIG. 2. Partial U II term diagram containing the odd low levels in U II used as initial states in this study. Also shown are the upper excited even levels between 16 706 and 28 507  $\text{cm}^{-1}$ , for which lifetimes have been determined. The 12 629  $\text{cm}^{-1}$  level is discussed in the text.

the great differing of recent measurements,<sup>18,19</sup> and  $^{238}\text{U I}$  is chosen as another test case because several experimental data are available.<sup>8,9,18-20</sup> A typical decay curve is shown in Fig. 3. The solid curve represents the convolution

$$F(t') = \int_{-\infty}^{\infty} g(t)E(t'-t)dt, \quad (4)$$

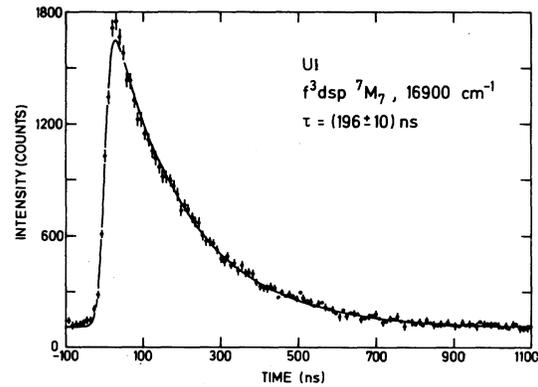


FIG. 3. Decay of the 16 900  $\text{cm}^{-1}$  level in  $^{238}\text{U I}$ , obtained after charge exchange of 50-kV  $\text{U}^+$  in Na vapor. An excited-state lifetime of  $(196 \pm 10)$  ns is found.

where

$$E(t) = \begin{cases} 0 & \text{for } t < m, \\ \exp(t/\tau) & \text{for } t \geq m, \end{cases} \quad (5)$$

is the exponential decay with lifetime  $\tau$  and  $g(t)$  is the prompt yield of the experiment.  $g(t)$  can be obtained experimentally by tuning the laser off any resonance and detecting the scattered light at the laser wavelength. The function  $g(t)$  is found to be well represented by a Gaussian with full-width at half maximum (FWHM) of  $\sigma$  and positioned at  $m$ . Thus  $F(t)$  is given by

$$F(x) = \text{const} \left[ 1 + \text{erf} \left( \frac{x-m}{\sigma\sqrt{2}} - \frac{\sigma}{\tau\sqrt{2}} \right) \right] \times \exp \left[ \frac{\sigma^2}{2\tau^2} - \frac{x-m}{\tau} \right], \quad (6)$$

erf being the error function.

The results obtained in  $^{90}\text{Zr I}$  and  $^{238}\text{U I}$  are shown in Table III. Good agreement is found

TABLE III. Lifetimes in neutral Zr and U.

Element	Level	Energy ( $\text{cm}^{-1}$ )	$\tau$ (ns)	$\tau$ (ns)	Other references
Zr I	$z^3G_5^0$	22 564	$118 \pm 5$	$107 \pm 2$	(Ref. 18)
				$450 \pm 25$	(Ref. 19)
U I	$f^3dsp^7M_7$	16 900	$196 \pm 10$	$190 \pm 10$	(Ref. 18)
				$205 \pm 20$	(Ref. 9)
				$232 \pm 25$	(Ref. 19)
				$255 \pm 25$	(Ref. 8)
				$205 \pm 20$	(Ref. 20)
U I	$J=6$	23 572	$85 \pm 4$	$80 \pm 8$	(Ref. 9)

TABLE IV. Measured radiative lifetimes in  $^{238}\text{U II}$ , even levels.

Upper level ( $\text{cm}^{-1}$ ) <sup>a</sup>	Classification <sup>a</sup>		Lifetimes (ns) <sup>c</sup>	
16 706.300	$f^2ds^2$	$^4K_{11/2}$	15	(B)
17 392.211	$f^2d^2s$	$^6K_{9/2}$	1370	(B)
17 434.364		$J = 11/2$	3800	(B)
18 200.004 <sup>b</sup>	$f^2ds^2$	$^4I_{9/2}$	520	(A)
18 827.009	$f^2d^2s$	$^6K_{11/2}$	1600	(B)
21 710.767 <sup>b</sup>		$J = 13/2$	330	(A)
21 975.588	$f^2d^3$	$^6K_{13/2}$	1170	(B)
22 429.069 <sup>b</sup>		$J = 9/2$	162	(A)
22 642.478 <sup>b</sup>		$J = 9/2$	78	(A)
22 868.033		$J = 9/2$	288	(A)
22 917.451 <sup>b</sup>		$J = 11/2$	70	(A)
23 241.034		$J = 9/2$	1000	(B)
23 241.363 <sup>b</sup>		$J = 11/2$	160	(A)
23 315.090 <sup>b</sup>	$f^3sp$	$^6K_{9/2}$	85	(A)
23 553.976 <sup>b</sup>		$^6I_{11/2}$	107	(A)
23 911.637 <sup>b</sup>		$J = 9/2$	260	(A)
24 288.003 <sup>b</sup>	$f^3sp$	$J = 11/2$	84	(A)
25 163.902		$J = 13/2$	124	(A)
25 492.917		$J = 11/2$	171	(B)
25 713.632		$J = 9/2$	178	(A)
25 714.049 <sup>b</sup>	$f^3sp$	$J = 13/2$	34	(A)
26 191.309	$f^3dp$	$^6M_{13/2}$	$2 < \tau < 10$	
26 716.697		$J = 13/2$	78	(A)
28 154.450 <sup>b</sup>	$f^3dp$	$^6L_{11/2}$	$2 < \tau < 10$	
28 507.890		$J = 11/2$	69	(A)

<sup>a</sup>Reference 1.<sup>b</sup>Possibly observed in the solar photospheric spectrum.<sup>c</sup>(A) $\pm$ 5%, (B) $\pm$ 10%.

between the lifetimes of this work and those resulting from other selective schemes,<sup>9,18</sup> whereas a major disagreement is found with the results based on fast-ion sputtering.<sup>19</sup>

The lifetimes measured in  $^{238}\text{U II}$  are given in Table IV. A typical decay curve is shown in Fig. 4. A total of 25 excited-state lifetimes have been determined. For lifetimes of the same order as the width of the exciting laser pulse, the lower limit has been determined directly from the spectral width of the resonances. In no case was found a linewidth larger than 60–70 MHz. These results represent the first direct lifetime measurements in the uranium ion, and the present results show a very large range of lifetimes.

However, not only lifetimes of the excited states can be determined by means of the present technique. A limit can be set to the lifetime of the initial odd-parity levels. From Table II it is seen that

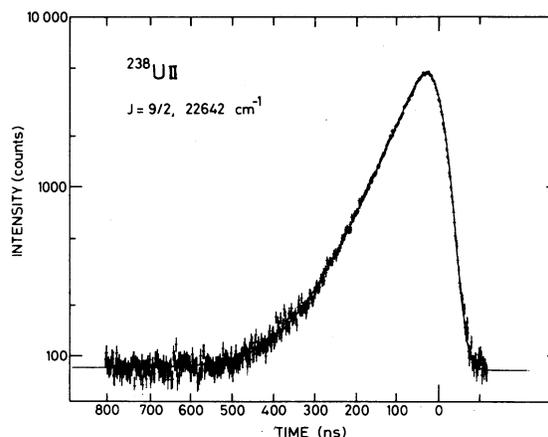


FIG. 4. Decay of the  $22\,642\text{ cm}^{-1}$  level in  $^{238}\text{U II}$ , obtained after laser excitation at  $5800.1363\text{ \AA}$  [ $V_0 = (74.769 \pm 0.001)\text{ kV}$ ], and detection was at  $4472\text{ \AA}$  ( $22\,642\text{--}22\,289$ ). An excited-state lifetime of 78 ns is found.

a transition, starting from the odd level of  $11\,544\text{ cm}^{-1}$ , is induced. This level decays to the low-lying even level of  $4663\text{ cm}^{-1}$ , also shown in Fig. 2. The transit time through the apparatus is  $\simeq 25\text{ }\mu\text{s}$ . Thus the  $11\,544\text{ cm}^{-1}$  and other low-lying levels have lifetimes longer than  $25\text{ }\mu\text{s}$ . The transition classified as  $12\,629\text{--}29\,476\text{ cm}^{-1}$ , which in LA-4501 (Ref. 1) is listed as a strong transition, is *not* seen in the present work. Thus the  $12\,629\text{ cm}^{-1}$  level may have a lifetime considerably shorter than  $25\text{ }\mu\text{s}$ , otherwise it is erroneously classified, as suggested by the fact that that LA-8251-MS (Ref. 2) does not observe this transition either.

### C. Absolute oscillator strengths

The experimentally determined lifetimes can be converted into absolute oscillator strengths (gf values) assuming the branching ratios are known. Reliable branching ratios are not yet available, but a correction can be found to the results of Corliss and Bozman,<sup>21</sup> using their relative gf values determined in classical total-emission studies of a gas in local thermodynamical equilibrium (LTE). In the case where Corliss and Bozman do not list all branches from a given upper level, the remaining transitions, in most cases contributing less than 10% to the total transition strength, are found<sup>1</sup> in LA-4501 with an adjustment to take into account the different temperatures in the two light sources. In Fig. 5 are shown the obtained corrections to Corliss and Bozman's lifetime values, ranging from a factor of 8 to 2 within  $12\,000\text{ cm}^{-1}$  of the excitation energy. The correction can be expressed as

$$\log_{10}(\tau^{\text{CB}}/\tau) = C_1 + C_2 E,$$

where  $\tau^{\text{CB}}$  is the lifetimes of Corliss and Bozman and  $E$  is the upper-level excitation energy ( $\text{cm}^{-1}$ ).

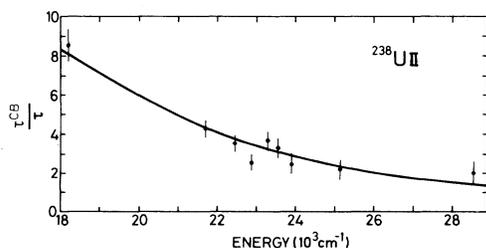


FIG. 5. Corrections to the lifetimes of Corliss and Bozman (Ref. 21), showing a pronounced dependence on the upper-level energy. Only levels observed strongly by Corliss and Bozman are used in this presentation.

A least-squares fit to the experimental data yields  $C_1 = +2.32$  and  $C_2 = -7.78 \times 10^{-5}$ .

## IV. DISCUSSION

The transition energies measured in the work (Table II) clearly demonstrate that high absolute precision can be obtained. Within our uncertainty, good agreement is found with the classical techniques.<sup>1-4</sup> In spectra investigated in less detail than U II, the present technique could be used to identify part of a spectrum by systematically tuning the laser frequency, or Doppler shifting the transitions into resonance, followed by a determination of all decay channels for each excitation. As an example, detailed spectroscopic data such as isotope shifts (IS) and hyperfine data for selected U II lines could be used to synthesize the solar spectrum to deduce the  $^{235}\text{U}$ -to- $^{238}\text{U}$  abundance ratio. Such data could be used to clarify aspects of the nuclear synthesis taking place in the sun.

Oscillator strengths have been measured in many neutral atoms, recently in uranium by Miron *et al.*<sup>9</sup> by means of selective laser excitation, but only a few and scattered data have been available for the ions, and for the  $\text{U}^+$  ions, no data at all. From an astrophysical point of view, the first ionized spectra are of particular interest due to their appearance in the solar spectrum. This work, together with the recent work of Hannaford and Lowe,<sup>18</sup> are examples of lifetime studies where selective methods have been used which can be applied to test the solar abundance of the heavy elements.<sup>22</sup>

Generally, oscillator strengths can be measured directly by observing the emission spectrum of a gas in LTE. This has been done for a wide variety of elements by Corliss and Bozman<sup>21</sup> at NBS. However, problems associated with the determination of the absolute temperature of a gas and the ensuring of LTE make these studies difficult.<sup>23</sup> Often large corrections have to be applied. Such emission spectra, which may yield fairly precise branching ratios, can be combined with direct lifetime measurements to yield precise absolute oscillator strengths.

Many other nonselective methods have been used to obtain oscillator strengths of heavier elements with rather complex spectra. The beam-foil technique is not well suited because severe cascade problems, scattering and energy loss in the foils, and large Doppler broadening can render unique spectral identification difficult. The beam-foil technique, on the other hand, has its strength when

we are dealing with highly charged ions, where other methods do not work at all.<sup>24</sup> More useful is pulsed-electron excitation,<sup>20</sup> where a certain selectivity can be obtained by a proper choice of electron energy. Still problems such as radiation trapping and pressure broadening are factors to be taken into account. Recently, a fast-beam sputtering technique was reported,<sup>19</sup> where the heavy-ion sputtering process is claimed to be selective. However, the technique is Doppler limited, affected by cascading as well as by complicated light-source plasma dynamics. The fast-beam Hanle technique, which depends on the anisotropy present in the heavy-ion—atom collision, has been used successfully in a few rare-earth ions.<sup>25</sup> It gives some problems, however, such as cascading of alignment and pressure broadening.

From the results of the present work and other selective-excitation schemes, compared to nonselective methods, it may be concluded that, taking the complex nature of many heavy atoms and/or ions into consideration, the selective schemes are preferable.

The transitions induced and the lifetimes measured in the present work represent only a small fraction of the possible excitations in the U II spectrum, using collinear fast-beam—cw-dye-laser spectroscopy. Attempts have been made to cover as wide a range of upper levels as possible with our

present laser. However, with optics to tune either blue or red, virtually all levels up to  $34\,000\text{ cm}^{-1}$  can be studied Doppler-free, including a determination of the upper-level lifetime. Stepwise excitation by means of two tunable cw-dye lasers would allow studies of Rydberg sequences, which could be extrapolated to the ionization limit.

By tuning the isotope separator to  $^{235}\text{U}^+$ , isotope shifts have been determined together with Doppler-free hyperfine spectra for transitions starting from the 12 lowest metastable odd levels in  $^{235}\text{U II}$ . Together with proper eigenfunctions, it is hoped that these data<sup>26</sup> can yield information about the relativistic one-electron integrals.

## V. CONCLUSION

Results are presented which make it possible to bring the oscillator strengths of heavy complex atoms and ions, in this case the U II ion, on an absolute scale. The method of collinear fast-beam—cw-dye-laser modulation spectroscopy is fast and convenient. The ease with which a large number of heavy atoms and ions are produced makes the method usable in many cases. Also studies of fine and hyperfine structure can conveniently be performed due to the high resolution inherent in this technique.

- 
- <sup>1</sup>D. W. Steinhaus, L. J. Radziemski, Jr., R. D. Cowan, J. Blaise, G. Guelacvili, Z. Ben Osman, and J. Vergés, Los Alamos Scientific Laboratory Report No. LA-4501 (unpublished).
- <sup>2</sup>B. A. Palmer, R. A. Keller, and R. Engleman, Jr., Los Alamos Scientific Laboratory Report No. LA-8251-MS (unpublished).
- <sup>3</sup>J. Blaise and L. J. Radziemski, Jr., *J. Opt. Soc. Am.* **66**, 644 (1976).
- <sup>4</sup>S. Gerstenkorn and P. Luc, *Astron. Astrophys.* **58**, 255 (1977).
- <sup>5</sup>H. -D. W. Böhm, W. Michaelis, and C. Weitkamp, *Opt. Commun.* **26**, 177 (1978).
- <sup>6</sup>W. J. Childs, O. Poulsen, and L. S. Goodman, *Opt. Lett.* **4**, 35 (1979).
- <sup>7</sup>W. J. Childs, O. Poulsen, and L. S. Goodman, *Opt. Lett.* **4**, 63 (1979).
- <sup>8</sup>L. R. Carlson, J. A. Paisner, E. F. Worden, S. A. Johnson, C. A. May, and R. W. Solars, *J. Opt. Soc. Am.* **66**, 846 (1976).
- <sup>9</sup>E. Miron, R. David, G. Erez, S. Lavi, and L. A. Levin, *J. Opt. Soc. Am.* **69**, 256 (1979).
- <sup>10</sup>W. H. Wing, G. A. Ruft, W. E. Lamb, and J. Spezeski, *Phys. Rev. Lett.* **36**, 1488 (1976).
- <sup>11</sup>H. Winter and M. L. Gaillard, *J. Phys. B* **10**, 2739 (1977).
- <sup>12</sup>K. R. Anton, S. L. Kaufman, W. Klempt, G. Moruzzi, R. Neugart, E. -W. Otten, and B. Schinzler, *Phys. Rev. Lett.* **40**, 642 (1978).
- <sup>13</sup>S. L. Kaufman, *Opt. Commun.* **17**, 309 (1976).
- <sup>14</sup>P. Ceyzeriat, D. J. Pegg, M. Carré, M. Druetta, and M. L. Gaillard, *J. Opt. Soc. Am.* **70**, 901 (1980).
- <sup>15</sup>J. J. Snyder and J. L. Hall, *Proceedings of the Second International Conference on Laser Spectroscopy, Megève, 1975* (Springer, Heidelberg, 1975), p. 6.
- <sup>16</sup>J. L. Hall and S. A. Lee, *Appl. Phys. Lett.* **29**, 367 (1976).
- <sup>17</sup>S. Gerstenkorn and P. Luc, *Opt. Commun.* **36**, 322 (1981).
- <sup>18</sup>P. Hannaford and R. M. Lowe, *J. Phys. B* **14**, L5 (1981).
- <sup>19</sup>P. S. Ramanujam, *Phys. Rev. Lett.* **39**, 1192 (1977).
- <sup>20</sup>J. Z. Klose and P. A. Voigt, *Phys. Rev. A* **16**, 2032 (1977).
- <sup>21</sup>C. H. Corliss and W. R. Bozman, *NBS Monograph No. 53* (1962).

<sup>22</sup>E. Biemont and N. Grevesse, *Phys. Scr.* **16**, 39 (1977).

<sup>23</sup>C. H. Corliss and J. L. Tech, *J. Res. Natl. Bur. Stand. Sect. A* **80**, 787 (1976).

<sup>24</sup>B. Denne and O. Poulsen, *Phys. Rev. A* **23**, 1229

(1981).

<sup>25</sup>T. Andersen, O. Poulsen, A. P. Petkov, and P. S. Ramanujam, *Sol. Phys.* **44**, 257 (1975).

<sup>26</sup>O. Poulsen (unpublished).