# Mean lives and $gf$ values in U I†

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Determinations of the mean lives of the 27887- and 16900-cm$^{-1}$ levels in U I have been made, each at a single vapor density, using electronic excitation and a method of delayed coincidence. The lifetime values were obtained by optically detecting the decays of the 3584.9- and 5915.4-Å resonance transitions, respectively. Using branching ratios obtained from known relative $f$ values, the averages of the measured lifetimes were corrected for imprisonment of the resonance radiation giving the following results: $\tau_0^{27887} = 10.9 \pm 0.8$ ns and $\tau_0^{16900} = 205 \pm 20$ ns. These mean lives were then used to place a system of relative $gf$ values on two independent absolute scales, which were found to be in very close agreement. The lifetimes and $gf$ values are presented in comparison with the experimental results of others.

## I. INTRODUCTION

In a recent paper[1] a determination of the mean life of the $f^3d^2p\ ^7N_7$ (27 886.992 cm$^{-1}$) level in U I [2] using the 3584.878-Å resonance transition was reported. This lifetime was subsequently used by several workers[3-5] to place their systems of relative transition probabilities on absolute scales. We report here a redetermination of this lifetime with improved techniques yielding a substantially different result. We also measured the mean life of the $f^3dsp\ ^7M_7$ (16 900.387 cm$^{-1}$) level[2] to provide the basis for an independent-comparison absolute scale. These lifetimes have been used to place the set of relative transition probabilities of Voigt[3,4] on absolute scales. The two scales were found to be in very close agreement and to provide upper limits on lifetimes which are in good accord with lifetimes measured by others.[6-8]

## II. EXPERIMENTAL METHOD

The present lifetime measurements were carried out by the same method as those reported previously for the 3585-Å transition.[1] In this method atoms in the vapor state are excited by a pulsed beam of low-energy electrons. The decay times of individual events are measured by a time-to-time pulse-height converter and are stored in a pulse-height analyzer. An electron energy of 100 eV and system time calibrations of $1.79 \pm 0.03$ and $16.7 \pm 0.2$ ns per analyzer channel were used in the measurements to be reported here.

The uranium atoms in the vapor state were obtained in a diffuse beam from a Knudsen effusion device, which has as its central element a tantalum or tungsten effusion cell. This source has been described in detail in an earlier report.[9] The sample of uranium used was National Bureau of Standards Standard Reference Material 960, which is of normal isotopic composition, is 99.975% pure, and has other metallic impurities present in amounts not exceeding 50 ppm.

The mean lives of the 27 887- and 16 900-cm$^{-1}$ levels were obtained here using the 3584.878- and 5915.385-Å resonance transitions. Thus, the measured values had to be corrected for possible imprisonment of the emitted resonance radiations. The corrections were made in the manner described previously for low optical densities.[1] They depend on the geometry of the source and exciting beams, the absorption coefficients at the centers of the Doppler-broadened resonance lines, the dimensions of the source, and the branching ratios of the resonance transitions. Since high temperatures are necessary to produce an appreciable vapor of uranium, sufficiently low optical densities were readily obtained. At such low densities the corrections are small, as will be seen below, and the other important possible density effect, competing collisionally induced decay, is negligible.

## III. ANALYSIS

Since the electron excitation energy of 100 eV used here is much greater than the 2.1- and 3.5-eV threshold excitation energies of the 16 900- and 27 887-cm$^{-1}$ levels, one would expect multiexponential decays due to cascading into the levels of interest from simultaneously excited higher levels. However, the decay-time distributions of the lifetime runs showed no obvious evidence of multiexponential decays. Nevertheless, the runs were analyzed using a least-squares computer program fitting two exponentials and a background to the raw data. The computer analyses gave pairs of exponentials with lifetimes of approximately 11 and 100 ns for the 3585-Å data, but could make only single-exponential fits to the 5915-Å runs. Thus, the results of the analyses support the initial findings of single-exponential decays.

The absence of detectable cascading effects can be taken as an indication that the cascading transitions are much weaker than the transitions of interest. Since no transitions terminating in the levels of interest are identified in the latest available experimental line lists,[10,11] one can infer that the cascading transitions are weak. Nevertheless, the possibility of a cumulative cascading effect due to a number of weak transitions cannot be dismissed.

The runs were obtained with a monochromator slit setting of 1 mm, representing a bandpass of 16 Å. Although U I has a very dense line spectrum, only a few strong lines could be observed under our experimental conditions. Thus, this setting was adequate to isolate the 3585-Å line from its nearest neighbor at 3567 Å, as determined from spectrographic exposures made with the lifetime apparatus under typical conditions of a lifetime run.[1] No exposures were made involving the 5915-Å line, which, from relative $gf$ values[3,4] adjusted to an oven temperature of 2500 K, was found to be nearly 30 times as intense as the 3585-Å line. To reduce the scattered background light from the uranium oven in the lifetime runs, a Corning 7-54 glass filter was used at 3585 Å, and a 5893-Å interference filter with a 100-Å bandwidth was used at 5915 Å. For the 3585-Å runs, the use of the filter provided a higher signal-to-background ratio than was obtained in the previous work at this wavelength.

Five measurements were used to obtain the final result for the mean life of the 27 887-cm$^{-1}$ level. The runs were made consecutively at the same vapor density and yielded an average value for the lifetime of 11.1 ns with an uncertainty of 0.8 ns. For the 16 900-cm$^{-1}$ level, four runs at the same vapor density were used to obtain a final lifetime result of 208 ns with an uncertainty of 20 ns. These average values must now be corrected for the imprisonment of the resonance radiations at 3585 and 5915 Å, respectively.

## IV. CORRECTION FOR IMPRISONMENT

At the low optical densities used here, the theories of resonant photon imprisonment give[12,13]

$$\tau_m = \tau_0(1 + ak_0LB_r)\,, \tag{1}$$

where $\tau_m$ is the measured lifetime, $\tau_0$ is the natural (zero vapor density) lifetime, $a$ is a constant determined by the geometry of the source and exciting beams, $k_0$ is the absorption coefficient at the center of the Doppler-broadened resonance line, $L$ is a characteristic dimension of the source, and $B_r$ is the branching ratio of the resonance transition. The previously chosen values of $L = 0.85$ cm and $a = 1.0$ for this source[1] were applied here. It should be noted that the correction to $\tau_m$ is highly insensitive to the choice of $a$. For example, $a$ may be varied as much as 45% without producing a change greater than 1% in the corrected value of $\tau_m$.

The branching ratios were determined from the relative $gf$ value measurements in U I of Voigt, both previously reported[3,4] and newly reported below. As many as 66 allowed downward transitions from the 27 887-cm$^{-1}$ level and 21 such transitions from the 16 900-cm$^{-1}$ level can be derived from the level lists of Blaise and Radziemski.[2] However, only 11 and 15, respectively, of these transitions have been identified.[10,11] Of the observed spectral lines for the 27 887-cm$^{-1}$ level, two are much stronger than the others, and only these two, at 3585 and 4620 Å, were of sufficient intensity to be included in the $gf$ value measurements. Thus, the branching ratio had to be determined on the basis of these two transitions and was found to be 0.6. Similarly, the branching ratio for the 16 900-cm$^{-1}$ level had to be determined from only the two transitions at 5915 and 7632 Å and was found to be 0.9. An uncertainty of $\lesssim 10\%$ is indicated in the branching ratios due to statistical and systematic errors in the relative $gf$ values.[3] Further systematic errors could arise from the omission of transitions above 7640 Å, the maximum wavelength of the $gf$ value measurements. From the intensities of the observed transitions, this seems unlikely for the 27 887-cm$^{-1}$ level, but may be somewhat more likely for the 16 900-cm$^{-1}$ level.

To evaluate $k_0$ the particle density of unexcited uranium atoms in the beam, $n$, must be determined.[1] Referring to Ramsey[14] and Klose,[1] we obtain the expression

$$n = \frac{2}{3\pi}\frac{1}{l_0^2}\frac{p'}{kT}r_s^2 \tag{2}$$

for an effusion cell with a very thin lid. In Eq. (2), $l_0$ is the distance from the cell to the center of the observation window, $p'$ is the pressure inside the cell, $T$ is the absolute temperature, and $r_s$ is the radius of the aperture in the lid of the cell. For the present configuration of the beam source, $l_0 = 5.35$ cm and $r_s = 0.1$ cm.

In determining the values of $n$, the uranium vapor pressures inside the cell had to be obtained from expressions giving the pressure as a function of temperature. The temperatures were measured by sighting an optical pyrometer on the aperture in the lid of the cell. The second column in Table I gives the brightness temperatures $T_B$ measured through a 7056 glass window. Column three gives the brightness temperatures corrected for reflec-

TABLE I. Experimental parameters used in correcting the measured mean lives for radiative imprisonment.

| $\lambda$ (Å) | $T_B$ (K) | $T_{BC}$ (K) | $T$ (K) | $p'$ ($10^{-5}$ atm) | $n$ ($10^9$ cm$^{-3}$) | $v_0$ ($10^3$ cm s$^{-1}$) | $\tau_m$ ($10^{-9}$ s) | $T_0$ ($10^{-9}$ s) | $A_{3585}$ ($10^7$ s$^{-1}$) | $A_{4620}$ ($10^7$ s$^{-1}$) | $A_{5915}$ ($10^7$ s$^{-1}$) | $A_{7632}$ ($10^7$ s$^{-1}$) |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| 3584.88 | 2156 | 2174 | 2376 | 1.23[a] | 2.82 | 4.33 | 11.1 | 10.9 | 5.6 | 3.6 | ... | ... |
| | | | | 1.20[b] | 2.75 | | | 10.9 | 5.6 | 3.6 | ... | ... |
| 5915.39 | 2171 | 2189 | 2394 | 1.48[a] | 3.36 | 4.35 | 208 | 205 | ... | ... | 0.446 | 0.042 |
| | | | | 1.45[b] | 3.30 | | | 205 | ... | ... | 0.446 | 0.042 |

[a]Reference 16. [b]Reference 17.

tions at the window surfaces, $T_{BC}$. The true temperatures, obtained by correcting for the nonblackbody character of the glowing cell, are given in the fourth column. These corrections were made using information given by Kaspar.[15] Vapor-pressure data taken from two sources[16,17] were utilized to obtain values for the pressures of uranium inside the cell, given in column five of Table I. Finally, the particle densities in the beam were calculated from Eq. (2) and are given in column six of the same table. The differences in the particle densities as determined from the two sources of vapor pressure are indications of the magnitudes of possible systematic errors in $n$.

Also, in evaluating $k_0$ it was necessary to determine the quantity $v_0$, the component of velocity in the observing direction of an atom in the beam with the most probable speed. The half-angle divergence of the beam, $\theta$, determined from the configuration of the source was 9.96°. Since the divergence angle was small and the angular distribution was continuous, all atoms in the beam were assigned a half-angle divergence of $\frac{1}{2}\theta = 4.98°$. Referring to Ramsey,[14] the value of $v_0$ is given by the expression

$$v_0 = \left(\frac{3}{2}\right)^{1/2}\left(\frac{2RT}{M}\right)^{1/2}\sin(\tfrac{1}{2}\theta) \qquad (3)$$

and is listed in column seven of Table I.

Finally, using the values given above for $L$, $a$, and $B_r$ and evaluating $k_0$ as previously described,[1] Eq. (1) was solved for the natural mean lives given in column nine of Table I. Columns 10–13 display the values of the probabilities obtained here for 3585-, 4620-, 5915-, and 7632-Å transitions. We see from the table that the results for the mean lives and transition probabilities are the same for each pair of values of the vapor pressures inside the cell.

## V. LIFETIME RESULTS

Table II gives the natural mean lives as determined in the present experiment, with other results for comparison. Throughout the present work the temperatures were maintained constant with a reproducibility in measurement of ±2 absolute degrees. Thus, the particle densities of the atomic beam were assumed to be constant with maximum variations ranging from 0.055 to $0.065 \times 10^9$ cm$^{-3}$ as determined from the maximum variation in the temperatures. The electron beam was also stable over the durations of the runs which ranged in length from 3 to 67 min.

The error given with each of the present results in Table II is the root sum of squares of the standard deviation and the spread in the values which

TABLE II. Natural mean lives of the 27 887-cm$^{-1}$ and 16 900-cm$^{-1}$ levels in U I.

| Authors | Method | $\tau_0^{27887}$ (ns) | $\tau_0^{16900}$ (ns) |
|---|---|---|---|
| Present work | Delayed coincidence | 10.9 ± 0.8 | 205 ± 20 |
| Carlson *et al.*[a] | Stepwise photoionization | ... | 255 ± 25 |
| Voigt and Kornblith[b] | Arc | ... | 150[c] |
| Corliss[d] | Arc | ... | 330[c] |
| Klose[e] | Delayed coincidence | 7.3 ± 1.1 | ... |
| Corliss and Bozman[f] | Arc | 11 | 960 |

[a] Reference 7.
[b] Reference 4.
[c] Determined from absolute transition probabilities based on the mean life of the 27 887-cm$^{-1}$ level from Ref. 1.
[d] Reference 5.
[e] Reference 1.
[f] Reference 18.

can be fitted to the raw data by the computer program. In addition, a systematic error of ≈1% due to possible nonlinearities in the time scale of the system is assigned to the present results. Other possible systematic errors in $L$, $a$, $B_\gamma$, and $n$ are difficult to evaluate. In this connection it should be emphasized that errors in these four quantities affect only the corrections to the measured lifetimes of ≲2%, which are small compared to the total uncertainties of 7 to 10%. No estimates of possible errors due to cascading have been attempted, since no downward transitions terminating in the levels of interest are identified in the latest available experimental line lists.[10,11]

The comparison values given in Table II can be separated into two groups, those of Carlson *et al.*[7] and Klose,[1] which were obtained from direct lifetime measurements, and those of Voigt and Kornblith,[4] Corliss,[5] and Corliss and Bozman,[18] which were obtained from arc measurements of relative $gf$ values. The result of Corliss for the 16 900-cm$^{-1}$ level was obtained by placing a temperature-corrected system of the relative $gf$ values of Corliss and Bozman on an absolute scale determined by the 7.3-ns lifetime for the 27 887-cm$^{-1}$ level.[1] Voigt and Kornblith used this same lifetime to establish an absolute scale for their system of relative $gf$ values. These three arc results for the 16 900-cm$^{-1}$ level should be regarded as upper limits on the lifetime since only three downward transitions were used in the two related works and only one such transition was used in the third work to derive the values.

For the 16 900-cm$^{-1}$ level the result of Carlson *et al.*, obtained by a process of two-step photoionization with tuned lasers, is seen to be in near agreement with the present work. For the 27 887-cm$^{-1}$ level the early result of Corliss and Bozman agrees with the present work although their result for the 16 900-cm$^{-1}$ level is at wide variance with the present work. The two later arc results provide a comparison of values for the lifetime of the 16 900-cm$^{-1}$ level obtained from systems of relative transition probabilities whose absolute scales were determined by the same measured lifetime. It is seen that these two systems give values for the 16 900-cm$^{-1}$ level which bracket the lifetime obtained in the present work. Finally, it is felt the discrepancy in the results between the present and earlier delayed coincidence works for the 27 887-cm$^{-1}$ level is due primarily to the improved signal-to-background ratio of the present work. However, we note here that twice the stated errors in the two results overlap and that these errors are essentially standard deviations. Thus, if a confidence limit approaching 100% is taken as the criterion for agreement, the results do not disagree.

## VI. ABSOLUTE $gf$ VALUES

Table III lists the relative $gf$ values of Voigt[3] and Voigt and Kornblith[4] placed on absolute scales based on the mean lives of the 27 887- and 16 900-cm$^{-1}$ levels determined in the present work. In addition, upper limits on the lifetime of each level as determined from the two sets of absolute transition probabilities are presented. As before, only an upper limit can be determined since probabilities are available for just one or two downward transitions from each level.

Referring to the discussion of the errors in the relative $gf$ values,[3,4] we combine those errors with the errors in the present measured mean lives to obtain a total uncertainty of 20% in the present tabulated $gf$ values. Thus, one sees that the two sets of $gf$ values based on the lifetime determinations of the present work are in agreement. Also, it is seen that the upper limits on the mean lives are generally consistent with lifetimes

TABLE III. Absolute $gf$ values and derived upper limits on mean lives for transitions and levels in U I.

| Upper level ($cm^{-1}$) | $J$ | λ (Å) | $gf$ ($\tau_0^{27887}$ = 10.9 ns) | $\tau_{\mu l}$ | $gf$ ($\tau_0^{16900}$ = 205 ns) | $\tau_{\mu l}$ | $gf_{LLL}$ | $\tau_{LLL}$ (ns) | $\tau_{AVCO}$ [a] (ns) |
|---|---|---|---|---|---|---|---|---|---|
| 16 900.387 | 7 | 5915.39 [c] | 0.36 | 200 | 0.35 | 205 | | 255 [e] | |
| | | 7631.74 [d] | 0.057 | | 0.055 | | | | |
| 17 070.469 | 6 | 6077.29 [d] | 0.061 | 380 | 0.059 | 390 | | | |
| | | 7533.93 [d] | 0.20 | | 0.19 | | | | |
| 17 361.895 | 6 | 5971.50 [d] | 0.073 | 640 | 0.071 | 660 | | 390 [e] | |
| | | 7639.54 [d] | 0.058 | | 0.056 | | | | |
| 19 885.515 | 7 | 5027.38 [b] | 0.35 | 160 | 0.34 | 170 | | | |
| 20 464.525 | 7 | 4885.14 [c] | 0.11 | 470 | 0.11 | 490 | | | |
| 21 584.695 | 6 | 4631.62 [b] | 0.30 | 110 | 0.29 | 110 | | | |
| | | 4768.66 [c] | 0.080 | | 0.078 | | | | |
| 22 056.302 | 6 | 4532.58 [d] | too weak | 330 | too weak | 340 | | | 300 |
| | | 4663.75 [c] | 0.13 | | 0.12 | | | | |
| 22 582.654 | 6 | 4426.93 [b] | 0.094 | 170 | 0.091 | 180 | | | 130 |
| | | 4551.97 [c] | 0.13 | | 0.13 | | | | |
| 22 754.061 | 6 | 4393.59 [b] | 0.40 | 67 | 0.39 | 69 | 0.39 [f] | 65 [f] | |
| | | 4516.72 [b] | 0.17 | | 0.16 | | | | |
| 22 862.451 | 6 | 4372.76 [d] | blended | 420 | blended | 430 | | | 135 |
| | | 4494.71 [c] | 0.094 | | 0.091 | | | | |
| 22 918.555 | 7 | 4362.05 [b] | 0.49 | 86 | 0.48 | 89 | 0.50 [f] | 68 [f] | |
| 22 543.508 | 7 | 4246.26 [b] | 0.27 | 150 | 0.26 | 160 | | | |
| 23 572.086 | 6 | 4241.11 [d] | blended | 84 | blended | 86 | | | 60 |
| | | 4355.74 [c] | 0.44 | | 0.43 | | | | |
| 24 066.566 | 7 | 4153.97 [b] | 0.46 | 84 | 0.45 | 87 | | | |
| 24 671.388 | 6 | 4790.10 [b] | 0.11 | 390 | 0.11 | 400 | | | |
| 25 319.274 | 5 | 4047.61 [b] | 0.31 | 88 | 0.30 | 90 | | | |
| 25 348.977 | 6 | 3943.82 [b] | 0.66 | 46 | 0.64 | 47 | | | |
| 25 577.725 | 4 | 4005.69 [b] | 0.15 | 140 | 0.15 | 140 | | | |
| 25 580.751 | 6 | 4005.21 [b] | 0.21 | 150 | 0.21 | 150 | | | |
| 25 672.465 | 7 | 3894.12 [b] | 0.29 | 120 | 0.28 | 120 | | | |
| 25 825.565 | 6 | 3871.03 [b] | 0.57 | 51 | 0.55 | 53 | | | |
| 25 938.232 | 6 | 3854.22 [b] | 0.37 | 79 | 0.36 | 81 | | | |
| 26 225.569 | 6 | 3811.99 [b] | 0.47 | 60 | 0.46 | 61 | | | |
| 27 477.553 | 8 | 4222.37 [b] | 0.62 | 48 | 0.60 | 50 | | | |
| | | 5164.14 [b] | 0.48 | | 0.47 | | | | |
| 27 886.992 | 7 | 3584.88 [b] | 1.6 | 10.9 | 1.6 | 11 | | | |
| | | 4620.21 [b] | 1.7 | | 1.7 | | | | |
| 28 499.859 | 4 | 3585.84 [c] | 0.22 | 79 | 0.21 | 81 | | | |
| 28 503.448 | 5 | 3507.34 [c] | 0.60 | 34 | 0.58 | 35 | | | |
| 29 837.643 | 7 | 3839.63 [b] | 1.3 | 26 | 1.2 | 27 | | | |

[a] Reference 6.
[b] Reference 3.
[c] Reference 4.
[d] Previously unreported values.
[e] Reference 7.
[f] Reference 8.

measured by others, who obtained their results by observing the yield of isotope ions as a function of the delay between exciting and ionizing laser pulses.[6-8] The single exception to this is the result of Carlson *et al.* of 255 ns for the 16 900-$cm^{-1}$ level.[7] In two cases where transition probabilities could not be determined because of blends, the comparison lifetimes indicate that the blended lines could be significant in reducing the upper limits on the mean lives. Finally, two comparison $gf$ values determined by Carlson *et al.*[8] using a method of time-resolved optical pumping are seen to be in agreement with the $gf$ values based on the mean lives measured in the present work.

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[1]J. Z. Klose, Phys. Rev. A 11, 1840 (1975).
[2]J. Blaise and L. J. Radziemski, Jr., J. Opt. Soc. Am. 66, 644 (1976).
[3]P. A. Voigt, Phys. Rev. A 11, 1845 (1975).
[4]P. A. Voigt and R. L. Kornblith, J. Opt. Soc. Am. 66, 492 (1976).
[5]C. H. Corliss, J. Res. Natl. Bur. Stand. (U.S.) A 80, 1 (1976).
[6]G. S. Janes, I. Itzkan, C. T. Pike, R. H. Levy, and L. Levin, IEEE J. Quantum Electron. QE-12, 111 (1976).
[7]L. R. Carlson, J. A. Paisner, E. F. Worden, S. A. Johnson, C. A. May, and R. W. Solarz, J. Opt. Soc. Am. 66, 846 (1976).
[8]L. R. Carlson, S. A. Johnson, E. F. Worden, C. A. May, R. W. Solarz, and J. A. Paisner, Opt. Commun. 21, 116 (1977).
[9]J. Z. Klose, Proc. Soc. Photo-Opt. Instrum. Eng. 76, 150 (1976).
[10]F. Guyon, doctoral thesis (University of Paris, 1972) (unpublished).
[11]D. W. Steinhaus, L. J. Radziemski, Jr., R. D. Cowan, J. Blaise, G. Guelachvili, Z. Ben Osman, and J. Vergés, Los Alamos Scientific Laboratory Report No. LA-4501, 1971 (unpublished).
[12]G. M. Lawrence and H. S. Liszt, Phys. Rev. 178, 122 (1969).
[13]H. K. Holt, Phys. Rev. A 13, 1442 (1976).
[14]N. F. Ramsey, *Molecular Beams* (Oxford U.P., London, 1956), Chap. II.
[15]J. Kaspar, in *American Institute of Physics Handbook*, edited by D. E. Gray (McGraw-Hill, New York, 1972), 3rd. ed., Chap. 6k.
[16]R. J. Ackermann and E. G. Rauh, J. Phys. Chem. 73, 769 (1969).
[17]E. K. Storms, *The Refractory Carbides* (Academic, New York, 1967), Chap. XI.
[18]C. H. Corliss and W. R. Bozman, *Experimental Transition Probabilities for Spectral Lines of Seventy Elements*, Natl. Bur. Stand. (U.S.), Monogr. No. 53 (U.S. GPO, Washington, D. C., 1962), pp. 464–469.