Transition probability of the 358.48-nm transition in neutral uranium from the line-absorption method

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The transition probability of the 358.487-nm transition in neutral uranium has been measured by the line-absorption method using a single hollow-cathode discharge lamp. Our value agrees with that reported by Klose and Voigt [Phys. Rev. A **16**, 2032 (1977)] but is surprisingly about four times larger than the value reported by Bieniewski [J. Opt. Soc. Am. B **1**, 300 (1984)].

INTRODUCTION

In the existing data on the transition probabilities of neutral uranium transitions,[1] the values obtained by Klose and Voigt[2] and Bieniewski[3,4] are considered to be the most accurately measured values. The absolute transition probability of the 358.487-nm transition was used in both of these works as a reference value. However, the transition probability of the 358.487-nm transition reported by Klose and Voigt based on lifetime measurements was about three times larger than that reported by Bieniewski using an absorption method. To resolve this discrepancy, we measured the relative transition probability of the 358.487-nm transition by our recently reported[5,6] line-absorption technique using a single hollow-cathode discharge lamp (HCDL). The relative transition probability of the 358.487-nm transition is placed on an absolute scale using the reported value[6] of the transition probability of the 436.205-nm transition. Our value of the transition probability of the 358.487-nm transition is closer to that reported by Klose and Voigt, but is about three and a half times larger than the value reported by Bieniewski. Since the transition probability of the 358.487-nm transition obtained by us is three and a half times larger than that of Bieniwski, we wanted to check whether such a large difference in the values is a stray case or whether it is systematic. Therefore, we measured the absolute transition probability of another U I line at 356.659 nm. This was done by first measuring the relative transition probability of the 356.659-nm U I transition, involving the lowest metastable state (620 cm^{-1}), and placing it on an absolute scale using the reported value[6] of the transition probability of the 435.574-nm transition, also ending on the 620-cm^{1} level. It is found that for this transition also, our value is again three and a half times larger than that reported by Bieniewski. In this Brief Report we present our experimental observations and their comparison with the results of previous workers.

EXPERIMENTAL PROCEDURE

The experimental setup is the same as in earlier reported works.[5,6] The uranium HCDL used in these studies was designed and fabricated in our laboratory. The cathode is a 30-mm-long 10-mm-diam natural uranium cylinder having a bore of 7 mm diam. A standard vacuum system allows easy replacement of the carrier gas and regulation of its pressure in the discharge lamp. The HCDL is continuously cooled with water at 5 °C. The absorption coefficients k_0 for all the transitions are estimated[6] from Eq. (1) by measuring the transmitted light intensity I_t, the direct light intensity I_d, and the calibration factor R:

$$A_\alpha = 1 - \frac{I_t}{RI_d} = \frac{\sum_{n=1}^{\infty} \frac{(-1)^{n+1}(k_0 l)^n}{n!\sqrt{n}} \left[2 - n! \sum_{i=0}^{n} \frac{1}{i!(n-i)!} \right]}{\sum_{n=1}^{\infty} \frac{(-1)^{n+1}(k_0 l)^n}{n!\sqrt{n}}} , \quad (1)$$

where A_α is the line absorption.

The transition probability of a transition is estimated from the measured absorption coefficients with the help of the following relation:

$$gA_s = \frac{k_{0s}}{k_{0r}} (\lambda_r / \lambda_s)^3 gA_r , \quad (2)$$

where subscripts s and r represent, respectively, the transition for which the transition probability is to be evaluated and the reference transition.

To estimate the experimental error in the gA_{3584} value, all the measurements are repeated eight times in different discharge conditions. The absorption coefficient ratio $[k_0(3584)/k_0(4362)]$ listed in Table I in different discharge conditions and the reference transition probability value[6]

$$gA_{4362} = (1.645 \pm 0.049) \times 10^8 s^{-1}$$

give the average transition probability value

$$gA = (1.018 \pm 0.079) \times 10^9 \ s^{-1} ,$$

where the stated uncertainty is one rms variance.

COMPARISON WITH OTHER MEASUREMENTS

Before comparing our results with other measurements reported in the literature the experimental uncertainties affecting our results should be evaluated. The line-absorption method[7] is valid if the natural damping ratio $a \leq 0.01$ and the product $k_0 l \leq 3.0$. The natural damping

TABLE I. The ratios of the absorption coefficients of the 358.487- and 436.205-nm transitions of neutral uranium obtained with an effective spectral resolution of 1.0×10^5.

Buffer gas	Pressure (Torr)	Discharge current (mA)	k_0l 3584	k_0l 4362	$\frac{k_0(3584)}{k_0(4362)}$
Argon	1.1	70.0	2.31	0.60	3.88
		80.0	2.38	0.71	3.37
		90.0	2.48	0.75	3.31
	1.5	90.0	1.43	0.38	3.81
Neon	1.5	70.0	2.81	0.92	3.06
		90.0	2.91	0.92	3.17
	2.0	70.0	2.52	0.73	3.48
		90.0	2.75	0.81	3.41
Argon	1.1	60.0	3.05	0.79	3.86[a]
Mean value					3.436
rms variance					0.268

[a]This value was obtained using a 6-mm bore HCDL at an effective spectral resolution of 2.0×10^5 and is not included in averaging.

ratio is defined as

$$a=\sqrt{\ln 2}\frac{\Delta\nu_L}{\Delta\nu_D},$$

where $\Delta\nu_L$ is the total Lorentzian width and $\Delta\nu_D$ is the Doppler width. The HCDL is operated at very small pressure (1–3 Torr); therefore pressure broadening effects on the natural linewidth of the 358.487-nm transition ($\tau=10.9$ ns) can be ignored. For the 358.487-nm transition in the natural damping ratio in a HCDL is about 0.01 as the Doppler width in the HCDL at the 358.487-nm line is about 1.1 GHz.[5] It can be seen in Table I that the value of the product k_0l in all the discharge conditions is always less than 3.0. The other experimental uncertainty in this investigation could be caused by the interference of neighboring lines. All the intensities were recorded using a monochromator (Jobin Yvon model THR-1500) with an effective resolution $\nu/\Delta\nu=1.0\times10^5$ at 358 nm. But to verify that the desired line is not blending with any neighboring lines, the spectrum in the neighborhood of the 358.487-nm U I line was recorded with the same monochromator with an effective resolution $\nu/\Delta\nu=2.0\times10^5$ and it was possible for us to resolve all the spectral lines[8] in the neighborhood of the 358.487-nm U I line. It can be seen in Fig. 1 that there is no neighboring line that can significantly affect the absorption coefficient of the 358.487-nm transition. We also measured the absorption coefficient ratio $[k_0(3584)/k_0(4362)]$ at higher resolution (2.0×10^5) and it can be seen in Table I that the value obtained at this resolution agrees with those obtained at lower resolution (1.0×10^5). The error due to the interference of the minor isotope (^{235}U, 0.7 wt. %) will be one order of magnitude smaller than the experimental error and therefore can be ignored. In our experiment we have assumed the emission length and absorption length in a HCDL to be equal but in the actual situation there always exists diffusion of the sputtered atoms out of the cathode bore

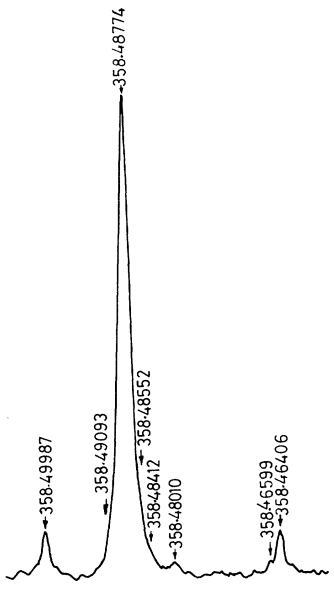

FIG. 1. Uranium hollow-cathode emission spectrum in the neighborhood of the 358.487-nm U I transition recorded with effective spectral bandwidth of 0.0018 nm in our experimental setup.

TABLE II. The absolute transition probabilities of the 358.487- and 356.659-nm U II transitions of neutral uranium.

Wavelength (nm)	gA_{3584} (10^9 s^{-1}) This work	Ref. 2[a]	Ref. 3
358.487	1.018±0.79	0.82	0.28±0.03
356.659	1.210±0.120		0.26±0.03

[a]The error is around 20%.

TABLE III. Transition probabilities of the 599.731-, 358.487-, and 462.021-nm U I transitions estimated by the "bowtie" method from the least-squares adjustment of the relative intensities and absorption measurements of these U I transition measured with a spectral bandwidth of 0.0018 nm.

Wavelength (nm)	Intensity I (arb. units)	Absorption coefficient k_0 (cm^{-1})	Relative gA value	gA[a] (10^8 s^{-1})
436.205	49.11+0.60	0.292±0.005	1.00	1.645[b]
599.731	7.13 ±0.14	0.024±0.008	0.20	0.328
358.487	52.27±0.29	0.946±0.013	5.83	9.591
462.021	7.65 ±0.18	0.135±0.006	1.10	1.810

[a]Error is less than 10%.
[b]Reference value from Ref. 6.

on both sides. Thus the absorption length in a HCDL will always be slightly greater than the emission length. If an inverse-square dependence is assumed for the expansion of the material from the cathode bore, the ratio of optical opacities[9] ($\alpha = k_0' l' / k_0 l$) in emission and absorption will be at most about 0.9 instead of 1.0. We calculated the relative optical opactiy of 358.487 nm with respect to 436.205 nm by using $\alpha = 0.9$ in Eq. 2 of Ref. 9 and found that it is about 3% less than the value we have obtained by using $\alpha = 1.0$. Therefore, we can conclude that the systematic error due to the difference in the emission and absorption lengths will not be more than 3%. The experimental error in the measurement of line absorption A_α will be given as

$$\Delta A_\alpha = \frac{\Delta I_t}{R I_d} + \frac{I_t \Delta R}{R^2 I_d} + \frac{I_t \Delta I_d}{R I_d^2} .$$

By substituting the typical values of I_t, I_d, and the calibration factor R and their respective uncertainties in the above equation, we found that the experimental error in A_α is about 3%. This error in A_α will cause about 6% error in the relative optical opacity of 358.847 nm with respect to 436.205 nm. It shows that all the systematic errors added together will not cause more than 10% error in our values and it can be noticed in Table II that, within experimental error, our value is in good agreement with that of Klose and Voigt[2] and is about three and half times larger than the value reported by Bieniewski.[3]

As there exists a discrepancy between our value and that of Bieniewski, we also measured the absolute transition probability of the transition by the "bowtie" method.[9] This was done by measuring the relative emission intensities (self-absorption-free)[6] and absorption coefficients of the 436.205-, 599.731-, 358.487-, and 462.021-nm transitions. The transitions 436.205 and 599.731 nm share the common upper level 22 918 cm^{-1} and the transitions 358.487 and 462.021 nm share the common upper level 27 887 cm^{-1}. The transitions 599.731 and 462.021 nm share the common lower level 6249 cm^{-1}. The relative intensities were put on a common scale by calibrating our optical detection system using a standard tungsten ribbon lamp. It can be noticed in Table III that the transition probability value obtained from the bowtie method for least-squares adjustment of relative itensities and absorption measurements agrees excellently with the values obtained by the line-absorption method. We also measured the relative transition probability of the 356.659-nm transition and placed in on an absolute scale using the transition probability value[6]

$$gA_{4355} = (1.172 \pm 0.062) \times 10^8 \ s^{-1}$$

of the 435.574-nm transition. The absolute transition probability of the 356.659-nm transition is given in Table II. It is surprising that the relative value of the 358.487-nm with respect to the 356.659-nm transition obtained by us is in excellent agreement with that reported by Bieniewski but the absolute value of the 356.659-nm transition is again about three and a half times larger.

DISCUSSION

In Klose and Voigt's work the absolute transition probability of the 358.487-nm transition is estimated from the measured lifetime of the 27 887-cm^{-1} level and branching ratio of the same transition. The branching ratio was estimated from the measured intensities of different transitions starting from the 27 887-cm^{-1} level. Since these intensity estimates are not very accurate, we cannot fully rely upon the absolute gA value estimated by Klose and Voigt. However, if we estimate the absolute gA value of the 358.487-nm transition from its relative gA value obtained by Klose and Voigt, by making use of the absolute gA values of the 436.205- and 439.358-nm U I transition reported by Carlson *et al.*,[11] we obtain nearly the same value. A comparison in Table II, between the transition probability value of the 358.487-nm transition obtained by us with that obtained by Klose and Voigt, implies that in their work self-absorption dominated over the blending effect for the 358.487-nm transition.

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