Simple Technique to Measure Relative Transition Probabilities: Application to UI Transitions

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A novel and relatively simple technique based upon the line absorption method to measure relative transition probabilities is reported. The absorption measurements are performed with the use of a single hollow cathode discharge lamp, which serves as a light source as well as an absorption cell. The technique has been applied to measure the relative transition probabilities of a few ground-state transitions in UI. This technique can also lead to the determination of excitation temperature in the discharge without the aid of data from other sources.

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There are two basic methods for measuring the relative transition probabilities of atoms. Some methods make use of the emission spectrum of atoms in arcs[1,2] or discharge lamps[3,4] and others of the absorption spectrum.[5–7] The emission methods are inherently error prone because of the self-absorption of transitions and deviation of the plasma from local thermodynamic equilibrium. The absorption methods require a fully resolved absorption profile to measure the absorption coefficient of each transition, which can be obtained by either a high-resolution spectrograph[5,6] or a tunable cw laser.[7] Line absorption with use of an emission line source and an absorption cell is another method.[8] For measuring absorption coefficients by this method one needs to know the ratio of the Doppler widths of the spectral lines (if Doppler broadening dominates over other broadenings) in the light source and absorption cell and also the ratio of their absorption coefficients at the line center. To measure these ratios one has to rely on some other technique.[9] This problem can be solved if the same discharge lamp acts as the source of light as well as the absorption cell. The hollow cathode discharge lamp (HCDL) provides a simple and very reliable method of producing neutral atoms.[10,11] Absorption studies[12] in HCDL's have revealed that even at a current of 100 mA a high number density of sputtered atoms (10^{12}–10^{13} atoms cm^{-3}) can be produced which results in considerable absorption. In this Letter we describe a novel and relatively simple technique based upon line absorption to measure relative transition probabilities using a single HCDL.

The line absorption A_α has been defined as[8]

$$A_\alpha = 1 - \frac{I_t}{I_0} = \frac{\int E_\nu [1 - \exp(-k_\nu l)] d\nu}{\int E_\nu d\nu}, \tag{1}$$

where I_t is the transmitted light intensity, I_0 is the incident light intensity, E_ν is the spectral distribution of the line source, k_ν the absorption coefficient, and l is the length of the absorbing medium along the line of sight. For a self-absorbing source E_ν can be expressed as

$$E_\nu = S_\nu [1 - \exp(-k'_\nu l)], \tag{2}$$

where S_ν is the Planck function, assumed to be constant over the line profile, and k'_ν is the absorption coefficient in the light source. If Doppler broadening dominates over other broadening mechanisms, and the light source and the absorbing cell are the same, by the substitution of Eq. (2) in Eq. (1) the measured line absorption A_α can be related to the absorption coefficient k_0 at the line center by means of the relation[9]

$$A_\alpha = \frac{\sum_{n=1}^{\infty} (-1)^{n+1} [(k_0 l)^n / n! \sqrt{n}] \{2 - n! \sum_{i=0}^{n} 1/i!(n-i)!\}}{\sum_{n=1}^{\infty} (-1)^{n+1} [(k_0 l)^n / n! \sqrt{n}]}, \tag{3}$$

where

$$k_0 = \frac{1}{4\pi} (\ln 2/\pi)^{1/2} \frac{g_2 A \lambda_0^2 N}{g_1 \Delta\nu_{\mathrm{D}}}. \tag{4}$$

In Eq. (4) g_1 and g_2 are the statistical weights of the lower and upper level, respectively, A is the transition probability, λ_0 is the wavelength at the line center, N is population in the lower level, and $\Delta\nu_{\mathrm{D}}$ is the Doppler width. In Fig. 1 A_α is plotted against $k_0 l$. It can be seen from the curve that the dynamic range for k_0 approximately corresponds to the value of $A_\alpha = 0.0$ to 0.7. Beyond $A_\alpha \approx 0.7$, the accuracy of the evaluation of k_0, and therefore A, becomes poorer. However, the experimental conditions can always be chosen so that $A_\alpha < 0.7$. By the measurement of k_0 for different transitions starting from the same lower level, relative transition probabilities of these transitions can be obtained.

The above analysis is for a single line. In the presence of hyperfine-structure and/or isotopic components Eq. (1) can be rewritten as[8]

$$A_\alpha = \frac{\int E_\nu^1 [1 - \exp(-k_\nu^1 l)] d\nu + \int E_\nu^2 [1 - \exp(-k_\nu^2 l)] d\nu + \cdots}{\int E_\nu^1 d\nu + \int E_\nu^2 d\nu + \cdots}, \tag{5}$$

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where superscripts denote separate hyperfine-structure components or different isotopic components. With use of Eq. (2), Eq. (5) can be rewritten as

$$A_a = \frac{\sum_{n=1}^{\infty} (-1)^{n+1}[(k_0 l)^n/n!\sqrt{n}](1+a^n+b^n+\cdots)\{2-n!\sum_{i=0}^{\infty} 1/i!(n-i)!\}}{\sum_{n=1}^{\infty} (-1)^{n+1}[(k_0 l)^n/n!\sqrt{n}](1+a^n+b^n+\cdots)}, \tag{6}$$

where $a,b,c,\ldots,$ are the relative intensities of different hyperfine or isotopic components. Experimentally or theoretically determined values of $a,b,c,\ldots,$ can be used to calculate k_0 from the measured value of A_a. For a spectral line with hyperfine structure the value of k_0 is given as

$$k_0(1+a+b+c+\cdots) = \frac{1}{4\pi}(\ln 2/\pi)^{1/2}\frac{g_2 A \lambda_0^2 N}{g_1 \Delta\nu_D}. \tag{7}$$

The above equation is a result of an assumption that is justified theoretically, namely, that all the hyperfine levels of a resonance level have the same lifetime. However, for unresolved hyperfine components and/or isotopic components this method is not applicable.

Figure 2 shows the schematic of the experimental setup. The uranium HCDL used in these studies was designed and fabricated in our laboratory. The hollow cathode is a 30-mm-long, 10-mm-diam natural uranium cylinder having a bore of 8 mm diam. Argon is filled to 1.1 Torr and the lamp is operated at 90.0±0.1-mA current. The HCDL is continuously cooled with water at 10 °C.

The height of the entrance slit of the monochromator (model THR-1500 of M/S Jobin Yvon with an effective resolution $\nu/\Delta\nu = 1.25\times10^5$ at 500.0 nm) is adjusted in such a way that only light from the central part of the lamp can enter the slit. Lens L_2 focuses the light from the HCDL on a mirror M and the light retraces its path through the lamp and is focused by lens L_1 on the entrance slit of the monochromator. The monochromator is coupled to a Hamamatsu model R955 photomultiplier tube with a current amplifier. The photomultiplier output signal is simultaneously fed to a lock-in amplifier (PARC model 124) and a boxcar averager with dc input coupling (PARC model 165/162). The chopper provides both the reference signal for the lock-in-amplifier and the trigger signal for the boxcar averager. The gate position of the boxcar averager with respect to the photomultiplier tube signal is shown in Fig. 3. The lock-in amplifier gives a voltage signal I_t proportional to the intensity of light reflected from mirror M and transmitted through the hollow cathode, while the boxcar averager

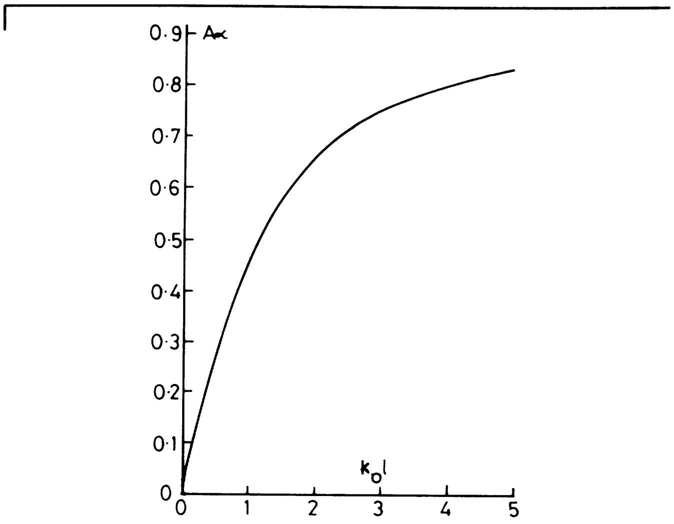

FIG. 1. Plot of A_a vs $k_0 l$.

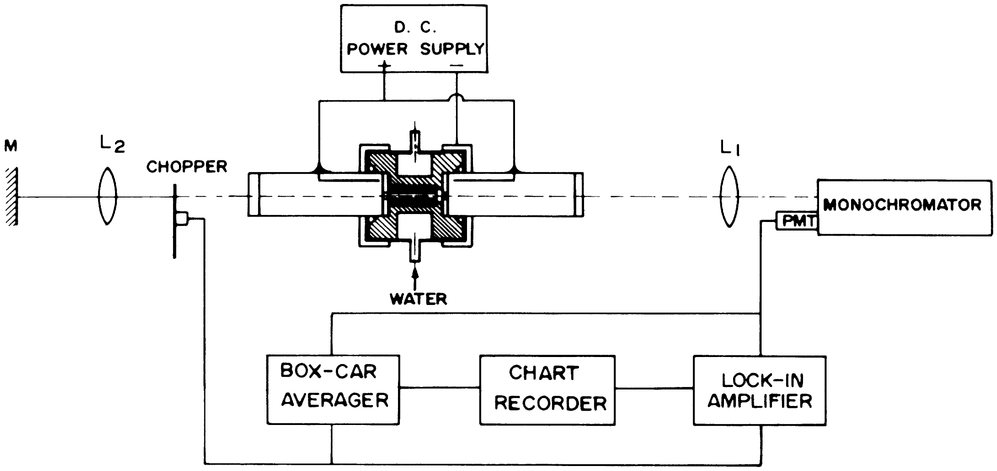

FIG. 2. Schematic of the experimental setup.

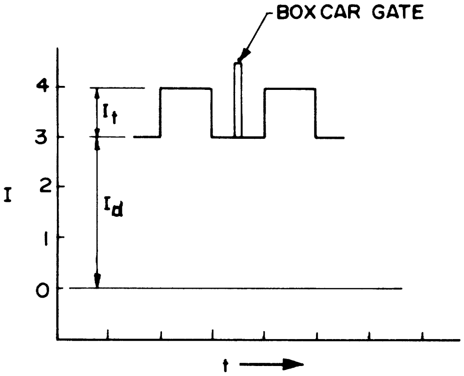

FIG. 3. Boxcar averager gate position with respect to the photomultiplier signal output.

gives a voltage signal I_d proportional to the direct light intensity. Both of these signals are simultaneously recorded on a chart recorder as functions of wavelength. The spectral bandwidth of the monochromator (0.160 cm^{-1}) is 5 to 6 times larger than the Doppler width of the spectral lines (0.03 cm^{-1}) emitted from HCDL and so the peak value of the recorded line can be assumed to be proportional to the true intensity of the emitted line. The direct light falling on the entrance slit of the monochromator is proportional to the light falling on the mirror M; therefore signal I_0, proportional to the intensity of light incident on the HCDL, can be obtained as

$$I_0 = RI_d,$$

where the proportionality constant R depends only on the optical geometry and the detection system used. For weak transitions with negligible absorption

$$I_t = I_0 = RI_d, \quad R = I_t/I_d.$$

The value of R is obtained by the measurement of the ratio of I_t and I_d for a few weak transitions at different currents. In our setup the value of R was 0.144 ± 0.001. By the measurement of I_t and I_d for all the transitions listed in Table I, the absorption coefficients k_0 are calculated with use of Eq. (3). We ignored the contribution of the lesser abundant isotope (U^{235} 0.7%). The relative gA values of all the transitions are calculated with reference to the 591.538-nm uranium line. The gA values listed in Table I for different transitions are calculated using the value of $gA = (4.71 \pm 0.7) \times 10^7\ \mathrm{s}^{-1}$ for the 591.538-nm transitions.[5] The values of k_0 and gA given in columns 2 and 3 are the average of three independent measurements. In these measurements, the HCDL was assumed to be a homogeneous source of sputtered atoms. However, the radial inhomogeneity in atom distribution in the lamp can be a source of error in the measurement of the relative values of k_0 because of its nonlinear dependence on A_α. The extent of radial inhomogeneity was estimated by our measuring the value of A_α using two different apertures (6 mm and 2 mm diam) in front of the lamp. It was found that the radial variation in the value of A_α was about 10% and within this variation the value of k_0 will change linearly with A_α (Fig. 1). Therefore, possible errors due to radial inhomogeneity in the relative k_0 values will be negligible. It can be seen that the average experimental error in the values of k_0 is not more than 3%. The large errors in gA values are predominantly because of the large errors in the reference values we have taken. Obviously, with more accurate reference values we can definitely obtain more accurate gA values with our simple technique than those obtained from more involved techniques.[1,2]

In conclusion, we report a novel and simple technique to measure relative transition probabilities, which can be applied, in general, to atoms of any metal and, in particular, to refractory metal atoms of astrophysical interest. These in turn can lead to a better estimate of their abundance in solar and other stellar objects. The advantage of recording an emission spectrum with this technique is that one can measure the extent of self-absorption present in different spectral lines[15] and can also find evi-

TABLE I. Transition probabilities in UI.

Wavelength of trans. (nm)	Absorption coefficient k_0 (cm^{-1})	gA ($10^8\ \mathrm{s}^{-1}$) This work	Ref. 1	Ref. 13[a]	Ref. 14
424.626	0.429 ± 0.004	1.37 ± 0.25	1.10	1.00	
436.205	0.559 ± 0.007	1.69 ± 0.32	1.60	1.72	1.76 ± 0.3
437.275	0.302 ± 0.007	0.91 ± 0.18	0.25		
439.358	0.499 ± 0.01	1.49 ± 0.29	1.20	1.38	1.36 ± 0.2
463.161	0.292 ± 0.004	0.78 ± 0.14	0.68	0.93	
488.514	0.135 ± 0.001	0.33 ± 0.06	0.11	0.31	
502.738	0.297 ± 0.01	0.677 ± 0.14	0.44	0.92	
591.538	0.286 ± 0.005	0.471 ± 0.07[b]	0.35	0.68	
639.542	0.0514 ± 0.0003	0.072 ± 0.013	0.11		

[a] Uncertainties for these values are quoted as ± 30.
[b] Value taken from Ref. 5.

dence of population inversion ($k_0 < 0$) in any level in the discharge. Since the self-absorption-free intensities and relative transition probabilities can be obtained with the method, the excitation temperature in the discharge can be measured without the aid of data from other sources.

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