DIRECT MEASUREMENT OF MEAN LIVES OF ATOMIC STATES

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Abstract

A method is developed for measuring the mean lives of atomic states by means of the thermal motion of excited atoms in two forms of resonance lamps. The methods are used to determine the mean life of the cadmium 2^3P_1 state for which state the more accurate method gives $T=2.5\times10^{-6}$ sec. with a probable error which is difficult to determine but can hardly be in excess of 10 percent.

IT is obvious that measurements of the mean life of atomic states by means of the thermal motion of excited atoms in a resonance lamp are possible if the mean lives are long enough to make the distance which an excited atom

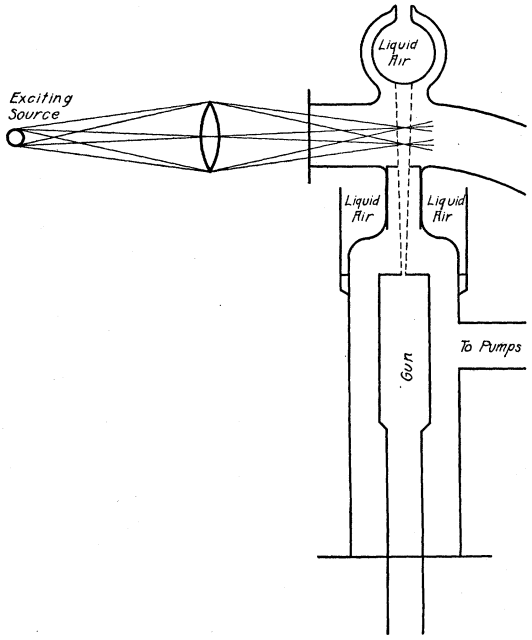

Fig. 1. Preliminary system. The only changes made throughout the entire procedure were to enter stops defining the exciting beam of radiation and to connect a second pump to the upper liquid air trap.

will move between excitation and emission observable by means of the camera with which the radiation is photographed.[1] Since the cadmium 2^3P_1

[1] Dunoyer attempted this in 1913. He shot a beam of sodium atoms through a beam of radiation of the sodium D lines. The mean lives of the excited states—the 2^2P states—were too short, however, to cause any observable motion of the excited atoms out of the excitation region. Dunoyer, Le Radium **10**, 400 (1913).

state was believed to have a mean life of sufficient length to permit such a measurement—Ellett obtaining the value 2.3×10^{-6} sec. by the method of depolarization of resonance radiation in weak magnetic fields—the following system was adopted to endeavor to measure the mean life of this state by a

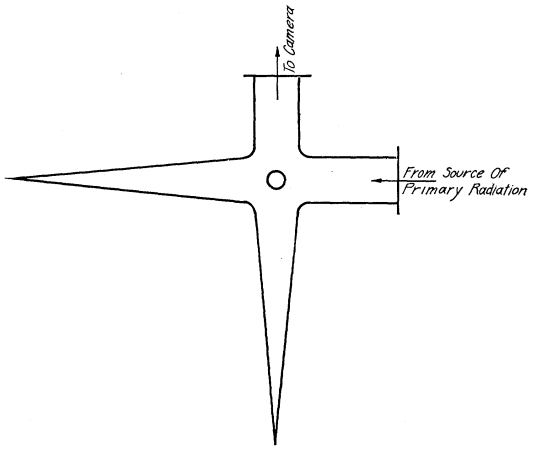

Fig. 2. Cross section of experimental chamber, as seen from above.

method depending upon the thermal motion of excited atoms. The apparatus, with the stops used to define the beam of exciting radiation removed, is shown in Figs. 1 and 2. The cadmium vapor effusing from a small hole in the top of a boiler—the gun of the diagram—was frozen out of the system with liquid air except for the small portion passing through the hole in an aluminum tube set in the constriction between the gun chamber and the upper chamber. The image of a cadmium discharge was focused upon the cadmium beam and the resonance radiation photographed along a line perpendicular to both cadmium beam and direction of exciting light ray.

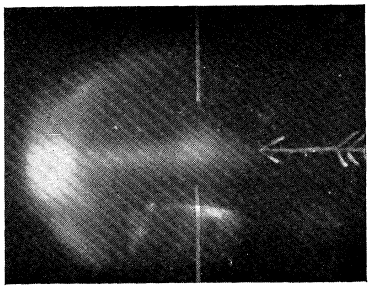

Fig. 3. Print from plate taken in preliminary run.

The print shown in Fig. 3 is from a preliminary run taken with the apparatus, and is given because it illustrates the coarser characteristics of the method better than the plates taken later would. The arrow indicates the direction of the incident light ray, the broken line the direction of the cad-

mium beam. Collisions between the cadmium atoms in the beam and air molecules in the experimental chamber caused sufficient vapor pressure of cadmium to develop in the experimental chamber to define the path of the exciting light ray by resonance radiation from this vapor. Before the more accurate work was attempted, a second pump was added to the system to eliminate this effect. At the position where the cadmium beam passed through the exciting ray the radiation is noticeably more intense and extends about 1.5 mm above the upper edge of the exciting beam.

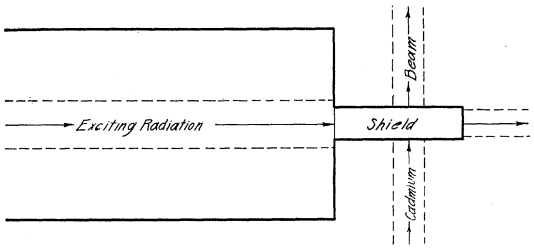

Fig. 4. Cross sections of slit system used for first method of determining the mean life.

To make use of such a system, however, it is necessary, first, to make the intensity of the exciting radiation break off sharply at the upper edge of the excitation region as we move along the beam; second, to obtain a system which will show a definite place to start from in making measurements on the films; third, to demonstrate that the effect is due to motion of excited atoms and not secondary resonance radiation. To meet these requirements the slit system of Fig. 4 was inserted into the experimental chamber of the apparatus. An improvement in the pumping system was made at the same time and the gas pressure in the experimental chamber reduced to less than 5×10^{-6} mm Hg.

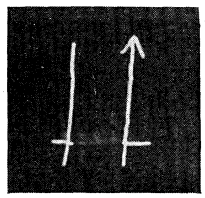

Fig. 5. Print of plate taken for first method of determining mean life.

In this system the exciting radiation from the discharge[2] strikes a slit and then passes behind a shield which hides the path of the radiation from the camera. This shield was made so as to extend 0.5 mm beyond the edge of the excitation region in order to make adjustment easier and give complete assurance that the path of the exciting radiation was hidden from the camera. The cadmium beam passed through the exciting radiation behind this shield.

[2] The discharge was of the type described by Ellett in connection with his earlier work on the depolarization of resonance radiation in magnetic fields. Ellett, Jour. Opt. Soc. Am. **10**, 427 (1925).

In Fig. 5 the upright lines mark the edges of the cadmium beam and the arrow indicates the direction of motion of the atoms. The lines drawn out to the side from these indicate the top of the shield. These marks were drawn on the plate. The radiation photographed is observed to start at the marks indicating the upper edge of the shield and to extend upward, decreasing in intensity quite rapidly as we go up along the beam.

This radiation was now entirely due to motion of the excited atoms since the entire excitation region was hidden from the camera and the failure of any radiation to appear below the shield eliminates the possibility of the effect being due to secondary resonance radiation.

Microphotometer curves of plates obtained by this method were taken. Assuming that the blackening on these plates was proportional to the intensity of radiation and that all the atoms were moving with the arithmetic mean velocity, the intensity of radiation outside the excitation region should be $I=I_0e^{-t/\tau}$ where t is the time the atom has been out of the excitation region and I_0 the intensity at the edge of the excitation region. Then if x is the distance from the edge of the excitation region along the direction of motion of the cadmium atoms, $t=x/v$ and by our assumption v is the arithmetic mean velocity.

Then

$$I = I_0e^{-x/v\tau}.$$

And the mean velocity of the atoms may be calculated from the temperature of the boiler which was measured by a copper-constantin thermocouple inserted in the base of the boiler for the number of atoms having a velocity between v and $v+dv$ effusing from the boiler is proportional to

$$e^{-mv^2/2kT}v^3dv$$

and if the probability that one of these is excited is proportional to $1/v$ the number of excited atoms having velocities between v and $v+dv$ is proportional to

$$e^{-mv^2/2kT}v^2dv.$$

So for the excited atoms

$$v(\text{mean value}) = \frac{\int_0^\infty e^{-mv^2/2kT}v^3dv}{\int_0^\infty e^{-mv^2/2kT}v^2dv}.$$

The assumption that the probability of excitation is proportional to $1/v$ is justified by the fact that the time any of these atoms remains in the excitation region is short and proportional to $1/v$. By fitting curves of this form to the microphotometer curve, the mean life of cadmium 2^3P_1 was calculated to be $\tau=1.3\times10^{-6}$ sec. The assumptions used, however, are not sufficiently accurate to regard this as more than a measurement of the order of magnitude, probably correct to within a factor of 2.

In this regard some justification must be given for giving this value to the 2^3P_1 state. Except in the case of very high excitation, 2^3P_1 and 2^1P_1 are the only states excited to any noticeable degree. It is a matter of experience with resonance radiation excited by a discharge of the type used that after the discharge has operated for some time its ability to excite the 2^1P_1 state decreases probably due to the formation of a very thin layer of cadmium glass in the quartz section from which the exciting radiation is taken. Since the radiation obtained in the cadmium beam was of very low intensity, the exposure time for the films was about four hours, 2^3P_1 and 2^1P_1 are the only states which could be excited to any degree, and 2^3P_1 is chosen because of the characteristics of the source of exciting radiation.

The difficulties met here in connection with the faults of the plate can be overcome if we can obtain a system in which the position of the maximum intensity is a function of the mean life τ and variables depending upon the geometry of the apparatus and the temperature of the cadmium atoms. In the system just discussed the greatest intensity was at $x=0$ regardless of either the mean life or the gun temperature. To obtain a set up of this type, let us consider a system in which the number of excited atoms having a velocity between v and $v+dv$ and position between x and $x+dx$ is N where x is measured along the atomic beam from an arbitrary plane perpendicular to the beam. Then since there is no appreciable secondary resonance radiation we may assume that the actual system consists of a group of independent systems each having a definite velocity—or range of velocities—and the contribution of the group to the intensity of radiation will be the sum of the contributions of the individual systems.

Then $(N/\tau)dtdx$ is the number radiating between $x=x$ and $x=x+dx$, $t=t$ and $t=t+dt$; and if $f(x)dxdt$ is the probability that an atom will be excited between x and $x+dx$, t and $t+dt$, the number excited within these limits will be $f(x)(N_g/v)dxdt$. Here N_g is the number of atoms leaving the gun and entering the beam with a velocity between v and $v+dv$ in one second, and in any experiment we can carry out $N_g/v \gg N$.

Now if we follow a group of atoms along the beam and disregard forced radiation due to light falling upon excited atoms $(dN/dt)dx=f(x)(N_g/v)dx - (N/\tau)dx$, or considering the forced radiation

$$(dN/dt)dx = f(x)(N_g/v)dx - (N/\tau)dx - Bf(x)Ndx$$

where B is the B_{2-1} coefficient of Einstein's radiation laws.

But $Bf(x)$ is small compared to $1/\tau$ so

$$\frac{\partial N}{\partial x} v + \frac{\partial N}{\partial t} = f(x)\frac{N_g}{v} - \frac{N}{\tau}$$

and N is independent of t so $\partial N/\partial t = 0$

$$\frac{dN}{dx} + \frac{N}{v\tau} = \frac{N_g}{v^2} f(x), \quad N = e^{-x/v\tau}\int_0^x e^{x/v\tau}\frac{N_g}{v^2} f(x)dx$$

if $N=0$ at $x=0$.

That is, if the excitation region starts at $x=0$.

Let $f(x)$ be a continuously decreasing function finite at $x=0$ and 0 for $x \geqq a$.

Then since $N=0$ at $x=0$ and is decreasing and greater than 0 at $x=a$, there must be a maximum value of N between $x=0$ and $x=a$. The function N must be decreasing at $x=a$ since N must reduce to $kf(x)$, (k a constant) for small values of $v\tau$ or values of x large compared to $v\tau$.

For the system actually used $f(x)=A(1-ax)$, $a=1.15$. The slit system used is shown in Fig. 6.

Two straight edges were set so as to form an acute angle and make the portion of the cadmium beam exposed to the exciting radiation triangular. Then if we observe the region between two planes perpendicular to the direc-

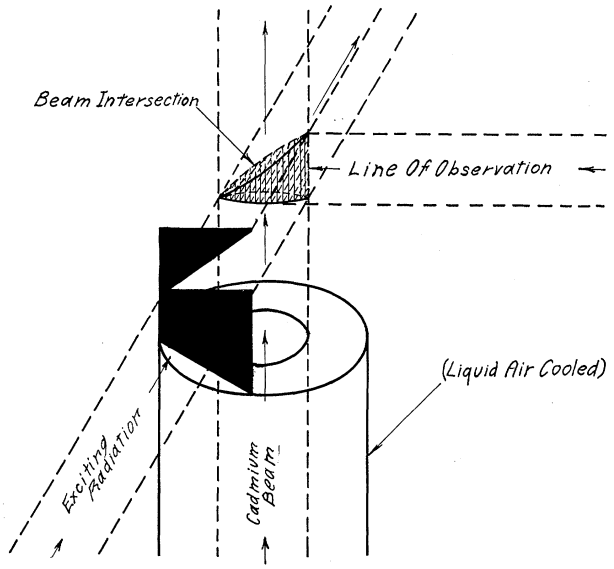

Fig. 6. Cross sections of slit system for second method.

tion or the exciting radiation and near the mid part of the cadmium beam as seen by the camera, the number of atoms excited per second between $x=x$ and $x=x+dx$ is proportional to the depth—measured along the line of sight—of the portion of the cadmium beam subject to exciting radiation. This is then proportional to $(b-cx)$ where b is the greatest width of the portion of the cadmium beam subject to excitation and c is the slope of the upper straight edge referred to the line of sight.

After the exciting radiation has passed through the cadmium beam, it struck an upright. This upright—a small wire nail—was heavily blackened to prevent it from scattering too much light. Then the portion of this nail which was illuminated by the exciting radiation was photographed with the resonance radiation and used to define the lower edge of the excitation region, the point chosen as our zero value of x.

Fig. 7 shows the print of a plate taken with this apparatus. The upright on the left is the slit. The upright on the right is the exciting ray defining nail.

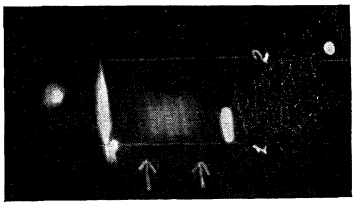

Fig. 7. Print of plate taken for second method of determining mean life.

Two arrows drawn on the plate show the direction of the cadmium beam. The radiation is seen above and between the arrows.

A microphotometer curve was taken for this plate and is shown as Fig. 8.

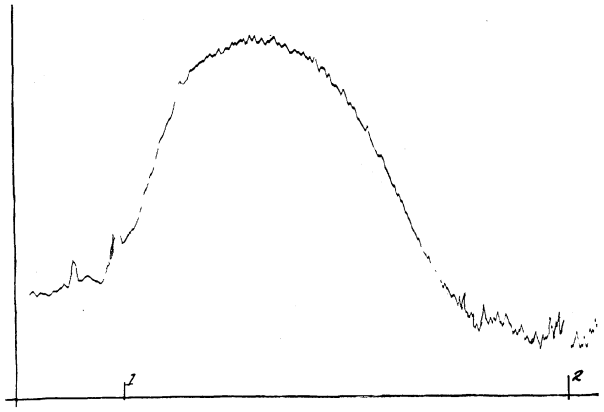

Fig. 8. Photometer curve taken off same plate as Fig. 7. Instrument run in direction of increasing x—direction of arrows on Fig. 7.

Now for the system represented here

$$I = N/\tau = A \int_0^\infty e^{-h m v^2} v^3 e^{-x/v\tau} \int_0^x e^{-x/v\tau} \frac{(1 - ax)}{v^2} dx dv$$

where $h = 1/2kT$, $a = c/b = 1.15$, $T = 690°\text{K}$.

$$N/\tau = I = A \int_0^\infty e^{-h m v^2/\tau}(- v\tau + 1.15 x v\tau - 1.15 v^2\tau^2) v dv$$

$$+ A \int_0^\infty e^{-h m v^2/\tau} e^{-x/v\tau}(v\tau + 1.15 v^2\tau^2) v dv.$$

For a maximum

$$dI/dx = 0 = A \left[1.15 \int_0^\infty e^{-h m v^2} v^2 dv - \int_0^\infty e^{-h m v^2} e^{-x/v\tau}(v/\tau)(1 + 1.15 v\tau) dv \right]$$

The first integral is known and the second was evaluated by assuming it equal to

$$\int_a^\infty e^{-h\,mv^2}(v/\tau)(1 + 1.15v\tau)dv$$

for large values of v and integrating mechanically over smaller values of v.

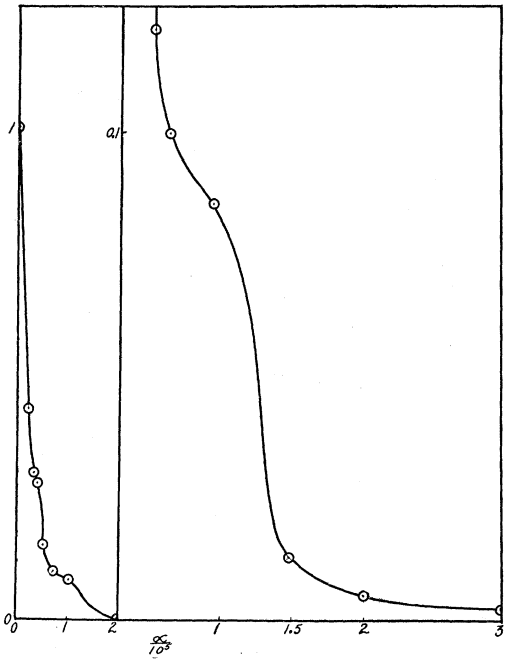

Fig. 9.

$$\frac{\int_0^{10^5} e^{-hmv^2}e^{-\alpha/v}v^2dv}{\int_0^\infty e^{-hmv^2}v^2dv}$$

plotted against $\alpha_0/10^5$.

For the mechanical integration the integral

$$\int_0^{10^5} e^{-h\,mv^2}e^{-\alpha/v}vdv,\quad \alpha = x/\tau$$

and the integral

$$\int_0^{10^5} e^{-h\,mv^2}e^{-\alpha/v}v^2dv$$

were integrated by plotting curves for $\alpha = 10^4$, 2×10^4, 3×10^4, 4×10^4, 5×10^4, 7×10^4, 10^5, 1.5×10^5, 2×10^5, 3×10^5 and integrating with a planimeter. The value of the integral was then plotted as a function of α—Figs. 9 and 10—and

the value of X, 0.46 cm—was taken from the photometer curves—Fig. 8. This made τ the only variable of the equation for the position of the maximum.

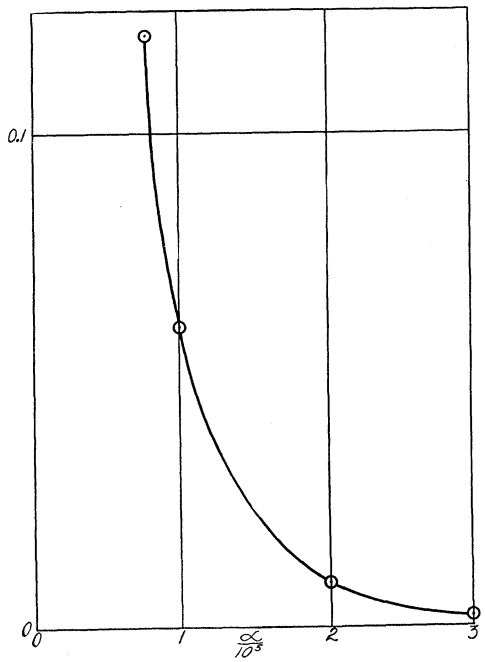

Fig. 10.

$$\frac{\int_0^{10^5} e^{-hmv^2} e^{-\alpha/v} v^2 dv}{\int_0^{\infty} e^{-hmv^2} v^2 dv}$$

plotted against $\alpha/10^5$.

The mean life was then determined by graphing

$$Y = \tau \int_0^{\infty} e^{-h\,mv^2} v^2 dv - \int_0^{\infty} \frac{e^{-h\,mv^2} e^{-x/v\tau}}{1.15} v dv$$

$$- \tau \int_0^{\infty} e^{-h\,mv^2} e^{-x/v\tau} v^2 dv$$

as a function of τ. The zero value of this function gave the desired value of τ, being $\tau = 2.5 \times 10^{-6}$ seconds.

In conclusion the authors wish to acknowledge an obligation to Professor Tate of the University of Minnesota who placed the microphotometer used in this work at their disposal.

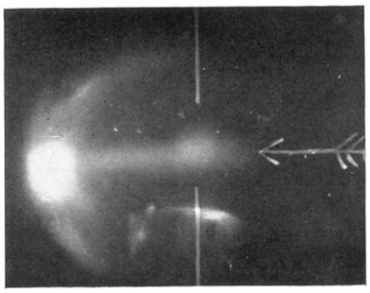

Fig. 3. Print from plate taken in preliminary run.

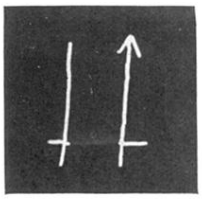

Fig. 5. Print of plate taken for first method of determining mean life.

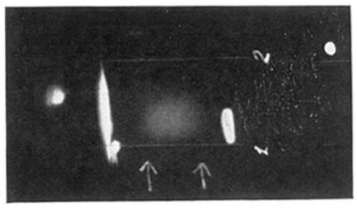

Fig. 7. Print of plate taken for second method of determining mean life.