ATTEMPT TO OBSERVE THE SPECTRUM OF DOUBLY EXCITED HELIUM

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ABSTRACT

An attempt to observe the spectrum of doubly excited helium by excitation by electron impact, with energies between 300-600 electron volts, is made without success. This and other failures are discussed by considering of the life times of the doubly excited states. An approximate calculation gives for $2s\,3s\,^3S$ a life time of the order 10^{-15} sec, corresponding to a natural width of the level of the order 1000 cm^{-1}. While this accounts for the absence of a line spectrum of doubly excited helium, it renders the former suggestion that the corona lines may be due to it untenable.

RECENTLY there have been various attempts(1) to calculate theoretically the energy states of helium atom with both of the electrons excited. These calculations are of some interest because i) they furnish knowledge of the approximate positions of doubly excited helium and ii) the spectrum of doubly excited helium might account for the well known corona lines whose origin remains as yet unexplained. On the basis of the unusually short life times that one would expect for these doubly excited states, Goudsmit and Wu(2) suggested that the corona lines, which are rather diffuse, may be due to such highly excited states as the doubly excited states of helium. An attempt to observe the spectrum of doubly excited helium in the laboratory has been made by Rosenthal(3) but without success. On the other hand, the lines at 320.4 Å and 357.5 Å

(1) Fender and Vinti, Phys. Rev. **46,** 77 (1934); Wu, Phys. Rev. **46,** 239 (1934); Wu and Ma, Phys. Rev. **48,** 917 (1935); Wu and Ma, Jour. Chinese Chem. Soc. **4,** 344 (1936); Wilson, Phys. Rev. **48,** 536 (1935).

(2) Goudsmit and Wu, Astrophys. Jour. **80,** 154 (1934); also Rosenthal, Z. f. Astrophysik **1,** 115 (1930).

(3) Rosenthal, Z. f. Phys. **48,** 794 (1933)

observed in helium by Kruger[4] and Compton and Boyce[5] are explained as arising from transitions between doubly excited states and singly excited states. From the magnetic spectrum of electrons scattered by helium atoms, Priestley and Whittington[6] found definitely electrons whose energy losses during collisions with helium atoms were 59.25 and 62.27 volts. These energy losses have been explained as due to excitation of both electrons. Hence it seems that under favorable conditions, there is still a possibility of observing the spectrum of doubly excited helium.

Massey and Mohr[7] recently calculated the probabilities of single and double electron excitation in helium by electron impact for different electron energies. It is found that while the probabilities of double electron excitations are in general very much smaller than, of the order of one hundredth of, those of single electron excitations, they have their greatest values for electrons possessing energies of about 300-400 electron volts. Thus failures to observe the spectrum of doubly excited helium may be due to the insufficiency of the electron energy under ordinary discharge conditions in which the electron energy is limited by collisions with helium atoms at relatively high pressure. In the following work, an attempt to produce the spectrum of doubly excited helium is made by direct electron impact, with electrons of energies between 300-600 electron volts.

Experimental

The best conditions for the excitation of the spectrum of doubly excited helium are *i*) high electron energies which can be obtained by running a discharge under low gas pressure, *ii*) high electron density which can be attained by furnishing a high electron current by means of a hot cathode. For this purpose, a discharge tube as shown in Fig. 1 was constructed.

(4) Kruger, Phys. Rev, **36,** 855 (1930)

(5) Compton and Boyce, Jour. Frank. Inst. **205,** 497 (1928)

(6) Priestley and Whittington, Proc. Roy. Soc. **A 145,** 462 (1934); Proc. Leeds. Phil. Soc. **3,** 81 (1935)

(7) Massey and Mohr, Proc. Camb. Phil. Soc. **31,** 604 (1935)

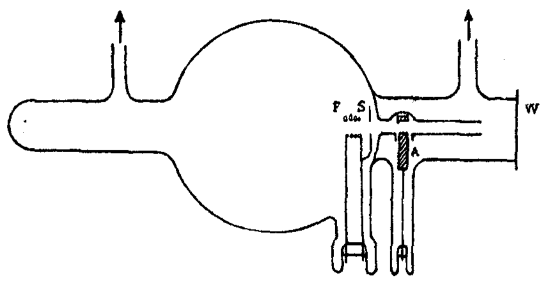

Fig. 1 Discharge tube

F—Filament cathode A—Anode
S—Shield W—Quartz window

Electrons were emitted by a hot cathode consisting of an oxide coated spiral tungsten filament. These electrons were accelerated in the direction of the axis of the spiral filament to a nickel anode at a distance of 2.5 cm., which was bored with a hole 3 mm. in diameter so that excitation can take place between the cathode and the anode and beyond the anode. A tube of 3 mm. bore extended from the hollow anode to confine the excitation process within it in order to increase the intensity of the radiation, which was observed end-on in the direction of the accelerating field. A nickel diaphragm in front of the filament prevented the light of the filament from entering the spectrograph.

Spectroscopic helium furnished by Hilger was admitted into the vacuum system after the system was baked out. The gas was circulated by means of a mercury pump through the discharge tube and two charcoal traps immersed in a mixture of solid carbon dioxide and acetone. After running the discharge for some time, the system was "cleaned up", as shown by the absence of even the strongest lines of such impurities as CO, NO. The pressure of helium was kept within 0.07 and 0.12 mm. of mercury so that the electron mean free path was of the order of 1 cm. The accelerating voltage was varied between 1,000 and 1,600 volts. The electron energies on impact with the helium atoms were of the order of 300-600 volts. By varying the filament current, the tube current could be varied from 20 to 100 milliamperes. Exposures, however, were made with a tube current of about 40 milliamperes so that the filament was not too bright to produce fogging of the photographic plate.

The strongest lines of the spectrum of doubly excited helium may be expected to arise from transitions between the lower states. Their positions may be estimated from the values of energy states obtained by the variational method[8]. Table I gives some of the lower states and the possible transitions.

Table I.

Calculated Energy States and Spectrum of Doubly Excited Helium.

State	Energy in R	Transitions	cm^{-1}
$2s2p\ ^3P$	-1.5043	$2s^2\ ^1S$-$2s2p\ ^1P$	15,800
$2s^2\ ^1S$	-1.4440	$2s2p\ ^1P$-$2s3s\ ^1S$	17,100
$2p^2\ ^3P$	-1.3976	$2s2p\ ^1P$-$2p^2\ ^1S$	9,100
$2p^2\ ^1D$	-1.326		
$2s2p\ ^1P$	-1.3000	$2p^2\ ^1D$-$2p3p\ ^1P$	22,300
$2p^2\ ^1S$	-1.217	$2p^2\ ^1S$-$2s3p\ ^1P$	10,320
$2s3s\ ^3S$	-1.163	$2s2p\ ^1P$-$2s3d\ ^1D$	20,770
$2s3s\ ^1S$	-1.144		
$2s3p\ ^3P$	-1.132	$2s2p\ ^3P$-$2p^2\ ^3P$	11,700
$2s3p\ ^1P$	-1.123	$2s2p\ ^3P$-$2s3s\ ^3S$	37,400
$2s3d\ ^3D$	-1.114	$2s2p\ ^3P$-$2s3d\ ^3D$	43,050
$2s3d\ ^1D$	-1.111	$2p^2\ ^3P$-$2s3p\ ^3P$	30,180

Of these the lines $2s^2\ ^1S$-$2s2p\ ^1P$, $2s2p\ ^1P$-$2s3s\ ^1S$ and $2s2p\ ^3P$-$2s3s\ ^3S$ approximately at 6320 Å, 5840 Å and 2670 Å are probably the strongest. These lines were looked for, the spectrum of the discharge tube being taken with a Hilger constant deviation glass spectrograph and a Hilger small quartz spectrograph. Exposures were limited to within ten hours because of the excessive sputtering and evaporation of the filament cathode. They failed to reveal any faint lines at the expected positions, although they were about 500 times longer than what would be necessary to photograph the lines of the ordinary line spectrum of helium.

(8) Wu and Ma, Jour. Chinese Chem. Soc. **4**, 344 (1936)

Discussion

While the failure to observe the transitions between doubly excited states may still be due to inappropriate conditions of excitation or insufficient length of exposure time, it seems that there may be more profound reason for their escaping observation, such as the unusually short life times, and consequently great natural widths, of these states. The life time of a doubly excited state is given by $\tau = \frac{1}{\Sigma P}$ where ΣP is the sum of the probabilities of (1) transitions to other doubly excited states, (2) transitions to singly excited states and (3) auto-ionization. The contributions due to these processes have been recently calculated by Kreisler[9] who found that while the probabilities of the processes (1) and (2) are of the same order of magnitude, uamely, 10^8/sec, the probability of autoionization is of the order 10^{14}/sec. Thus the life time is mainly determined by the process of autoionization. While Kreisler's calculation is straight forward, it is thought desirable to have an independent check of the probability of autoionization, as this seems to be the only case in which theoretical calculation has been carried out.

Consider the state $2s3s^3S$ which is in a position to undergo autoionization as it has the same parity as the continuum corresponding to the limit of $1sns^3S$. The wave function of $2s3s^3S$ is

$$\Psi(2s, 3s) = \frac{1}{\sqrt{2(1-c^2)}} [\psi(2s, \alpha r_1)\, \psi(3s, \beta r_2) - \psi(2s, \alpha r_2)\, \psi(3s, \beta r_1)]$$

where

$$\psi(2s, \alpha r) = \sqrt{\frac{\alpha^3}{\pi}}\, (1 - \alpha r)\, e^{-\alpha r}$$

$$\psi(3s, \beta r) = \frac{1}{3}\sqrt{\frac{\beta^3}{\pi}}\, (3 - 6\beta r + 2\beta^2 r^2)\, e^{-\beta r}$$

and

$$c = \int \psi(2s, \alpha r)\, \psi(3s, \beta r)\, d\tau$$

and the wave function for the continuum is

(9) Kreisler, Phys. Acta Polonica **4**, 1-2, 151 (1935)

$$\Psi(1s,E) = \frac{1}{\sqrt{2}} [\psi(1s, r_1)\,\psi(E, r_2) - \psi(1s, r_2)\,\psi(E, r_1)]$$

where

$$\psi(1s) = \sqrt{\frac{8}{\pi}}\, e^{-2r},$$

and for the ejected electron, the spherical wave employed by Wentzel[10] for the photoelectric effect was used instead of the continuous wave function of hydrogen employed by Kreisler, *i.e.*, in ordinary units,

$$\psi(E, r) = \sqrt{\frac{2}{\pi}\frac{dK}{dE}} \cdot \frac{\cos(Kr - \gamma)}{r},$$

where

$$K = \frac{2\pi}{h}\sqrt{2mE}\,.$$

The parameters α, β have been determined by the variational method[8] to be

$$\alpha = 1.03, \qquad \beta = 0.47.$$

and hence $C = 0.248.$

The probability of autoionization, i. e., a radiationless transition into the continuum, can be calculated by the method of variation of constants of Dirac to be

$$P = \frac{4\pi^2}{h^2} |V|^2$$

where

$$V = e^2 \int \Psi^*(2s, 3s) \frac{1}{r_{12}} \Psi(1s, E)\, d\tau_1\, d\tau_2$$

The energy E is given by[8]

$$E(2s3s\,^3S) - E(1s^2S) = 2.837$$

The constant γ is so determined that the continuous wave function is finite at the origin, i. e., $\gamma = -\frac{\pi}{2}$. Evaluation of the interaction integral can be carried out in a straight forward manner. The result is

(10) Wentzel. Z. f. Phys. **43,** 524 (1927)

$$P = 1.5 \cdot 10^{15} \text{ sec}^{-1}$$

Because of the approximate nature of the wave functions and also because of the fact that the result is obtained as the difference between terms that are nearly equal, this value does not claim to be more than an estimate of the order of magnitude. Although the agreement with Kreisler's value is not very good, it is seen from both calculations that P is probably of the order of 10^{+15}-10^{+14} sec^{-1}. The life time would then be of the order 10^{-15}-10^{-14} sec., and the uncertainty ΔE in the energy $\frac{h}{2\pi\tau}$ or 500-5000 cm^{-1}. The natural widths of other doubly excited states are certainly of the same order of magnitude. Transitions such as $2s^2\,{}^1S$-$2s2p\,{}^1P$, $2s2p\,{}^1P$-$2s3s\,{}^1S$ would give rise to continuous bands a few hundred Angstroms wide. Thus one would not expect to observe a "line" spectrum in the visible region. Also the suggestion that the corona lines may be due to doubly excited helium seems to be untenable, on the basis of this estimation of the life times of the states.

The lines observed by Kruger(4) and Compton and Boyce(5) at 320.4 Å and 357.5 Å, if they arise from transitions between doubly and singly excited states, would according to this calculation have a natural width of 0.5-5 Å, corresponding to the limits 10^{-15}-10^{-14}sec. for the life times. While the observed widths were not mentioned in their papers, they seem to be as sharp as other helium lines from the reproductions of their spectrograms. It is impossible to say whether the calculations of Kreisler and ours are incorrect or the explanation of the lines 320.4 Å and 357.5 Å must be sought elsewhere. While we are waiting for a more accurate calculation of the life time of doubly excited states, an another attempt is now being made to observe the transitions between doubly excited states under the same conditions in which the lines 320.4 Å and 357.5 Å were observed by Kruger, i.e., from a glow discharge in helium at low pressure in a Paschen hollow cathode.

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(11) For example, the Dirac formula for the transition probability may not be applicable in this case, since the value of the interaction matrix V is large.