Low States of the Heaviest Elements

With the aid of Fermi's potential for positive ions,[1] the energies of the $5f$, $6d$, $7s$ and $7p$ states of the atoms and ions built upon the radon core have been calculated from Schrödinger's equation by the approximation method of Wentzel-Kramers-Brillouin. It leads to the "phase integral" of the classical quantum theory with half-integer quantum numbers, except for the f states of uranium and singly ionized uranium (and Ac, Ac^+, Th, Th^+, Pa, Pa^+), in which cases the eigenvalues are given by an expression obtained from the quantum mechanical problem of two unequal potential minima.[2] The energy ϵ has to satisfy the relation

$$\int_{x_1}^{x_2} d\rho(\epsilon-v)^{\frac{1}{2}} = (n-l-\tfrac{1}{2})\pi - \tan^{-1}\left[\tfrac{1}{4}\exp\left\{-2\int_{x_2}^{x_3} d\rho(v-\epsilon)^{\frac{1}{2}}\right\}\tan\int_{x_3}^{x_4} d\rho(\epsilon-v)^{\frac{1}{2}}\right]$$

where $x_1<x<x_2$, $x_3<x<x_4$ are the two regions of possible classical motion and $x_2<x<x_3$ is the region under the potential hill.

The table below gives the results of the calculation of the energies in terms of the Rydberg constant as unity.

	U^{+++++}	U^{++++}	U^{+++}	U^{++}	U^{+}	U
$5f$	−3.22	−2.35	−1.45	−0.74	−0.256	−0.0625
$6d$	−3.15	−2.40	−1.70	−1.10	−0.55	−0.14
$7s$			−1.38	−0.90	−0.54	−0.20
$7p$			−1.30	−0.85	−0.47	−0.165

Calculations show that the energies of the different states depend chiefly on the degree of ionization and change very little with change of nuclear charge from $Z=92$ to 89. Hence the relative position of the levels in the uranium ions will be very similar to that for the corresponding ions of Pa, Th and Ac. The calculations involved are only approximations but nevertheless they give information as to which configurations can be expected to form the lowest states of the four heaviest elements; they are

89	Ac	Radon core +	$6d^2$	$7s$
			$6d$	$7s^2$
90	Th	Radon core +	$6d^3$	$7s$
			$6d^2$	$7s^2$
91	Pa	Radon core +	$6d^4$	$7s$
			$6d^3$	$7s^2$
92	U	Radon core +	$5f$	$6d^4$ $7s$
			$5f$	$6d^3$ $7s^2$
				$6d^5$ $7s$
				$6d^4$ $7s^2$

The interesting point is that with U the $5f$ state begins to be prominent, in contradiction with the older calculations of Sugiura and Urey.[3] Because of the approximate nature of the calculations it is not certain whether the lowest state of uranium already contains a $5f$ electron. However, beginning with element 93 we have to consider the six-fold ionized atom to obtain the ion with the radon core and in this ion the $5f$ is certainly the lowest level. This makes it very probable that the neutral atom 93 in its normal state contains at least one $5f$ electron. This points to the possible beginning of a series of elements similar to the rare earths in their relation to the periodic table, though the outer electron configurations and thus the chemical properties of these hypothetical elements will be different from those of the rare earths.

It is obvious that the results concerning the low states of the heaviest elements have to be confirmed by the analysis of the spectra of their ions, which has been started in this laboratory.

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[2] This has also been applied to the calculation of the quantum defects of states of heavy elements and shows why the quantum defects of penetrating orbits are in several cases near to integers. Details will be published shortly.

[3] Y. Sugiura and H. C. Urey, Kgl. Danske Vid. Selsk. **7**, 13 (1926).

The Separation of Hydrogen Isotopes by Fractional Desorption

Eyring[1] has recently emphasized the significance of zero point energy in the separation of hydrogen isotopes and has linked this with the observation of Washburn and Urey[2] on the separation by electrolysis and with the concept of activated adsorption and desorption of gases on surfaces developed by Taylor.[3] This theoretical approach lent especial importance to an experimental study of the possible separation of hydrogen isotopes by desorption from surfaces on which they are adsorbed with an activation energy. The experiments of Taylor and Sherman[4] on the para-hydrogen conversion at surfaces as a criterion of activated adsorption of hydrogen suggested at once that active charcoal surfaces would be the best for such experiments. Accordingly we have examined the desorption of electrolytic hydrogen, purified by passage over platinized asbestos and suitable drying agents, from an active adsorbent charcoal maintained at liquid air temperatures. We have examined, by the method developed by Bleakney,[5] the final samples of hydrogen desorbed from charcoal under these circumstances. A one stage desorption process yielded

a final sample of 35 cc from an initial adsorption of 5025 cc. This revealed a three-fold enrichment of H^1H^2. A two stage desorption was then made in which, from 5715 cc adsorbed, an end desorption was secured amounting to 440 cc, which was then readsorbed and fractionally desorbed to give a final sample of 90 cc. This sample showed a five-fold enrichment of H^1H^2 over that present in the hydrogen after passing the purification train. While this enrichment may not be as large as that to be expected theoretically on the basis of zero-point energy and activated adsorption, it appears to be much larger than would be expected from a simple distillation process of separation. Since, theoretically, this method of separation by desorption involving zero-point energy will be enormously more efficient at the temperature of liquid hydrogen we are taking steps to extend our experiments to these lower temperature ranges.

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[1] H. Eyring, Proc. Nat. Acad. Sci. **19**, 78 (1933).
[2] E. W. Washburn and H. C. Urey, Proc. Nat. Acad. Sci. **18**, 496 (1932).
[3] H. S. Taylor, J. Am. Chem. Soc. **53**, 578 (1931).
[4] H. S. Taylor and A. Sherman, Trans. Faraday Soc. **28**, 247 (1932).
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Scattering and Absorption of Neutrons

In these experiments the neutron source consisted of beryllium powder in one or more glass bulbs 4 mm diameter filled with radon, in amounts up to 1200 millicuries. Alpha-radiation equivalent to 3600 millicuries of polonium was thus utilized, making a much more powerful neutron source than hitherto reported, although de Broglie, le Prince-Ringuet, Thibaud and La Tour used a radon-beryllium source without stating the amount of radon.[1] Neutrons were ejected from the beryllium by the alpha-particles from radon, radium *A* and radium *C'*, to a number around 10^5 per second. To measure the intensity of the neutron stream a small ionization chamber was used, connected to an amplifier-oscillograph system described previously, which registered ionizations by single high-speed particles above the unavoidable background of gamma-radiation.[2] The ionizing particles were atoms projected from the paraffin front and the other materials of the chamber by neutron impact.

For the scattering measurements, annular ring scatterers were used, the direct beam being largely blocked out of the chamber by a cylinder of lead 19 cm long, and the scattering, measured by the increase in the number of neutrons registered with the scattering ring in place, was determined for positions giving several scattering angles.

TABLE I.

Material	*Angle of Scattering*				
	46±15	51±20	82±27	125±25	151±15
Paraffin	3.68	3.65	2.18	1.48	0.21
Water	3.59	3.45	2.50	1.48	0.56
Carbon	2.96	2.17	2.01	1.62	0.79
Lead	2.75	2.52	2.49	1.71	0.58

Table I shows the relative scattering by rings 15.5 cm mean radius and square cross section of 25 cm^2 in numbers of neutrons registering in ionization chamber per minute, the distance from source to chamber being 31.5 cm.

As might be expected, paraffin and water, containing much hydrogen, show small backward scattering, no more than would be indicated for the carbon or oxygen contained, while carbon and lead show more nearly uniform scattering.

The computation of the scattering coefficients requires approximations as to geometrical quantities and absorption. The following figures are probably relatively correct to within ±15 percent: fraction scattered (per unit volume of scatterer) from an incident beam of unit intensity into unit solid angle in the specified direction: lead 2.28×10^{-2} at 45°, 1.2×10^{-2} at 125°; carbon 2.12×10^{-2} at 45°, 0.96×10^{-2} at 125°.

Since the absorption of neutrons is largely scattering, the greatest absorption will be shown by an absorber just filling the geometrical path, because any material outside the direct beam scatters neutrons back into the chamber. For example, paraffin in the form of a cylinder 3 cm diameter and 4 cm long shows twice as much absorption as a large plate of paraffin 4 cm thick. Absorbers 3 cm in diameter were uniformly used in this work. Absorption curves were taken for carbon, lead, water and paraffin, the neutron beam having been filtered through 4 cm of lead to reduce the gamma-radiation. These curves are approximately exponential, and appear to approach a minimum which is probably the residual scattering from the room. A study was also made of the absorption by cylinders, 4 cm long, of various materials, as shown in Fig. 1. Copper has the highest absorption per cm of any material yet tested.

As to correlation of the atomic absorption with atomic weight, the simple hypothesis of elastic sphere collision between the neutron and the nucleus, with a definite radius for the neutron and a radius for the nucleus proportional to the cube root of its atomic weight, can be made to fit the absorption results very well, a radius of 4.6×10^{-13} cm being required for the neutron and one of 7.8×10^{-13} cm