

A Hydrogen Isotope of Mass 2

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The proton-electron plot of known atomic nuclei shows some rather marked regularities among atoms of lower atomic number¹. Up to O¹⁶ a simple step-wise figure appears into which the nuclear species H², H³ and He⁶ could be fitted very nicely. Birge and Menzel² have shown that the discrepancy between the chemical atomic weight of hydrogen and Aston's value by the mass spectrograph could be accounted for by the assumption of a hydrogen isotope of mass 2 present to the extent of 1 part in 4500 parts of hydrogen of mass 1.

It is possible to calculate with confidence the vapor pressures of the pure substances H¹H¹, H¹H², H¹H³, in equilibrium with the pure solid phases. It is only necessary to assume that in the Debye theory of the solid state, Θ is inversely proportional to the square root of the masses of these molecules and that the rotational and vibrational energies of the molecules do not change in the process of vaporization. These assumptions are in accord with well-established experimental evidence. We find that the vapor pressures for these three molecules in equilibrium with their solids should be in the ratio of $p_{11} : p_{12} : p_{13} = 1 : 0.37 : 0.29$. The theory of the liquid state is not so well understood but it seems reasonable to believe that the differences in vapor pressure of these molecules in equilibrium with their liquids should be rather large and should make possible a rapid concentration of the heavier isotopes, if they exist, in the residue from the simple evaporation of liquid hydrogen near its triple point.

Accordingly two samples of hydrogen were prepared by evaporating large quantities of liquid hydrogen and collecting the gas which evaporated from the last fraction of the last cubic centimeter. The first sample was collected

¹Urey, J. Am. Chem. Soc. **53**, 2872 (1931); Johnston, *ibid.*, **53**, 2866 (1931).

²Birge and Menzel, Phys. Rev. **37**, 1669 (1931).

from the end portion of six liters of liquid evaporated at atmospheric pressure, and the second sample from four liters evaporated at a pressure only a few millimeters above the triple point. The process of liquefaction has probably no effect in changing the concentration of the isotopes since no appreciable change was observed in the sample evaporated at atmospheric pressure.

These samples were investigated for the atomic spectra of H^2 and H^3 in a hydrogen discharge tube run in Wood's so-called "black stage" by using the second order of a 21 foot grating with a dispersion of 1.31A per mm. With the sample evaporated at the boiling point no concentration so high as had been estimated was detected. We then increased the exposures so that the ratio of the time of exposure to the minimum required to get the H^1 lines on our plates was about 4500 : 1. Under these conditions we found in this sample as well as in ordinary hydrogen faint lines at the calculated positions for the lines of H^2 accompanying $H_\beta, H_\gamma, H_\delta$. These lines do not agree in wave-length with any molecular lines reported in the literature³. However they were so weak that it was difficult to be sure that they were not ghosts of the strongly overexposed atomic lines.

The sample of hydrogen evaporated near the triple point shows these lines greatly enhanced, relative to the lines of H^1 , over both those of ordinary hydrogen and of the first sample. The relative intensities can be judged by the number and intensity of the symmetrical ghosts on the plates. The wave-lengths of the H^2 lines appearing on these plates could be easily measured within about 0.02A. The following table gives the mean of the observed displacements of these lines from those of H^1 and the calculated displacements:

Line	H_α	H_β	H_γ	H_δ
$\Delta\lambda$ calc.	1.793	1.326	1.185	1.119
$\Delta\lambda$ obs.				
Ordinary hydrogen	—	1.346	1.206	1.145
1st sample	—	1.330	1.199	1.103
2nd sample	1.820	1.315	1.176	—

The H^2 lines are broad, as is to be expected for close unresolved doublets, but they are not as broad and diffuse as the H^1 lines probably due to the smaller Döppler broadening. Although their intensities relative to the ghosts

³Gale, Monk and Lee, *Astrophys. J.* **57**, 89 (1928); Finkelburg, *Z. Physik* **52**, 57 (1928); Connelly, *Proc. Phys. Soc.* **42**, 28 (1929).

of the respective H^1 lines appear nearly constant for any one sample of hydrogen, they are not ghosts for their intensities relative to the known ghosts are not the same in the case of ordinary hydrogen and of the 1st sample as they are in the case of the second sample. They are not molecular lines for they do not appear on a plate taken with the discharge tube in the "white stage" with the molecular spectrum enhanced ($H^2\gamma$ was found as a slight irregularity on a microphotometer curve of this plate). Finally the $H^2\alpha$, line is resolved into a doublet with a separation of about 0.16A in agreement with the observed separation of the $H^1\alpha$ line.

The relative abundance in ordinary hydrogen, judging from relative minimum exposure time is about 1 : 4000, or less, in agreement with Birge and Menzel's estimate. A similar estimate of the abundance in the second sample indicated a concentration of about 1 in 800. Thus an appreciable fractionation has been secured as expected from theory⁴.

No evidence for H^2 has been secured, but its lines would fall on regions of our plates where the halation is bad.

The distillation was carried out at the Bureau of Standards by one of us (F.G.B.), who is continuing the fractionation to secure more highly concentrated samples. The spectroscopic work was done at Columbia University by the other two (H.C.U. and G.M.M.) who are working on the molecular spectrum.

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⁴Keesom and van Dijk, Proc. Acad. Sci. Amsterdam **34**, 52 (1931).