PRECISION DETERMINATION OF THE HYPERFINE STRUCTURE OF THE GROUND STATE OF ATOMIC HYDROGEN, DEUTERIUM, AND TRITIUM*

L. Wilmer Anderson† and Francis M. Pipkin‡
Lyman Laboratory, Harvard University, Cambridge, Massachusetts

and

James C. Baird, Jr.
Mallinckrodt Laboratory, Harvard University, Cambridge, Massachusetts

(Received November 12, 1959)

The hyperfine splittings of the hydrogen isotopes have been measured by an optical polarization method.[1-3] In this experiment a small quantity of one of the hydrogen isotopes was placed in a 500-cm^3 spherical flask together with a buffer gas and rubidium metal. Rubidium resonance radiation produced in an electrodeless radio-frequency light source was circularly polarized, passed through an interference filter to select the D_1 line, and then used to polarize the rubidium. A pulsed 30-Mc/sec oscillator was used to dissociate some of the hydrogen molecules into atomic hydrogen. This atomic hydrogen became oriented by spin exchange collisions with the polarized rubidium. The polarization of the rubidium was monitored by observing the transmitted resonance radiation with a 935 phototube. When a radio-frequency field was applied so as to destroy part of the hydrogen polarization, there was a decrease in the amount of light incident upon the photocell.

The following system was used to generate and measure precisely the frequencies for observing various transitions. The 1-Mc/sec output of an Atomichron, operated and standardized by J. A. Pierce of the Cruft Laboratory, was multiplied in a Gertsch AM-1 and used to phase lock the Gertsch FM-4 at a high frequency. The internal low-frequency oscillator of the AM-1 was not used. A T-116/APT-5A radar jamming oscillator was used to drive the hyperfine transitions. An auxiliary output of this oscillator was beat with the output of the Gertsch FM-4 obtaining a beat frequency between 3 and 4 Mc/sec. This frequency was amplified and compared in a phase-detecting circuit with a variable frequency General Radio 616D oscillator, which was very stable. This beat frequency was measured with a Northeastern Engineering 14-20 electronic counter, whose 100-kc/sec time base was supplied by the Atomichron. The output of the phase detection circuit was used to phase lock the APT-5. A relay was used to square-wave modulate the output of the APT-5 which induced the radio-frequency transitions. The output of the photocell was amplified and fed to a lock-in detector with an output time constant of 0.1 sec.

The field-independent 0 → 0 transition in hydrogen and tritium could not be detected in this fashion. This is probably due to the manner in which the spin exchange collisions populate the magnetic sublevels. For these isotopes it was necessary to measure the two field-dependent hyperfine transitions ($F=0$, $m=0 \rightarrow F=1$, $m=\pm 1$) and to average the results to obtain a field-independent quantity. A typical line profile for hydrogen is shown in Fig. 1. Each of the field-dependent hyperfine lines is asymmetrical; this asymmetry, however, occurs on the high-frequency side for one line and on the low-frequency side for the other line. It is assumed in this experiment that when the frequencies of the two lines are averaged, the effect of the asymmetry cancels out. All of the measurements on these

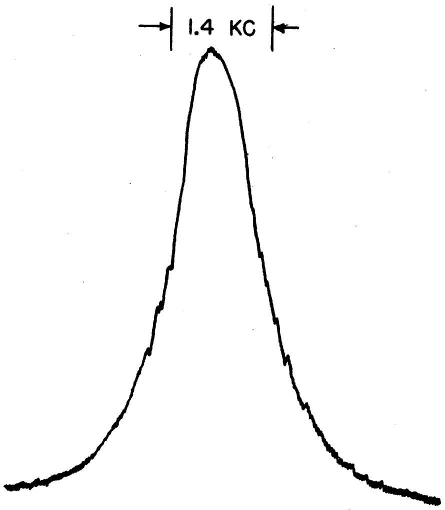

FIG. 1. Typical line profile for the ($F=0$, $m=0$ $\rightarrow F=1$, $m=1$) hyperfine transition of atomic hydrogen in a molecular hydrogen buffer gas. The lock-in-detector time constant was 0.1 sec.

two isotopes were made between the hours of 1 and 5 a.m. when the magnetic activity (i.e., trolleys) in the neighborhood was a minimum. In deuterium, however, there are two transitions ($F=3/2$, $m=1/2 \rightarrow F=1/2$, $m=-1/2$) and ($F=3/2$, $m=-1/2 \rightarrow F=1/2$, $m=1/2$) which are almost field independent (Fig. 2). These two transitions differ by twice the product of the nuclear moment and the field. These two lines could easily be resolved in the field of 0.15 gauss which was used in the experiment and had a linewidth of about 175 cycles/sec. This residual width was produced by exchange collisions between the deuterium atoms. To determine the hyperfine splitting of deuterium, both these lines were measured and the Zeeman transitions ($\Delta F=0$) were used to compute the second order correction.

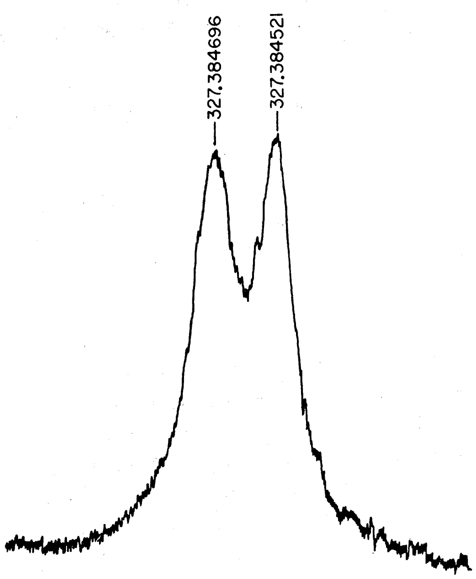

FIG. 2. Typical line profile for the ($F=1/2$, $m=-1/2 \rightarrow F=3/2$, $m=1/2$) and ($F=1/2$, $m=1/2 \rightarrow F=3/2$, $m=-1/2$) hyperfine transitions of atomic deuterium in a neon buffer gas. The separation of the two peaks is equal to twice the nuclear resonance frequency for the deuteron in a field of 0.15 gauss. The markers were made with only one direction of frequency sweep and serve only to mark the separation of the two peaks and the linewidth. The lock-in-detector time constant was 0.1 sec.

Since the value of the hyperfine splitting observed depends upon the pressure and nature of the buffer gas used, several bulbs were made at various pressures for each gas. Each of these bulbs was then measured on a number of different nights. A least-squares fit was used to determine the best straight line through these points. In this manner the pressure shift for each gas could be determined and the measured hyperfine splitting could be extrapolated to zero pressure. Table I shows the pressure shift obtained for the various buffer gases. Table II shows the values of the hyperfine splitting obtained by extrapolating to zero pressure. In the case of deuterium, where the observed transitions are field independent, the errors appear to be purely statistical. The value obtained for the zero-field hyperfine splitting is

$$\Delta\nu(\mathrm{H}^2) = 327.384349 \pm 0.000005\ \mathrm{Mc/sec}.$$

In the case of hydrogen and tritium, where field-dependent lines are measured, the errors do not appear to be statistical. The amount by which

Table I. The pressure shift data. The pressure in each bulb was measured at room temperature with an oil manometer when the bulb was prepared; the measurement of the hyperfine splittings were made with the bulb at 50°C. In computing the pressure shifts at 50°C the perfect-gas law was used to correct the pressure. The error in the pressure shift is estimated to be about 20%.

Hydrogen isotope	Buffer gas	Pressure shift (cycles/sec mm Hg)	Pressure shift/$\Delta\nu$ (mm Hg)$^{-1} \times 10^9$
H^1	A	-3.59	-2.53
H^1	Ne	+1.62	+1.14
H^1	$H_2{}^1$	-0.24	-0.17
H^1	He	+2.83	+2.00
H^2	A	-0.63	-1.94
H^2	Ne	+0.44	+1.34
H^3	A	-1.98	-1.31
H^3	Ne	+2.99	+1.98

Table II. The measured hyperfine splitting obtained by extrapolating to zero pressure.

Hydrogen isotope	Buffer gas	Number of bulbs at different pressure	Number of observations per bulb	Hyperfine splitting in Mc/sec
H^1	A	4	40	1420.405712
H^1	Ne	8	20	754
H^1	H_2	4	40	721
H^1	He	4	10	698
Average				1420.405726
H^2	A	3	20	327.384352
H^2	Ne	4	15	347
Average				327.384349
H^3	A	3	40	1516.701382
H^3	Ne	3	40	410
Average				1516.701396

the lines determined for the various buffer gases fail to meet in a point at zero pressure has been taken as a measure of the error in the hyperfine splittings of these two isotopes. The results for hydrogen and tritium are

$$\Delta\nu(H^1) = 1420.405726 \pm 0.000030 \text{ Mc/sec},$$

$$\Delta\nu(H^3) = 1516.701396 \pm 0.000030 \text{ Mc/sec}.$$

The result for hydrogen agrees very well with the most recent atomic beam result of Kusch[4] and the paramagnetic resonance result of Wittke and Dicke[5] with no pressure shift correction. The value for deuterium is slightly outside the quoted error in the atomic beam measurement of Kusch.[4] The value for tritium disagrees with the atomic beam measurement of Prodell and Kusch.[6]

---

[*] This research was supported in part by a grant from the Research Corporation.

[†] National Science Foundation Predoctoral Fellow 1959-60.

[‡] Alfred P. Sloan Fellow 1959-61.

[1] H. G. Dehmelt, Phys. Rev. 109, 381 (1958).

[2] Anderson, Pipkin, and Baird, Phys. Rev. Letters 1, 228 (1958).

[3] R. Novick and W. W. Holloway, Bull. Am. Phys. Soc. 3, 371 (1958).

[4] P. Kusch, Phys. Rev. 100, 1188 (1955).

[5] J. P. Wittke and R. H. Dicke, Phys. Rev. 103, 620 (1956).

[6] A. G. Prodell and P. Kusch, Phys. Rev. 106, 87 (1958).

NUCLEAR RESONANCE ABSORPTION AND NUCLEAR ZEEMAN EFFECT IN Fe^{57}[†]

G. DePasquali, H. Frauenfelder, S. Margulies, and R. N. Peacock
University of Illinois, Urbana, Illinois
(Received December 14, 1959)

Mössbauer recently demonstrated that the 129-kev gamma ray in Ir^{191} underwent resonance absorption when the osmium source and iridium absorber were both at low temperatures.[1] The resulting absorption peak displayed the natural line shape.[2] Mössbauer explained this effect by adapting a theory of Lamb.[3] If the emitting atom is bound, at a low ambient temperature, in a solid of high Debye temperature, it cannot always recoil freely. In a certain fraction of the transitions, the recoil momentum is given to the entire solid. The emitted photon then suffers no Doppler shift, but possesses the full transition energy and can be resonantly absorbed or scattered by another nucleus of the same type, also bound in a solid. Mössbauer's experiments have been repeated and extended[4,5] and the theory has been refined.[6,7]