Magnetic Moment of the Triton in Units of the Magnetic Moment of the Proton*

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High-resolution nuclear magnetic resonance techniques were used to perform a precise measurement of the ratio of the Larmor frequencies of tritons and protons in a sample of 20% tritiated water. The measured ratio ω_T/ω_H is 1.066 639 75(2). From this frequency ratio and an estimate of the magnetic shielding correction, a value of 1.066 639 86(11) is obtained for the moment of the triton in units of the proton moment. From this value and the hfs measurements of Kusch, and of Prodell and Kusch, one obtains a hfs anomaly of $(-5.62\pm0.13)10^{-6}$ for tritium. This observed anomaly is compared with the theoretical predictions of Adams. The longitudinal relaxation time T_1 of the protons and tritons in the sample was measured and found to be 1.02 ± 0.10 sec. and 0.83 ± 0.10 sec., respectively.

THE triton moment in units of the proton moment was previously measured by Bloch, Graves, Packard, and Spence[1] and found to be 1.066 636(10). Kusch[2] measured the hfs splitting of the ground state of hydrogen with a precision of five parts in 10^8, and more recently Prodell and Kusch[3] measured the hyperfine splitting of tritium in the ground state with a precision of 7 parts in 10^8. The hyperfine anomaly in tritium may be computed if both the hfs splittings of tritium and hydrogen and the ratio of triton moment and proton moment are known. A calculation by Adams[4] using several different theoretical assumptions predicts an anomaly of the order of one part in 10^5. The previous moment measurement was not sufficiently precise to establish the presence of an anomaly of this magnitude. The work presented here was undertaken to arrive at a more accurate determination of the moment ratio by the techniques of high-resolution magnetic resonance, which were developed subsequent to the first measurement of this quantity.

For a precise determination of the moment ratio, both nuclear species were resonated in the same magnetic field and the ratio of Larmor frequencies corresponding to this field was measured. Field changes which may arise when samples are interchanged were avoided by using a single sample containing both species. A sealed Pyrex-glass vessel containing 20% tritiated water (H_2O, HTO, T_2O) was used.

The previous measurement of Bloch *et al.*[1] demonstrated that the ratio of Larmor frequencies, ω_T/ω_H, was 16/15 to within about four parts in 10^5. The apparatus for this experiment was designed to take advantage of that result. Frequencies in the exact ratio of 16/15 were easily generated by using two frequency-multiplier chains, both of which were excited by a common crystal oscillator. The output signals from the multipliers were then audio-frequency modulated. The frequencies of the sideband signals thus generated were adjusted simultaneously with the dc field until both nuclear species were resonating at a common field. By the use of this technique, the ratio of unmodulated radio frequencies remained fixed at 16/15, independent of oscillator drift. In addition, even a relatively inaccurate measurement of the frequency yielded a very accurate result for the moment ratio.

The large permanent magnet, designed and constructed by Arnold[5] at Stanford University, was used for the experiments. His method of using a gated rf system and thus a single transmitter-receiver coil[6] was likewise employed. The use of this system permits the construction of a relatively simple single-coil induction head, eliminates the problem of balancing out rf leakage which occurs in a crossed-coil head, and makes the detected signal independent of fluctuations in the transmitter rf level.

A quantity of tritium water of a dangerously large (200-curie) activity was required in order to obtain adequate signal-to-noise ratio for the triton signal. The triton decays by beta-emission to He^3 with a half-life of 12.5 years, emitting an electron with a maximum energy of 11 kv. For the purpose of this experiment the sample could remain sealed in a glass tube which adequately shielded this activity. If the tube should be broken, however, a lethal amount of tritium water vapor might be released in the laboratory. As a precautionary measure the Pyrex tube containing the sample was mounted between the pole faces of the magnet in a sealed copper box. The sample tube consisted of a $4\frac{1}{2}$-inch section of 0.200-inch o.d. containing the water, sealed onto a wider 4-inch upper section which allowed for gas pressure build-up of the hydrogen, hydrogen peroxide, and He^3 decay product produced in the water. The tube was inserted into an air turbine in order to obtain line-narrowing by rotation of the sample.[7,8] The single rf coil was wound on an acrylic-

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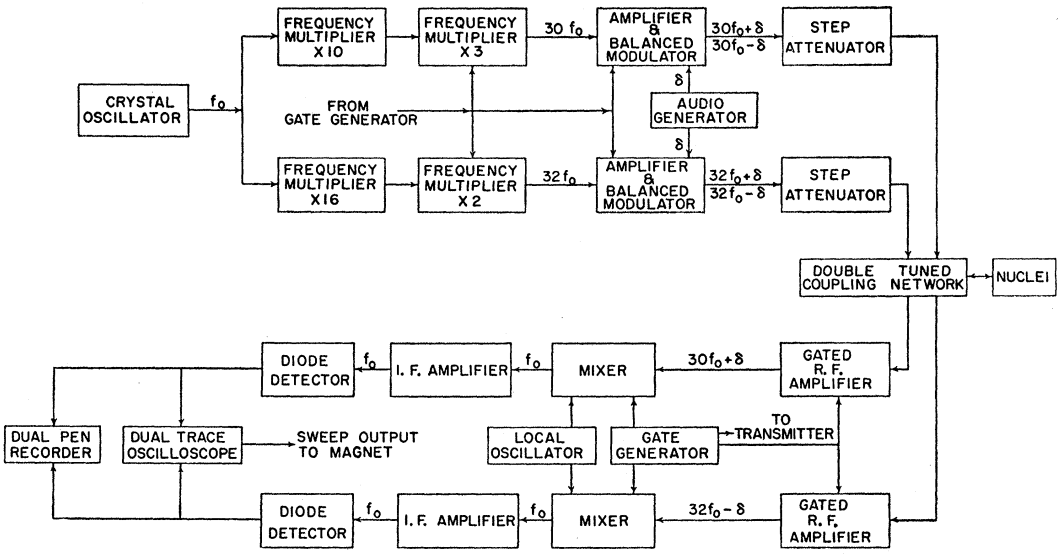

FIG. 1. Block diagram of the electronic system.

plastic cylinder concentric with the sample tube. The coil formed part of a double-tuned coupling network which was mounted in a shielded box immediately adjacent to the copper box. A $\frac{1}{4}$-hp diaphragm-type compressor, with a suitable air filter for damping out the pulsations in the air stream, provided a closed air system to run the turbine. This system was needed because of the danger of the presence of tritium water vapor in the air exhausted from the turbine, if the sample tube should break. An ionization chamber was inserted into the air line and the amplified output voltage from the chamber provided a continuous monitor on the presence of tritium water vapor in the closed system. The tritium water was provided by the Lawrence Radiation Laboratory at Livermore, and prepared by Dr. Henry Otsuki of that laboratory.

The electronic system (Fig. 1) consisted of a crystal-controlled, dual-frequency, gated, balanced-modulated transmitter and two fixed-frequency, gated superheterodyne receivers, as well as an appropriate gate generator, attenuators, oscilloscope, and recorder.

A 1-Mc/sec Meacham-bridge crystal oscillator provided a frequency source stable to better than 1 part in 10^8 over time intervals long compared to the time of an experimental run. The oscillator excited two conventional frequency-multiplier chains providing output signals at about 30 and 32 Mc/sec. These, in turn, excited amplifiers which were balanced-modulated at a variable audio frequency δ, providing output signals at about $(30\times10^6)\pm\delta$ and $(32\times10^6)\pm\delta$ cps. The final stages of both channels were gated. Two fixed-tuned rf amplifiers, each consisting of a Cascode input stage and a single pentode, provided the initial amplification for the two nuclear signals. Each rf amplifier was appropriately gated and the amplified nuclear signal was fed to a pentagrid-mixer tube driven with a local oscillator signal that was gated in synchronism with the rf gates. The mixers fed the rf amplifiers whose detected outputs were displayed on a dual-trace oscilloscope or a dual-pen recorder. Direct diode detection was used, giving a detected signal voltage proportional to $(u^2+v^2)^{\frac{1}{2}}$, where u and v represent the dispersive and absorptive contributions to the signal, respectively, which one would separately observe in a phase-sensitive detector. This method of detection was chosen in preference to phase-sensitive detection because of the uncertainty in the latter method with regard to complete elimination of the unwanted mode.

By using a wide field-sweep and a suitable value of the modulating audio-frequency, the two resonances, corresponding to the lower sideband of the higher frequency and the upper sideband of the lower frequency, were brought to coincidence. After optimizing field homogeneity over the sample as well as the sweep rate and the rf level, the line widths of both nuclear species were observed to be from 1 to 2 cps with a signal-to-noise ratio of at least 30 to one. Measurement of the audio frequency and the radio frequency with an electronic counter yielded the ratio of Larmor frequencies, using the relation

$$\frac{\omega_T}{\omega_H}=\frac{32\nu_0-\delta_0}{30\nu_0+\delta_0}\approx\frac{16}{15}-\frac{31}{450}\frac{\delta_0}{\nu_0}, \tag{1}$$

where δ_0 is the audio modulating frequency at which coincidence is observed, and ν_0 is the crystal oscillator frequency of about 1 Mc/sec. Quadratic and higher order terms in δ_0/ν_0 give a negligibly small contribution and are therefore omitted in this formula.

The measurements were recorded by a dual-channel pen recorder. The procedure consisted of sweeping through the resonance lines, putting a simultaneous

marker-voltage deflection on each pen shortly after passage through both lines, and noting the value of δ on the recorder for that run. Coincidence was indicated by a simultaneous occurrence of the recorded signals, while noncoincidence was observed if one signal preceded or followed the other. A number of such data were recorded for which δ was deliberately high or low or apparently at the correct value. The measured relative displacement of triton and proton resonance peaks was then plotted *versus* δ. From the graph, the audio frequency corresponding to coincidence was determined to have the value 397.15 ± 0.2 cps. The crystal oscillator frequency, ν_0, was measured and found to be 1.016 61(1) Mc/sec. If this frequency is multiplied by 30, using the known gyromagnetic ratio for the protons, this corresponds to a field of about 7160 gauss. Both frequencies were measured with an electronic counter. From Eq. (1), a Larmor frequency ratio of 1.066 639 75(2) was obtained.

Because of its intrinsic interest and in order to ascertain that relaxation affects the resonance of tritons and protons in a similar manner, the longitudinal relaxation time, T_1, was measured for each species. An adiabatic fast passage through the resonance line was employed to turn the macroscopic moment vector through 180°, and then the longitudinally decayed magnetization vector was sampled by rapidly sweeping back through the line, after a time delay of the order of T_1. The value of T_1 was obtained from a logarithmic plot of the amplitude of the second signal as a function of the delay time. It was found to be 1.02 ± 0.10 second for protons and 0.83 ± 0.10 second for tritons. The measured value for protons is less than the value of T_1 in ordinary distilled water by a factor of about 2.5. This reduction in relaxation time may have been caused by the presence of free radicals, produced by the beta activity in the tritiated water sample. The presence of paramagnetic impurities was ruled out since the sample was prepared by distillation.

While the ratio of the resonance frequencies, ω_T/ω_H, quoted above, was ascertained with an accuracy of about two parts in 10^8, it is not possible to obtain an equal accuracy for the ratio of nuclear moments since this would require that the magnetic shielding corrections were known for the two species. Only an estimate can be given for the relative correction.

The chemical shift for protons in water was found to be -2.7×10^{-5} by Gutowsky and Hoffman.[9] Although the shift for tritons is expected to be closely equal to this value, there exists the possibility of a small isotope effect. Such an effect can be expected on theoretical grounds by considering the difference in the vibration amplitudes of nonidentical hydrogen isotopes in the same molecular configuration. It may be estimated to give a correction which is quadratic in the vibration amplitudes, and of the order of magnitude of a part in 10^7. This expectation is supported by the measurements of Wimett,[10] who observed the difference in shielding for H_2 and D_2 gas to be given by $\sigma(D_2)-\sigma(H_2)=(0.65\pm0.59)\times10^{-7}$. A somewhat larger value would be expected for $\sigma(T_2)-\sigma(H_2)$ corresponding to the larger reduced mass of tritium. Unfortunately, there are at present no published data on the relative shielding of tritons and protons in water. The plausible assumption will be made here that $\sigma(HTO)-\sigma(H_2O)\approx\sigma(T_2)-\sigma(H_2)\approx(1\pm1)\times10^{-7}$, in view of Wimett's measurement. The uncertainty in this value represents the principal source of error in the measurement described here. The moment of the triton in units of the proton moment is thus quoted to be 1.066 639 86(11).

The hyperfine anomaly in tritium may be defined as the ratio of the difference between the splitting calculated from the relation of Fermi[11] for a point moment and the observed hfs splitting in the ground state, divided by the observed splitting. Using the value of Kusch[2] for the hydrogen hfs splitting, the value of Prodell and Kusch[3] for the tritium hfs splitting, and the above value for the triton moment in units of the proton moment, one obtains a hfs anomaly of $(-5.62\pm0.13)\times10^{-6}$. This value falls within the range of values calculated by Adams[4] on several theoretical assumptions. Only the number which he calculated on the basis of a phenomenological interaction-moment theory which excludes space exchange differs markedly from the experimental value. A more refined calculation would be required for a comparison of the above experimental value with the underlying assumptions of the theories.

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