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MEASUREMENTS OF THE HYDROGEN-DEUTERIUM ATOMIC MAGNETIC MOMENT RATIO AND OF THE DEUTERIUM HYPERFINE FREQUENCY*

D. J. Larson,† P. A. Valberg,‡ and N. F. Ramsey

Lyman Laboratory of Physics, Harvard University, Cambridge, Massachusetts 02138

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The ratio of the hydrogen atomic magnetic moment to that of deuterium has been measured by spin-exchange detection of Zeeman ($\Delta F=0$, $\Delta m_F=-1$) transitions in an atomic hydrogen maser. The value of this ratio is found to be 1.000 000 009 4(14) in agreement with a recent theoretical calculation. The spin-exchange technique has also been used to measure the zero-field ground-state deuterium hyperfine frequency. The result is $\Delta\nu(D) = 327\,384\,352.51(5)$ Hz.

Atomic magnetic-resonance experiments on stored particles¹⁻³ are characterized by their long interaction times and some degree of motional averaging. The resulting narrow resonance widths make possible experiments of unprecedented accuracy. The hydrogen maser^{4,5} is a device utilizing this storage principle, and its spectral purity and frequency stability are well known. In addition to its application as a time standard, the hydrogen maser can also be used for atomic spectroscopy.^{6,7}

It is the purpose of this note to report a high-precision measurement of the atomic magnetic moment of hydrogen relative to that of deuterium. A comparison with a recent theoretical calculation is made. Also reported is a new determination of the zero-field deuterium hyperfine frequency. This determination is more precise than the earlier measurement by Crampton, Robinson, Kleppner, and Ramsey⁶ using a similar technique.

The hydrogen maser was operated at 7.6 G on a field-dependent π transition. This transition is between levels whose population difference is directly related to the electron-spin polarization of the hydrogen atoms. If both atomic hydrogen

and atomic deuterium are present in the storage bottle of the maser, spin-exchange collisions⁸⁻¹⁰ couple the electron polarizations of the two systems. The spin-exchange process can induce transitions between hyperfine states; however, it conserves the z component of the total angular momentum. The two systems will come to a spin-exchange equilibrium which in the limit of high densities corresponds to the electron-spin polarization of each approaching the same value. Inducing transitions in the hyperfine spectrum of deuterium will disturb the equilibrium, and this will be reflected in the amplitude of the hydrogen maser oscillation.

The Breit-Rabi¹¹ equation gives the dependence of the energy of the hyperfine levels on the applied magnetic field. If the resonance frequency of a Zeeman transition ($\Delta F=0$, $\Delta M_F=\pm 1$) is measured, it can be used to obtain the product $g_J H_0$, where g_J is the gyromagnetic ratio of the atom and H_0 is the applied magnetic field. Spin-exchange coupling of the Zeeman level populations to the maser oscillation makes possible measurement of Zeeman frequencies for both hydrogen and deuterium. Since both atoms average the same magnetic-field region, H_0 dependence can-

cells out when the ratio of the $g_J H_0$ factors is taken. The experiment thus gives a value for $g_J(\text{H})/g_J(\text{D})$ which is equal to the ratio of atomic magnetic moments.

A diagram of the apparatus is given in Fig. 1. A beam of atomic hydrogen and deuterium passes through a hexapole state-selecting magnet which focuses atoms in the upper hyperfine states into the stem of a Teflon-coated quartz bulb. This storage bulb is located in the center of a spherical microwave cavity whose TE_{011} mode is tuned to the ($F=1, m_F=1$) to ($F=0, m_F=0$) hydrogen hyperfine transition. This transition is at approximately 1431 MHz when the field is 7.6 G. The perpendicular orientation of static and oscillatory magnetic fields couples to the $\Delta m_F = -1$ transition. With the available flux of atoms, power losses in the walls of the microwave cavity can be overcome, and self-excited oscillation occurs. A cylindrical solenoid with correction coils up to fourth order provides a highly homogeneous field over the region of the storage bulb. Cylindrical magnetic shields reduce ambient fields.

Cavity mistuning can shift the frequency of the maser oscillation.⁴ The necessity for accurate cavity tuning was avoided by not using this frequency directly in the determination of $g_J(\text{H})H_0$.

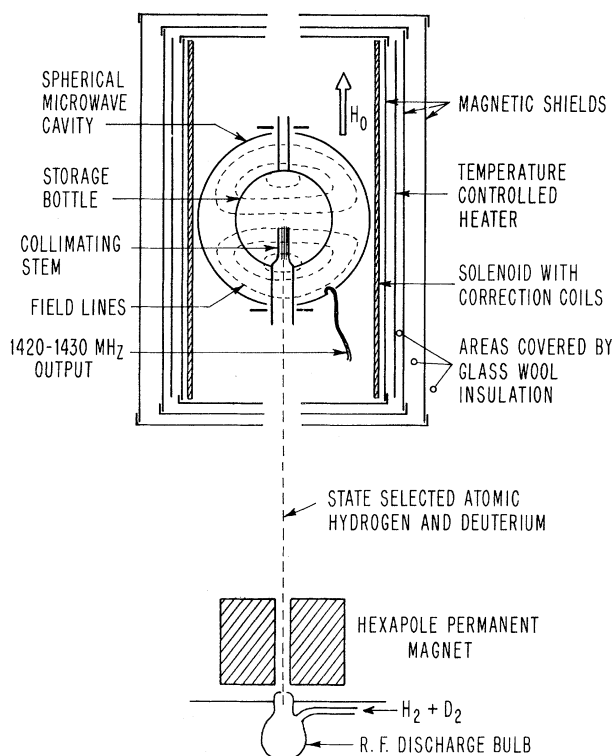


FIG. 1. Schematic diagram of the π maser.

Instead, this field-dependent frequency was used to stabilize the magnetic field by phase-locking it to a local crystal standard. Zeeman transitions ($\Delta F=0, \Delta m_F=-1$) were used in the g_J ratio determination. These were induced by applying an rf signal to a single loop of wire inside the microwave cavity. The amplitude of the maser oscillation served as an indicator of resonance for the applied rf fields.

The hydrogen Zeeman frequency was approximately 10.73 MHz, and the total linewidth for this experiment was typically 2 Hz full width at half-maximum power. The frequency of the deuterium Zeeman lines was about 7.42 MHz and the full width at half-maximum power for these was nearly 2.5 Hz. The primary source of the larger linewidth in the latter case was power broadening necessary for a good signal-to-noise ratio. A trace of the deuterium signal is shown in Fig. 2.

The ratio $g_J(\text{H})/g_J(\text{D})$ is obtained from the measured frequencies by means of the Breit-Rabi equation. The auxiliary constants necessary for this calculation are displayed in Table I. The uncertainties in these constants contribute negligible error.

There are two possible sources of systematic error that must be considered. The first arises from the spin-exchange effect that couples the two systems. This mechanism gives rise to a

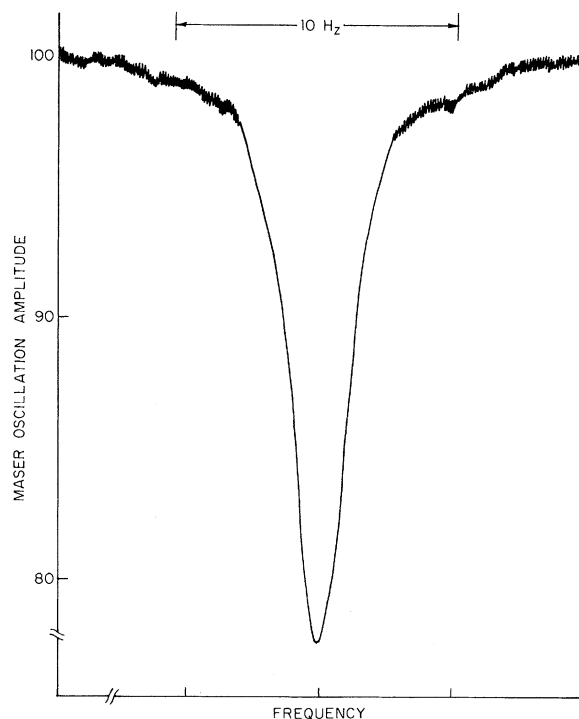


FIG. 2. Deuterium Zeeman resonance at 7.42 MHz.

Table I. Values of atomic constants used in calculating $g_J(\text{H})/g_J(\text{D})$.

Atom	Hyperfine separation $\Delta\nu$ (Hz)	Nuclear to atomic g -factor ratio g_I/g_J (10^{-4})
H	1420 405 751.8 ^a	15.192 703 ^b
D	327 384 352.51	2.332 176 ^c

^aS. B. Crampton, D. Kleppner, and N. F. Ramsey, *Phys. Rev. Letters* **11**, 338 (1964).

^bP. F. Winkler, F. G. Walther, M. T. Myint, and D. Kleppner, in Proceedings of the Arnold Sommerfeld Centennial Memorial Meeting and Symposium on Physics of One- and Two-Electron Atoms, Munich, Germany, 9-14 September 1968 (North-Holland Publishing Company, Amsterdam, The Netherlands, to be published).

^cObtained from hydrogen g_I/g_J using $g_J(\text{D}) = g_J(\text{H})$ and the ratio $\mu_I(\text{D})/\mu_I(\text{H})$ taken from the nuclear-moment table in N. F. Ramsey, *Molecular Beams* (Oxford University Press, London, England, 1956).

small shift in the hyperfine energy levels from their free-space values. The shift depends on the spin-exchange collision rate and the level populations. The values of the parameters necessary for a precise calculation of the shift from the theory of spin-exchange collisions are difficult to determine. However, it can be assumed that the shift does not depend on the applied magnetic field. Thus the spin-exchange contribution can be evaluated by making measurements at different values of magnetic field. A spin-exchange shift equal to four times the statistical error was found, but its value could be determined to 25% by taking data at two magnetic-field values differing by a factor of 70.

Brenner has discovered an inhomogeneity frequency shift that may affect resonance experiments in motional-averaging systems.¹² Such shifts can arise if the magnetic-field-averaged volume contains a poorly communicating subvolume where the magnetic field differs significantly from the volume average. The entrance stem of the usual hydrogen maser bulb fits into this category. However, the shift can be reduced by placing the collimating stem internal to the bulb (in a region of high magnetic-field homogeneity) instead of as an appendage external to it. This scheme worked in Brenner's experiment to eliminate the shift, and this type of bulb was also used in the present experiment. Moreover, a fortuitous cancellation between the decreased field dependence of the deuterium frequency and

the decreased velocity of the deuterium atom relative to that of the hydrogen atom indicates that any such shift would be about the same in both deuterium and hydrogen Zeeman lines. Consequently, when the ratio is taken, the expected size of any such effect is reduced by a factor of 10 from that for either atom frequency alone. The possibility of such shifts still being significant was investigated by broadening the lines both with a high-order correction coil and with the rf power. No effects were found.

The uncorrected value of $g_J(\text{H})/g_J(\text{D})$ was found to be 1.000 000 005 3(8). The spin-exchange contribution measured at low field is 0.000 000 004 1(11), and thus the final result is

$$g_J(\text{H})/g_J(\text{D}) = 1.000\,000\,009\,4(14).$$

This is considerably more precise than an earlier determination¹³ where $g_J(\text{H})/g_J(\text{D}) = 0.999\,999\,9(2)$. A theoretical value for the difference between the g_J factors of hydrogen and deuterium has been given in a recent calculation by Hegstrom.¹⁴ The theoretical value is 1.000 000 0097, in very good agreement with the experimental value.

The hyperfine frequency of deuterium was also measured at low magnetic field (0.11 G) by inducing the ($F = \frac{3}{2}, m_F = \frac{1}{2}$) to ($F = \frac{1}{2}, m_F = -\frac{1}{2}$) hyperfine transition. At low magnetic field, this transition is field independent to first order so that the magnetic-field correction can be made to high accuracy. The shift in the hyperfine frequency of deuterium due to collisions with the storage-bottle wall^{3,4} can be estimated from the wall shifts of hydrogen¹⁵ and tritium.¹⁶ For the 6-in. storage bottle used, it should be about -0.006 Hz. Spin-exchange shifts in the field-independent lines of deuterium occur only because of D-D collisions. There is no contribution from H-D collisions.¹⁷ The size of the shift can be estimated by assuming that the ratio of shift to linewidth is about the same as in hydrogen.¹⁷ The deuterium linewidth from deuterium collisions is estimated from the difference in the hydrogen Zeeman linewidths measured with and without deuterium in the maser. This is roughly 0.5 Hz. The maximum likely shift is about 10% of this linewidth or 0.05 Hz. This estimate gives the limit of error in the deuterium hyperfine-frequency measurement since the statistical error is 5 times smaller. The final result is

$$\Delta\nu(\text{D}) = 327\,384\,352.51(5)$$

in agreement with the previously most precise measurement.⁶

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†National Science Foundation Graduate Fellow.

‡Present address: Physics Department, Amherst College, Amherst, Mass.

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SOUND ABSORPTION IN LIQUID HELIUM BELOW 0.6 K

R. K. Wehner and R. Klein

Laboratories RCA Ltd., Zürich, Switzerland

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The temperature dependence of ultrasonic attenuation in superfluid ⁴He below 0.6 K is studied. It is shown that vertex corrections in the phonon self-energy improve the agreement with experiment, especially near the transition between the collisionless and collision-dominated regions. The introduction of one average lifetime for the relevant thermal phonons is critically examined.

The theoretical interpretation^{1,2} of recent experimental results^{3,4} on the temperature and frequency dependence of the velocity and the attenuation of high-frequency sound waves in liquid ⁴He at temperatures below 0.6 K is still incomplete. In this Letter we report microscopic calculations of the ultrasonic attenuation for values of the frequency ω and temperature T such that $\omega\tau(T)$ is not too far from 1. Here, τ is an average lifetime of those phonons which are the predominant partners for the interaction of the sound-wave phonons. This region of values for $\omega\tau$ is of particular interest since it represents the transition from the collision-free case ($\omega\tau \gg 1$), where the bubble-diagram result of Pethick and ter Haar¹ is valid, to the collision-dominated region ($\omega\tau \ll 1$), where the vertex renormalization is of primary importance.^{5,6} In the latter case sound absorption is usually calculated from a phenomenological Boltzmann equation.⁷ In the microscopic theory this equation corresponds to the integral equation for the vertex part.^{5,6} Starting from low temperatures, where $\omega\tau$ is large, and going to higher temperatures, the contribution from vertex corrections becomes more and more important.

The ultrasonic attenuation due to three-phonon scattering processes can be calculated from the following expression for the self-energy of the sound-wave phonon⁸:

$$\Pi_q(\omega) = 6i\hbar\beta \sum_{\vec{q}'} V(-\vec{q}, \vec{q}_1, \vec{q}_2) n_{q'}(n_{q'} + 1) \left\{ -1 + \frac{\omega}{\omega - \vec{q} \cdot \vec{v}_{q'} + i2\Gamma_{q'}} \right\} \Gamma_{\vec{q}'\vec{q}}(\omega). \quad (1)$$

Here, $V(-\vec{q}, \vec{q}_1, \vec{q}_2)$ is the parameter of anharmonic coupling between the long-wavelength phonon \vec{q} and two thermal phonons $\vec{q}_{1,2} = \pm\vec{q}' + \frac{1}{2}\vec{q}$, and $n_{q'}$ is the thermal-phonon occupation number. Within the hydrodynamic model^{9,10} for liquid ⁴He we assume a linear dispersion law $|\vec{v}_q| = c$, $\omega_q = cq$.¹¹

Using displacement modes instead of density fluctuations as normal coordinates we write the Hamiltonian as in Eq. (2.1) of Ref. 8. For the three-phonon coupling parameters in Eq. (1) we obtain

$$V(\vec{q}_1, \vec{q}_2, \vec{q}_3) = \left(\frac{\hbar q_1 q_2 q_3 c_0^4}{288 V \rho c^3} \right)^{1/2} [2u - 1 + (\hat{q}_1 \cdot \hat{q}_2)^2 + (\hat{q}_2 \cdot \hat{q}_3)^2 + (\hat{q}_3 \cdot \hat{q}_1)^2], \quad (2)$$