Measurement of the hydrogen-tritium g_J-factor ratio*

D. J. Larson and N. F. Ramsey

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

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A hydrogen maser operating at fields of up to 16 G has been used as a spin-exchange detector of Zeeman transitions in the hyperfine spectra of hydrogen and tritium. The maser oscillates on the field-dependent π transition which is sensitive to the state of colliding atoms. Atomic magnetic moment or g_J-factor ratios were obtained from the measured Zeeman frequencies with the aid of the Breit–Rabi formula. Measurements were made at different values of magnetic field and an extrapolation was done to eliminate the effects of spin-exchange frequency shifts. The result is $g_J(\mathrm{H})/g_J(\mathrm{T}) = 1 + (10.7 \pm 1.5) \times 10^{-9}$, in agreement with the theoretical predictions.

I. INTRODUCTION

The precision of measurements of atomic g-factor ratios has been dramatically improved in the last few years primarily owing to the use of high-homogeneity magnetic fields and coated-wall storage bulbs[1,2] which allow averaging of residual magnetic field inhomogeneities. At the same time improvements in calculations have added substantial interest to the measurements.[3,4] This paper reports a measurement of the hydrogen-tritium g_J-factor ratio with a precision that represents an improvement of more than two orders of magnitude over the last reported measurement.[5] Also, this is the first measurement of the ratio to differ significantly from unity.

II. EXPERIMENTAL METHOD

Many of the details of the experimental method have been reported elsewhere.[6,7] The description given here emphasizes those unique aspects of the present measurements which have not been previously described.

A. Basic design

A hydrogen maser oscillating on the $(F = 1, m_F = 1)$ to $(F = 0, m_F = 0)$ or π hyperfine transition is the basic piece of experimental apparatus. A schematic diagram of the maser is shown in Fig. 1. This maser is equipped with a solenoid and correction coils which provide magnetic fields of up to 16 G with homogeneity on the order of 5 ppm in a 10-cm-diam region. The 10-cm-diam Teflon-coated storage bulb used in these measurements was of a special design with the collimating entrance stem placed in the center of the bulb. This was done to place the entrance stem, which is a region of poor communication with the rest of the bulb, in a position of high magnetic field homogeneity and therefore to reduce problems associated with the averaging of the inhomogeneous field found at the normal entrance stem external to the bulb.[8] In order to obtain oscillation with a reasonably low density of hydrogen atoms in the rather small storage bulb, a feedback circuit was used to multiply the Q of the spherical microwave cavity.

Transitions between the states of interest in determining g_J factors are made observable in the maser through the mechanism of spin exchange. If both atomic hydrogen and atomic tritium are present in the storage bulb of the maser, spin-exchange collisions will couple the electron polarizations of the two systems. 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 tritium disturbs the equilibrium and this is reflected in the amplitude of the maser oscillation. This method of detection requires that the maser be oscillating on the π and not on the σ transition since the spin-exchange relaxation of the σ transition is not first-order dependent on the state of the colliding atoms.

Cavity mistuning can shift the frequency of the maser oscillation. The difficult problem of cavity tuning was avoided by not using the oscillation frequency in the g_J-ratio determination. Rather, this field-dependent frequency is used to stabilize the magnetic field by phase locking it to a frequency derived from a frequency-standard maser and feeding back to a correction solenoid.

The measurement is made by comparing Zeeman transitions in each type of atom. These transitions are induced by applying rf power to a loop of wire around the storage bulb inside the microwave cavity. Since the maser amplitude serves as the indicator of resonance, great care was taken to ensure

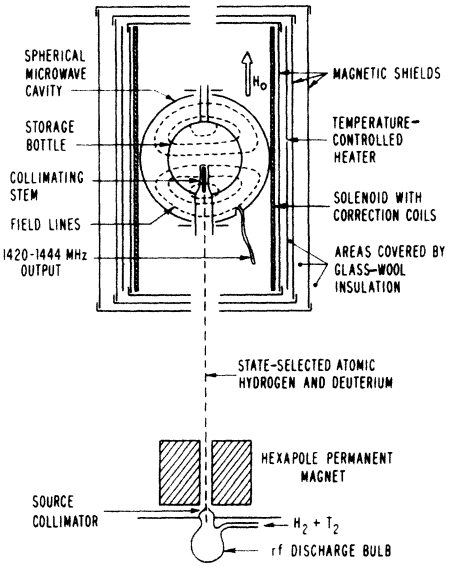

FIG. 1. Schematic diagram of the π maser.

that in the absence of applied signals the maser oscillation amplitude was stable. Even with the cavity-Q multiplying network, which added some instability, fluctuations in the amplitude of as little as one part in three hundred over a period of several minutes were attained. A least-squares fit to a single pass over a hydrogen Zeeman line is shown in Fig. 2.

With the aid of the Breit-Rabi formula and the auxiliary constants $\Delta\nu$ (hyperfine separation) and g_I/g_J (nuclear to electronic g-factor ratio), the measured Zeeman frequency can be used to obtain the product $g_J\mu_B H_0/h$ for each type of atom. Since both species sample the same magnetic field region, when the ratio is taken the H_0 cancels out except for small corrections to be discussed later.

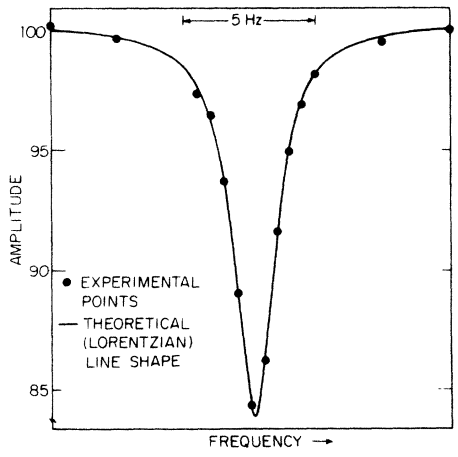

FIG. 2. Zeeman transition in hydrogen.

The experiment thus gives a value for $g_J(\mathrm{H})/g_J(\mathrm{T})$. Measurements were also done to determine the value of $g_J(\mathrm{H})/g_J(\mathrm{D})$ and this provided a check[9-11] that the apparatus was functioning properly, allowing conservation of the limited amount of tritium available, and also provided a test for systematic errors by comparison of the deuterium and tritium results. The values of the auxiliary constants used in the calculation of the g_J-factor ratios are given in Table I.

B. Correction for spin-exchange frequency shifts

Since the frequency resolution of the measurements (on the order of a few millihertz) was such that spin-exchange frequency shifts might well contribute significantly, a major objective of the experimental design was the elimination and/or measurement of such shifts. A technique for the elimination of the shifts from the data resulted from the observation that if we assume (as discussed later) that the spin-exchange frequency shifts are independent of the value of the magnetic field H_0, the true g_J-factor ratio can be obtained by extrapolating from the results of measurements at different values of the field. This can be seen by writing

$$\left(\frac{g_J(\text{species 1})}{g_J(\text{species 2})}\right)_{\text{obs}} = \frac{\nu_g(2)}{\nu_g(1)} \,. \tag{1}$$

The quantity ν_g is obtained from the uncorrected Zeeman frequency by using Eq. (5) below and, in

TABLE I. Values of atomic constants used in calculating g_J-factor ratios.

$\Delta\nu$ (H)	1 420 405 751.768(3) Hz [a]
g_I (H)/g_J (H)	$15.192\,703\,35(15)\times10^{-4}$ [b]
$\Delta\nu$ (D)	327 384 352.5222(17) Hz [c]
g_I (D)/g_J (D)	$2.332\,172\,5(7)\times10^{-4}$ [d]
$\Delta\nu$ (T)	1 516 701 470.9064(16) Hz [e]
g_I (T)/g_J (T)	$16.205\,143(5)\times10^{-4}$ [f]

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[e] B. S. Mathur, S. B. Crampton, D. Kleppner, and N. F. Ramsey, Ref. 12.

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the absence of frequency shifts, is equal to $g_J\mu_B H_0/h$. [At low field ν_g equals $(2I+1)\nu_z$.] If the uncorrected Zeeman frequency is shifted by spin exchange, the derived frequency ν_g will also be shifted and thus will not exactly equal $g_J\mu_B H_0/h$. We can represent this by writing

$$\nu_g = g_J\mu_B H_0/h + \delta \,, \tag{2}$$

where δ is the contribution from the spin-exchange shifts. Therefore, including spin-exchange contributions explicitly, we have

$$\left[\frac{g_J(1)}{g_J(2)}\right]_{\rm obs} = \frac{g_J(1)[1+\delta_1 h/g_J(1)\mu_B H_0]}{g_J(2)[1+\delta_2 h/g_J(2)\mu_B H_0]} \approx \left(\frac{g_J(1)}{g_J(2)}\right)\left[1+\left(\frac{\delta_1}{g_J(1)}-\frac{\delta_2}{g_J(2)}\right)\frac{h}{\mu_B H_0}\right]. \tag{3}$$

Thus if the measured g_J-factor ratio is plotted as a function of $1/H_0$, the intercept $(1/H_0=0)$ of a straight-line fit to the data gives the ratio free of errors introduced by spin exchange.

The linear extrapolation described above is clearly only valid if δ_1 and δ_2 are independent of magnetic field. Since the spin-exchange shifts are functions of atom densities and of the population differences between the various hyperfine states, these must be unchanged in making measurements at different values of the magnetic field. This is done by providing the same fluxes of atoms from the source and by maintaining the same maser oscillation conditions (cavity Q, atomic linewidth, etc.) at the different fields. The reproducibility of measurements at a given field provides an indication of whether or not the spin-exchange conditions can be kept the same from measurement to measurement.

One effect that could conceivably lead to a change in δ as the magnetic field is changed is the recoupling of the low-field hyperfine states that occurs as the field is increased. An argument against such an effect is that since both δ and ν_g are electron effects, a change in coupling to the nuclear moment should not affect the relationship between the two frequencies. This argument can be made explicit by observing that since δ is a small change in ν_g due to a shift of the uncorrected Zeeman frequency,

$$\delta = \Delta\nu_g = \left(\frac{\partial\nu_g}{\partial\nu_z}\right)\Delta\nu_z = \left(\frac{\partial\nu_g}{\partial\nu_z}\right)\beta \,, \tag{4}$$

where β is the spin-exchange shift of the uncorrected Zeeman frequency. Now, for the $(F=I+\frac{1}{2}, m_F=-I+\frac{1}{2})$ to $(F=I+\frac{1}{2}, m_F=-I-\frac{1}{2})$ transition used in all the measurements in this experiment, we have

$$\nu_g = \nu_z + 2I\Delta\nu\left(\frac{\nu_z - N}{\Delta\nu + (2I+1)(\nu_z - N)}\right). \tag{5}$$

Here $N = g_I\mu_B H_0/h$. Therefore

$$\frac{\partial\nu_g}{\partial\nu_z} = 1 + \frac{2I\Delta\nu^2}{[\Delta\nu + (2I+1)(\nu_z - N)]^2} \,. \tag{6}$$

It is shown in the Appendix that for the transition under consideration the spin-exchange shift β is proportional to

$$\Delta\langle\sigma_z\rangle \equiv \langle F=I+\tfrac{1}{2}, m_F=I+\tfrac{1}{2}|\,\sigma_z\,|F=I+\tfrac{1}{2}, m_F=I+\tfrac{1}{2}\rangle - \langle F=I+\tfrac{1}{2}, m_F=-I-\tfrac{1}{2}|\,\sigma_z\,|F=I+\tfrac{1}{2}, m_F=-J-\tfrac{1}{2}\rangle = \left[1+\frac{2I\Delta\nu^2}{[\Delta\nu+(2I+1)(\nu_z-N)]^2}\right]^{-1} \,, \tag{7}$$

which is the change in electron polarization in the transition. (Here σ_z is the z component of the electron-spin operator.) Upon taking the product of the expressions in Eqs. (7) and (6), we find that the dependence upon H_0 cancels out. However, the fact that β (and thus δ) depends upon the electron polarization in the colliding species of atom has not as yet been taken into account. This fact is indeed a basis for a change in the spin-exchange shift with magnetic field. The average electron polarization provided by the state-selecting magnet is dependent upon the field in the storage bulb. The loss of polarization due to coupling of the electrons to the nucleus gets smaller at higher fields and this can cause increased spin-exchange shifts. To first order in $\nu_z/\Delta\nu$ the fractional increase in polarization is $[8I^2/(2I+1)](\nu_z/\Delta\nu)$. The largest effect in the hydrogen isotopes occurs in deuterium, where at 16 G this term becomes approximately $\frac{1}{9}$. This means that the average electron polarization in the bulb could at most be increased by about 5% since the polarization carried by the hydrogen atoms is changed by a much smaller amount. A 5 or even 10% nonlinearity in the spin-exchange extrapolation is not significant in the present measurements.

C. Handling of tritium

The fact that tritium is radioactive is a source of difficulty in that only a limited amount is usually made available and careful containment of the gas is necessary. Recirculation of tritium is the usual technique by which the total amount necessary to carry out an experiment is held to an acceptable level. However, in a previous maser experiment, done to measure the tritium hyperfine structure, many difficulties were encountered with loss and contamination of tritium in the recirculation system.[12] In the present experiment the necessity of recirculation was avoided by re-

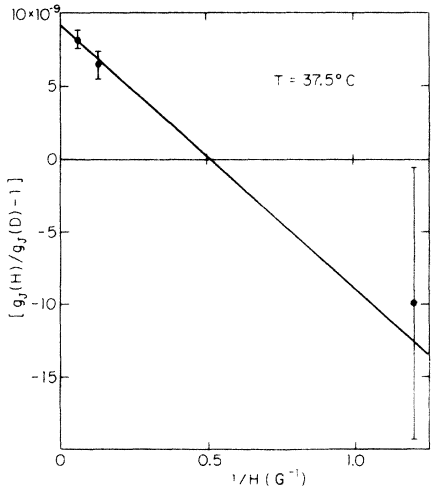

FIG. 3. $g_J(\mathrm{H})/g_J(\mathrm{D})$ as a function of magnetic field.

ducing the amount of tritium flux required. The total flux needed to give the required net flux to the storage bulb was reduced by simply adding a multichannel glass collimator to the source. The net flux required for a given density in the storage bulb was reduced by use of the small 10-cm storage bulb. The loss of tritium due to trapping of atoms on the walls of the discharge bulb was well documented by Mathur.[13] He found that the loss in a quartz bulb was significantly less than that observed with Pyrex. For this reason, the discharge bulb used on the present maser was made of quartz. One curie of tritium provided at least one hour of running time, implying that the total flux was not more than 5×10^{15} atoms/sec. A total of 15 Ci was used.

Since there was no need to recirculate the tritium through the maser, it was decided that the safest and easiest procedure would be simply to pump the gas with the titanium pumps. A small fraction of the total tritium was collected in the VacIon pump in the storage-bulb region. The rest was pumped by the Orb-Ion pump in the source region. The absorbed tritium provided no safety problem during the course of the experiment. The body of the Orb-Ion pump has been removed and stored for possible future use with tritium. This method of handling the tritium made it hardly more difficult to use than hydrogen or deuterium except for one thing. When the maser was opened, a significant amount of tritium was found to be dissolved in the stainless-steel walls of the vacuum system. Extensive cleaning with acid and high-temperature baking was required to eliminate measurable amounts of the tritium.

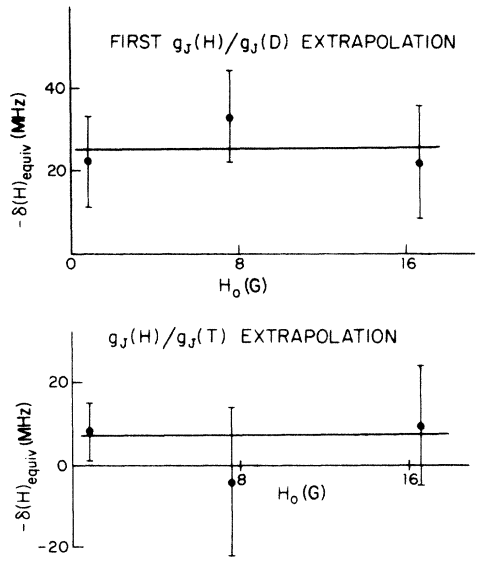

FIG. 4. Measured spin-exchange shift as a function of magnetic field.

III. RESULTS AND INTERPRETATION

A. Experimental results

One use of the deuterium measurements in the present experiment is to verify the spin-exchange-shift extrapolation described in Sec. II B. One such extrapolation is shown in Fig. 3. Of course doing such an extrapolation allows one to measure the relative spin-exchange frequency shift between the hydrogen and deuterium or tritium Zeeman lines. This shift can be obtained by comparing the extrapolated g-factor ratio with the results at any one field. The shift should be independent of the field at which it is measured if the assumptions in Sec. II B are correct. Figure 4 shows the equivalent spin-exchange shift (that shift of the hydrogen Zeeman line which would be necessary to give the observed g_J-factor ratio if the deuterium or tritium line were unshifted) as a function of magnetic field for two separate extrapolations.

One startling result of these measurements was the observation that the equivalent spin-exchange shift was a function both of temperature and of time. (The measured g_J-factor ratio was not a function of temperature or time.) This observation led to the hypothesis that much of the spin exchange was occurring at the wall of the storage bulb. This point is discussed in more detail elsewhere.[14]

The result of the measurement on tritium was

$$g_J(\mathrm{H})/g_J(\mathrm{T}) = 1 + (10.7 \pm 0.7)\times10^{-9}\,,$$

where the error given includes only statistical errors.

B. Error analysis

In addition to the spin-exchange frequency shifts, a primary source of possible systematic error is provided by the residual inhomogeneities in the magnetic fields, both static and oscillatory. Effects due to inhomogeneous oscillatory fields were checked by making two measurements with the rf power driving the tritium transition increased by a factor of 10. No significant shift was observed. In addition the line was broadened by about a factor of 2 by decreasing the static field homogeneity and four separate measurements were made. These measurements gave a result which differed from the result with reduced linewidth by -0.8 ± 0.7 parts in 10^9.

Since most of the data was taken by finding the mean between two points of equal amplitude on the Zeeman line, the symmetry of the line was of primary importance. A possible source of asymmetry, besides magnetic field inhomogeneities, is the changed spin-exchange shift of the hydrogen hyperfine transition (which is used to maintain a constant field) which occurs when a Zeeman transition is driven. Different levels on the line correspond to different amounts of net electron polarization and this could be converted by means of the spin-exchange shift and the field-locking network to different values of magnetic field. A computerized data taking system became available during the last part of these measurements and the symmetry of both hydrogen and deuterium Zeeman lines was tested by least-squares fitting to Lorentzian line shapes with a dispersive component. No significant asymmetry was found.

A measurement of g_I/g_J in deuterium was also made as a part of the search for systematic errors. This was done to see if the present experimental arrangement and measurement techniques would duplicate the result indicated in Table I, which was obtained from a combination of $g_I(\mathrm{H})/g_J(\mathrm{H})$ measured in a hydrogen maser[15] and $g_I(\mathrm{D})/g_I(\mathrm{H})$ measured by nuclear magnetic resonance.[16] The measurement could be done with a reasonable accuracy because the $(F=\frac{3}{2}, m_F=\frac{1}{2})$ to $(F=\frac{1}{2}, m=-\frac{1}{2})$ transition involves a change of nuclear but not electron orientation at low magnetic field. Thus the measurement was done by comparing the frequency of the above transition with the frequency of a Zeeman transition. Measurements were made at 0.8 and 16.6 G to allow elimination of the constant-frequency errors contributed by spin exchange and the wall shift. The result was $0.233\,217\,17(35)\times10^{-3}$, in good agreement with the NMR result which is $0.233\,217\,25\times10^{-3}$. The indicated error makes no allowance for systematic effects and represents a frequency resolution comparable to that obtained in the g_J-factor measurements.

C. Conclusions

Combining the statistical error and an allowance for possible magnetic field averaging error the result is

$$g_J(\mathrm{H})/g_J(\mathrm{T}) = 1 + (10.7 \pm 1.5)\times10^{-9} ,$$

where the error corresponds to 1 standard deviation. This result is in agreement with the theoretical predictions[3,4] of $1+9.7\times10^{-9}$ and with the results of measurements on the hydrogen-deuterium g_J-factor ratio,[9-11] assuming that the deviation from unity is a term that is inversely proportional to the nuclear mass.

IV. APPENDIX

The result obtained in Sec. II B depends upon the fact that the spin-exchange shift is proportional to the change of electron polarization in the transition. For collisions involving atoms of two different types the validity of this statement is immediately demonstrated by examining the form of the spin-exchange equations derived by Valberg.[17] The frequency shift due to colliding atoms of polarization ϵ in the density-matrix equations for atoms undergoing a transition from state c to state d is

$$\begin{aligned}\Delta\nu &= v N \sigma_{\mathrm{SF}} k[\langle c|\sigma_z|c\rangle - \langle d|\sigma_z|d\rangle]\rho_{cd} \\ &= v N \sigma_{\mathrm{SF}} k[\Delta\langle\sigma_z\rangle]\rho_{cd} \ . \end{aligned} \tag{A1}$$

For collisions involving the same type of atom an additional term can contribute. This is of the form

$$\Delta\nu' = \tfrac{1}{2} v N \sigma_{\mathrm{SF}} k\left[\sum_j |\langle c|\sigma_j|d\rangle|^2\right](\rho_{cc}-\rho_{dd})\rho_{cd} \ . \tag{A2}$$

For the transitions $(F=I+\frac{1}{2}, m_F=-I+\frac{1}{2})$ to $(F=I+\frac{1}{2}, m_F=-I-\frac{1}{2})$ used in the present measurements, this term can also be shown to be proportional to the change of electron polarization in the transition. Since

$$|F=I+\tfrac{1}{2}, m_F=-I-\tfrac{1}{2}\rangle = |m_J=-\tfrac{1}{2}, m_F=-I\rangle \tag{A3}$$

we have, using now the $m_J m_I$ basis,

$$\begin{aligned}&\sigma_x|-\tfrac{1}{2},-I\rangle = i\sigma_y|-\tfrac{1}{2},-I\rangle = |+\tfrac{1}{2},-I\rangle , \\ &\sigma_z|-\tfrac{1}{2},-I\rangle = -|-\tfrac{1}{2},-I\rangle \ . \end{aligned} \tag{A4}$$

If we write

$$|F=I+\tfrac{1}{2}, m_F=-I+\tfrac{1}{2}\rangle = c_1|+\tfrac{1}{2},-I\rangle + c_2|-\tfrac{1}{2},-I+1\rangle \tag{A5}$$

then, dropping reference to F since we always have $F = I + \frac{1}{2}$,

$$\sum_j |\langle m_F = -I + \tfrac{1}{2} | \sigma_j | m_F = -I - \tfrac{1}{2} \rangle|^2 = 2c_1^2 \ . \tag{A6}$$

Now

$$\langle m_F = -I + \tfrac{1}{2} | \sigma_Z | m_F = -I + \tfrac{1}{2} \rangle - \langle m_F = -I - \tfrac{1}{2} | \sigma_Z | m_F = -I - \tfrac{1}{2} \rangle = c_1^2 - c_2^2 + 1 = 2c_1^2 \ .$$

Therefore

$$\sum_j |\langle m_F = -I + \tfrac{1}{2} | \sigma_j | m_F = -I - \tfrac{1}{2} \rangle|^2 = [\langle m_F = -I + \tfrac{1}{2} | \sigma_Z | m_F = -I + \tfrac{1}{2} \rangle - \langle m_F = -I - \tfrac{1}{2} | \sigma_Z | m_F = -I - \tfrac{1}{2} \rangle] = \Delta \langle \sigma_Z \rangle \ . \tag{A8}$$

The use of the arbitrary coefficients c_1 and c_2 in the above derivation indicates that it is valid for any value of field (i.e., for any amount of recoupling of the low-field states). This verifies the assumption made earlier about the spin-exchange shift.

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