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DETERMINATION OF THE QUADRUPOLE COUPLING CONSTANT IN THE N¹⁴ ATOMIC GROUND STATE*

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The zero-field hyperfine frequencies in the ground state of atomic N¹⁴ have been measured in a hydrogen maser with sufficient accuracy to resolve the quadrupole coupling constant B . The result is $B = +1.32 \pm 0.20$ Hz.

Previous observations of the N¹⁴ ground-state hyperfine transition frequencies by paramagnetic resonance absorption¹ and spin-exchange optical pumping²⁻⁶ determined the magnetic hyperfine coupling constant but were unable to resolve the quadrupole coupling constant. We report a re-determination⁷ of the N¹⁴ hyperfine transition frequencies which does resolve the quadrupole coupling constant. The nitrogen resonances were detected by their effect on the oscillation level of a hydrogen maser oscillating on one of the $\Delta m_F = \pm 1$ hyperfine transitions in ground-state atomic hydrogen. Essentially the same detection technique and apparatus were used previously to measure the ground-state hyperfine structure of atomic deuterium and have been described in detail elsewhere.⁸ The particular apparatus used in this experiment was a hydrogen maser previously modified to study the effects of added paramagnetic gases on the hydrogen radiative lifetimes and has also been described in detail elsewhere.⁹ Nitrogen atoms dissociated in the maser second source⁹ were admitted unpolarized to the maser storage bottle, where they were polarized by electron-spin-exchange collisions with hydrogen atoms oscillating on one of the magnetic-field-dependent $\Delta m_F = \pm 1$ ground-state hydrogen hyperfine transitions. A second rf magnetic field driven by a single turn of wire

around the maser microwave cavity depolarized the nitrogen atoms by stimulating nitrogen hyperfine transitions. On resonance this loss of electron polarization by the nitrogen atoms increased the rate at which nitrogen-hydrogen spin-exchange collisions coupled electron polarization away from the oscillating hydrogen atoms and thereby reduced the maser oscillation level. The observed nitrogen transition frequencies were corrected to zero magnetic field using the observed field-dependent hydrogen oscillation frequencies and the previously determined gyromagnetic ratios.^{1,10}

The effective Hamiltonian for a ground-state nitrogen-14 atom in a magnetic field H is¹¹

$$\mathcal{H} = A \vec{I} \cdot \vec{J} - g_J \mu_0 \vec{H} \cdot \vec{J} - g_I \mu_0 \vec{H} \cdot \vec{I} \\ + \frac{1}{6} B [3(\vec{I} \cdot \vec{J})^2 + \frac{3}{2} \vec{I} \cdot \vec{J} - (\vec{I})^2 (\vec{J})^2],$$

where A is the magnetic hyperfine coupling constant, g_J is the atomic gyromagnetic ratio, g_I is the nuclear gyromagnetic ratio, B is the quadrupole coupling constant, \vec{I} is the nuclear spin, and \vec{J} is the atomic angular momentum. B includes all interactions which have the quadrupole-interaction symmetry, including the second-order hyperfine interaction.

The ground state has nuclear spin 1 and atomic angular momentum $\frac{3}{2}$, so that there are three

sets of hyperfine levels characterized in low magnetic field by total angular momentum $F=I+J=\frac{5}{2}, \frac{3}{2}$, and $\frac{1}{2}$, respectively. In terms of the zero-magnetic-field hyperfine frequencies

$\nu(\frac{5}{2} \rightarrow \frac{3}{2})$ and $\nu(\frac{3}{2} \rightarrow \frac{1}{2})$ the hyperfine and quadrupole coupling constants are

$$A = \frac{3}{10} \nu(\frac{5}{2} \rightarrow \frac{3}{2}) + \frac{1}{6} \nu(\frac{3}{2} \rightarrow \frac{1}{2})$$

Table I. Nitrogen-14 hyperfine-transition frequency readings corrected to zero magnetic field. Each determination is the mean of two series having equal but opposite magnetic field corrections in low magnetic field. A.1 time scale (Ref. 15).

	Nitrogen Transition (F, m_F) \rightarrow (F, m_F)	Nitrogen Transition Frequency
1.	(5/2, 5/2) \rightarrow (3/2, 3/2)	26 127 324.15 \pm .32 Hz.
	(3/2, -3/2) \rightarrow (5/2, -5/2)	26 127 324.20 \pm .35
		26 127 324.18 \pm .47 Hz.
2.	(5/2, 5/2) \rightarrow (3/2, 3/2)	26 127 324.59 \pm .26 Hz.
	(3/2, -3/2) \rightarrow (5/2, -5/2)	26 127 324.29 \pm .36
		26 127 324.44 \pm .45 Hz.
3.	(3/2, 3/2) \rightarrow (5/2, 5/2)	26 127 324.60 \pm .29 Hz.
	(5/2, -5/2) \rightarrow (3/2, -3/2)	26 127 324.06 \pm .27
		26 127 324.33 \pm .39 Hz.
4.	(3/2, 3/2) \rightarrow (5/2, 5/2)	26 127 324.91 \pm .24 Hz.
	(5/2, -5/2) \rightarrow (3/2, -3/2)	26 127 323.59 \pm .23
		26 127 324.25 \pm .33 Hz.
	$\nu(5/2 \rightarrow 3/2) =$	26 127 324.30 \pm .28 Hz.
1.	(3/2, 3/2) \rightarrow (1/2, 1/2)	15 676 389.85 \pm .27 Hz.
	(1/2, -1/2) \rightarrow (3/2, -3/2)	15 676 391.04 \pm .29
		15 676 390.45 \pm .40 Hz.
2.	(3/2, 3/2) \rightarrow (1/2, 1/2)	15 676 389.90 \pm .24 Hz.
	(1/2, -1/2) \rightarrow (3/2, -3/2)	15 676 391.24 \pm .26
		15 676 390.57 \pm .36 Hz.
3.	(1/2, 1/2) \rightarrow (3/2, 3/2)	15 676 390.71 \pm .29 Hz.
	(3/2, -3/2) \rightarrow (1/2, -1/2)	15 676 390.64 \pm .24
		15 676 390.68 \pm .38 Hz.
4.	(3/2, 3/2) \rightarrow (1/2, 1/2)	15 676 389.69 \pm .27 Hz.
	(1/2, -1/2) \rightarrow (3/2, -3/2)	15 676 391.20 \pm .36
		15 676 390.45 \pm .46 Hz.
5.	(1/2, 1/2) \rightarrow (3/2, 3/2)	15 676 390.35 \pm .24 Hz.
	(3/2, -3/2) \rightarrow (1/2, -1/2)	15 676 390.64 \pm .21
		15 676 390.50 \pm .32 Hz.
6.	(1/2, 1/2) \rightarrow (3/2, 3/2)	15 676 390.69 \pm .22 Hz.
	(3/2, -3/2) \rightarrow (1/2, -1/2)	15 676 390.79 \pm .22
		15 676 390.74 \pm .31 Hz.
7.	(3/2, 3/2) \rightarrow (1/2, 1/2)	15 676 390.46 \pm .21 Hz.
	(1/2, -1/2) \rightarrow (3/2, -3/2)	15 676 391.10 \pm .23
		15 676 390.78 \pm .31 Hz.
8.	(3/2, 3/2) \rightarrow (1/2, 1/2)	15 676 390.87 \pm .22 Hz.
	(1/2, -1/2) \rightarrow (3/2, -3/2)	15 676 390.57 \pm .23
		15 676 390.72 \pm .32 Hz.
	$\nu(3/2 \rightarrow 1/2) =$	15 676 390.63 \pm .27 Hz.

and

$$B = \frac{1}{5} \nu(\frac{5}{2} \rightarrow \frac{3}{2}) + \frac{1}{3} \nu(\frac{3}{2} \rightarrow \frac{1}{2}).$$

Because of the large number of nitrogen hyperfine levels dividing the electron polarization carried into the system by state-selected hydrogen atoms, the signal-to-noise ratio of the change of oscillation amplitude due to any one nitrogen hyperfine transition was not very high. The best signal-to-noise ratio among upper ($F = \frac{5}{2} \rightarrow F = \frac{3}{2}$) hyperfine transitions was about 15 to 1 on the $(\frac{5}{2}, \frac{5}{2}) \rightarrow (\frac{3}{2}, \frac{3}{2})$ transition with width about 10 Hz. The best signal-to-noise ratio among lower ($F = \frac{3}{2} \rightarrow F = \frac{1}{2}$) hyperfine transitions was about 10 to 1 on the $(\frac{3}{2}, \frac{3}{2}) \rightarrow (\frac{1}{2}, \frac{1}{2})$ transition with width about 20 Hz.

A series of observations of one of these transitions was followed immediately by a series of observations of the transition having equal but opposite magnetic-field correction in low magnetic field. The means of the two series were averaged to provide one determination of the zero magnetic-field hyperfine splitting. The results of four such determinations of the upper hyperfine splitting over two days and eight determinations of the lower hyperfine splitting over three days are shown in Table I. $\Delta m_F = +1$ and -1 nitrogen transitions were observed using the $\Delta m_F = +1$ and -1 hydrogen transition oscillations, respectively.

Although each individual observations has been corrected to zero magnetic field before being averaged into its series, the mean of a series for one hyperfine transition often disagrees by a cycle or two from the mean for the opposite hyperfine transition. The direction of the discrepancy is not consistent with a spin-exchange frequency-shift effect, and the discrepancies do not vary consistently with the linewidth or signal to noise. The discrepancies are thought to be due to the fact that because of their lower velocities and larger gyromagnetic ratios the nitrogen atoms average the magnetic field in the storage-bottle exit stem differently than do the hydrogen atoms. This effect has been discussed recently by Brenner.¹² The predicted effect is the right order of magnitude and should vary from day to day as the magnetic-field gradients vary. The error is equal and opposite for nitrogen hyperfine transitions having equal but opposite magnetic field corrections. The averages of means of series for equal but opposite magnetic field corrections should therefore be reproducible from day to day, as observed, and should be unshifted by this effect from the true zero magnet-

ic-field hyperfine splitting within the precision of this experiment.

The errors shown in Table I for each series represent one standard deviation from the mean. The combined limits of error for the hyperfine frequencies include the following: (1) The statistical 90% confidence limits. (2) A limit of error of 0.15 Hz due to calibration of the nitrogen frequency signal generator. (3) A limit of error of 0.2 Hz due to spin-exchange shifts^{13, 14} of the nitrogen transitions. Spin-exchange shifts not corrected by averaging equal but opposite transitions are presumed to be only a few percent of the approximately 5 Hz spin-exchange contribution to the linewidth.

The mean upper and lower nitrogen hyperfine frequencies combine to give for the hyperfine and quadrupole coupling constants and their limits of error

$$A = 10\,450\,929.06 \pm 0.19 \text{ Hz},$$

$$B = +1.32 \pm 0.20 \text{ Hz}.$$

The result for B disagrees with the predicted⁴ value

$$B = -3.4 \pm 1.6 \text{ Hz}.$$

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