types seem to be implied. This behavior is different from that observed in octupole bands of rare-earth nuclei,[5,6] where the octupole collectivity is weaker and appears to be destroyed at rather low spin values. The concept of the Coriolis force aligning an angular momentum vector along the rotation axis has previously been applied to one-particle states in weakly deformed nuclei,[7] and to two-particle states in strongly deformed even-even nuclei.[8] One sees from the ^{238}U case that this idea may apply to any angular momentum vector in a rotating system, including, in some circumstances, collective angular momenta.

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Observation of Magnetic Octupole and Scalar Spin-Spin Interactions in I_2 Using Laser Spectroscopy*

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Magnetic octupole and scalar spin-spin interactions have been observed in $^{127}I_2$ by precisely measuring (line centers located to 1 part in 10^{11}) the hyperfine spectrum on the $P(13)$, 43-0 line using laser–molecular-beam techniques. The values of the coupling strengths obtained from fitting the spectrum are, for the electric quadrupole, $eQq' = -554\,094 \pm 13$ kHz and $eQq'' = -2\,448\,025 \pm 10$ kHz; for the spin-rotation, $C' - C'' = 186.71 \pm 0.10$ kHz; for the tensor spin-spin, $D_t' - D_t'' = -100.5 \pm 1.0$ kHz; for the scalar spin-spin, $D_s' - D_s'' = -2.72 \pm 1.0$ kHz; and for the magnetic octupole, $\Omega m' - \Omega m'' = -2.17 \pm 0.70$ kHz.

We report the observation of magnetic octupole and scalar spin-spin interactions in the optical spectrum of I_2 at 5145 Å. The line spacings were measured with an accuracy of 5 kHz (1 part in 10^{11}) using a heterodyne technique employing two argon-ion lasers individually stabilized to I_2 hyperfine lines excited in molecular beams.

After the first observation of I_2 hyperfine structure[1,2] Kroll explained the results in terms of nuclear electric quadrupole and magnetic spin-rotation interactions.[3] Subsequently, Hanes *et al.*,[4] Hänsch, Levenson, and Schawlow,[6] and Ruben *et al.*[7] studied the hyperfine structure associated with a number of I_2 transitions. With further improvements in the measured data, Bunker and Hanes[8] introduced a tensor nuclear spin-spin coupling term to fit seven components of the $R(127)$ line to within a standard deviation of approximately 60 kHz.

In this Letter we report precision measurements of hyperfine transitions on the $P(13)$ 43-0, $B\,^3\Pi$-$X\,^1\Sigma$ line in $^{127}I_2$. Figure 1 shows the spec-

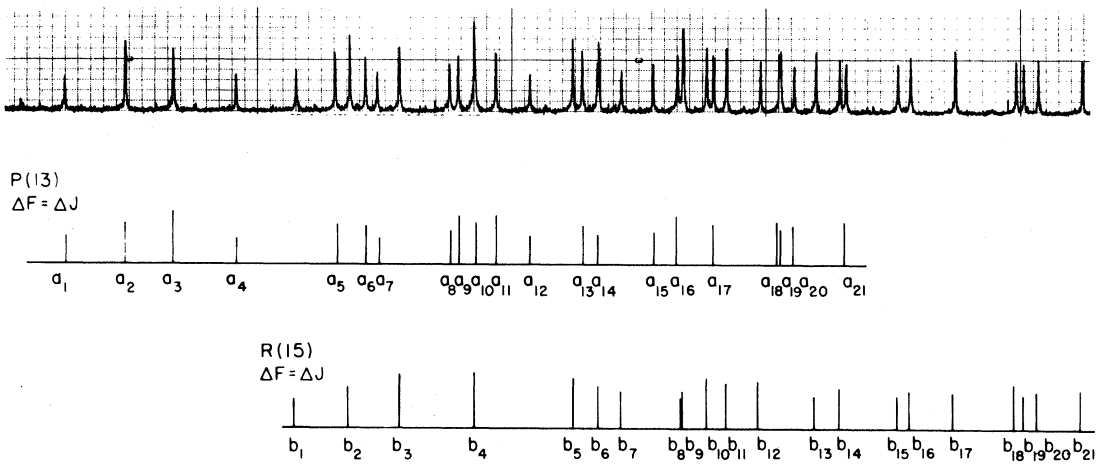

FIG. 1. Complete hfs on $P(13)$ and $R(15)$ lines. The $P(13)$ transitions are labeled by a_1 through a_{21} and the $R(15)$ transitions are labeled by b_1 through b_{21}. Typical linewidth is 650 kHz and the time constant is 30 msec. The frequency scale is 15 MHz/div.

trum with the $P(13)$ hyperfine components labeled by a_1 through a_{21}. The components of the $R(15)$ 43-0 line, shown in the figure, are labeled by b_1 through b_{21} but are not analyzed at this time. These hyperfine components follow the selection rule $\Delta F = \Delta J$ and are the strongest of the allowed hyperfine transitions. The weak $\Delta F = 0$ transitions, a few of which are observable in the figure, have also been measured and will be discussed in detail in a future paper.

The experimental setup is similar to that described earlier[9] where two single-frequency 5145-Å argon-ion lasers were individually stabilized to hyperfine transitions excited in two independent molecular beams of I_2. The line positions were precisely determined by counting the beat frequency between the two lasers.

In these measurements the angular divergence of the I_2 molecular beam was set at 2×10^{-3} rad giving a Doppler-broadened linewidth of about 500 kHz. The natural linewidth of the $P(13)$ lines is estimated to be 70 kHz.[1,10] In high-resolution line-shape experiments[11] we have observed linewidths as narrow as 150 kHz using a beam divergence of 10^{-4} rad.

Measurements were made of 16 of the 21 possible $P(13)$ hyperfine components and the results are shown in Table I. Lines a_{10}, a_{13}, a_{14}, a_{18}, and a_{19} were not included because a_{10} and a_{14} overlapped with $R(15)$ lines, a_{18} and a_{19} overlapped with each other, and a_{13} was found to coincide almost with a weak $\Delta F = 0$, $P(13)$ line. All but three lines were measured directly using line a_7 as the arbitrary zero. Lines a_{17}, a_{20}, and a_{21} were measured with respect to an additional line and then referenced to a_7. The measured values were repeatable to within a standard deviation of 5.2 kHz.

In order to describe the hyperfine structure accurately, it was necessary to construct a Hamiltonian which included the following interactions:

$$H_{\mathrm{hfs}} = H_{\mathrm{NEQ}} + H_{\mathrm{SR}} + H_{\mathrm{TSS}} + H_{\mathrm{SSS}} + H_{\mathrm{NMO}}.$$

The first three terms of the Hamiltonian have been included in previous analyses of iodine hyperfine spectra. These are, respectively, the nuclear electric quadrupole,[3,4] the magnetic spin-rotation,[3] and the tensor spin-spin[8] interactions. Whereas previously it was sufficient to calculate the quadrupole energy to second order,[4,7] in the present work, we also considered effects of third-order contributions to the interaction by including matrix elements off diagonal by $J' = J \pm 4$. This expanded the energy matrix to 105 $\times$105. However, by sorting the states with common F, a block-diagonal matrix is obtained in which the largest submatrix is only 11×11.

The fourth term in the Hamiltonian is the scalar part of the nuclear spin-spin interaction which results from the indirect electron-coupled spin-spin interaction. This term has the form $\vec{I}_1 \cdot \vec{I}_2$ and was first discussed by Ramsey and Purcell.[12] The matrix elements are

$$\langle JIF|H_{\mathrm{SSS}}|JIF\rangle = \tfrac{1}{2}D_s[I(I+1) - 2I_1(I_1+1)],$$

where D_s is the scalar spin-spin coupling strength and J, I, and F are the rotational, nuclear-spin, and total-angular-momentum quantum numbers respectively. In the case of iodine the nuclear spin I_1 takes on the value $\frac{5}{2}$.

The last term of the Hamiltonian, the magnetic octupole interaction, results from the coupling of the nuclear magnetic octupole moment with the third derivative of the molecular vector potential. The form of the matrix elements was discussed by Cassimir and Karreman,[13] and the first observation of a nuclear magnetic octupole interaction was made by Jaccarino *et al.*[14] in the ground state of atomic iodine. In the case of molecular iodine, the matrix elements of the magnetic octupole interaction are taken as[15]

$$\langle JIF|H_{\mathrm{NMO}}|JIF\rangle = \tfrac{1}{2}\Omega m(-1)^{J+I+F}\left[\frac{21(I+1)(I+2)(2I+1)(2I+3)}{I(I-1)(2I-1)(2J+3)(2J-1)}\right]^{1/2} J(J+1)(2J+1)\begin{Bmatrix} F & J & I \\ 3 & I & J \end{Bmatrix}\begin{Bmatrix} J & 2 & J \\ 3 & J & 1 \end{Bmatrix},$$

where Ωm is defined as the magnetic octupole coupling strength and the Wigner $6j$ symbols take on their usual meaning.

With use of the above matrix elements the coupling strengths associated with every term in the Hamiltonian were varied in a least-squares computer program to obtain the best fit to the data.

Table I shows the improvement in the fit as the various interactions are included in the Hamiltonian. The first column gives the line numbers and the second column gives the measured line positions relative to line a_7. The remaining columns labeled $F1$ through $F5$ give the measured minus calculated transition frequencies for the various fits. Also listed is the standard deviation for each fit. The hyperfine coupling strengths obtained from the best fit (column $F5$) are given in Table II where eQq, C, and D_t are the electric quadrupole, spin-rotation, and tensor spin-spin coupling strengths, respectively.

From Table I we see that the inclusion of the tensor nuclear spin-spin interaction (column $F2$) dramatically reduced the standard deviation of the fit to 12.2 kHz. The addition of the scalar

TABLE I. Measured frequencies of the hyperfine structure on the $P(13)$ 43-0 line in $^{127}I_2$ and fits. Column $F1$ includes H_{NEQ} (to second order) and H_{SR}; column $F2$ adds H_{TSS}; column $F3$ adds H_{SSS}; column $F4$ adds H_{NMO}; and column $F5$ adds H_{NEQ} (to third order). (a) Frequencies measured relative to this component. (b) Line not included in fit because of overlapping structure.

Line	Meas. Freq. (kHz)	Meas. - Calc. (kHz) F1	F2	F3	F4	F5
a_1	-386,761	-577	-10	-5	-1	0
a_2	-314,901	+693	0	-3	+5	+2
a_3	-254,994	-646	-2	+3	-8	-4
a_4	-178,956	-323	-8	0	-1	-1
a_5	-51,776	+467	+2	-3	+8	+6
a_6	-14,224	+388	+5	+1	-4	-5
a_7	0 (a)	-66	-1	+9	+6	+5
a_8	83,696	+145	0	+2	-4	-5
a_9	94,711	-148	-15	-6	+1	+1
a_{10}	(b)					
a_{11}	138,964	-196	+29	+16	+9	+11
a_{12}	180,592	-351	+12	-5	-2	0
a_{13}	(b)					
a_{14}	(b)					
a_{15}	332,387	+203	+4	+10	+5	+4
a_{16}	361,739	+19	-14	-8	-1	-2
a_{17}	405,908	+73	+7	-3	-3	-2
a_{18}	(b)					
a_{19}	(b)					
a_{20}	502,614	+241	-2	+6	+3	+2
a_{21}	562,302	+80	-10	-11	-11	-10
Std. dev. (kHz)		395	12.2	8.5	6.8	6.3

TABLE II. Molecular hyperfine structure coupling strengths (in units of kHz) obtained from the best fit ($F5$). Uncertainties in constants are 2σ. Primed constants are associated with the upper $^3\Pi$ state. Double-primed constants are associated with the lower $^1\Sigma$ state. C'', D_t'', and D_s'' were held fixed in the least-squares fit.

eQq'	$-554\,094 \pm 13$
eQq''	$-2\,448\,025 \pm 10$
C'	186.71 ± 0.10
C''	0.0
D_t'	-100.8 ± 1.1
D_t''	0.26
D_s'	-2.72 ± 1.0
D_s''	0.0
$\Omega m' - \Omega m''$	-2.17 ± 0.70

nuclear spin-spin term (column $F3$) improved the fit to 8.5 kHz, and the nuclear magnetic octupole interaction (column $F4$) improved the fit further to 6.5 kHz. Extending the quadrupole calculations to third order (column $F5$) improved the fit slightly to 6.3 kHz.

We performed a statistical F test[16] on the merits of including the scalar spin-spin and magnetic octupole interactions in the theoretical model. The results of the F test gave a better than 99% confidence level that the improvement in the fit for each of these terms was indeed real and not just statistical. The third-order corrections to the quadrupole interaction were included because they affected the final values of the quadrupole coupling strengths by as much as 60 kHz even though their improvement of the fit was small.

In the case of the scalar spin-spin interaction, the matrix elements, which depend only on I, are identical for the states connected by the transition, so that the effects of this interaction are due solely to the difference in the scalar coupling strengths for the upper and lower states. Hence, it is not possible to determine D_s' and D_s'' from the fit alone. Since the scalar spin-spin interaction is an electron-coupled interaction, we expect little contribution from the $^1\Sigma$ ground state so that the calculated value of $D_s' - D_s''$ is due primarily to D_s'.

The observation of the magnetic octupole interaction, which to our knowledge is the first such observation in excited electronic states, determines a coupling strength[17] which is the difference between upper- and lower-state values. An improvement in the data of at least an order of magnitude would be necessary in order to determine the constants for each state. Such an improvement is certainly possible since the present long-term stability of the molecular-beam-stabilized argon laser is 1×10^{-13} for integration times of 200 sec.[18]

With the advent of dye lasers, it should now be possible to perform highly precise measurements similar to the ones discussed here over the entire optical region of the spectrum. Dye-laser-excited hyperfine structure in a molecular beam of I_2 has already been observed[19] with a resolution of 1 part in 10^9. The measurement of the coupling strengths due to higher-order nuclear moments would therefore make possible a more complete study of electric- and magnetic-field distributions in excited electronic states of atoms and molecules.

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