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Search for the Nuclear Electric Hexadecapole Moment of ^{115}In by Molecular-Beam Electric-Resonance Spectroscopy*

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This experiment with a molecular-beam electric-resonance spectrometer determined the hyperfine structure of indium fluoride, $^{115}\text{In}^{19}\text{F}$, to an accuracy of 1 part in 10^6 . The experiment was an attempt to observe the interaction of a nuclear electric hexadecapole moment of ^{115}In that a simple order of magnitude calculation and a previous experiment showed might barely be observable. Although a hexadecapole interaction energy larger than 500 Hz could have been detected, no evidence of the interaction was found. To understand this result, the electric hexadecapole moment was calculated on the basis of a model in which the nuclear electric charge density was assumed to be uniform within a surface defined by $r(\theta, \phi) = R_0[1 + \beta_2 P_2(\cos\theta) + \beta_4 P_4(\cos\theta)]$, where β_2 and β_4 are taken to have the same values as the corresponding parameters for the nuclear potential. The value of the hexadecapole interaction energy in $^{115}\text{In}^{19}\text{F}$ predicted by the model is 10 Hz, well below the limit of detectability with the spectrometer employed in this experiment.

I. INTRODUCTION

We used a high-resolution molecular-beam electric-resonance spectrometer to measure the hyperfine structure (hfs) of the $J=1$ and $J=2$ rotational states of indium fluoride, $^{115}\text{In}^{19}\text{F}$. The experiment was undertaken because there seemed to be a good chance that the interaction between the electric hexadecapole moment of the ^{115}In nucleus and the fourth derivative of the electric potential in the InF molecule was observable. A spectroscopically measured hexadecapole interaction would be of interest because the hexadecapole moment, which could be calculated from the interaction, is closely related to a Y_{40} deformation in the nuclear shape. This deformation has recently come under both theoretical¹ and experimental² investigation.

Experiments have shown that in $^1\Sigma$ molecules

the hfs that arises from the electric quadrupole interaction of nuclei with spin $I \leq \frac{3}{2}$ is very well understood; thus, one expects that the effect of a nuclear electric hexadecapole interaction could be detected if the hfs of a $^1\Sigma$ molecule containing a nucleus with $I \geq 2$ were carefully measured. On the basis of measurements on TlCl ,³ TlBr ,⁴ and TlI ,⁵ one concludes that a hexadecapole interaction could be identified if that interaction shifted the hfs transition frequencies by 500 Hz or more. Because a nucleus must have spin angular momentum $I \geq 2$ in order to exhibit an electric hexadecapole moment, only TlI of the three molecules mentioned might show a nuclear hexadecapole interaction. The interaction was not observed in TlI, but an upper limit of 500 Hz was placed on the interaction constant.

An estimate⁶ shows that the ratio of the hexadecapole interaction constant (ehH) to the quadru-

pole constant (eqQ) in the $^{115}\text{In}^{19}\text{F}$ molecule should be on the order of 10^{-6} . The estimate includes antishielding factors⁷ for both the eqQ and the ehH interactions. On this basis one would expect $ehH \approx 10^3$ Hz.

Other evidence for an observable hexadecapole moment in the In nucleus had been reported by Mahler, James, and Tantilla.⁸ They observed ultrasonically induced, $\Delta m = \pm 3$, ^{115}In nuclear spin transitions in an InAs single crystal. Although $\Delta m = \pm 3$ transitions can result from a number of effects including magnetic octupole and electric hexadecapole interactions, Mahler and his colleagues argued from interaction-energy considerations that the transitions were due to the hexadecapole interaction. Assuming an antishielding factor of 10^5 , they estimated that ^{115}In nucleus has a hexadecapole moment of about 10^{-47} cm⁴. A moment of this magnitude would generate an ehH of about 10^4 Hz in $^{115}\text{In}^{19}\text{F}$, and hyperfine interaction constants of this size are readily measured with the Michigan molecular-beam electric-resonance spectrometer.

II. MOLECULAR HYPERFINE STRUCTURE

A. Hamiltonian

Since the fluorine nucleus has spin $I_2 = \frac{1}{2}$, the appropriate Hamiltonian for $^{115}\text{In}^{19}\text{F}$ in the $^1\Sigma$ state is

$$H = F_{v,J} + Q^{(2)} : V^{(2)} + H^{(4)} : V^{(4)} + c_1 \vec{I}_1 \cdot \vec{J} + c_2 \vec{I}_2 \cdot \vec{J} + c_3 \vec{I}_1 \cdot d(\vec{J}) \cdot \vec{I}_2 + c_4 \vec{I}_1 \cdot \vec{I}_2 - \vec{\mu}_e \cdot \vec{E} - g_J \mu_o \vec{J} \cdot \vec{H} - \mu_N \vec{H} \cdot (g_1 \vec{I}_1 + g_2 \vec{I}_2). \quad (1)$$

The $F_{v,J}$ term is the usual operator for a vibrating

$$\begin{aligned} \langle (I_1, J) F_1 I_2 F, M_F | Q^{(2)} : V^{(2)} | (I_1, J) F_1 I_2 F, M_F \rangle &\equiv eqQ F(I_1, J, F_1), \\ \langle (I_1, J) F_1 I_2 F, M_F | H^{(4)} : V^{(4)} | (I_1, J) F_1 I_2 F, M_F \rangle &\equiv ehH G(I_1, J, F_1). \end{aligned}$$

Here q and h are the expectation values of the second and fourth derivative, respectively, of the electric potential at the site of the nucleus; eqQ and ehH are the spectroscopically observed quadrupole and hexadecapole moments of the nucleus; F and G are known functions of the quantum numbers I_1, J , and F_1 .

C. Intrinsic and Spectroscopic Nuclear Electric Moments

The intrinsic multipole moments, Q_0 and H_0 , are not equal to the spectroscopically observed moments, Q and H . This quantum mechanical effect arises from the rotation of the intrinsic frame

rotor. The next two terms represent the interaction of the electric quadrupole and electric hexadecapole moments of the indium nucleus with the second and fourth derivatives of the potential. The terms c_1 and c_2 contain the interaction of the nuclear magnetic moment of the indium and fluorine with magnetic field which arises from the rotation of the molecule. The sixth and seventh terms are operators for the tensor and scalar parts of the nuclear spin-spin interaction. The remaining terms represent the interaction of the molecule with external electric and magnetic fields.

The magnetic octupole interaction is too small to show up in the hyperfine spectrum of TlCl and TlBr as observed with our spectrometer.^{3,4} Since the octupole moments of the chlorine and bromine nuclei are comparable to the octupole moment of indium,⁹ we feel justified in omitting the octupole term from the Hamiltonian of Eq. (1).

B. Matrix Elements

The dominant feature of the hyperfine structure of $^{115}\text{In}^{19}\text{F}$ is the electric quadrupole interaction that couples the spin of the indium nucleus (I_1) to the molecular rotation (J). The matrix elements, expressed in the appropriate

$$|(I_1, J) F_1, I_2, F, M_F\rangle$$

representation, have been given in detail elsewhere.^{5,10} We write the matrix elements of the nuclear electric multipole interactions in order to introduce the symbols for the quadrupole and hexadecapole interaction constants, eqQ and ehH :

about the laboratory frame. Mottelson¹¹ showed that

$$Q = Q_0 (2I+1) (-1)^{I-K} \begin{pmatrix} I & 2 & I \\ -I & 0 & I \end{pmatrix} \begin{pmatrix} I & 2 & I \\ -K & 0 & K \end{pmatrix}, \quad (2)$$

where K is the projection of I , the nuclear spin, on the intrinsic z axis, and the terms in parentheses are 3- j symbols. For most nuclei in the ground state $K=1$, hence

$$Q = Q_0 \frac{I(2I-1)}{(I+1)(2I+3)}. \quad (3)$$

Note that $Q=0$ when $I=\frac{1}{2}$ regardless of the value of Q_0 and that Q is smaller than Q_0 by a factor of

$\frac{1}{5}$ for $I = \frac{3}{2}$ and $\frac{6}{11}$ for $I = \frac{5}{2}$. Löbner, Vetter, and Honig¹² used this equation in the calculation of β_2 from spectroscopic quadrupole moments.

The similar relation between the intrinsic and spectroscopic hexadecapole is¹³:

$$H = H_0(2I+1)(-1)^{I-K} \begin{pmatrix} I & 4 & I \\ -I & 0 & I \end{pmatrix} \begin{pmatrix} I & 4 & I \\ -K & 0 & K \end{pmatrix}. \quad (4)$$

Note that $H = 0$ when $I < 2$ regardless of the value of H_0 . If $K = 1$, the spectroscopic H is smaller than the intrinsic H_0 by a factor of $\frac{1}{42}$ for $I = \frac{5}{2}$, $\frac{7}{99}$ for $I = \frac{7}{2}$, and $\frac{18}{143}$ for $I = \frac{9}{2}$.

D. Antishielding and Pseudo-hexadecapole Effects

Antishielding is an enhancement of the quadrupole (or hexadecapole) interaction caused by excitations in the electron core when one considers the quadrupole (or hexadecapole) moment of the nucleus as a perturbation.⁷ The measured quadrupole and hexadecapole coupling constants can be written:

$$eqQ = eq'(1 - \gamma_\infty)Q,$$

$$ehH = eh'(1 - \eta_\infty)H,$$

where the effects of the electron core excitations are contained in the antishielding factors γ_∞ and η_∞ . The antishielding factors are typically negative and much larger than unity. The expectation values of the second and fourth derivatives of the electric potential (neglecting the contributions from the electron core excitation) are denoted by q' and h' . Q and H are the spectroscopically observable moments as discussed in Sec. IIC above.

The antishielding factors, as they have been described above, do not change the I_1 , J , and F_1 dependence of the quadrupole or hexadecapole interactions. However, Sternheimer¹⁴ has pointed

out that the quadrupole antishielding effects generate, in second order, contributions to the hyperfine energy that have the same dependence on I_1 , J , and F_1 as does the $H^{(4)} : V^{(4)}$ term in the Hamiltonian. The strength of this pseudo-hexadecapole effect, which Sternheimer denotes as $H_{\text{ind. by } Q}$, is proportional to Q^2 . Sternheimer estimates that

$$\frac{H_{\text{ind. by } Q}}{(1 - \eta_\infty)H} \approx 0.1 \frac{Q^2}{H}.$$

If $H \approx 10^{-47} \text{ cm}^4$, this ratio is roughly 10^{-2} so that $H_{\text{ind. by } Q}$ can be neglected. However, if $H \approx 10^{-49} \text{ cm}^4$ (as estimated in Table II), $H_{\text{ind. by } Q}$ must be considered for the proper identification of a true nuclear hexadecapole interaction in an observed spectrum.

E. Energy Levels

Perturbation theory is used to calculate the hyperfine energy levels of the molecule. Contributions to the energy from the electric quadrupole and magnetic dipole interactions of the indium nucleus are carried out to third order; contributions from the magnetic dipole interactions of the fluorine nucleus are carried to second order. We have determined by experiment that the contributions from Stark and Zeeman effects to the final data are small enough to be neglected, so operators for the interactions with external fields are not included in the Hamiltonian used for our analysis.

F. Isolation of the Hexadecapole Effect

The isolation and measurement of the hexadecapole interaction is attempted as follows: First, a least-squares fit to $J = 2$, $v = 0$ spectrum is made with five constants (eqQ , c_1 , c_2 , c_3 , and c_4), and

TABLE I. Hyperfine interaction constants of $^{115}\text{In } ^{19}\text{F}$ in kHz.

State	eqQ	$c_1(\text{In})$	$c_2(\text{F})$	c_3	c_4
This experiment (molecular-beam electric resonance)					
$J = 1, v = 0$	-723 799.6(2)	17.50(1)	18.77(10)	2.62(3)	-2.15(3)
$J = 1, v = 1$	-717 115(50)	17.50 ^a	18.77 ^a	2.62 ^a	-2.15 ^a
$J = 1, v = 2$	-710 490(50)	17.50 ^a	18.77 ^a	2.62 ^a	-2.15 ^a
$J = 2, v = 0$	-723 789.6(2)	17.50(1)	18.76(3)	2.62(1)	-2.11(1)
$J = 2, v = 1$	-717 062(50)	17.50 ^a	18.76 ^a	2.62 ^a	-2.11 ^a
Previous experiment ^b (microwave absorption)					
$J = 1 \rightarrow 2, v = 0$	-723 740(150)	17(4)			
$J = 1 \rightarrow 2, v = 1$	-717 100(150)	17(4)			
$J = 1 \rightarrow 2, v = 2$	-710 460(150)	17(4)			

^a The constants c_1 , c_2 , c_3 , and c_4 in the higher vibrational states were assumed to be the same as in the $v = 0$ state so that eqQ could be calculated.

^b J. Hoefl, F. J. Lovas, E. Tiemann, and T. Törring, Z. Naturforsch. 25a, 1029 (1970).

the goodness of this fit is compared to the one found if the least-squares fit is made with six interaction constants (eqQ , ehH , c_1 , c_2 , c_3 , and c_4). Next, a least-squares fit to the $J=1$, $v=0$ hyperfine spectrum is made using the Hamiltonian with five interaction constants eqQ , c_1 , c_2 , c_3 , and c_4 . (We note that the hexadecapole contribution vanishes in the $J=1$ state.) To present the hfs as convincing evidence for an electric hexadecapole moment of the indium nucleus, we would demand that the addition of the hexadecapole term to the Hamiltonian significantly improve the fit of theoretical predictions to the $J=2$ spectrum. We would also demand that all the other hyperfine constants as determined from the $J=2$ spectrum be consistent with (or differ only in a clearly understandable way from) those determined from $J=1$.

III. EXPERIMENT

The molecular-beam spectrometer used for this study is described in detail in other reports.^{13, 15, 16} Only a few important features are given here.

InF was produced as it was being used in the spectrometer by the reaction of In and MgF_2 in a tantalum tube oven heated to a temperature of about 1300°K. The molecular beam issues from a 0.01-cm-diam hole in the side of the tube. The detector, 1.62 m from the oven, is a hot tungsten-wire surface ionizer. The signal from state selected molecules is about 7×10^{-13} A on a background of about 1×10^{-13} A. The most intense transition results in a detector current change of about 1×10^{-14} A. The raw signal-to-noise ratio for these intense transitions is about 5:1. Averaging the results over several measurements improves the signal-to-noise by a factor of 4 or so. The linewidth is approximately 500 Hz.

IV. RESULTS

A. Hyperfine Interaction Constants

We were able to measure eight lines in the $J=1$, $v=0$ spectrum and nine lines in the $J=2$, $v=0$ spectrum of $^{115}\text{In}^{19}\text{F}$. From these lines and the Hamiltonian without a hexadecapole term, we calculated the hyperfine interaction constants. The constants (in kHz) are given in Table I. The errors quoted for the constants are estimated from changes in the constants when the observed transition frequencies are given various weights in the least-squares calculation. The values of the constants given are calculated with all lines weighted equally. We note that the quadrupole interaction constant changes by 10 kHz between

the $v=0$, $J=1$ and the $v=0$, $J=2$ states, but this relatively large shift is in qualitative agreement with theoretical estimates¹⁷⁻¹⁹ for the effect of centrifugal stretching on eqQ . The hfs constants, c_1 , c_2 , c_3 , and c_4 show no significant dependence on rotational state.

B. Upper Limit for Hexadecapole Moment of Indium

The interaction constants given in Table I predict the measured hyperfine spectrum within 80 Hz for $J=2$. When the hexadecapole term is included in the least-squares calculation for the constants, one finds $ehH = 228$ Hz, while the values of the other constants differ insignificantly from those given in Table I; one also finds that the fit to the nine lines in the $J=2$ spectrum is now within 45 Hz. This improvement is not surprising, since the same number of transitions are now being fitted with six constants instead of five. Since the lack of a detailed line-shape analysis and the spectrometer noise lead to an uncertainty on the order of 100 Hz in the frequency assigned to each of the observed transitions, however, we cannot consider the slight improvement in fit as clear evidence for the observation of the hexadecapole interaction. We conclude that a Hamiltonian without a hexadecapole term adequately describes the molecule's hyperfine structure.

If the hexadecapole interaction were as large as 2000 Hz, on the other hand, the result would be a shift in the line frequencies by something on the order of 200 Hz, large enough to be clearly observed with our spectrometer. We conclude that ehH is less than 2000 Hz. To interpret this in terms of a nuclear hexadecapole moment, we take the field gradient to have the value $h' = 7 \times 10^{30}$ esu,²⁰ and use the value of hexadecapole anti-shielding $\eta_\infty = -3680$ ²¹ to conclude that the intrinsic hexadecapole moment of the indium nucleus is less than 1×10^{-48} cm⁴.

V. DISCUSSION

The possible evidence for a large ($\sim 10^{-47}$ cm⁴) electric hexadecapole moment of the ^{115}In nucleus was reported by Mahler, James, and Tantilla,⁸ who assumed an antishielding factor $\eta_\infty = -10^5$. If the recently calculated antishielding factor²¹ of $\eta_\infty = -3.7 \times 10^3$ is used, the hexadecapole moment of ^{115}In implied by the work of Mahler and his colleagues becomes $\sim 2 \times 10^{-46}$ cm⁴. It seems unlikely that the present experiment could have missed the effect of a hexadecapole moment as large as that, some 200 times larger than the upper limit reported in Sec. IV B.

To aid in the selection of a future candidate for a hexadecapole search and, indeed, to help decide if future searches are justified at all, a calculation of the size of the hexadecapole moments for various nuclei is described here.

A. Model for Nuclear Electric Multipole Moments

In a coordinate frame (r, θ, ϕ) fixed in the nucleus, assume that the nuclear electric charge density is uniform inside the surface given by

$$r(\theta, \phi) = \lambda R_0 [1 + \beta_2 P_2(\cos\theta) + \beta_4 P_4(\cos\theta)] \quad (5)$$

and zero outside that surface. R_0 , the average radius of a nucleus with A nucleons, is given by $R_0 = r_0 A^{1/3}$, where r_0 is 1.3×10^{-13} cm. The volume enclosed by the surface is

$$V = \frac{4}{3} R_0^3 \lambda^3 (1 + \frac{3}{5} \beta_2^2 + \frac{1}{3} \beta_4^2 + \dots), \quad (6)$$

where terms on the order of β_2^3 and beyond are negligibly small. Since this volume is to be equal to the volume of the equivalent sphere, $\frac{4}{3} \pi R_0^3$, the parameter λ is given by

$$\lambda = (1 + \frac{3}{5} \beta_2^2 + \frac{1}{3} \beta_4^2 + \dots)^{-1/3}. \quad (7)$$

The charge density everywhere within the nuclear surface is simply $\rho = eZ/\frac{4}{3}\pi r_0^3 A$. Thus, the intrinsic electric quadrupole and hexadecapole moments, as defined by $Q_0 = \frac{2}{3} \int \rho r^2 P_2(\cos\theta) d\tau$ and $H_0 = \frac{8}{3} \int \rho r^4 P_4(\cos\theta) d\tau$, become¹³:

$$Q_0 = \frac{6}{5} Z r_0^2 A^{2/3} (1 - \frac{3}{5} \beta_2^2 - \frac{1}{3} \beta_4^2)^{5/3} \times \alpha_2 (1 + \frac{20}{35} \beta_2 + \frac{40}{35} \beta_4) + \frac{200}{593} \beta_4^2 + \dots \quad (8)$$

$$H_0 = 4 Z r_0^4 A^{4/3} (1 - \frac{3}{5} \beta_2^2 - \frac{1}{3} \beta_4^2)^{7/3} \times \beta_4 (\frac{2}{3} + \frac{80}{77} \beta_2 + \frac{324}{1051} \beta_4) + \frac{36}{35} \beta_2^2 + \dots \quad (9)$$

To calculate the nuclear electric multipole moments from Eqs. (8) and (9), one needs values for the deformation parameters β_2 and β_4 that characterize the charge distribution. In the absence of adequate information about β_2 and β_4 , the present model uses the assumption that deformation parameters for the nuclear potential also describe the electric charge distribution. Thus, values of the nuclear potential parameters α_2 and α_4 , as calculated by Seeger²² on the basis of the Nilsson and liquid-drop models, are taken as the values of β_2 and β_4 .

B. Quadrupole Moments as a Test of the Model

The adequacy of the above model can be tested by comparing its predictions with the experimental values for the intrinsic electric quadrupole moments. Löbner, Vetter, and Honig¹² used a model similar to the one described here (albeit with β_4

= 0) together with experimental values²³ of quadrupole moments to obtain values of β_2 for a wide variety of nuclei. We have extended their calculations to ¹¹⁵In and ¹²¹Sb. The parameters β_2 are usually within 30% of being consistent with Seeger's values for α_2 ; the worst cases are less than a factor of 3 apart. This is a promising indication that the nuclear potential parameters can be used to make reasonable estimates of the nuclear electric multipole moments.

C. Model Predictions for Hexadecapole Moments

Effects ascribed to nuclear electric hexadecapole interactions were first reported for the nuclei of antimony (¹²¹Sb, ¹²³Sb)²⁴ and indium (¹¹⁵In),⁸ but reliable values for the hexadecapole moments cannot be obtained from these early experiments. However, recent Coulomb-excitation experiments have determined the intrinsic hexadecapole moments of ^{152, 154}Sm,²⁵ and of ²³⁰Th and ²³⁸U.²⁶ Using a uniform-charge model similar to the one described in Sec. V A above, those authors calculate charge deformation parameters that are within 50% of Seeger's values of the nuclear potential parameters.

In addition, the theoretical values of the nuclear potential parameters α_4 agree relatively well (~50%) with α -particle scattering experiments done by Hendrie *et al.*² and by Moss *et al.*²⁷ Thus, it is reasonable to suppose that theoretical parameters of the nuclear potential can be used to get reasonable estimates for the electric hexadecapole moments of other nuclei.

Table II shows the predictions of the spectroscopic hexadecapole moments and (when available) the antishielding factors⁷ for stable nuclei that are expected to have hexadecapole interactions larger than that of indium. If we take 7×10^{90} esu as a typical value for h' in the host molecule, and if H is expressed in units of 10^{-48} cm⁴, then $\eta_\infty H$

TABLE II. Predictions of spectroscopic nuclear electric hexadecapole moments.

Nucleus	$ H $ (10^{-48} cm ⁴)	$ \eta_\infty $ (Ref. 7)
¹¹⁵ In	0.008	3680
^{161, 163} Dy	0.18	
¹⁶⁵ Ho	0.16	
¹⁶⁷ Er	0.12	
¹⁷⁶ Yb	0.03	
¹⁷⁵ Lu	0.06	
¹⁸¹ Ta	0.008	
^{185, 187} Re	0.015	~50 000
²³⁵ U	0.52	
^{241, 243} Am	0.18	

must be 3000 or larger [i.e., $eh'(1 - \eta_\infty)H \geq 10^3$ Hz] if the interaction is to be observed with the resolution of the present spectrometer. The factor $\eta_\infty H$ is 30 for indium, about 2 orders of magnitude too small; the factor is 750 for rhenium, still a factor of 4 too small. The hexadecapole interaction may be observable in uranium if the antishielding factor, η_∞ , were 6000 or larger, and in dysprosium or americium if the antishielding factor were 15 000 or larger. Unfortunately, the antishielding factors for these last-named nuclei are yet to be calculated.

Note added in proof: Recent atomic beam magnetic resonance experiments provide evidence for nuclear electric hexadecapole interactions in isotopes of holmium and dysprosium. Penselin,

Dankwort, and Ferch²⁸ report hexadecapole moments that have a magnitude on the order of 0.5×10^{-48} cm⁴ (uncertain to about 30%); these are in good agreement with the model predictions given in Table II.

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