For explicit calculations, we have averaged (A8) over all directions of the vector q; this gives

$$\langle I_q \rangle_{av} = 2J^2 Z(\sin qb/qb). \tag{A9}$$

Here, J is the exchange integral between two neighboring spins, and b is the lattice parameter. It may be worthwhile remarking that this usual averaging procedure, which is consistent with the experimental situation, has the nontrivial effect of destroying the periodicity of I_q in different Brillouin zones.

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Nuclear Hexadecapole Interactions

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The nuclear hexadecapole matrix elements for the static and the one-phonon nuclear interactions are developed and are evaluated for $\Delta m = \pm 3$ and $\Delta m = \pm 4$ nuclear transitions involving a spin-$\frac{9}{2}$ nucleus in a crystal with $\bar{4}3m$ symmetry. An expression for the saturation factor for a general interaction which gives rise to nuclear-spin transitions involving the change in the z component of the spin by any amount $\Delta m = \pm n$ is developed and is used to derive the angular variation of the one-phonon, $\Delta m = \pm 3$ and $\Delta m = \pm 4$ nuclear hexadecapole interactions. Finally a way to end the speculation about the observation of the hexadecapole interaction is presented.

1. INTRODUCTION

SINCE the first observation of the interaction of the nucleus with its environment through its electric moment, many experiments have been done to investigate both the static and the time-dependent effects of this phenomenon. In 1948, Pound[1] demonstrated that the time-dependent quadrupole interaction was responsible for the relaxation of the Br^{79} and Br^{81} nuclei in solution. He then observed the static effect through the splitting of the nuclear-resonance lines in a crystal with lower than cubic symmetry.[2] In 1956, Proctor and Tanttila[3] observed externally induced Cl^{35} quadrupole transitions between the degenerate quadrupole levels in $NaClO_3$.

In 1955, Wang[4] postulated that an unexplained shift in the pure quadrupole spectra in Sb^{121} and Sb^{123} was due to the static nuclear-electric hexadecapole interaction. In 1966, externally induced hexadecapole transitions between magnetically split In^{115} levels in InAs were believed to have been observed.[5]

The nuclear-electric moments are coupled to their electronic environment through the electric-field gradients of the electronic charge. To first approximation in an ionic crystal, the electronic charge is symmetric about the nucleus; thus there is no coupling between the nucleus and its surrounding electrons. In this approximation, the electric-field gradients arise solely from charges external to the ion. However, there is a distortion from this spherical symmetry due to the interaction with external charges and with the nuclear-quadrupole moment, which gives rise to an additive coupling characterized by an antishielding factor γ. Sternheimer and others have calculated these antishielding factors for both the quadrupole[6–10] and the hexadecapole interaction.[11,12] In addition to the antishielding factor, there is an additional contribution to the hexadecapole coupling due to the perturbation of the ion by the field of the nuclear-quadrupole moment.[13]

2. THEORY

The interaction energy of a nuclear-charge distribution $\rho_N(r_N)d\tau_N$ and an electron-charge distribution $\rho_E(r_E)d\tau_E$ can be written

$$U = \int_N \int_E \rho_N(r_N) \frac{\rho_E(r_E)}{|\mathbf{r}_E - \mathbf{r}_N|} d\tau_N d\tau_E. \tag{1}$$

Assuming the electron does not penetrate the nucleus,

[1] R. V. Pound, Phys. Rev. **73**, 1247 (1948).
[2] R. V. Pound, Phys. Rev. **79**, 685 (1950).
[3] W. G. Proctor and W. H. Tanttila, Phys. Rev. **101**, 1757 (1956).
[4] T. C. Wang, Phys. Rev. **99**, 566 (1955).
[5] R. J. Mahler, L. W. James, and W. H. Tanttila, Phys. Rev. Letters **16**, 259 (1966).
[6] R. M. Sternheimer, Phys. Rev. **80**, 102 (1950).
[7] R. M. Sternheimer, Phys. Rev. **84**, 244 (1951).
[8] H. M. Foley, R. M. Sternheimer, and D. Tycko, Phys. Rev. **93**, 734 (1954).
[9] R. M. Sternheimer and H. M. Foley, Phys. Rev. **92**, 1460 (1953).
[10] R. M. Sternheimer and H. M. Foley, Phys. Rev. **102**, 731 (1956).
[11] R. M. Sternheimer, Phys. Rev. Letters **6**, 190 (1961).
[12] R. M. Sternheimer, Phys. Rev. **123**, 870 (1961).
[13] R. M. Sternheimer, Phys. Rev. **127**, 812 (1962).

we can expand the integrand of Eq. (1) to obtain

$$U=\sum_{l=0}^{\infty}\sum_{n=-l}^{+l}N_l^nE_l^{n*}, \quad (2)$$

where

$$N_l^n=\left[\frac{4\pi}{2l+1}\right]^{1/2}\int_N r_N^l\rho_N(r_N)Y_l^{n*}(\theta_N,\phi_N)d\tau_N, \quad (3)$$

and

$$E_l^{n*}=\left[\frac{4\pi}{2l+1}\right]^{1/2}\int_E \frac{Y_l^n(\theta_E,\phi_E)}{r_E^{l+1}}\rho_E(r_E)d\tau_E.$$

If we choose suitable nuclear wave functions and express the N_l^n as expectation values of nuclear operators, odd values of l will be forbidden since stationary nuclear states have definite parity. The five operators for $l=2$ are the nuclear-quadrupole operators, and the nine operators corresponding to $l=4$ are the hexadecapole operators. We require that operator expressions for the N_4^n terms have the same matrix elements, aside from a constant, as the Hermitian operators formed by the spin operators, $I_\pm$, I_z,[14] which results in the following hexadecapole operators:

$$\begin{aligned}
M_{16}^0&=AB^0,\\
M_{16}^{\pm1}&=5^{1/2}AB^{\pm1},\\
M_{16}^{\pm2}&=\tfrac{1}{2}(10)^{1/2}AB^{\pm2}, \quad (4)\\
M_{16}^{\pm3}&=(35)^{1/2}AB^{\pm3},\\
M_{16}^{\pm4}&=\tfrac{1}{2}(70)^{1/2}AB^{\pm4},
\end{aligned}$$

where

$$A=eM_{16}/16I(I-1)(2I-1)(2I-3),$$

$$eM_{16}=\langle II|\sum_{i=1}^{A}(35z_i^4-30r_i^2z_i^2+3r_i^4)|II\rangle,$$

$$B^0=[35I_z^4-30I^2I_z^2+25I_z^2-6I^2+3I^4]_{\text{op}},$$

$$B^{\pm1}=[(7I_z^3-3I^2I_z-I_z)I_\pm+I_\pm(7I_z^3-3I^2I_z-I_z)]_{\text{op}},$$

$$B^{\pm2}=[(7I_z^2-I^2-5)(I_\pm)^2+(I_\pm)^2(7I_z^2-I^2-5)]_{\text{op}},$$

$$B^{\pm3}=[I_z(I_\pm)^3+(I_\pm)^3I_z]_{\text{op}},$$

$$B^{\pm4}=[(I_\pm)^4]_{\text{op}},$$

$$I_\pm=(I_x\pm iI_y).$$

In the above, the $M_{16}^{\pm n}$ are the hexadecapole operator equivalents of the N_l^n terms for $l=4$. (Expressions similar to the $B^{\pm n}$ have appeared in the literature.[15])

Proceeding along similar lines, we can express the electronic terms in Eq. (2) as

$$\begin{aligned}
E_{16}^0&=2V^0,\\
E_{16}^{\pm1}&=\tfrac{4}{5}5^{1/2}V^{\pm1},\\
E_{16}^{\pm2}&=\tfrac{2}{5}(10)^{1/2}V^{\pm2}, \quad (5)\\
E_{16}^{\pm3}&=(4/35)(35)^{1/2}V^{\pm3},\\
E_{16}^{\pm4}&=(1/35)(70)^{1/2}V^{\pm4},
\end{aligned}$$

where

$$\begin{aligned}
V^0&=(1/24)(V_{zzzz}),\\
V^{\pm1}&=(1/12)(V_{zzzx}\pm iV_{zzzy}),\\
V^{\pm2}&=(1/24)(V_{zzxx}-V_{zzyy}\pm 2iV_{zzxy}),\\
V^{\pm3}&=(1/12)[V_{zxxx}-3V_{zxyy}\pm i(3V_{zxxy}-V_{zyyy})],\\
V^{\pm4}&=(1/48)[V_{xxxx}+V_{yyyy}-6V_{xxyy}\\
&\qquad\pm 4i(V_{xxxy}-V_{xyyy})].
\end{aligned}$$

In the above expressions,

$$V_{xyzx}\equiv[(\partial^4/\partial x\partial y\partial z\partial x)V]_0,$$

where V is the electronic potential, and $[\ ]_0$ signifies the evaluation of the quantity in the bracket at the nuclear site.

The terms in Eq. (2) corresponding to the hexadecapole Hamiltonian then reduce to the simple expression

$$\begin{aligned}
H_{16}&=\sum_{n=-4}^{n=+4}M_{16}^nE_{16}^{n*}=\sum_{n=-4}^{n=+4}M_{16}^nE_{16}^{-n}\\
&=A(B^0V^0+B^{+1}V^{-1}+\cdots+B^{+4}V^{-4}+B^{-4}V^{+4}). \quad (6)
\end{aligned}$$

The first term in the expansion in Eq. (6) gives rise to a small static shift in the energy levels and was used to explain the small perturbation on the pure quadrupole spectra[4] of Sb^{121} and Sb^{123}. The $n=1$, 2, 3, and 4 terms will be shown to give rise to the interaction of phonons with the spin system, corresponding to one-phonon $\Delta m=\pm1$, ±2, ±3, and ±4 nuclear spin transitions, respectively. It should be expected that all the terms in the expansion are extremely small, and since there are competing $\Delta m=\pm1$ dipole and $\Delta m=\pm1$ and $\Delta m=\pm2$ quadrupole interactions, one should not be able to detect these phonon-induced hexadecapole transitions. For the $\Delta m=\pm3$ transitions, there is a competing magnetic-octupole process that has been shown to be weaker than the hexadecapole interaction.[5] Finally, for the $\Delta m=\pm4$ transitions, there is no other interaction involving nuclear moments of lower order that will give rise directly to such a transition.

3. HEXADECAPOLE-PHONON INTERACTIONS

In order to investigate the interaction of phonons with the spin system, it will be necessary to introduce phonon operators into the expression for the interaction energy, Eq. (6). This is done by expanding the terms

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[14] See, for example, A. Abragam, *The Principles of Nuclear Magnetism* (Oxford University Press, London, 1961), pp. 159–166.

[15] K. W. H. Stevens, Proc. Phys. Soc. (London) **A65**, 209 (1952); E. Ambler, J. C. Eisenstein, and J. F. Schooley, J. Math. Phys. **3**, 118 (1962).

in the expansion in a Taylor series about the equilibrium position of the nucleus. If ξ_i^α denotes the ith component of the displacement of the αth particle from its equilibrium position, the Taylor series can be written

$$H_{mm'}(n)=AB^{\pm n}V^{\mp n}$$
$$=[AB^{\pm n}V^{\mp n}]_0+AB^{\pm n}\sum_{i,\alpha}\left[\frac{\partial}{\partial\xi_i^\alpha}V^{\mp n}\right]_0\xi_i^\alpha$$
$$+\frac{1}{2!}AB^{\pm n}\sum_{\substack{i,\alpha\\ j,\beta}}\left[\frac{\partial^2}{\partial\xi_i^\alpha\partial\xi_j^\beta}V^{\mp n}\right]_0\xi_i^\alpha\xi_j^\beta+\cdots, \quad (7)$$

where $n>0$ and $\sum_\alpha$ denotes a lattice sum. From this point we have two approaches to the problem. The first approach involves expanding the displacements ξ in terms of phonon operators. Such an expansion clearly shows that the second term on the right side of Eq. (7) describes the creation or annihilation of one phonon coupled with an energy conserving nuclear-spin transition, and the third term describes the interaction of two phonons with the spin system coupled with an energy conserving nuclear transition. For $n=1$, the energy conserving nuclear transition is a $\Delta m=\pm1$ transition; similarly, $n=2$, 3, and 4 correspond to $\Delta m=\pm2$, 3, and 4 transitions, respectively. Since we are interested only in the interaction between a longitudinal phonon of a particular frequency and direction of propagation with the nuclear-spin system, the problem is simplified a great deal, but the result of such an approach yields a solution in terms of internal-energy density of externally added phonons, antishielding factors, and lattice terms of which we have, at best, only scant knowledge. We choose, therefore, a second approach which deals with lattice symmetries. This approach yields as a result the angular variation of both the static and hexadecapole-phonon interaction matrix elements.

4. ANGULAR DEPENDENCE IN A CRYSTAL WITH $\bar{4}3m$ SYMMETRY

In Fig. 1 is shown the unit-cell structure of a crystal with $\bar{4}3m$ symmetry, as well as a set of axes which we will define as the [1,0,0] frame. A nucleus of type A is located at the origin, surrounded by 4 nearest neighbors of type B located at (g,g,g), $(g,-g,-g)$, $(-g,-g,g)$, and $(-g,g,-g)$, where g is one-half the usual lattice constant. For such cubic symmetry, $(V_{xx})_0=(V_{yy})_0=(V_{zz})_0$, and supposedly the Laplacian is zero when evaluated at the nuclear site; hence the static quadrupole interaction is zero.

If we consider only the $n=0$ term in Eq. (6) we obtain

$$H_{16}{}^0=AB^0(m^2)V^0.$$

It can be seen that this splitting will be degenerate in $\pm m$ (just as in the quadrupole case). For the particular case of a spin-$\frac{9}{2}$ nucleus in a crystal with $\bar{4}3m$ symmetry,

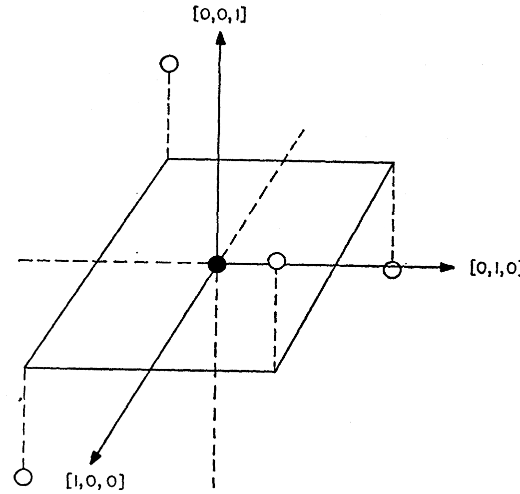

FIG. 1. Unit cell structure of a crystal with $\bar{4}3m$ symmetry. The axes shown define the [1,0,0] frame; the (1,0,0), (0,1,0), and (0,0,1) axes are along the x, y, z axes, respectively.

the evaluation of $B^0(m^2)$ is shown in Fig. 2. Note that the $\pm\frac{1}{2}$ and $\pm\frac{9}{2}$ levels fall on each other. Of course, the zero-field splitting due to the hexadecapole moment is purely academic, since the interaction is normally extremely weak. A more realistic case would be a spin-$\frac{9}{2}$ nucleus in a crystal with lower than cubic symmetry. In this case, the M_{16} interaction would be a small perturbation on the quadrupole split levels, as indicated in Fig. 3.

For the case of a cubic crystal in a uniform magnetic field we have

$$H=H_m+H_{16}{}^0, \quad (8)$$

where $H_m=-h\nu_L m$, ν_L is the nuclear Larmor frequency, and it will be assumed that $H_m\gg H_{16}{}^0$. The magnetic field splits the nuclear-energy levels into $2I+1$ equally spaced levels, and the hexadecapole interaction can be regarded as a small perturbation on these levels.

In the [1,0,0] frame (as defined by Fig. 1), there are

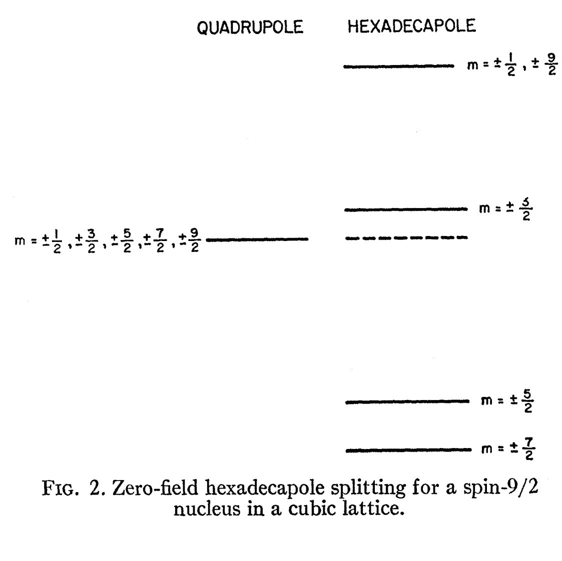

FIG. 2. Zero-field hexadecapole splitting for a spin-9/2 nucleus in a cubic lattice.

21 nonzero terms of the fourth-rank tensor V_{ijkl}. Of these 21 terms there are three equal terms of the form V_{iiii} which will be denoted by a, and 18 equal terms of the form V_{iijj} (and permutations) which will be denoted by b. An arbitrary rotation about $a(0,1,0)$ axis yields

$$V'_{zzzz}=a[1-((a-3b)/2a)\sin^2 2\theta], \qquad (9)$$

where the prime denotes the rotated frame, and θ is the angle between the external magnetic field (taken to be along the z' axis in the rotated frame) and the (0,0,1) crystalline axis. If the potential satisfies Laplace's equation, one can show that $a=-2b$. The shift in the energy levels due to the static hexadecapole interaction becomes

$$H_{16}^0=\frac{eM_{16}a[35m^4-30I(I+1)m^2+25m^2-6I(I+1)+3I^2(I+1)^2]}{384I(I-1)(2I-1)(2I-3)}\left[1-\left(\frac{(a-3b)}{2a}\right)\sin^2 2\theta\right]. \qquad (10)$$

The angular dependence for rotation about other crystalline directions may be calculated in a straightforward manner but tends to be slightly more complex.

In order to calculate the angular dependence of the hexadecapole one-phonon interaction, we write the displacement ξ as

$$\xi=\xi_0 kg(l',m',n')(lL+mM+nN), \qquad (11)$$

where (L,M,N) are the direction cosines of the nuclear sites, (l,m,n) are the direction cosines for the wave vector $\mathbf{k}$, and (l',m',n') are the direction cosines of the polarization, all with respect to the [1,0,0] frame. We must now evaluate the rotational properties of the fourth-rank tensor

$$V_{jklm}{}^p=\sum_i\left\{\sum_\alpha\left[\frac{\partial}{\partial\xi_i{}^\alpha}V_{jklm}\right]_0\xi_i{}^\alpha\right\}.$$

In general, in the [1,0,0] frame, the frame displaying the maximum symmetry, there will be 81 nonzero components of the fourth rank $V_{jklm}{}^p$ tensor. Of these 81 terms only 21 are nonzero after the lattice sum, and of these nonzero terms only two are distinct and will be denoted by $\alpha[V_{iiii}{}^p, i=x, y, z(3)]$, and $\beta[V_{iijj}{}^p$, and permutations with $i\neq j(18)]$.

We can simplify the problem by assuming that we are dealing with longitudinally polarized phonons with wave vectors parallel to the x axis. With this assumption, only 21 tensor components of the $V_{jklm}{}^p$ tensor will be nonzero after the lattice sum is taken. We will evaluate the tensor for one particular experimental arrangement since the calculation is somewhat lengthy.

If we define the [1,1,1] frame as the x axis along the (1,1,1) direction, the y axis along the $(-1, 1,0)$ direction, and the z axis along the $(-1, -1, 2)$ crystalline direction, and assume that the longitudinal phonons are propagated along the x axis in this frame, we can evaluate the 21 nonzero components of the $V_p{}^{\pm n}$ tensor in this [1,1,1] frame in terms of the two independent values, α and β, by performing a transformation which consists of two rotations, first about the (0,0,1) direction, then about the $(-1, 1,0)$ direction. An arbitrary rotation about the $(-1, 1,0)$ crystalline direction will yield the angular dependence of the $\Delta m=\pm 3$ and $\Delta m=\pm 4$ one-phonon hexadecapole matrix elements

$$V_p{}^{\pm 3}(\theta)=vD\sin^3\theta\cos\theta, \qquad (12a)$$

$$V_p{}^{\pm 4}(\theta)=-vD\sin^4\theta, \qquad (12b)$$

where

$$D=(7kg\chi/864)[\alpha-3\beta], \qquad (12c)$$

and θ is the angle between the direction of phonon-propagation vector and the external magnetic field.

In deriving Eqs. (12) we assumed that the amplitude of the displacement (ξ_0) is proportional to the driving voltage (v) impressed on the phonon generator, the transducer, or $\xi_0=\chi v$. Actually, χ is not constant, more probably a Lorentzian function of frequency for a given transducer, and varies with the transducer thickness and material.[16] However, this fact will not affect our results.

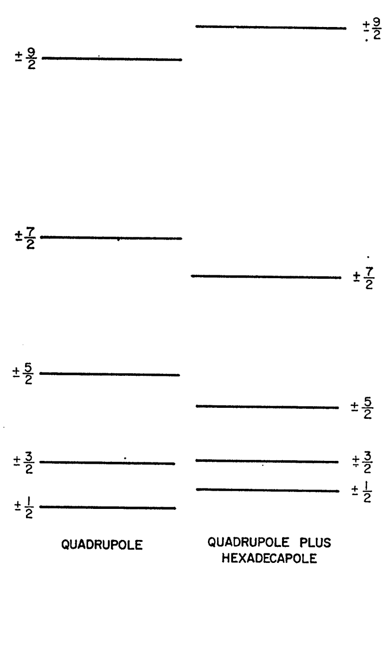

FIG. 3. Zero-field quadrupole and quadrupole plus hexadecapole splitting for a spin-9/2 nucleus in a crystal with lower than cubic symmetry.

5. SATURATION FACTOR

The transition probability per unit time $W_{mm'}$ for a $\Delta m=\pm n$ transition is given by time-dependent first-

[16] R. J. Mahler and W. H. Tanttila, J. Acoust. Soc. Am. 38, 429 (1965).

order perturbation theory as

$$W_{mm'}(n)=(1/\hbar^2)\rho_n(\nu)|H_{mm'}|^2,$$

where $H_{mm'}$ is the perturbation matrix element connecting the m and m' states, which in general will be different for each value of m, and $\rho_n(\nu)$ is the density of transitions defined such that $\rho_n(\nu)d\nu$ is the number of possible transitions in a frequency range $d\nu$ normalized such that

$$\int_{-\infty}^{+\infty}\rho_n(\nu)d\nu=1.$$

If we assume that $\rho_n(\nu)$ is a Gaussian distribution,[17]

$$\rho_n(\nu)=(1/\pi^{1/2}\Delta\nu_n)\exp\{-[(\nu-\nu_{mm'})/\Delta\nu_n]^2\},$$

$\Delta\nu_n$ can be evaluated experimentally by measuring the half-width of the absorption line at a point where the saturation is e^{-1} of its maximum value. Evaluating $W_{mm'}(n)$ at maximum saturation, we find

$$W_{mm'}(n)=(1/\pi^{1/2}\hbar^2)|H_{mm'}|^2(1/\Delta\nu_n). \quad (13)$$

In Eq. (7) we have written the one-phonon hexadecapole interaction Hamiltonian $H_{mm'}$ as

$$H_{mm'}=AB^{\pm n}\sum_{i,\alpha}\left[\frac{\partial}{\partial\xi_i{}^\alpha}V^{\mp n}\right]_0\xi_i{}^\alpha,$$

where only $B^{\pm n}$ is a function of the z component of the nuclear spin. This type of separation is always possible when the total wave function can be written as a product of a spin and a space function. In what follows, we will not restrict ourselves to hexadecapole interactions when we derive a general expression for the saturation factor. We merely require that the interaction Hamiltonian can be written as a product of a spin and space function,

$$W_{mm'}(n)=W_n|B^{\pm n}(m)|^2, \quad (14a)$$

where W_n includes the spatial part of the transition probability (and constants), and the explicit dependence on m is given by $|B^{\pm n}(m)|^2$. In the case of the one-phonon hexadecapole interaction,

$$W_n=\frac{A^2|V_p{}^{\mp n}|^2}{\pi^{1/2}\hbar^2\Delta\nu_n}. \quad (14b)$$

We will further assume that $B^{\pm n}$ contains nothing more than products of the matrix elements of the nuclear raising and lowering operators $[I_\pm]_{\rm op}$,

$$\langle m\pm1|[I_\pm]_{\rm op}|m\rangle=[(I\mp m)(I\pm m+1)]^{1/2}, \quad (15a)$$

and the operator $[I_z]_{\rm op}$,

$$\langle m|[I_z]_{\rm op}|m\rangle=m. \quad (15b)$$

If p_m represents the population of the $I_z=m$ level, we can write

$$dp_m/dt=-W_{m(m+n)}(p_m-p_{m+n})-W_{m(m-n)}(p_m-p_{m-n})+[(SL)_m+(SS)_m]p_m, \quad (16)$$

where $(SL)_m$ represents the spin-lattice and $(SS)_m$ represents the spin-spin contributions, $I\geq(m+n)\geq-I$, and $I\geq(m-n)\geq-I$. If we write down the $2I+1$ equations corresponding to the levels labeled by m, multiply each by the appropriate value of $\gamma\hbar m$, and add all the equations, we obtain

$$\begin{aligned}\gamma\hbar\sum_m m\frac{dp_m}{dt}&=\frac{d}{dt}\langle M_z\rangle\\&=-W_n\gamma\hbar n\{\sum_{m=1/2}^{m=I}|B^{-n}(m)|^2(p_m-p_{-m})\\&\quad-\sum_{\substack{\text{all}\\m>n>0}}|B^{+n}(m-n)|^2(p_{(m-n)}-p_{-(m-n)})\}\\&\quad+\gamma\hbar[\sum_m m(SL)_mp_m+\sum_m m(SS)_mp_m], \quad (17)\end{aligned}$$

where we have assumed half-integer spin and M_z is the magnetization. Since in a strong magnetic field, the spin-spin interactions cannot change the energy of the spin system, they must leave $\langle M_z\rangle$ unchanged, or

$$\sum_m m(SS)_mp_m=0. \quad (18a)$$

We assume that the spin system approaches equilibrium with the lattice exponentially with a single relaxation time T_1, or

$$\gamma\hbar\sum_m m(SL)_mp_m=-(M_z-M_0)/T_1, \quad (18b)$$

where M_0 is the maximum magnetization with no perturbation applied. We then assume that the spin-spin interaction constantly maintains a Boltzmann distribution between the spin levels, or

$$p_m-p_{-m}=2m(p_{1/2}-p_{-1/2}). \quad (18c)$$

Finally, we write

$$\begin{aligned}\langle M_z\rangle=\gamma\hbar\sum_{m=-I}^{I}mp_m&=\gamma\hbar\sum_{m=1/2}^{m=I}2m^2(p_{1/2}-p_{-1/2})\\&=\tfrac{1}{3}I(I+1)(2I+1)\gamma\hbar(p_{1/2}-p_{-1/2}). \quad (18d)\end{aligned}$$

Solving for $(p_{1/2}-p_{-1/2})$ in Eq. (18d), substituting Eqs. (18a), (18b), (18c), and (18d) into Eq. (17), and assuming $(d/dt)\langle M_z\rangle=0$ at equilibrium, one obtains

$$\frac{M_z}{M_0}=\left[1+\frac{6nT_1W_n}{I(I+1)(2I+1)}\{\sum_{m=I}^{m>0}m|B^{-n}(m)|^2-\sum_{\substack{\text{all}\\m>n>0}}(m-n)|B^{+n}(m-n)|^2\}\right]^{-1}. \quad (19)$$

[17] E. Brun, R. J. Mahler, H. Mahon, and W. L. Pierce, Phys. Rev. 129, 1965 (1963).

This equation gives the saturation factor for any interaction which gives rise to spin transitions involving a change in the z component of the spin (m) by any amount ($\Delta m=\pm n$).

Since it would be almost impossible to observe $\Delta m=\pm 1$ and ± 2 hexadecapole transitions because of the much stronger quadrupole transitions, we only need to evaluate $B^{\pm 3}$ and $B^{\pm 4}$. By repeated use of the raising and lowering operator one obtains

$$\langle m\pm 3|[B^{\pm 3}]_{\rm op}|m\rangle = (2m\pm 3)[(I\mp m)(I\mp m-1)(I\mp m-2) \times (I\pm m+1)(I\pm m+2)(I\pm m+3)]^{1/2}, \quad (20a)$$

and

$$\langle m\pm 4|[B^{\pm 4}]_{\rm op}|m\rangle = [(I\mp m)(I\mp m-1)(I\mp m-2)(I\mp m-3)(I\pm m+1) \times (I\pm m+2)(I\pm m+3)(I\pm m+4)]^{1/2}. \quad (20b)$$

One can immediately see from Eq. (20a) that hexadecapole transitions are forbidden between $m=\pm\frac{3}{2}$ levels, quite analogous to the forbidden quadrupole transitions between $m=\pm\frac{1}{2}$ levels.

6. CONCLUSION

We have thus far calculated the matrix elements for the $\Delta m=\pm 3$ and $\Delta m=\pm 4$ hexadecapole-phonon interaction and evaluated them for a crystal with $\bar{4}3m$ symmetry and a nucleus with arbitrary spin I. Let us now assume we have a sample containing spin $I=\frac{9}{2}$ nuclei, and that it is cut so that longitudinally polarized phonons may be added to the lattice along a (1,1,1) crystalline direction. Experimentally, we add phonons of a frequency which corresponds to the $\Delta m=\pm 3$ or $\Delta m=\pm 4$ nuclear transition frequency and monitor the magnetization to determine the effect of these phonons on the spin system as the angle between the phonon-propagation direction and the external magnetic field is varied.

Theoretically, we evaluate Eq. (19) using Eqs. (20), (14), (13), (12), and (4) to obtain the angular dependence of the $\Delta m=\pm 3$ and $\Delta m=\pm 4$ hexadecapole-phonon interaction. For the $\Delta m=\pm 3$ transitions, we obtain

$$M_z/M_0=[1+7.26\times 10^{-4}(eM_{16}Dv/\hbar)^2 \times (T_1/\Delta\nu_3)\sin^6\theta\cos^2\theta]^{-1}, \quad (21a)$$

and for the $\Delta m=\pm 4$ transitions

$$M_z/M_0=[1+8.62\times 10^{-5}(eM_{16}Dv/\hbar)^2 \times (T_1/\Delta\nu_4)\sin^8\theta]^{-1}, \quad (21b)$$

where θ is the angle between the direction of phonon propagation [the (1,1,1) crystalline direction] and the external magnetic field [rotating about a (1,1,0) crystalline axis], v is the voltage applied to the transducer, T_1 is the spin-lattice relaxation time, $\Delta\nu_n$ is the $\Delta m=\pm n$ hexadecapole linewidth, eM_{16} is the hexadecapole moment, and D is given in Eq. (12c). The angular variation of Eqs. (21) is shown in Fig. 4.

Experimentally, one would like to show that in a crystal with $\bar{4}3m$ symmetry, the angular variation of the $\Delta m=\pm 3$ and $\Delta m=\pm 4$ saturation corresponds to Fig. 4. It would also be extremely valuable to prove that the observed saturation was a result of the hexadecapole-phonon interaction. The $\bar{4}3m$ symmetry crystal is not suitable to prove this, but was chosen for this paper because $\Delta m=\pm 3$ transitions have been observed in InAs,[5] a crystal of this structure. It should be noted that in this experiment, phonons were added to the lattice along the (1,1,1) direction, but the rotation direction was unknown. The maximum saturation was observed near 60° which corresponds very well with Fig. 4.

One would like next to prove that the saturation was due to the hexadecapole-phonon interaction. This can be done by again observing In^{115} $\Delta m=\pm 3$ and $\Delta m=\pm 4$ transitions in a crystal with lower than cubic symmetry in which the static quadrupole splitting would be nonzero. One could then split the degenerate quadrupole levels with an external magnetic field and attempt to observe the $\Delta m=\pm 3$ and $\Delta m=\pm 4$ transitions between the various levels. If these transitions are observable, but the forbidden transition between the $\pm\frac{3}{2}$ levels are not observable, then one would have almost conclusive proof that the transitions are caused by the hexadecapole-phonon interaction, since only $\Delta m=\pm 3$ electric-hexadecapole transitions are forbidden between these levels.

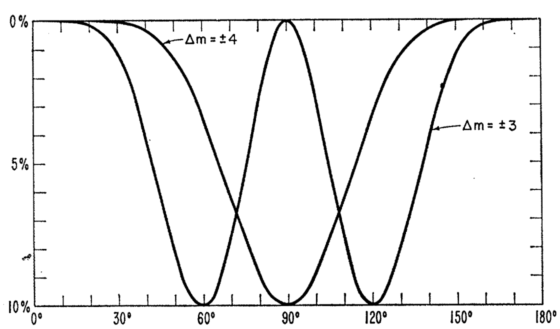

FIG. 4. Angular dependence of the nuclear-spin saturation due to the $\Delta m=\pm 3$ and $\Delta m=\pm 4$ nuclear hexadecapole-phonon interaction in a crystal with $\bar{4}3m$ symmetry. The direction of phonon propagation is along the (1,1,1) crystalline direction, rotation is about a (−1, 1,0) direction, and θ is the angle between the phonon propagation direction and the external magnetic field.