Dynamic shift of the frequency of a quadrupole-split NMR spectrum

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Equations describing the motion of the nuclear magnetic system of a ferromagnet are derived allowing for the quadrupole and inhomogeneous hyperfine interactions. A study is made of the change in the NMR line profile on increase in the dynamic frequency shift. It is shown that the interaction of quadrupole-split components of the NMR spectrum increases considerably the relative amplitude of the low-frequency line. A qualitative explanation of the published experiments is given.

It is well known[1-3] that the main features of NMR in magnetic materials are governed by the hyperfine interaction between electron and nuclear spins. Another important factor for nuclear spins in the range $I > 1/2$ is frequently the quadrupole interaction. An additional important feature is the microinhomogeneity of the hyperfine fields at the nuclei without allowance for this feature it is not possible to describe transient and some steady-state processes in a nuclear magnetic system. In many experiments the situation is such that all three factors are manifested simultaneously (see, for example, Refs. 4 and 5). Therefore, in the first section below we shall obtain equations for the motion of the nuclear magnetization allowing for the quadrupole and inhomogeneous hyperfine interactions. We shall then consider small fluctuations near an equilibrium state and the change in the NMR spectrum on transition from a small dynamic frequency shift to a large one. In the second section we shall carry out such an analysis of a homogeneous electron-nuclear magnetic system, whereas in the third section we shall allow for an inhomogeneity of the hyperfine interaction. All the calculations will be made for a ferromagnetic sample and a nuclear spin $I = 1$. In the section headed Conclusions we shall discuss the results of an experimental investigation of King *et al.*,[6] who investigated in detail the topics of interest to us.

1. EQUATIONS OF MOTION

We shall write the Hamiltonian of the electron-nuclear magnetic system in the form[1]

$$\hat{\mathcal{H}}=\hat{\mathcal{H}}_e+\hat{\mathcal{H}}_{en}+\hat{\mathcal{H}}_q,$$

$$\hat{\mathcal{H}}_{en}=-\sum_i \mathcal{A}_i\hat{\mathbf{S}}_i\hat{\mathbf{I}}_i, \quad \hat{\mathcal{H}}_q=\hbar Q\sum_i \hat{I}_{iz}^2, \tag{1}$$

where $\hat{\mathcal{H}}_e$ is the Hamiltonian of the electron system without allowance for the hyperfine interaction; $\hat{\mathcal{H}}_{en}$ describes the hyperfine interaction with an inhomogeneous constant $\mathcal{A}_i$; $\hat{\mathcal{H}}_q$ is the quadrupole interaction in a system of coordinates linked to the symmetry axis; $\hat{\mathbf{S}}$ and $\hat{\mathbf{I}}$ are the operators of the electron and nuclear spins; no allowance is made directly for the interaction of nuclei with an external magnetic field. We shall assume specifically that $\mathcal{A}_i > 0$ (which corresponds to the real situation for the majority of magnetic materials) and that $Q > 0$.

The motion of an electron ferromagnetic system can be described phenomenologically.[2] Separating the operators $\hat{\mathbf{S}}$ and $\hat{I}$, we obtain an expression for the density of the macroscopic hyperfine energy $\mathcal{H}_{en}$:

$$\mathcal{H}_{en}=-N_n\langle\mathbf{S}\rangle\langle\mathcal{A}\mathbf{I}\rangle=-\mathbf{H}_I\mathbf{M}, \quad \mathbf{H}_I=-\gamma_n\hbar N_n\langle A\mathbf{I}\rangle. \tag{2}$$

Here, $\mathbf{H}_I$ is the effective hyperfine field acting on electrons; $\mathbf{M} = -\gamma_e\hbar N_e\langle\mathbf{S}\rangle$ is the macroscopic electron magnetization; N_e and N_n are the concentrations of the electron and nuclear spins; γ_e is the modulus of the electron gyromagnetic ratio; γ_n is the nuclear gyromagnetic ratio (it is assumed that $\gamma_n > 0$); $A = \mathcal{A}/(\gamma_e\gamma_n\hbar^2 N_c)$ is the phenomenological hyperfine interaction constant: the angle brackets denote averaging over a volume of radius r_b ($r_0 \ll r_b \ll r_\alpha$, where r_0 is a characteristic size of the inhomogeneity of the hyperfine interaction, which is regarded as small compared with the correlation radius of the exchange interaction r_α). We shall assume that: a) in an equilibrium state a sample is magnetized antiparallel to the z axis; b) a circularly polarized magnetic field $\mathbf{h}(t)$ causes the vector $\mathbf{M}$ to execute small oscillations about its equilibrium position; c) the field frequency ω is close to the NMR frequency and it is low compared with the FMR frequency ω_e. If these conditions are satisfied, the solution of the Landau-Lifshitz equations for $\mathbf{M}$ can be written in the form

$$M_z=-M, \quad \mathbf{M}_\perp=\chi(\mathbf{h}+\mathbf{H}_{I\perp}), \tag{3}$$

where χ is the transverse static electron susceptibility, which we shall assume to be a scalar quantity (in this case we have $\chi = \gamma_e M/\omega_e$).

The Hamiltonian of the nuclear magnetic system after decoupling of $\hat{\mathbf{S}}$ and $\hat{\mathbf{I}}$ splits into a sum of the Hamiltonians $\hat{\mathcal{H}}_i(A_i)$:

$$\hat{\mathcal{H}}_i(A_i)=\gamma_n\hbar A_i\mathbf{M}\hat{\mathbf{I}}_i+\hbar Q\hat{I}_{iz}^2. \tag{4}$$

Since inhomogeneous motion in a system of nuclear spins is related only to the inhomogeneity of A, we can omit the index i in Eq. (4). The dynamics of an ensemble of nuclear spins with a fixed value A is governed by the evolution of the density matrix $\rho(A, t)$. Bearing in mind that $r_0 \ll r_b$, we find that the expression for $\langle AI\rangle$ in Eq. (2) can be written in the form

$$\langle A\mathbf{I}\rangle=\int_{-\infty}^{\infty} A\mathbf{I}(A,t)g(A)\,dA, \quad \mathbf{I}(A,t)=\mathrm{Tr}\{\hat{\mathbf{I}}\rho(A,t)\}, \tag{5}$$

where $g(A)$ is the distribution function.

We shall use the index p ($1 \leqslant p \leqslant 2I+1$) for a state char-

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acterized by $I_z = I + 1 - p$, i.e., $p = 1$ corresponds to the maximum value of $I_z = I$. The familiar relationships for the matrix elements of the operator $\hat{\mathbf{I}}$ readily yield the equation of motion for the density matrix ρ:

$$-i\dot{\rho}_{pj}=\gamma_n A\{M_z(p-j)\rho_{pj}+{}^1/_2M_+[j(2I+1-j)]^{1/2}\cdot\rho_{p,j+1}$$
$$+{}^1/_2M_-[(j-1)(2I+2-j)]^{1/2}\rho_{p,j-1}$$
$$-{}^1/_2M_+[(p-1)(2I+2-p)]^{1/2}\rho_{p-1,j}$$
$$-{}^1/_2M_-[p(2I+1-p)]^{1/2}\rho_{p+1,j}\}+Q[(I+1-j)^2-(I+1-p)^2]\rho_{pj}. \quad (6)$$

Substituting here the expressions in Eq. (3) and allowing for Eqs. (2) and (5), we shall obtain the required system of integrodifferential equations describing the motion of nuclear spins.

2. HOMOGENEOUS NUCLEAR MAGNETIC RESONANCE

The eigenfrequencies of a homogeneous nuclear system of a magnetic material deduced allowed for the quadrupole interaction were first considered by Kurkin and Parfenova[7] (see also Ref. 1). It was shown that an indirect (Suhl-Nakamura) interaction between nuclear spins induces a dynamic shift D of the NMR spectrum and an analysis was made of two limiting cases: $D \ll Q$ and $D \gg Q$. In the former case ($D \ll Q$) there is a shift of quadrupole-split NMR frequencies. In the latter case ($D \gg Q$) there is a change to collective motion of nuclear spins with a single frequency which is equal to the shifted NMR frequency in the absence of quadrupole interactions. Investigations[8-10] dealt in detail with the case when $D \ll Q$ and calculations were made of the spectrum of nuclear spin waves and of relaxation characteristics of the nuclear system. To the best of our knowledge, the intermediate range $D \sim Q$ has not yet been investigated, so that this section will be devoted to homogeneous NMR in this range.

We shall consider small fluctuations of the nuclear magnetic system about its equilibrium position. We shall assume that $h_+ = -h\exp(-i\omega t)$. We shall adopt a coordinate system linked to the field $\mathbf{h}$ (with the x axis antiparallel to $\mathbf{h}$). Bearing in mind that $A = \text{const}$, we obtain a system of two linear equations for the spin $I = 1$:

$$\dot{s}_k+i(\omega_k-\omega)s_k+\Gamma_n s_k-i(D_1s_1+D_2s_2)=iq. \quad (7)$$

Here,

$$s_k=\rho_{k+1,k}/n_k^0, \quad n_k=\rho_{kk}-\rho_{k+1,k+1}, \quad \omega_{1,2}=\omega_n\mp Q,$$
$$\omega_n=\gamma_n AM, \quad D_k=\gamma_n\hbar N_n\chi A^2 n_k^0, \quad q=2^{-1/2}\gamma_n hA\chi, \quad k=1,2; \quad (8)$$

the index "0" means that the corresponding quantity is considered in a state of equilibrium and, moreover, a Bloch relaxation term $\Gamma_n s_k$ is introduced. We shall assume that $D_k, Q \ll \omega_n \ll T/\hbar$. Writing down the equilibrium density matrix in the high-temperature approximation $\rho^0 \propto [1 - \hat{\mathscr{H}}^0(A)/T]$ and omitting from the equations of motion the terms $\sim D_k Q/\omega_n$, we obtain $n_{1,2}^0 = \gamma_n{}^2\hbar AM/3T$ and, therefore, the index k can be omitted from n_k^0 and D_k.

Substituting $\omega = q = 0$ into Eq. (7), we obtain the complex frequencies of natural oscillations $\tilde{\omega}_\mp = \omega_\mp - i\Gamma_n$, where

$$\omega_\mp=\omega_n+\Delta_\mp, \quad \Delta_\mp=-D\mp(Q^2+D^2)^{1/2}. \quad (9)$$

Clearly, the system described by Eq. (7) is fully equivalent to two interacting oscillators. The only special feature is that the oscillator frequencies are $\omega_n \mp Q - D$, i.e., they are shifted by an amount $-D$, which is also the parameter of the interaction between the oscillators. The frequencies $\omega_\mp$ decrease monotonically on increase in D; however, the frequency ω_+ remains always larger than ω_n. If $D \ll Q$ and $D \gg Q$, we have, respectively,

$$\Delta_\mp \approx \mp Q - D \mp D^2/2Q; \quad \Delta_- \approx -2D - Q^2/2D, \quad \Delta_+ \approx Q^2/2D. \quad (10)$$

Hence, it is clear that the interaction between oscillators enhances the shift of ω_- and weakens the shift of ω_+, and that on increase in D the value of Δ_- approaches $-2D$, which corresponds to the dynamic shift of the NMR frequency in the absence of the quadrupole interaction, whereas Δ_+ approaches zero.

We shall now consider the ratio of amplitudes s_1 and s_2 for each type of natural oscillation. For frequencies $\omega_\mp$ we have, respectively,

$$(s_2/s_1)_\mp=[\pm(Q^2+D^2)^{1/2}-Q]/D. \quad (11)$$

In the limit $D \to 0$ the frequency ω_- corresponds to a transition 1 (between levels 1 and 2), whereas ω_+ corresponds to a transition 2 (between levels 2 and 3). For finite values of D we obtain two modes of coupled oscillations and in the case of $\omega = \omega_-$ the oscillations due to the transitions 1 and 2 are in phase, whereas for $\omega = \omega_+$ they are in antiphase. If $D \gg Q$, we have $(s_2/s_1)_\mp \approx \pm 1$. This means that the frequencies $\omega_\mp$ correspond to cophasal and antiphasal oscillations with the same contributions of s_1 and s_2. We shall now express the transverse component of the nuclear spin I_+ in terms of s_1 and s_2. To within terms of the order of $\sim Q/\omega_n$, we have

$$I_+=2^{1/2}n^0(s_1+s_2). \quad (12)$$

Hence, it is clear that if $D \gg Q$, then natural oscillations of frequency ω_+ are not related to the motion of the nuclear spin ($I_+ = 0$). In other words, if $D \gg Q$, we have the same situation as in the absence of quadrupole interactions: there is only one collective oscillation mode I_+ of frequency $\omega_n - 2D$; this is in full agreement with the familiar results given in Refs. 1 and 7.

We shall now consider the line profile of homogeneous NMR. The NMR signal is governed by the transverse component of the electron magnetization induced by the nuclear system, which we shall denote by $\Delta\mathbf{M}_\perp$. It follows from Eqs. (2) and (3) that

$$\Delta M_+=-\gamma_n\hbar\chi AN_nI_+, \quad (13)$$

where I_+ is given by Eq. (12). Subsituting $\dot{s}_k = 0$ into Eq. (7), we readily obtain the steady-state solution. The absorption line profile governed by the component I_y can be described conveniently by a dimensionless function $F(\omega) = I_y/I_{y0}$, where Y_{y0} is the value of I_y corresponding to $D = Q = 0$ and $\omega = \omega_n$:

$$F(\omega)=\Gamma_n{}^2\left[\frac{\alpha_-}{(\omega-\omega_-)^2+\Gamma_n{}^2}+\frac{\alpha_+}{(\omega-\omega_+)^2+\Gamma_n{}^2}\right], \quad (14)$$
$$\alpha_\mp=\mp\Delta_\mp(\Delta_+-\Delta_-)^{-1}.$$

It should be pointed out that the expression obtained de-

scribes signals with Lorentzian profile and can be used only for qualitative analysis.

We shall now consider the behavior of $F(\omega)$. If $D=0$, we can readily show that for $Q^2<\Gamma_n^2/3$, there is no quadrupole splitting, i.e., $F(\omega)$ has just one maximum at the point $\omega=\omega_n$. If Πp and $Q^2>\Gamma_n^2/3$, then the curve $F(\omega)$ has two peaks. It follows from numerical analysis that in the former case ($Q^2<\Gamma_n^2/3$) an increase in D shifts the one-peak NMR signal by an amount $2D$ exactly as in the absence of the quadrupole interaction. A more interesting situation occurs in the latter case. Figure 1 (case *a*) shows the behavior of $F(\omega)$ on increase in D for $Q=5\Gamma_n$. It is clear from this figure that apart from the shift of the signals toward lower frequencies, there is also transfer of intensity from the high- to the low-frequency signal. It follows from Eq. (14) that the amplitudes of the signals α_- and α_+ satisfy the relationships

$$r=\alpha_+/\alpha_-=-\Delta_+/\Delta_-, \quad \alpha_++\alpha_-=1. \tag{15}$$

Therefore, an increase in D reduces the ratio of the amplitudes α_+ and α_-, but their sum remains constant. If $D\ll Q$ and $D\gg Q$, we have, respectively,

$$r\approx 1-2D/Q, \quad r\approx (Q/2D)^2. \tag{16}$$

It should be pointed out that the interaction between the transitions 1 and 2 has two effects: an additional shift of the NMR frequency and the transfer of intensities. However, comparing Eqs. (10) and (16), we can see that when $D\ll Q$ then the former effect is quadratic in respect of the parameter D/Q, whereas the latter is proportional to D/Q so that it is manifested more strongly than the former. It is clear from Fig. 1a that if $D/\Gamma_n=0.5$ and $Q/\Gamma_n=5$, the additional frequency shift amounting to $D^2/2Q\approx 0.025$ is practically undectable, whereas the ratio of the amplitudes of the signals differs considerably from unity: $r\approx 0.8$. The transfer of intensity from the high- to the low-frequency signal is clearly due to the fact that an increase in D gradually makes the two natural oscillation modes collective and the high-frequency mode then describes antiphase precession of spins due to the transitions 1 and 2, whereas the low-frequency mode describes the cophasal precession.

3. INHOMOGENEITY OF THE HYPERFINE INTERACTION

In the absence of the quadrupole interactions the change in the NMR line profile on increase in D depends strongly on the nature of the distribution function $g(A)$: if $g(A)$ decreases faster than the Lorentzian function, the NMR line becomes narrower on increase in D; in the opposite case, the line broadens, whereas for the Lorentzian profile the function remains constant.[11,12]

We shall now consider the influence of inhomogeneity on the line profile of a quadrupole-split NMR spectrum. Instead of the variable A it is convenient to use $\delta=\gamma_n M(A-\langle A\rangle)$ and, consequently, the distribution function $g(A)$ is conveniently replaced by a function $g(\delta)$ with a characteristic parameter Γ. Ignoring the terms $\sim DQ/\omega_n, D\Gamma/\omega_n$, and $q\Gamma/\omega_n$ we obtain the system of equations (7), where

$$\omega_k\to\omega_k+\delta, \quad D_k s_k\to D\langle s_k\rangle, \tag{17}$$

and in the expressions for ω_n, D, and q the quantity A is

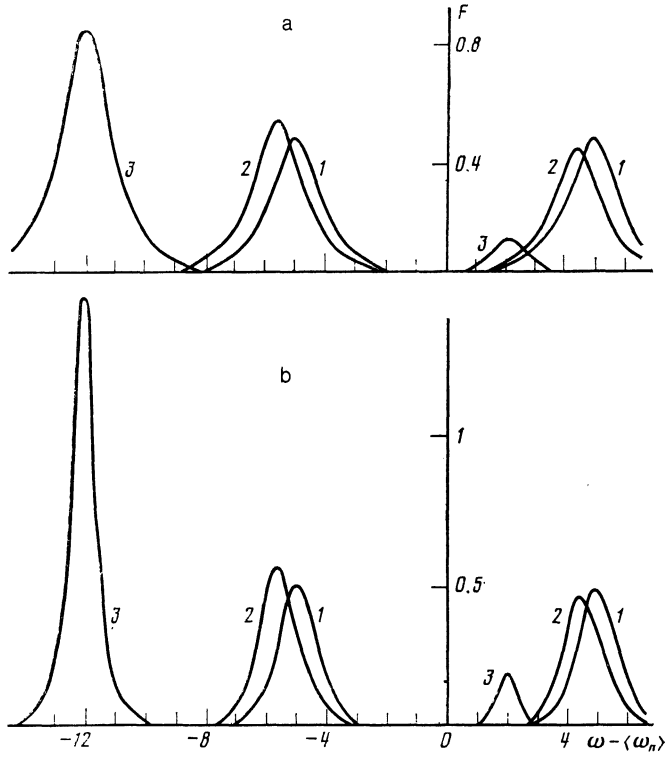

FIG. 1. Graphs of the function $F(\omega)$ plotted for $Q=5$. Curves denoted by 1 correspond to $D=0$ and curves denoted 2 and 3 correspond to $D=0.5$ and $D=5$, respectively. In the case shown in Fig. 1a all quantities are in units of Γ_n, whereas in Fig. 1b they are in units of Γ.

replaced with $\langle A\rangle$. Under steady-state conditions we have

$$s_k=-[q+D(\langle s_1\rangle+\langle s_2\rangle)][\omega-(\omega_k+\delta)+i\Gamma_n]^{-1}. \tag{18}$$

Multiplying the left- and right-hand sides of this equation by $g(\delta)$ and integrating with respect to δ, we obtain a closed system of algebraic equations for finding $\langle s_1\rangle$ and $\langle s_2\rangle$. Since we are assuming that $\Gamma\ll\omega_n$, it follows that the value of ΔM_+ is described approximately by the functions (12) and (13) where the quantities I_+, s_k, A, and n^0 are replaced by their averages ($I_+\to\langle I_+\rangle$, etc.).

The absorption line profile will be described by a dimensionless function $F(\omega)=\langle I_y\rangle/\langle I_{y0}\rangle$. Simple calculations show that

$$F(\omega)=\frac{1}{\Phi_0}\frac{\Phi''}{(1-D\Phi')^2+(D\Phi'')^2}, \tag{19}$$

where the function $\Phi=\Phi(\omega)$ is proportional to the transverse nuclear susceptibility in the absence of a dynamic frequency shift:

$$\Phi=\Phi'+i\Phi''=\sum_{k=1}^{2}\int_{-\infty}^{\infty}\frac{g(\delta)\,d\delta}{\omega_k-\omega+\delta-i\Gamma_n}. \tag{20}$$

In the case of a Lorentzian distribution function the expression for Φ (and, consequently, for F) differs from the homogeneous case by the substitution $\Gamma_n\to\Gamma_0=\Gamma_n+\Gamma$, i.e., the line profile of quadrupole-split signals does not change on increase in D and only a transfer of the intensity takes place, as shown in Fig. 1a.

We shall now analyze the situation for an arbitrary distribution function $g(\delta)$. If $D\ll\Gamma_0$, the NMR line is described

approximately by the function $\Phi''(\omega)$. In the case of interest to us when $Q \gg \Gamma_0$, this function has two maxima at frequencies $\omega = \omega_{1,2}$ with a half-width of the order of Γ_0. We shall now consider the situation when $D \gg \Gamma_0$ and both peaks are shifted by an amount considerably greater than Γ_0. In this case the positions of the maxima can be found approximately from the condition $\Phi'(\omega) = 1/D$. Assuming that $|\omega - \omega_{1,2}| \gg \Gamma_0$, we have

$$\Phi' \approx 1/(\omega_1 - \omega) + 1/(\omega_2 - \omega), \tag{21}$$

so that we obtain $\omega = \omega_{\mp}$. Therefore, the positions of resonance frequencies coincide approximately with the positions of natural frequencies of a homogeneous system and are independent of the nature of the distribution function $g(\delta)$. In the vicinity of the resonance points $\omega_{\mp}$, we have

$$F_{\mp}(\omega) \sim \Phi''(\omega_{\mp}) \{[(\omega - \omega_{\mp})\Phi_{\omega}'(\omega_{\mp})]^2 + [\Phi''(\omega_{\mp})]^2\}^{-1}, \tag{22}$$

where $\Phi_{\omega}' = d\Phi'/d\omega$. Hence, we obtain an approximate expression for the half-width of the shifted NMR peaks:

$$\Delta\omega/2 \approx \Phi''(\omega_{\mp})/\Phi_{\omega}'(\omega_{\mp}). \tag{23}$$

If the distribution function $g(\delta)$ is Lorentzian, then under the assumption that

$$\Phi'' \approx \Gamma_0[(\omega_1 - \omega)^{-2} + (\omega_2 - \omega)^{-2}], \tag{24}$$

we obtain $\Delta\omega/2 = \Gamma_0$, which is the same as the exact result. If for $D \ll \Gamma_0$ the NMR signals $\sim \Phi''(\omega)$ decrease faster in the wings than the Loretnzian profile, then on increase in D they should become narrower; in the opposite case, one should observe broadening. Therefore, the situation is analogous to that in the absence of the quadrupole interactions.

The ratio of the amplitudes of the high- and low-frequency signals is given by the expression

$$r \approx \Phi''(\omega_-)/\Phi''(\omega_+). \tag{25}$$

In the case of Lorentzian signals, we can use Eq. (24) to show that $r = -\Delta_+/\Delta_-$, which again is equal to the exact value. As pointed out in Sec. 2, an increase in D causes thue value of ω_- to drift away from its initial value ω_1 faster than ω_+ or ω_2. It therefore follows from Eq. (25) that if $r < 1$, then on increase in D this quantity decreases at the rate which becomes higher when the unperturbed NMR signals decrease more rapidly in the wings in the case $D \ll \Gamma_0$.

By way of illustration of general results, Fig. 1b shows plots of the function $F(\omega)$ for the quadratic Lorentzian function $g(\delta) \sim (\delta^2 + \Gamma^2)^{-2}$ when $\Gamma_n/\Gamma = 0.2$. We can see that on increase in D the two peaks become narrower. Therefore, the amplitude of the low-frequency peak increases faster and that of the high-frequency peak decreases more slowly than for a Lorentzian distribution function. The effect of intensity transfer is manifested more strongly than in Fig. la and the total amplitude of the peaks is naturally no longer conserved.

CONCLUSIONS

We shall consider briefly the experimental results of King *et al.*[6] in the light of the theory presented above. These authors[6] investigated a uniaxial antiferromagnet MnF_2 in a strong magnetic field $H \sim 90$ kOe, close to the spin-flopping field. The value of D was varied continuously by altering the field H. Two NMR spectra of $^{55}Mn(I = 5/2)$ were determined by a cw method and these corresponded to the two sublattices of the antiferromagnet; the two spectra varied in the same way on increase in D. For $D \ll Q$ each spectrum consisted of five quadrupole-split lines. For $D \gtrsim Q$ there was only a single NMR line (and also nuclear magnetostatic modes), which the authors of Ref. 6 interpreted as a transition to the collective behavior of the nuclear spins.

The transition from $D \ll Q$ to $D \gtrsim Q$ occurred as follows: the ratio of the amplitude of the low-frequency component of the spectrum to the amplitudes of four high-frequency components decreased monotonically. Moreover, the effect was observed clearly already for $D \ll Q$, when the dynamic frequency shift was described satisfactorily by a theory ignoring the interaction between different transitions. It was natural to assume that the observed changes in the ratio of the amplitudes represent the transfer of intensity to the low-frequency component of the spectrum considered above and associated with cophasal oscillations of nuclear spins. As pointed out already, this effect is proportional to D/Q and should appear earlier than the additional frequency shift which is a quadratic function of D/Q. King *et al.* reported also narrowing of the quadrupole-split NMR lines on increase in D in the range $\Delta\omega < D < Q$, but they assumed that the line width is governed by the Suhl-Nakamura interaction and not by the inhomogeneity of the hyperfine interaction.

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