Capture of quadrupole moment of a nucleus by a high-intensity acoustic wave

A. R. Kessel' and M. M. Shakirzyanov

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It is known that capture of the nuclear quadrupole moment takes place upon quantization of the nuclear spin in the field of an acoustic wave in a rotating coordinate system. This capture prevents the macroscopic mean value of the moment from decaying on account of spin-spin relaxation. The nuclear induction signal produced by resonant excitation of the quadrupole energy levels in the rotating coordinate system is calculated; these levels are due to quantization of the long-lived component of the quadrupole moment in the acoustic field.

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In all cases when the nuclei of the atoms of a crystal have a spin $I \geqslant 1$, their coupling to the sound is via the interaction ($\mathscr{H}'_Q$) of the electric quadrupole moments Q_μ^{-j} with the tensor-gradient of the electric field $G_\mu(\mathbf{j})$ produced by the sound at the locations of the nuclei $\mathbf{r}_j$. In a coordinate system (RCS) that rotates around the constant magnetic field at the frequency of sound, the tensor $G_\mu(\mathbf{j})$ has components that are independent of the time, and the static part $\mathscr{H}_Q^{\text{const}}$ of the operator $\mathscr{H}'_Q$ takes the form of the Hamiltonian of static quadrupole interactions in the lab (Ref. 1).

Changing over to the alternating-field representation, in which the operator $\mathscr{H}_Q^{\text{const}}$ is diagonal,

$$\mathscr{H}_Q^{\text{const}}=\hbar\omega_Q Q_0, \quad Q_0=\sum_j Q_j^0=\sum_j [3I_{zj}^2-I(I+1)], \qquad (1)$$

we can verify that the eigenvalues of the operator (1)

$$E_m=\hbar\omega_Q[3m^2-I(I+1)],$$

which determine the so called quasienergy levels,[2] are arranged exactly as the energy levels of the nuclear quadrupole resonance in the lab.[1]

The quasienergy spectrum appears if the interaction with the sound exceeds substantially the NMR line width in the lab, the latter being usually produced by the secular part ($\mathscr{H}_d$) of the magnetic dipole interaction of the nuclei ($\mathscr{H}_{dd}$), i.e., if $\mathscr{H}_Q^{\text{const}} \gg \mathscr{H}_d$. When this inequality is satisfied, the operator (1) becomes the principal Hamiltonian of the spin system in the RCS. The latter leads to a natural separation of the interaction $\mathscr{H}_d$ into a secular ($\mathscr{H}_d^s$) and nonsecular ($\mathscr{H}_d^{ns}$) part with respect to the quasi-energy spectrum, which play essentially different roles in the dynamics of the spin system. Just as in the lab (in the case of strong constant magnetic fields), the contribution to the dynamics of the spin system can be neglected, whereas $\mathscr{H}_d^s$ causes the quasienergy level widths, establishes a canonical distribution on them, and determines the components of the nuclear dipole and multipole moments that are not diagonal in this representation.[3] These processes evolve within times of the order of

$$T_2 \approx [\mathrm{Sp}(\mathscr{H}_d^s)^2\hbar^{-2}/\mathrm{Sp}\, I_z^2]^{-1/2},$$

which are much shorter than the times of the nuclear spin-lattice relaxation T_1 (Ref. 3). We note that the interaction does not change the Hamiltonian (1), inasmuch as by definition

$$[\mathscr{H}_d^s, \mathscr{H}_Q^{\text{const}}]=0.$$

Thus, the behavior of the spin system at times $t \gtrsim T_2$ is characterized, besides by the mean value $\langle \mathscr{H}_d^s \rangle$ also by the mean value $\langle Q_0 \rangle$ of the operator Q_0, and both mean values change only over times $t \gtrsim T_1$, because of spin-lattice relaxation processes. On the other hand, if the energy $\langle \mathscr{H}_Q^{\text{const}} \rangle$ does not exceed $\langle \mathscr{H}_d \rangle$, the breakdown of $\mathscr{H}_d$ into $\mathscr{H}_d^s$ and $\mathscr{H}_d^{ns}$ loses its physical meaning. Then, under the influence of $\mathscr{H}_d^{ns}$ the quantity $\langle Q_0 \rangle$ becomes small over the same times T_2 which characterize the damping of the mean values of the off-diagonal operators. It follows therefore that in strong resonant acoustic fields ($\mathscr{H}_Q^{\text{const}} \gg \mathscr{H}_d$) the quadrupole moment of the nucleus should become trapped (Q-trapping) and retained for a time $\sim T_1 \gg T_2$.

Recent experiments were performed aimed at direct observation of magnetic resonance on the quasi-energy levels that occur upon quantization of the spins in an alternating RF field.[4] In the same papers were discussed the prospects of using nonstationary NMR methods in the RCS. By virtue of the specifics of the acoustic methods of excitation, the most suitable procedure of studying Q-trapping is the pulsed nuclear induction method, a discussion of which as applied to the considered effect is presented below.

The Q-trapping phenomenon takes place under conditions of spin quantization in the field of an acoustic wave, investigated in earlier papers.[1] We shall use the results of these papers. We consider a cubic crystal (e.g., NaCl) located in a constant magnetic field $\mathbf{H}_0 \| \mathbf{Z}$. Let the longitudinal acoustical oscillations with frequency ω and with wave vector $\mathbf{k}$, parallel to the X axis in the lab, excite resonant transitions with $|\Delta m| = 2$. If the spin-sound interaction energy substantially exceeds the interaction $\mathscr{H}_d$, then in the case of nuclear spins $I = 3/2$ the Hamiltonian of the spin system in the alternating-field representation (at exact acoustic resonance $\omega = 2\omega_0 = 2\gamma_n H_0$) takes the form[1]

$$\mathscr{H}=\mathscr{H}_0+\mathscr{H}_d,\ \mathscr{H}_0=\sqrt{3}\hbar a\sum_j Q_0^j,\ Q_0^j=(P_{33}^j-P_{\bar{3}\bar{3}}^j)+(P_{11}^j-P_{\bar{1}\bar{1}}^j),$$
$$a=\frac{eq}{16I(2I-1)\hbar}S_{11}U_0,\ 3,\bar{3}\to m_z={}^3/_2,-{}^3/_2,\ 1,\bar{1}\to m_z={}^1/_2,-{}^1/_2, \qquad (2)$$

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where U_0 is the amplitude of the deformations, S_{11} is the spin-phonon coupling constant, eq is the electric quadrupole moment of the nucleus, P^j_{mn} are the projection operators in the basis of the eigenfunctions of the operator $\hat{I}_z$ in the lab, and m_z are the eigenvalues of $\hat{I}_z$. (Here and elsewhere, on changing over to a new coordinate frame, we use the so called "active" point of view[5]). The eigenvalues of the operator (2) yield two doubly degenerate quasienergy levels, separated by an interval $\hbar\Omega_0 = 2\sqrt{3}\hbar a$.

Let, in addition, the spin system be acted upon by an alternating magnetic field of low frequency (LF field) $\Omega \approx \Omega_0$, directed along the **Z** axis of the lab. In the alternating-field representation, the interaction with the LF field is described by the Hamiltonian

$$\mathcal{H}_M{}^t=\hbar\gamma_n H_1 \sum_j (B_j+K_j+K_j{}^+)\cos\Omega t,$$

$$B_j={}^1/_2[(P_{33}{}^j-P_{\bar{3}\bar{3}}{}^j)-(P_{11}{}^j-P_{\bar{1}\bar{1}}{}^j)], \tag{3}$$

$$K_j=-(P_{3\bar{1}}{}^j+P_{1\bar{3}}{}^j)e^{i\varphi_x^j}, \quad K_j{}^+=-(P_{\bar{1}3}{}^j+P_{\bar{3}1}{}^j)e^{-i\varphi_x^j},$$

where $\varphi_x^j = \mathbf{k}\cdot\mathbf{X}_j$ is the phase of the acoustic traveling wave at the location of the nucleus j.

Owing to the operators K_j and K_j^+ of the Hamiltonian (3), the pulsed LF field excites resonant transitions in the quasi-energy spectrum. The change of the mean values of the components of the dipole and multipole moments is described by systems of dynamic equations of motion,[6] one of which takes the form

$$d\langle Q_0\rangle/dt=i2\omega_1\{\langle K\rangle-\langle K^+\rangle\}\cos\Omega t,$$

$$d\langle K\rangle/dt=i\Omega_0\langle K\rangle+i\omega_1\langle Q_0\rangle\cos\Omega t, \tag{4}$$

$$d\langle K^+\rangle/dt=-i\Omega_0\langle K^+\rangle-i\omega_1\langle Q_0\rangle\cos\Omega t, \quad \omega_1=\gamma_n H_1.$$

The remaining systems of equations which do not contain $\langle Q_0\rangle$ need not be presented here, since they consist of linear homogeneous equations with zero initial conditions at the instant when the LF field is turned on. Such initial conditions arise when the start of the LF pulse is delayed compared with the start of the action of the acoustic field by a time $\Delta t > T_2$, as a result of which a canonical distribution has time to become established on the quasienergy levels.

Solving the system (4), we find that by the instant of termination of the action of LF pulse, the nonzero mean values are

$$\langle Q_0(t_1)\rangle=\langle Q_0(t_0)\rangle\cos(\omega_1\tau), \quad \langle Q_0(t_0)\rangle=-{}^1/_4\beta_s\hbar\omega_0,$$

$$\langle K(t_1)\rangle=i\langle Q_0(t_0)\rangle\sin(\omega_1\tau)\exp\{i\Omega(t-t_0)\}, \quad \tau=t_1-t_0, \tag{5}$$

$$\langle K^+(t_1)\rangle=-i\langle Q_0(t_0)\rangle\sin(\omega_1\tau)\exp\{-i\Omega(t-t_0)\},$$

$$\beta_s\approx 5\beta_0\omega_d{}^2/[5\omega_d{}^2+(2\sqrt{3}a)^2],$$

where t_0 and t_1 are the instants of the start and end of the LF pulse, β_s is the reciprocal spin temperature in the alternating-field representation,[1] and

$$\hbar^2\omega_d{}^2=\mathrm{Sp}\,\mathcal{H}_d{}^2/\mathrm{Sp}\,I_z{}^2.$$

Since the measurements are carried out in the lab, it is necessary to know the change of the spin mean values in this reference frame. In the lab, the nonzero spin variables mean values at the instant of time $t \geqslant t_1$ are

$$\langle I_z{}^j(t)\rangle=-2\langle Q_0(t_0)\rangle\sin(\omega_1\tau)\sin\Omega(t-t_0),$$

$$\langle Q_{\pm 2}{}^j(t)\rangle=\sqrt{3}\langle Q_0(t_0)\rangle\{\cos(\omega_1\tau)\pm i\sin(\omega_1\tau)$$

$$\times\cos\Omega(t-t_0)\}\exp[\pm i(2\omega_0 t-\varphi_x{}^j)],$$

$$\langle T_0{}^j(t)\rangle=2\langle Q_0(t_0)\rangle\{\cos\omega_1\tau-\sin(\omega_1\tau)\sin\Omega(t-t_0)\}, \tag{6}$$

$$T_0{}^j=(P_{33}{}^j-P_{\bar{3}\bar{3}}{}^j)-3(P_{11}{}^j-P_{\bar{1}\bar{1}}{}^j),$$

$$Q_{+2}{}^j=(I_+{}^j)^2=2\sqrt{3}(P_{3\bar{1}}{}^j+P_{1\bar{3}}{}^j), \quad Q_{-2}{}^j=(I_-{}^j)^2=2\sqrt{3}(P_{\bar{1}3}{}^j+P_{\bar{3}1}{}^j),$$

where T_0^j is the zeroth component of a third-rank tensor made up of the spin operators I_α^j. The change of the mean values (6) with time gives rise to electromagnetic radiation, since these mean values are proportional respectively to the components of the dipole, electric quadrupole, and multipole moments of third order. And although all these spin mean values (6) are of the same order of magnitude, only the magnetic dipole-moment radiation has an observable value

$$M_z{}^j(t)=\gamma_n\hbar\langle I_z{}^j(t)\rangle.$$

The damping of the transient excited by the LF pulse can be due to a number of causes. The most effective among them in our case is the inhomogeneity of the amplitude of the acoustic field, which leads to inhomogeneity $\Delta\Omega_0$ of the resonant frequency Ω_0. Assuming a normal distribution of $\Delta\Omega_0$ with a second moment σ^2_{ac}, we obtain after simple calculations[6] for the total magnetization of the sample

$$M_z(t)={}^1/_2\gamma_n\hbar N\beta_s\hbar\Omega_0\sin(\omega_1\tau)\sin\Omega(t-t_0)$$

$$\times\exp\{-\sigma_{ac}{}^2(t-t_1)^2/2\}. \tag{7}$$

When the condition $\mathcal{H}_Q^{\mathrm{const}} \gg \mathcal{H}_d$ is satisfied, the amplitude of the signal generated by the oscillations of the low-frequency magnetization (7) is comparable in magnitude with the low-frequency signal in experiments on direct observation of magnetic resonance in the RCS.[4] Thus, the nuclear-induction signals calculated here can be perfectly observable and can serve as proof of the existence of Q-trapping. Observation of this effect will also be direct proof of the quantization of the nuclear spins in the field of a high-intensity acoustic wave, the presence of which has been confirmed so far only by an indirect experimental method.[7] It must be noted that similar experiments are superior to the existing procedures for measuring the spin-phonon coupling constants, since they result in higher accuracy with respect to the low-frequency field.

[1]A. R. Kessel' and M. M. Shakirzyanov, Akust. Zh. **33**, 469 (1977) [Sov. Phys. Acoustics **23**, 263 (1977); Fiz. Tverd. Tela (Leningrad) **19**, 1535 (1977) [Sov. Phys. Solid State **19**, 899 (1977)].

[2]Ya. B. Zel'dovich, Zh. Eksp. Teor. Fiz. **51**, 1492 (1966) [Sov. Phys. JETP **24**, 1006 (1967)].

[3]A. Abragam, The Principles of Nuclear Magnetism, Oxford, 1961.

[4]A. E. Mefed and V. A. Atsarkin, Pis'ma Zh. Eksp. Teor. Fiz. **25**, 233 (1977) [JETP Lett. **25**, 215 (1977)]. Zh. Eksp. Teor. Fiz. **74**, 720 (1978) [Sov. Phys. JETP **47**, 378 (1978)].

[5]G. A. Korn and T. M. Korn, Mathematical Handbook for Scientists and Engineers, McGraw, 1961.

[6]A. R. Kessel' Yadernyĭ akusticheskiĭ rezonans (Nuclear Acoustic Resonance), Nauka, 1969.

[7]V. A. Kirsanov and V. F. Tarasov, Abstracts, Sixth International Symposium on NQR Spectroscopy, Moscow, 1981, p. 48.

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