SOVIET PHYSICS JETP VOLUME 26, NUMBER 2 FEBRUARY, 1968

NUCLEAR MAGNETIC RESONANCE IN HIGHLY VISCOUS LIQUIDS

I. A. CHABAN

Acoustics Institute

November 23, 1966; resubmitted April 10, 1967

Zh. Eksp. Teor. Fiz. 53, 556–564 (August, 1967)

The theory developed in[9] for the structure and kinetics of highly viscous liquids is used to explain some features of nuclear magnetic resonance in highly viscous liquids. The theory is based on the assumption that the liquid is a two-component medium consisting of a disordered liquid and of more or less ordered regions located in it. The degree of order of these regions changes upon variation of the external conditions in accordance with the diffusion law.

In highly viscous liquids, nuclear magnetic resonance (NMR) exhibits certain unique singularities at high viscosity and high temperature; these singularities become clearly manifest in the temperature variation of the longitudinal relaxation time T_1: Upon cooling, which is accompanied in the case of such liquids by a strong increase in the viscosity η, T_1 varies like $\sqrt{\eta}$. Such a dependence does not agree with the theories of Bloembergen, Purcell, and Pound[1] and of Kubo and Tomita[2], in which it is assumed that the motion of the molecules in the liquid constitutes Brownian rotation and displacement of individual molecules, giving $T_1 \sim \eta$ at large viscosities. Attempts by Favret and Meister[3] to eliminate the disparity between theory and experiment by introducing an entire spectrum of correlation times is a purely computational device and does not describe the physical picture of the phenomenon.

Skrotskiĭ and Kokin[4] attempted to explain the singularities of NMR in strongly-viscous liquids by assuming that a predominant role is played by translational motion of the molecules. Numerical estimates show, however, that this explanation is incorrect. Indeed, for glycerin, for example, assuming the distance between spins and the molecule radius to be 2.5×10^{-8} cm, and the volume per molecule to be 125×10^{-24} cm^3, and substituting these values into the formulas of[4], we find that the change to the half-integer dependence of the viscosity should be observed only at $\omega_0\tau_c = 3.4 \times 10^2$, whereas experiment yields for this transition $\omega_0\tau_c \approx 1$ (ω_0—Larmor frequency, τ_c—correlation time for Brownian rotation). This disparity is illustrated in Fig. 1 by the discrepancy between the experimental points and the dash-dot line, which was constructed by us in an absolute scale using the formulas of[4] for the high-frequency branch of the theoretical curve.

Hunt and Powles[5] proposed to explain the singularities of NMR in strongly viscous liquids on the basis of the "defect" diffusion picture previously advanced by Glarum[6] to describe the singularities in the propagation of electric magnetic waves: It is assumed that the molecule rotates either when a "defect" diffuses towards it, or as a result of ordinary Brownian motion. The half-integer dependence is the result of a mechanism connected with the diffusion of the "defects." An appropriate calculation, carried out in[5] for the one-dimensional case, gave satisfactory agreement between the obtained figures and the experimental data. This agreement, however, has a purely formal character: The indicated mechanism of the action of the "defects" leads in a real three-dimensional case, just as in the theory of[4], to a shift of the transition to the half-integer dependence far into the high-frequency region, thus completely violating the apparent agreement between the theory of[5] and experiment. The same objection pertains to[6].

As noted earlier, the singularities of NMR and the singularities of the propagation of sound, shear, and electromagnetic waves in strongly-viscous liquids can apparently be explained by a single mechanism. This is indicated by the fact that these phenomena have a common half-integer asymptotic dependence on the viscosity and on the frequency, and also that the characteristic times of the processes responsible for the dispersion and the anomalous absorption of all three types of waves are close to the correlation time in NMR. In this connection, we explain the behavior of strongly-viscous liquids in NMR by using the theory of the construction of strongly-viscous liquids[7-9], which has already provided an explanation for the singularities of the propagation of sound, shear, and electromagnetic waves. Such a unified analysis would be impossible in any of the theories previously proposed for this phenomenon.

The main premises of the theory of[7-9] are as follows: It is assumed that a strongly-viscous liquid is a

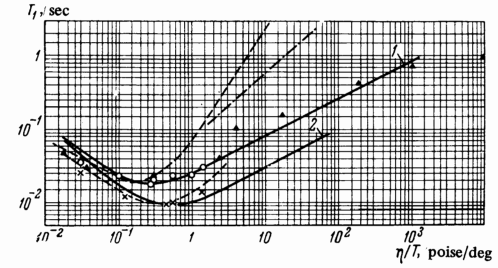

FIG. 1. Dependence of the longitudinal relaxation time T_1 on the ratio of the viscosity η to the absolute temperature T for glycerin (water content less than 1%). Circles and crosses – experimental points taken from [3] for Larmor frequencies 20 and 10 MHz respectively. Triangles – experimental points taken from [11] for 21.5 MHz. The viscosities are taken from [12]. The solid lines show the theoretical curves obtained from the proposed theory. The dashed lines show curves constructed in accordance with the theory of [1,2]. The dash-dot line shows the high-frequency branch of the theoretical curve plotted from the formulas of Skrotskiĭ and Kokin [4].

disordered medium in which more or less ordered regions are situated. The degree of ordering of these regions is characterized by a quantity ξ_1. Under different external conditions, the equilibrium values of ξ_1 will differ. If the external conditions change sufficiently rapidly, then ξ_1 will have a nonequilibrium value equal to $\xi_1^0 + \xi_1'$, where ξ_1^0 is the equilibrium value under the new external conditions, and ξ_1' is the deviation from the equilibrium value, which tends to zero with increasing time. For the disordered part of the liquid we introduce analogously the quantity ξ_2 ($\xi_2 = \xi_2^0 + \xi_2'$). Unlike ξ_1^0, the equilibrium value ξ_2^0 does not change when the external conditions change. The establishment of the equilibrium value ξ_1 and ξ_2 is via diffusion equalization of the values of ξ_1' and ξ_2'. The quantities ξ_1 and ξ_2 can be regarded in rough approximation as hole concentrations, and it can be assumed that the equilibrium values ξ_1^0 and ξ_2^0 are established by diffusion of the excess holes through the boundaries of the ordered regions. For simplicity, the ordered regions are assumed in the calculations to be spherical with identical radii a. The sphericity assumption is inessential, since it can be shown that the final results are not very sensitive to the shape of the ordered regions.=

It follows from the hole interpretation of the degree of ordering that the actions that lead to the realignment should change the volume per molecule. In order to get an idea of the order of magnitude of the quantities involved, we present the following figures. As shown in[7-9], ultrasound of intensity of the order of 0.1 W/cm^2 gives rise to realignment of the ordered regions. In glycerin, for example, the relative change in the volume is of the order of 10^{-6}, and the energy required for this purpose is 10^{-22} erg per molecule. In spite of the fact that this energy is negligibly small compared with kT (k is Boltzmann's constant and T is the absolute temperature), realignment does nevertheless take place, since, as shown by calculation, at the indicated change in volume a change takes place in the thermodynamic potential by an amount of the order of kT, owing to the change in entropy. It was shown in[7-9] that a shear wave of intensity 0.1 W/cm^2 also causes realignment. Estimating for glycerin the torque that orients the molecule in this case, we find it to be of the order of 10^{-21} dyne-cm.

Molecules of highly viscous liquids are diamagnetic, that is, they do not have any constant magnetic moments connected with the electron shell. Those with which we deal are made up of C, H, and O atoms, with the hydrogen atom having spin 1/2 and the others having zero spin. The spin system considered below is an aggregate of spins coupled by dipole-dipole interaction. This interaction depends on the orientation of the axis joining the spins and on the distance between spins, and relates the spin system with the molecular surrounding (lattice). If the spin system is somehow taken out of its equilibrium state, then the restoration of the equilibrium is a result of its interaction with the lattice.

The explanation proposed by us for the singularities of NMR in highly viscous liquids is based on the following physical picture. The spin flip which leads to restoration of the equilibrium magnetization, is brought about by thermal motion of the molecules both in the disordered part of the liquid and in the ordered regions. In the disordered part of the liquid, however, the spin flip does not change the equilibrium state of the lattice (the lattice can be regarded as a thermostat), and in the ordered region, we shall assume that the spin flip upsets the equilibrium of the lattice. This loss of equilibrium is the result of the change in the equilibrium volume per molecule, leading to a change in the equilibrium degree of ordering. The equilibrium state of the lattice is restored by realignment of the ordered regions.

To verify that spin flip does indeed lead to realignment of the ordered regions, let us estimate the change in the energy of the dipole-dipole interaction of the spins when one of the spins of the ordered region flips, per molecule of this region, and the torque rotating that molecule in which the spin reorientation took place. For glycerin at room temperature we obtain respectively 10^{-24} erg and 10^{-2} dyne-cm. From a comparison of these figures with those presented earlier it follows that spin flip can actually cause realignment of the ordered regions. Each spin-flip act in the ordered region causes realignment independently of any other act, since the time of realignment is much shorter than the average time interval between flips. Indeed, for example, for glycerin at room temperature, the ordered regions contain approximately 100 molecules, that is, 800 nuclei with nonzero spin, whereas the magnetization relaxation time is of the order of 10^{-2} sec. Thus, the average interval between successive spin-flip acts is of the order of 10^{-5} sec, which is much longer than the realignment time of the ordered region, which at room temperature is approximately 10^{-8} sec.[9]

Each spin-flip act can be regarded as instantaneous compared with the realignment time. Indeed, the spin-flip time must be regarded as equal to the time of the molecule-jumping process itself; this time, in accordance with the generally accepted picture of Brownian motion in the liquid, is small compared with the correlation time, that is, the average time between successive molecule jumps. On the other hand, the time of realignment of the ordered regions is of the same order as the correlation time observed in NMR.

The correlation of the position and orientation of the molecules in the ordered region, that is, their dependence at the instant τ on the position and orientation at the instant $\tau = 0$, will be determined by two processes: the realignment of the ordered regions and the usual Brownian motion. Both types of motion will be taken into consideration in calculating the relaxation time in the ordered region. On the other hand, in the disordered region, of the liquid we shall assume that the correlation is connected only with Brownian motion. We shall henceforth take into account only rotational Brownian motion, since its contribution exceeds the contribution of the translational Brownian motion.

Thus, on the basis of the picture described above, let us proceed to calculate the relaxation times of the magnetization in the ordered region. We shall take into account here only the interaction between spins belonging to one molecule, and assume the distances between spins to be equal; we shall likewise disregard the correlation and the motion of the different spin pairs belonging to one molecule. For the reciprocal values of the longitudinal and transverse relaxation times in the ordered region we have the following formulas[10]:

$$\frac{1}{T_1'} = \frac{9}{8}\gamma^4\hbar^2 n\,[J^{(1)}(\omega_0) + J^{(2)}(2\omega_0)], \tag{1}$$

$$\frac{1}{T_2'} = \frac{1}{T_2''} + \frac{3}{4}\gamma^4\hbar^2 n\left[\frac{3}{8}J^{(0)}(0) + \frac{15}{4}J^{(1)}(\omega_0) + \frac{3}{8}J^{(2)}(2\omega_0)\right], \tag{2}$$

where γ is the gyromagnetic ratio, $\hbar$ Planck's constant, n the number of spins in the molecule with which the given spin interacts,

$$J^{(q)}(q\omega_0) = \int_{-\infty}^{\infty} G^{(q)}(\tau)\,e^{-iq\omega_0\tau}\,d\tau, \tag{3}$$

$G^{(q)}(\tau) = \overline{F^{(q)}(t+\tau)F^{(q)*}(t)}$ the correlation function of the quantities $F^{(q)}$ and $F^{(q)*}$ (the bar denotes averaging over the ensemble), and $F^{(q)}$ is a function of the angles θ and φ of the axis joining the spin pair and the distance r_0 between these spins:

$$F^{(0)} = -\frac{1}{r_0^3}\frac{2}{\sqrt{5}}Y_2^{(0)}(\theta), \qquad F^{(1)} = \frac{1}{r_0^3}\sqrt{\frac{8\pi}{15}}\,Y_2^{(1)}(\theta,\varphi),$$

$$F^{(2)} = \frac{1}{r_0^3}\sqrt{\frac{32\pi}{15}}\,Y_2^{(2)}(\theta,\varphi),$$

where

$$Y_2^{(0)}(\theta) = \frac{\sqrt{5}}{2}(3\cos^2\theta - 1), \qquad Y_2^{(1)}(\theta,\varphi) = \sqrt{\frac{15}{8\pi}}\sin\theta\cdot\cos\theta e^{-i\varphi},$$

$$Y_2^{(2)}(\theta,\varphi) = \sqrt{\frac{15}{32\pi}}\sin^2\theta e^{-2i\varphi}$$

are normalized spherical functions. Unlike the formulas given in[10], we retained in the expression for $1/T_2'$ in (2) the first-order term $1/T_2''$, since it does not vanish in the ordered regions.

We assume for simplicity that the molecules have at the initial time a random orientation. This is possible, since allowance for the order and the regions under consideration only complicates the calculation, without changing the final result. The quantity $\Phi(\tau) = G^{(q)}(\tau)/D^{(q)}(0)$ is the reduced correlation function of the molecule orientation, which had at the initial instant of time a random orientation, and then changed the orientation both as a result of Brownian motion and as a result of realignment of the ordered region, due to the instantaneous change in equilibrium degree of ordering from the value $\xi_1^0(s_1)$ to the value $\xi_1^0(s_2)$ (s_1 and s_2 are the aggregates of the spin coordinates of the ordered region before and after the spin flip). We shall assume, just as in[10], that the reduced correlation function is the same for any q.

Realignment of the ordered regions upon instantaneous change in the degree of ordering from a value $\xi_1^0(s_1)$ to $\xi_1^0(s_2)$ can be described, as follows from[9], by means of the following equations:

$$\begin{gathered}\dot{\xi}_1'(r,\tau) - D\Delta\xi_1'(r,\tau) = [\xi_1^0(s_1) - \xi_1^0(s_2)]\,\delta(\tau),\\ \dot{\xi}_2'(r,\tau) - D\Delta\xi_2'(r,\tau) = 0\end{gathered} \tag{4}$$

subject to the boundary conditions

$$\xi_1'(r,\tau) = \xi_2'(r,\tau), \quad \operatorname{grad}\xi_1'(r,\tau) = \operatorname{grad}\xi_2'(r,\tau) \quad \text{for } r = a, \tag{5}$$

where r is the distance from the center of the ordered region, $\delta(\tau)$ is the δ-function, and D is the diffusion coefficient, which may or may not coincide with the ordinary diffusion coefficient.

These equations describe the realignment of the ordered regions on the average in all regions. In view of the linearity of the equations and of the boundary conditions, the solution can be represented in the form

$$\xi_1'(r,\tau) = [\xi_1^0(s_1) - \xi_1^0(s_2)]\,\xi_{1,\delta}'(r,\tau),$$

where $\xi_{1,\delta}'(r,\tau)$ is the solution of (4) under the condition $\xi_1^0(s_1) - \xi_1^0(s_2) = 1$, and consequently does not depend on s_1 and s_2.

The quantity $1 - \xi_{1,\delta}'(r,\tau)$ can be treated as the probability of realignment at a distance r by the instant of time τ, and

$$\overline{\xi}_{1,\delta}'(\tau) = \frac{3}{a^3}\int_0 \xi_{1,\delta}'(r,\tau)\,r^2\,dr \tag{6}$$

can be treated as the reduced correlation function connected with the realignment, averaged over all the molecules of the ordered region. Using the solution of (4) obtained in[9] with a periodic right-hand side, it is easy to obtain for $\overline{\xi_{1,\delta}'(\tau)}$ the following expression, which is convenient for further calculations:

$$\overline{\xi}_{1,\delta}'(\tau) = \frac{1}{2\pi}\int_{-\infty}^{\infty}\frac{F(\omega\tau_0) + i}{\omega}\,e^{-i\omega\tau}\,d\tau, \tag{7}$$

where

$$F(\omega\tau_0) = \frac{3}{2}\,\frac{\{1 + (1-i)\sqrt{\omega\tau_0}\}\{(1-i)\sqrt{\omega\tau_0} - \operatorname{th}[(1-i)\sqrt{\omega\tau_0}]\}}{(1-i)(\omega\tau_0)^{3/2}\{1 + \operatorname{th}[(1-i)\sqrt{\omega\tau_0}]\}}, \tag{8}$$

and

$$\tau_0 = a^2/2D = a^2\delta\eta/kT \tag{9}$$

is the time of realignment of the ordered region (δ is a constant). A plot of the function $F(\omega\tau_0)$ is given in[9]. When $\omega\tau_0 \ll 1$ we have

$$F(\omega\tau_0) = {}^4/_5\,\omega\tau_0 - {}^2/_3(\omega\tau_0)^{3/2} - i[1 - {}^2/_3(\omega\tau_0)^{3/2}]. \tag{10}$$

When $\omega\tau_0 \gg 1$ we have

$$F(\omega\tau_0) = \frac{3}{4}\,\frac{1-i}{\sqrt{\omega\tau_0}}. \tag{11}$$

The correlation function corresponding to the assumption that the molecule can become reoriented both as a result of realignment and as a result of Brownian rotation, is the product of the correlation function corresponding to the realignment by the correlation function corresponding to the Brownian rotation, that is,

$$\Phi(\tau) = \exp\{-|\tau|/\tau_c\}\,\overline{\xi_{1,\delta}'}(\tau). \tag{12}$$

Using (7) and (12), let us calculate the quantities $J^{(1)}(\omega_0)$ and $J^{(2)}(2\omega_0)$ which enter in (1) and (2). After simple transformations we obtain

$$J^{(q)}(\omega_0 q) = G^{(q)}(0)\left\{\frac{F(-\omega_0 q\tau_0 + i\tau_0/\tau_c) + i}{-\omega_0 q + i/\tau_c} + \frac{F(\omega_0 q\tau_0 + i\tau_0/\tau_c) + i}{\omega_0 q + i/\tau_c}\right\} \tag{13}$$

($q = 1, 2$). Substituting these expressions in (1) and calculating $G^q(0)$, we get an expression for the reciprocal of the relaxation time in the ordered region. Substituting (13) in (2) and noting that the term with $q = 0$ is determined only by the Brownian rotation, we obtain an expression for the reciprocal of the transverse relaxation time. We shall not write out these expressions, to save space, and present only the formula for the reciprocal of the longitudinal relaxation time for the entire liquid as a whole, that is, with allowance for the contribution from the unordered part of the liquid:

$$\frac{1}{T_1} = \frac{3}{20}\gamma^4\hbar^2 n\frac{1}{r_0^6}\left\{2\left[\frac{\tau_c}{1 + (\omega_0\tau_c)^2} + \frac{4\tau_c}{1 + 4(\omega_0\tau_c)^2}\right]\right.$$
$$\left. + \phi\left[\frac{F(-\omega_0\tau_0 + i\tau_0/\tau_c)}{-\omega_0 + i/\tau_c} + \frac{F(\omega_0\tau_0 + i\tau_0/\tau_c)}{\omega_0 + i/\tau_c}\right]\right.$$

$$+4\phi\left[\frac{F(-2\omega_0\tau_0+i\tau_0/\tau_c)}{-2\omega_0+i/\tau_c}+\frac{F(2\omega_0\tau_0+i\tau_0/\tau_c)}{2\omega_0+i/\tau_c}\right]\Bigg\}, \qquad (14)$$

ϕ is the relative volume occupied by the ordered regions. When $\omega_0\tau_c \gg 1$, this expression yields

$$\frac{1}{T_1}=\frac{3}{20}\nu^4\hbar^2 n\frac{1}{r_0^6}\left\{\frac{4}{\omega_0^2\tau_c}+\phi\frac{3}{2}\frac{1+\sqrt{2}}{\omega_0\sqrt{\omega_0\tau_c}}\sqrt{\frac{\tau_c}{\tau_0}}\right\}, \qquad (15)$$

and when $\omega_0\tau_c \ll 1$

$$\frac{1}{T_1}=\frac{3}{2}\gamma^4\hbar^2 n\frac{1}{r_0^6}\tau_c\left(1-\phi\frac{3\sqrt{2}}{4}\sqrt{\frac{\tau_c}{\tau_0}}\right), \quad \text{if} \quad \frac{\tau_0}{\tau_c}\gtrsim 1, \qquad (16)$$

$$\frac{1}{T_1}=\frac{3}{2}\gamma^4\hbar^2 n\frac{1}{r_0^6}\tau_c\left(1-\phi+\frac{4}{5}\phi\frac{\tau_0}{\tau_c}\right), \quad \text{if} \quad \frac{\tau_0}{\tau_c}\ll 1. \qquad (17)$$

The fundamental term in (15) is the second one, and thus T_1 changes when $\omega_0\tau_c \gg 1$ in proportion to $\eta^{1/2}T^{-1/2}\omega_0^{3/2}$. When $\omega_0\tau_c \ll 1$ the quantity T_1 varies in proportion to η/T and does not depend on the frequency, as follows from (16) and (17).

Let us compare now the results of the foregoing calculation with the experimental data. The circles and triangles in Fig. 1 represent the experimental points for the longitudinal relaxation time in glycerin at frequencies 20 and 21.5 MHz, as taken from [3,11]. The crosses denote the experimental points for 10 MHz from [3]. The solid lines show the theoretical curves at 20 MHz (curve 1) and 10 MHz (curve 2) obtained from formula (14). To plot these curves we assumed, as above, that $r_0 = 2.5 \times 10^{-8}$ cm, in agreement with the data on the lengths and angles of the bonds; τ_c was assumed equal to 8×10^{-9} sec at $\eta/T = 1.6$ poise/deg and to vary in proportion to η/T; τ_0 was obtained from acoustic measurements. The relative volume occupied by the ordered regions was estimated by starting from the assumption that it varies linearly from zero to unity when the temperature drops from the point of intersection of the elastic moduli (+153°C) to the vitrification temperature (−89°C). The dashed line in the same figure shows the curves given by the theory [1,2].

The circles and crosses in Fig. 2 show the experimental points for the longitudinal relaxation time in 1—3 butanediole at frequencies 20 and 10 MHz, respectively, taken from [3]. The continuous lines show the theoretical curves calculated from formula (14) for 20 MHz (curve 1) and 10 MHz (curve 2). To plot these curves, we assumed $r_0 = 2.6 \times 10^{-8}$ cm, which agrees with the data on the length and angles of the bonds, and τ_c was assumed equal to 8×10^{-9} sec at $\eta/T = 0.53$ poise/deg and to vary in proportion to η/T; τ_0 was taken from acoustic measurement. The relative volume occupied by the ordered regions was estimated in the same manner as for glycerin. For 1—3 butanediole the point of intersection of the elastic moduli is +116°C, and the vitrification temperature is not known exactly, but was assumed to be −120°C.

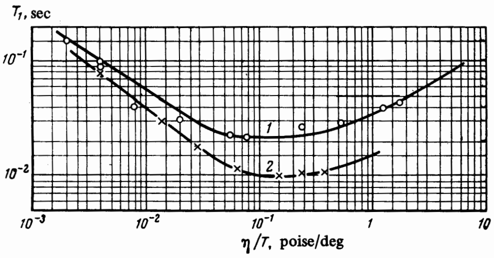

FIG. 2. Dependence of the longitudinal relaxation time T_1 on the ratio of the viscosity η to the absolute temperature T for 1 — 3 butanediole. Circles and crosses — experimental points taken from [3] for Larmor frequencies 20 and 10 MHz respectively. The viscosities are taken from [13]. The solid lines show the theoretical curves obtained from the proposed theory.

As seen from the figures, the proposed theory agrees well with experiment for the longitudinal relaxation time in the entire temperature range. It must be emphasized here that in constructing the theoretical curves, just as in [1,2], only two quantities, r_0 and τ_c, were taken from the experiment under consideration at a single temperature. Thus, the theory proposed in [7-9] for the construction and kinetics of strongly-viscous liquids, which has already made it possible to explain earlier the singularities in the propagation of acoustic, shear, and electromagnetic waves, turns out to be capable of explaining the singularities of NMR as well. In the derivation of (14) we assumed that the observed value of $1/T_1$ is an average taken over all molecules. In fact, in a number of experiments (see, for example, [14]) there is no such averaging, and under certain conditions it is possible to observe separately the relaxation of the magnetization of different groups of molecules. It is possible that in these experiments one can separate the relaxation given by the fraction of the molecules, the correlation of which is determined by the Brownian rotation, and the fraction of the molecules whose correlation is connected with the realignment[1)]. Here, however, the situation is aggravated by the fact that the molecules that are located at different distances from the center of the ordered region have different correlation times, so that the relaxation connected with the realignment cannot be described by a single exponential.

For an experimental verification of the hypotheses on which the theory of [7-9] is based, as well as for a check on the assumption that this theory is applicable to NMR, it would be interesting to attempt an observation of acoustic nuclear resonance, the appearance of which can be expected in highly viscous liquids, since periodic compressions and rarefactions in the acoustic wave lead to periodic realignments of the ordered regions, and consequently to periodic changes in the dipole-dipole interaction between the spins of the nuclei; in exactly the same way, one can expect the appearance of nuclear resonance when an alternating electric field is applied.

In conclusion, I am grateful to M. A. Isakovich for a discussion of the work.

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Translated by J. G. Adashko
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