Acoustic nuclear magnetic resonance in the case of direct generation of sound in metals

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The contribution to the NMR signal in a metallic plate due to acoustic waves generated by an external electromagnetic field is calculated for the case of mechanical resonance conditions. The relative magnitudes of the contributions to the NMR signal due to various sound absorption mechanisms in the nuclear spin system are estimated.

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In the observation of nuclear magnetic resonance (NMR) in conducting media, it is usually assumed that only the spins of nuclei located in the skin layer, of thickness δ, take part in the resonance absorption of energy of the electromagnetic field. However, it is known that in conductors placed in a constant magnetic field, weakly attenuated electromagnetic waves can be propagated in a direction parallel to the magnetic field.[1] It was shown theoretically by Antoniewicz and Rodriguez[2] that in the case of equality of the frequency of the helicon wave to the frequency of precession of the nuclear magnetic moment in a constant magnetic field, and at equality of the signs of the Hall coefficient of the sample and the nuclear magnetic moment, resonance absorption of energy of the helicon wave by the nuclear spins takes place. The first report on the experimental observation of such absorption was made by Shapoval.[3] It should be noted that the geometry of the experiments on NMR (the directions of the constant ($\mathbf{H}_0$) and the alternating ($\mathbf{H}_1$) magnetic fields) always corresponds to conditions of maximal generation of helicons in the conductor ($\mathbf{H}_0 \perp \mathbf{H}_1$).

The penetration of the electromagnetic field into the sample in the presence of a constant magnetic field is accompanied by direct generation of sound. The excited acoustical wave can in turn interact with the nuclear spins and acoustic NMR can be observed. Thus, in the observation of NMR in conductors, absorption of energy by the spins of the nuclei takes place not only in the skin layer, but throughout the entire volume of the sample, owing to the helicons and acoustic waves. Therefore, the experimentally observed NMR signal is the sum of the absorption signals. Depending on the experimental conditions, the maximal contribution to the NMR signal is made by the different absorption processes, and it is necessary each time to analyze carefully the nature of the observed signal.

Quinn,[4] has shown that the contribution to the NMR signal in metallic plates, due to the acoustic waves generated by the external electromagnetic field under conditions of mechanical resonance, can be several orders of magnitude larger than the contribution to the NMR from the skin layer. He has considered the magnetic dipole mechanism of sound absorption by the nuclear spins of the system, namely, the oscillations of the ions create a variable electromagnetic field at each point of the sample, and the nuclear spins interact with the magnetic components of this field. In the general case of arbitrary spin, the limited character of such a picture follows from the existence of electric quadrupole interaction of the nuclear spin system with the sound. But even for spin ½ there exists a specific mechanism of interaction with the sound through the modulation of dipole-dipole interactions. In the present work, the contribution of such mechanisms to the NMR signal is calculated and it is shown that it can be comparable in magnitude with the contribution from the magnetic dipole mechanism considered by Quinn.

The processes of propagation of electromagnetic and acoustic waves in a sample and their interaction with the nuclear spin system are described by the coupled system of equations of elastic waves, the Maxwell equations, and the equations that describe the evolution of the spin system. For the description of the NMR effect in conductors, we shall essentially follow the papers of Quinn and Rodriguez[4-7], which were devoted to the generation and interaction of helicons and phonons in conductors. Kaner and Fal'ko pointed out a limitation under which the Quinn theory of generation of acoustic waves is valid: $qR \ll 1$, where q is the wave number of the sound, R the cyclotron radius of the electron ($R = cmv_F/eH_0$, c is the velocity of light, m and e are the mass and charge of the electron, and v_F is the Fermi velocity). Under NMR conditions, $q = \gamma H_0/2\pi v_{ac}$, where γ is the gyromagnetic ratio and v_{ac} the velocity of sound. Then the condition $\gamma m v_F c/e\pi v_{ac} e \ll 1$ holds and is well satisfied for most metals.

We represent the meta by a model consisting of a lattice of positive ions of charge ζe and mass M and a gas of free electrons of charge $-e$ and mass m.[4-7] The short-range forces between the ions are described by elastic constants c_l and c_t associated with the compression displacements and the shear displacements, respectively. A phenomenological phonon relaxation time τ_p, due to non-electron damping of the sound is introduced.

The equation of motion for the i-th component of the ion displacement has the form

$$M(\ddot{\xi}+\dot{\xi}/\tau_p)_i=c_l(\nabla(\nabla\xi))_i-c_t[\nabla\times[\nabla\times\xi]]_i+\zeta eE_i+\frac{\zeta e}{c}[\dot{\xi}\times\mathbf{H}_0]_i+F_i+\frac{\partial\Sigma_{ik}}{\partial x_k}. \tag{1}$$

Here $\mathbf{F}$ represents the force of collision friction $\mathbf{F} = (-\zeta e/\sigma_0).\ (\mathbf{j}_e + \mathbf{j}_I)$, where $\mathbf{j}_e$ and $\mathbf{j}_I$ are the electron and ion current densities, and $\sigma_0 = ne^2\tau/m$ is the conductivity at constant current[4] (n is the density of the free electrons, τ the free path time of the electrons). The last term of Eq. (1) describes the force due to the existence of spin-phonon interaction,[10] Σ_{ik} is the elastic stress tensor.

The set of Maxwell equations is of the form

$$[\nabla\times\mathbf{H}]=\frac{4\pi}{c}\mathbf{J}+\frac{1}{c}\frac{\partial\mathbf{E}}{\partial t},\quad [\nabla\times\mathbf{E}]=-\frac{1}{c}\frac{\partial}{\partial t}(\mathbf{H}+4\pi\mathbf{M}). \tag{2}$$

The total current in the sample consists of the ion current, the electron current, and the fictitious surface current. For the Fourier components we have

$$\mathbf{J}_q=ine\omega\xi_q+\mathbf{j}_{eq}+\mathbf{j}_{0q}. \tag{3}$$

The expression for the electron current is of the form[11]

$$\mathbf{j}_{eq}=\hat{\sigma}_q\mathbf{E}_q-im\omega\hat{\sigma}_q\xi_q/e\tau+ne\hat{D}q(q\xi), \tag{4}$$

where $\hat{\sigma}_q$ is the conductivity tensor, $\hat{D}=(2E_F/3e^2n)(1+i\omega\tau)^{-1}\hat{\sigma}$ is the diffusion tensor, and E_F is the Fermi energy. The first term in (4) represents the conductivity, the second is the current due to collisions with ions, and the third is the diffusion current.

The expression for the nuclear spin magnetization M is found with the help of equations of the Bloch type for a spin system interacting with the electromagnetic field and with the sound. Such equations can be obtained by using the Wangsness-Bloch formula[12] for the mean values of the spins:

$$\frac{d\langle S_j\rangle}{dt}=-\frac{i}{\hbar}\langle[S_j,\mathscr{H}]\rangle+R(\langle S_j\rangle), \tag{5}$$

where $\mathscr{H}$ is the Hamiltonian of the spin system, which interacts with the external fields, and $R(\langle S_j\rangle)$ are the relaxation terms.

For a cubic crystal with the coordinates axes oriented along the three fourfold axes c_{4v}, the Hamiltonian of the spin-phonon interaction is of the form[13]

$$\begin{aligned}\mathscr{H}_{\rm sph}={}&{}^1/_3(\varepsilon_{xx}+\varepsilon_{yy}+\varepsilon_{zz})(G_{11}+2G_{12})HS\\&+{}^1/_6(\varepsilon_{xx}+\varepsilon_{yy}-2\varepsilon_{zz})(G_{11}-G_{12})(HS-3H_zS_z)\\&+{}^1/_4(\varepsilon_{xx}-\varepsilon_{yy})(G_{11}-G_{12})(H_xS_x-H_yS_y)+2G_{44}[\varepsilon_{xy}(H_xS_y\\&+S_xH_y)+\varepsilon_{xz}(H_xS_z+S_xH_z)+\varepsilon_{yz}(H_yS_z+S_yH_z)],\end{aligned} \tag{6}$$

where ϵ_{ij} (i, j = x, y, z) are the components of the deformation tensor, $G_{\alpha\beta}$ (α,β = 1, 2, 3) are the components of the spin-phonon interaction tensor in the Vogt notation. If the constant magnetic field $\mathbf{H}_0$ is directed along the z axis perpendicular to the surface of the sample, and the alternating magnetic field $\mathbf{H}_1$, as is usual in NMR experiments, is oriented in the xy plane, the transverse acoustic wave excited in the sample is propagated along $\mathbf{H}_0$.[11] Then

$$\mathscr{H}_{\rm sph}=H_0S_zG_{11}\varepsilon_{zz}+2H_0G_{44}(S_x\varepsilon_{xz}+S_y\varepsilon_{yz}), \tag{7}$$

and the total Hamiltonian of the spin system interacting with the electromagnetic and acoustic fields is

$$\begin{aligned}\mathscr{H}={}&-\gamma\hbar H_0S_z+H_0S_zG_{11}\varepsilon_{zz}+2H_0G_{44}(S_x\varepsilon_{xz}\\&+S_y\varepsilon_{yz})-\gamma\hbar(S_xH_x+S_yH_y).\end{aligned} \tag{8}$$

With the help of Eq. (5), we obtain a set of equations of the Bloch type for the average values of the spin components $\langle\tilde{S}_i\rangle$ in a system of coordinates which rotates with the frequency ω of the external electromagnetic field:

$$\begin{aligned}\frac{d\langle S_z\rangle}{dt}={}&\frac{iGH_0}{2\hbar}\int_{-\infty}^{\infty}iq(\langle\tilde{S}_+\rangle\xi_{q-}-\langle\tilde{S}_-\rangle\xi_{q+})e^{-iqz}\frac{dq}{2\pi}\\&+\frac{i\gamma}{2}\int_{-\infty}^{\infty}(\langle\tilde{S}_+\rangle H_{q-}-\langle\tilde{S}_-\rangle H_{q+})e^{-iqz}\frac{dq}{2\pi}-\frac{\langle S_z\rangle-S_0}{T_1};\\ \frac{d\langle\tilde{S}_+\rangle}{dt}={}&\langle S_+\rangle\left(-i\Delta-\frac{1}{T_2}\right)+\langle S_z\rangle\Big[-\frac{GH_0}{\hbar}\int_{-\infty}^{\infty}q\xi_{q+}e^{-iqz}\frac{dq}{2\pi}\\&+i\gamma\int_{-\infty}^{\infty}H_{q+}e^{-iqz}\frac{dq}{2\pi}\Big]\end{aligned} \tag{9}$$

and the conjugate equation for $\tilde{S}_-$. Here $\tilde{S}_\pm=e^{\mp i\omega t}S_\pm$, $S_\pm=S_x\pm iS_y$; $\Delta=\omega-\omega_0$, $\omega_0=-\gamma H_0$, T_1 and T_2 are the longitudinal and transverse relaxation times, and $S_0=-\frac{1}{2}\tanh(\hbar\omega_0/2k_BT)$. In the derivation of (9) we have represented the linearly-polarized field $\xi_{x,y}$ in the form of two fields with circular polarization $\xi_{+,-}$ and used the expansion of the components of the alternating fields in Fourier integrals. Similar expressions can be written for the components of the magnetic field.

Solving the system (9) for the steady state, we obtain expressions for the Fourier components of the nuclear magnetization:

$$\begin{gathered}M_{q\pm}=\chi_\pm H_{q\pm}e^{\pm i\omega t}+\varkappa_\pm q\xi_{q\pm}e^{\pm i\omega t},\\ \chi_\pm=\pm\frac{i\gamma T_2M_0(1\mp i\Delta T_2)}{1+\Delta^2T_2^2+T_1T_2\omega_1^2},\quad \varkappa_\pm=\mp\frac{T_2M_0a(1\mp i\Delta T_2)}{1+\Delta^2T_2^2+T_1T_2\omega_1^2},\\ \omega_1^2=\left[-a\int_{-\infty}^{\infty}q\xi_{q-}e^{-iqz}\frac{dq}{2\pi}+i\gamma\int_{-\infty}^{\infty}H_{q-}e^{-iqz}\frac{dq}{2\pi}\right]\\ \times\left[a\int_{-\infty}^{\infty}q\xi_{q+}e^{-iqz}\frac{dq}{2\pi}-i\gamma\int_{-\infty}^{\infty}H_{q+}e^{-iqz}\frac{dq}{2\pi}\right],\\ a=GH_0/\hbar,\quad M_0=\mu_nS_0.\end{gathered} \tag{10}$$

As is usual in the solution of Bloch equations, the terms containing ω_1^2 describe the saturation of the spin system. If the external fields are sufficiently weak ($T_1T_2\omega_1^2 < 1+\Delta^2T_2^2$), the saturation effects can be neglected.

The resultant set of equations (1)–(3), (11) differ from those used in[4,11] by the introduction of the last term in Eq. (1) and the presence of a dependence of the nuclear magnetization on the acoustic field (10). We solve this set, expressing $\mathbf{E}$ and ξ in terms of the surface current $\mathbf{j}_0$. We first find the expression for J_q in terms of E_q and ξ_q from the Maxwell equations (2):

$$\begin{gathered}J_{q\pm}=\pm i\sigma_0\vartheta_\pm E_{q\pm}\mp cq^2\frac{\varkappa_\pm}{\nu_\pm}\xi_{q\pm},\\ \nu_\pm=1+4\pi\chi_\pm,\quad \vartheta_\pm=\frac{c^2q^2}{4\pi\omega\sigma_0}\left(\frac{1}{\nu_\pm}-\frac{\omega^2}{c^2q^2}\right).\end{gathered} \tag{11}$$

Substituting $J_{q\pm}$ in the left side of (3), we get the expression for $E_{q\pm}$ in terms of $\xi_{q\pm}$ and $j_{0q\pm}$. In the "helicon" geometry used in the experiment ($H_0 \parallel z$, H_0 is perpendicular to the surface of the sample), under the assumption $\sigma_{xx}=\sigma_{yy}$, $\sigma_{xy}=-\sigma_{yx}$, $\sigma_{xz}=\sigma_{yz}=0$, it is convenient to introduce the circular components of the conductivity tensor $\sigma_\pm=\sigma_{xx}\mp i\sigma_{xy}$.[1] We obtain

$$\begin{gathered}E_{q\pm}=\pm\xi_{q\pm}\left(\frac{im\omega}{e\tau}\right)\frac{(\sigma_\pm-\sigma_0+i\lambda_\pm)}{(\sigma_\pm\mp i\sigma_0\vartheta_\pm)}-\frac{j_{0q\pm}}{(\sigma_\pm\mp i\sigma_0\vartheta_\pm)},\\ \lambda_\pm=\frac{cq^2e\tau}{m\omega}\frac{\varkappa_\pm}{\nu_\pm}.\end{gathered} \tag{12}$$

It is now necessary to use Eq. (1) to eliminate ξ. We first determine the form of $\partial\Sigma_{ik}/\partial x_k$. The elastic stress tensor Σ_{ik} is determined as the derivative of the free energy with respect to the elastic deformation tensor and is expressed in our case in terms of the magnetization of the spin system $\mathbf{M}$[10]:

$$\frac{\partial\Sigma_{xz}}{\partial z}=\frac{2GH_0}{\mu_nN}\frac{\partial M_x}{\partial z},\quad \frac{\partial\Sigma_{yz}}{\partial z}=\frac{2GH_0}{\mu_nN}\frac{\partial M_y}{\partial z}, \tag{13}$$

where N is the number of nuclei per unit volume. Substituting (12) and (13), which are expressed in terms of the circular components, in (1), we finally obtain the expression

$$E_{q\pm}=\left\{\mp\frac{i\zeta m\omega^2}{M\sigma_0\tau}\frac{[1-\sigma_0R_\pm(1+\psi_\pm)](1-\sigma_0R_\pm+iR_\pm\lambda_\pm)}{f_\pm(1\mp i\sigma_0R_\pm\vartheta_\pm)}-\frac{R_\pm}{1\mp i\sigma_0R_\pm\vartheta_\pm}\right\}j_{0q\pm}, \tag{14}$$

where

$$\begin{aligned}f_\pm={}&\left(\omega^2\mp\frac{i\omega}{\tau_p}-s^2q^2+\omega\Omega_c\pm\frac{\zeta ecq^2}{M\sigma_0}\frac{\varkappa_\pm}{\nu_\pm}-\frac{2iGH_0q^2}{M\mu_nN}\frac{\varkappa_\pm}{\nu_\pm}\right)\\&\times(\omega\mp i\omega\sigma_0R_\pm\vartheta_\pm)\pm\frac{i\omega^2\Omega_c}{\omega_c\tau}(1\mp i\vartheta_\pm+\psi_\pm)(1-\sigma_0R_\pm+iR_\pm\lambda_\pm),\end{aligned}$$

$$\psi_\pm=\frac{2GH_0cq^2}{\zeta e\mu_n N\omega}\frac{\chi_\pm}{\nu_\pm},\quad R_\pm=\frac{1}{\sigma_\pm},\quad \Omega_c=\frac{\zeta eH_0}{Mc},\quad s=\frac{c_t}{M}.$$

If we assume that the mechanisms of interaction of sound with the nuclear spin system that we have considered are ineffective, i.e., $G = 0$, the expression (14) transforms exactly into the corresponding Quinn expression.[4,11]

The surface impedance, which is determined by the formula

$$Z=\frac{4\pi}{c}\left[\frac{E(z)}{H(z)}\right]_{z=0} \tag{15}$$

is an experimentally observed quantity. In the case of the use of circular components of the field, it is customary to introduce the circular components of the surface impedance

$$Z_\pm={}^1/_2(Z_{xx}+Z_{yy})\mp iZ_{xy},\quad Z_\pm'={}^1/_2(Z_{xx}-Z_{yy}),\quad E_\pm=Z_\pm J_\pm+Z_\pm' J_\mp. \tag{16}$$

If $Z_{xx} = Z_{yy}$, Eq. (15) can be reduced to the form

$$Z_\pm=\pm i\frac{4\pi}{c}\left[\frac{E_\pm(z)}{H_\pm(z)}\right]_{z=0}; \tag{17}$$

$H_\pm(z)$ is found from the second of Maxwell's equations (2) and in our case, for $z = 0$, it takes the form

$$H_\pm(0)=\frac{2\pi}{c\nu_\pm}\left[\frac{2\zeta e\omega c}{M\sigma_0}\varkappa_\pm\int_{-\infty}^{\infty} j_0(q)q\frac{1-\sigma_0R_\pm(1+\psi_\pm)}{f_\pm}dq+1\right]_{z=0}. \tag{18}$$

Up to now, we have carried on the calculation for a semi-infinite metal with boundary at $z = 0$. We make the problem more specific by calculating the impedance of a metal plate of thickness d. In NMR experiments, the sample is placed in the antinode of an alternating magnetic field, i.e., in accord with the terminology of Bass, Blank, and Kaganov[14], asymmetric excitation of the plate is produced ($E_{0-} = -E_{d+}$, $E_{a\pm} = E(z = a \pm \epsilon)$, ϵ is infinitesimally small). If we use the condition of specular reflection of the electrons from the surface ($E_{0+} - E_{0-}$, $(\partial E/\partial\epsilon)_{0-} = -(\partial E/\partial z)_{0+}$), then the expression for the Fourier components of the surface current have the form (see[15])

$$\frac{4\pi i\omega}{c^2}\mathbf{j}_0(q)=\frac{2\pi}{d}\sum_{n=-\infty}^{\infty}e^{iqnd}\Delta\mathbf{E}_0'\delta_{q,(2l+1)\pi/d},$$

$$\Delta\mathbf{E}_0'=\left[\left(\frac{\partial\mathbf{E}}{\partial z}\right)_{0-}-\left(\frac{\partial\mathbf{E}}{\partial z}\right)_{0+}\right]=-2\left(\frac{\partial\mathbf{E}}{\partial z}\right)_{0+}. \tag{19}$$

where n and l are integers.

Substituting (19), (18), and (14) in Eq. (17), we obtain an expression for the surface impedance of a conducting plate in the presence of direct generation of sound in the plate by the external electromagnetic field:

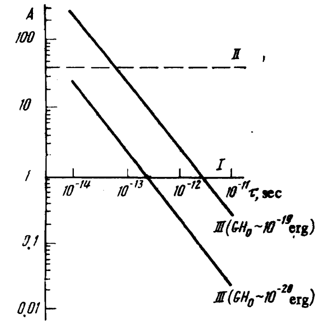
$$Z_\pm=\pm\left\{\frac{c\nu_\pm}{2\pi d}\sum_{q_l}\left[-\frac{R_\pm}{1\mp i\sigma_0R_\pm\vartheta_\pm}\mp\frac{i\zeta m\omega^2}{M\sigma_0\tau}\right.\right.$$
$$\left.\left.\times\frac{[1-\sigma_0R_\pm(1+\psi_\pm)](1-\sigma_0R_\pm+iR_\pm\lambda_\pm)}{f_\pm(1\mp i\sigma_0R_\pm\vartheta_\pm)}\right]\right\}$$
$$\times\left\{1-\frac{2ic^2\zeta e}{M\sigma_0 d}\sum_{q_l}q\frac{1-\sigma_0R_\pm(1+\psi_\pm)}{f_\pm}\right\}^{-1}. \tag{20}$$

It must be noted immediately that the denominator differs little from unity under ordinary NMR conditions.

We use Eq. (20) to estimate the relative contribution to the NMR signal from the various mechanisms. Tentatively, we can distinguish three mechanisms.

I. The ordinarily considered contribution of electromagnetic absorption without account of sound generation; it is described by the first term in the numerator of Eq. (20).

Relative magnitude (A) of contributions to the NMR signal from different mechanisms of absorption of energy of the nuclear spin system.



II. The contribution considered by Quinn and due to the mechanism of magnetic dipole absorption of the sound wave generated in the sample; it is described by the second term in the numerator of (20) if we set the spin-phonon interaction constant $G = 0$.

III. The contribution due to the sound-absorption mechanisms other than the magnetic-dipole mechanism; it is described by the part of the second term in the numerator of (20) which has dropped out in the previous case.

For estimates we use the following values of the physical quantities, which are typical for metals: $n \sim 10^{23}$ cm^{-3}, $N \sim 10^{23}$ cm^{-3}, $m \sim 10^{-27}$ g, $M \sim 10^{-24}$ g, $\gamma \sim 10^4$ cm$^{1/2}$g$^{1/2}$, $H_0 \sim 10^4$ Oe, $GH_0 \sim 10^{-19}$ erg. The free path time of the electrons τ depends essentially on the temperature, and ranges from $\sim 10^{-14}$ sec ($T = 300°K$) to $\sim 10^{-11}$ sec ($T = 4°K$). The parameter τ_p that determines the nonelectronic damping time of sound is estimated from the acoustic quality factor of the samples and for frequencies $\omega \sim 10^7-10^8$ sec^{-1} can be taken as $\tau_p \sim 10^{-3}-10^{-4}$ sec^{-1}.[4] The local approximation $\sigma_\pm = \sigma_0/(1 \mp \omega_c\tau)$ is used.

As was to be expected, the large contribution to the NMR signal is made by only one circular polarization (in our case, +). The results of the numerical estimates for this polarization are given in the figure. The Roman numerals label curves that describe the relative contributions to the NMR signal corresponding to different mechanisms of absorption. The value of the NMR signal under the assumption of the absence of sound generation was taken as unity.

The following conclusion can be drawn: 1) account of the generation of sound in a metal in the observation of NMR is necessary in most cases; 2) the contribution to the NMR signal due to mechanisms, other than the magnetic dipole mechanism of absorption of sound generated by the spin-system in the sample can be significant.

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