both of which are mainly delocalized. This result points up a key reason for the success of independent-electron models for solids, namely that the variable part of the electron-interaction energy is a very small part of the total interaction energy.

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# NUCLEAR ACOUSTIC RESONANCE IN ALUMINUM VIA COUPLING TO THE MAGNETIC DIPOLE MOMENT

J. Buttet and E. H. Gregory*
California Institute of Technology, Pasadena, California 91109

and

P. K. Baily†
University of California, Los Angeles, California 90024

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We have observed a new interaction mechanism between a sound wave and the nuclear spin system in a single crystal of aluminum. The coupling is via the oscillating magnetic field induced by the sound wave in the presence of a large applied magnetic field and the magnetic moment of the nucleus. The experimental results agree with the theoretical predictions based on this mechanism.

The acoustic excitation of nuclear-spin resonance in metals has been reported unambiguously in tantalum.[1] We have recently observed the nuclear acoustic resonance in single crystals of niobium.[2] The interaction mechanism for both tantalum and niobium is the coupling between the oscillating electric field gradient created by the sound wave and the electric quadrupole moment of the nucleus. This is indicated by the observation of the $\Delta m = 2$ transition as well as the $\Delta m = 1$ transition. We report in this Letter the observation of the acoustic excitation of the $\Delta m = 1$ nuclear spin transition in a single crystal of aluminum[3] (at 300 and 77°K) and we present evidence that the coupling mechanism is the interaction between the oscillating magnetic field induced by the sound wave in the presence of a large applied magnetic field and the magnetic moment of the nucleus.

According to the mechanism of Alpher and Rubin[4] the sound wave forces a mechanical motion of the charged particles in a conducting medium. In the presence of an applied magnetic field these charges are deflected and a transverse current is set up, which generates an electromagnetic field propagating in a metal at the velocity of the sound wave. The induced fields modify slightly the velocity[5] of the sound wave as well as the attenuation. Both effects are proportional to the square of the applied magnetic field and have been observed in aluminum.[6,7]

On the basis of this mechanism, we have calculated the acoustic attenuation coefficient for the absorption of energy by the nuclear spin system. The attenuation coefficient, $\alpha_n$, is defined as $\frac{1}{2}P_n/P_0$, where $P_0$ is the incident acoustic power per unit area and is equal to $\frac{1}{2}\rho v_s{}^3\epsilon^2$, where $\rho$ is the density of the metal, $v_s$ the velocity of the sound wave appropriate to shear or longitudinal wave propagation, and $\epsilon$ the peak value of the strain.[8] $P_n$ is the power per unit volume absorbed by the nuclear spin system and is given by

$$P_n = \frac{N(h\nu)^2}{(2I+1)kT}\sum_m W_{mm'}, \qquad (1)$$

where $N$ is the number of nuclear spins per unit volume, $\nu$ is the frequency of the sound wave, and $W_{mm'}$ the transition probability per unit time from the spin state $m$ to the state $m'$. For magnetic dipole coupling, $W_{mm'}$ is well known[9] and is proportional to the square of the oscillating magnetic field component perpendicular to the applied

magnetic field. In the present case, this oscillating field is that which is induced by the sound wave. Equation (1) ignores the effect of eddy currents; this may be important in a metal.

We have calculated the magnitude of the induced rf magnetic field $\vec{B}$, on the basis of the model given by Rodriguez[10,11] for the modification of the velocity of sound in metals by an applied magnetic field. Rodriguez calculates the relation between the self-consistent electric field and the displacement field $\vec{\xi}(r,t)$.[12] Through the Maxwell equations, we obtain the induced magnetic field. In general, the phase between the velocity of the particle motion and the electric field is given by an angle whose tangent $\beta$ is equal to $c^2q^2/4\pi\omega\sigma_0$, where $c$ is the speed of light, $q$ the magnitude of the sound wave vector, and $\sigma_0$ the dc conductivity. If $\beta$ is equal to zero, we can calculate the power absorbed by the spin system by using Eq. (1). The absorbed power will be proportional to the imaginary component $\chi''$ of the complex susceptibility $\chi$. If $\beta$ differs from zero, the component of the electric field which is not in phase with the particle velocity will give rise to an absorption of energy proportional to the real component $\chi'$ of the susceptibility. In the present calculation we have assumed that $\beta$ is zero.

We have chosen a Cartesian coordinate system such that the sound wave vector points in the $z$ direction and the applied magnetic field $\vec{B}_0 = (0, B_0\sin\theta, B_0\cos\theta)$ lies in the $y$-$z$ plane and forms an angle $\theta$ with $\vec{q}$. We have also assumed that the conditions $\omega\tau \ll 1$, $(\omega_c\tau)^2 \ll 1$, and $ql \ll 1$ are valid, where $\omega_c$ is the cyclotron frequency, $\tau$ the relaxation time, and $l$ the electron mean free path. For pure metals at room temperature or liquid-nitrogen temperature, with frequencies below 1000 MHz and with fields under 20 kG, these conditions are well satisfied. The amplitude of the component of $\vec{B}$ in a plane perpendicular to $\vec{B}_0$ is then given by

$$B_\perp = \epsilon B_0 f(\theta), \tag{2}$$

where $f(\theta)$ is an angular factor depending on the polarization of the sound wave. The value of $f(\theta)$ is given in Table I.

Table I. Angular dependence of the component of $\vec{B}$ perpendicular to $\vec{B}_0$.

| | $\vec{\xi}$ | $f(\theta)$ |
|---|---|---|
| Longitudinal | $(0,0,\xi)$ | $\sin\theta\cos\theta$ |
| Shear $z$-$x$ | $(\xi,0,0)$ | $\cos\theta$ |
| Shear $z$-$y$ | $(0,\xi,0)$ | $\cos^2\theta$ |

From Eqs. (1) and (2), we obtain

$$\alpha_\eta = \frac{\pi^2 h^2 NF(I)g(\nu)\nu^4}{4kT\rho v_s{}^3}[f(\theta)]^2, \tag{3}$$

where $g(\nu)$ is the normalized line-shape factor and $F(I)$ is a spin factor given by

$$F(I) = \frac{1}{2I+1}\left[2I^2(I+1) - \sum_{m=-I+1}^{I} m(m-1)\right]. \tag{4}$$

For the case of a 20-MHz transverse wave, propagating at room temperature along the [001] direction in aluminum, we calculate $\alpha_\eta = 1.7\times10^{-7}$ cm$^{-1}$, if a Gaussian line shape is assumed.

The measurements have been done with a marginal-oscillator ultrasonic spectrometer[13] which gives the derivative of the attenuation coefficient. The experiments have been performed at 77°K and at room temperature. In the best conditions (20 MHz, shear wave) the signal-to-noise ratio was greater than 20, if we used a time constant of 10 sec and a modulation field of 4 G peak-to-peak. The value of the resonance field for the $\Delta m = 1$ transition is in agreement with the known value. In order to verify the proposed interaction mechanism, we have studied the angular dependence of the absorption coefficient. Figures 1 and 2 show the measured angular dependence of the area under the absorption curve. This area should be proportional to $[f(\theta)]^2$. Within experimental error, these points fitted the predicted dependences for longitudinal and transverse waves. We have also compared the measured room-temperature values of $\alpha_\eta$ at 20 and 10 MHz. The measured ratio of these values is 33:1. The ratio predicted by Eq. (3) is 16:1, while that for the interaction between the nuclear electric quadrupole moment and the acoustically induced elec-

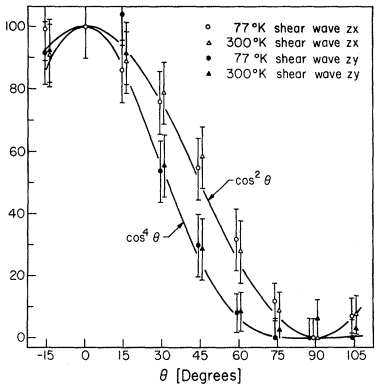


FIG. 1. Area under absorption curve, $\alpha_\eta$, as a function of angle between $\vec{B}_0$ and $\vec{q}$. The 20-MHz sound wave propagates along the [001] axis of the aluminum crystal and $\vec{B}_0$ is always in the (100) plane. The one nonzero value at 90° is probably due to a misorientation of the sample.

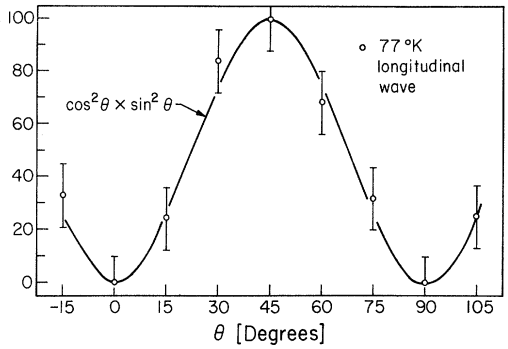


FIG. 2. Area under the absorption curve, $\alpha_n$, as a function of angle between $\vec{B}_0$ and $\vec{q}$. The 20-MHz sound wave propagates along the [001] axis of the aluminum crystal and $\vec{B}_0$ is always in the (100) plane. Experiments done at room temperature had a larger experimental error but were in agreement with these results.

tric field gradient is 4:1.[8] Our absolute measurements of $\alpha_n$ require knowledge of the background attenuation, as well as other parameters. Since these are not accurately known in this experiment, we feel that ratios of 33:1 and 16:1 are compatible within experimental error. The measured value of $\alpha_n$ for 20-MHz transverse waves at room temperature is approximately three times the calculated value. We are planning more accurate measurements. We have searched very carefully, but have been unable to detect the $\Delta m = 2$ transition resonance signal which would be expected if the coupling mechanism were the quadrupole interaction. This places an upper limit of $5\times10^{-8}$ cm$^{-1}$ on the value of the absorption coefficient due to the quadrupole interaction. Furthermore, the angular dependence of $\alpha_n$ for $z$-$y$ shear waves in $(\sin^2\theta-\cos^2\theta)^2$ for the $\Delta m = 1$ quadrupole transition.[8] This is not consistent with our results. We feel that these experiments confirm the coupling via the magnetic dipole moment.

The observed line shapes at 77 and 300°K are not the same. At 77°K, the resonance signal is symmetric. The absorption line shape changes with the orientation of the applied magnetic field. The linewidth is greater when $\vec{B}_0$ is along the [011] axis than when $\vec{B}_0$ is along the [001] axis. The variations in shape and width are the same as that observed by conventional nuclear magnetic resonance (NMR) in the skin depth of a single crystal after correction for the distortion due to eddy currents.[14] At room temperature, the resonance signal is asymmetric. The high-field side of the derivative of the absorption curve is higher and broader than the low-field side. The linewidth varies in the same manner as at 77°K. At room temperature the derivative of the absorption curve is similar to that observed in conventional NMR in metals before correcting for the distortion due to eddy currents. This line shape suggests that there is a term in the power absorption which is proportional to the real part of the susceptibility. At room temperature, the estimated value of $\beta$ is 0.25 and we cannot neglect the component of the electric field that is out of phase with the particle velocity. As discussed earlier, this introduces a $\chi'$ term into the power absorption. At 77°K, $\beta$ is approximately ten times smaller. The $\chi'$ term is then negligible and a symmetric line shape is expected.

This new interaction mechanism opens the possibility of studying single crystals of metals with nuclear spins of $\frac{1}{2}$ of with very weak quadrupole moments; investigations are currently in progress. We are also continuing to investigate the details of the interaction in aluminum, including the effect of a nonzero value of $\beta$ and that of low temperature where one can no longer assume a very short relaxation time.[15]

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*Present address: Hughes Aircraft Company, Culver City, Calif.

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