Nuclear-Acoustic-Resonance Absorption and Dispersion in Aluminum*

R. G. Leisure, D. K. Hsu, and B. A. Seiber
Department of Physics, Colorado State University, Fort Collins, Colorado 80521
(Received 16 April 1973)

Nuclear-acoustic-resonance dispersion has been observed for the first time in a metal. Absorption and dispersion signals in aluminum were investigated as a function of frequency, sound-wave velocity, and temperature. The line shapes of both absorption and dispersion signals show temperature-dependent asymmetries which are in striking agreement with theory.

We report, in this Letter, results of experimental studies of nuclear-acoustic-resonance (NAR) absorption and dispersion in aluminum. The observation of NAR dispersion in a metal has not previously been reported. We have studied the pure dispersion signal in aluminum over the temperature range of 70 to 275 K. We have also studied the pure absorption signal over the same temperature range. Our results are particularly pertinent to the various theories of NAR in metals.[1-5]

An acoustic wave propagating in a metal, in the presence of a static magnetic field, produces a transverse current which results in an electromagnetic field oscillating at the acoustic-wave frequency.[6] The coupling of this oscillating field to the nuclear magnetic-dipole moment results in the acoustic excitation of nuclear magnetic resonance, as was first observed in aluminum.[7] The NAR absorption signal in aluminum was found to be asymmetric. The asymmetry was qualitatively interpreted in terms of a mixture of χ' and χ'' (the real and imaginary parts of the complex nuclear susceptibility), the χ' part being related to the fact that there is a component of the oscillating field which is 90° out of phase with the acoustic wave. The actual theoretical expressions for the NAR absorption coefficient differ markedly in their dependences on χ' and χ''.[2,3] Recently, Fedders presented a comprehensive theory of the coupled system of acoustic waves, electrons, and nuclear spins. His results,[5] for both absorption and dispersion, are

$$\Delta\alpha=\frac{\omega B_0^2 S(\theta)}{2\rho v^3(1+\beta^2)^2}[(1-\beta^2)\chi''-2\beta\chi'], \tag{1a}$$

$$\frac{\Delta v}{v}=\frac{B_0^2 S(\theta)}{2\rho v^2(1+\beta^2)^2}[(\beta^2-1)\chi'-2\beta\chi'']. \tag{1b}$$

In these equations, ω is the angular frequency of the acoustic wave, B_0 is the static magnetic field, ρ is the density of the solid, v is the velocity of the acoustic wave, and $S(\theta)$ is a factor which depends on the angle θ between the propagation direction and the magnetic field direction. $\Delta\alpha$ and Δv are, respectively, the resonant change in attentuation (absorption) and resonant change in phase velocity (dispersion) of the acoustic wave due to its interaction with the nuclear spins. The factor β is equal to $\omega c^2/4\pi\sigma_0 v^2$ where c is the velocity of light and σ_0 is the dc electrical conductivity. Equation (1a) is the same as that obtained in Ref. 3 for the case $\theta=0$. Equations (1) were derived under the assumption that $\omega_c\tau$, $\omega\tau$, and $q\Lambda$ are all much less than 1, where ω_c is the cyclotron frequency, τ the electron relaxation time, q the acoustic wave vector, and Λ the electron mean free path. These assumptions are valid in aluminum for the frequencies, fields, and temperatures employed in the present work.

An experimental and theoretical study of the NAR absorption signal has been reported for niobium.[3] Equation (1a) was derived and the experimental results found to be consistent with that equation. Unfortunately, the comparison of experimental results with theory was difficult since the NAR absorption signal in niobium had a complex shape. This signal was attributed to the sum of two signals: a broad, symmetric line due to coupling between the acoustic wave and the nuclear electric-quadrupole moment, and a narrow, asymmetric line due to coupling with the nuclear magnetic-dipole moment. It was necessary to subtract the symmetric line before analyzing the asymmetric line, and the uncertainty in the decomposition process led to large errors. The present results are more direct. We have studied both the NAR absorption and dispersion signals in aluminum where previous work[7] has shown the coupling to be due to the dipolar interaction. The conclusion of dipolar coupling was based on the absence of a $\Delta m=2$ transition, which is expected for quadrupolar coupling, and the angular dependence of the absorption signal, which

clearly indicated dipolar coupling. Our angular dependence results, being in qualitative agreement with Eq. (21) of Ref. 5, also indicate dipolar coupling.

The measurements were made on a single-crystal aluminum cylinder 1.05 cm in length and approximately 1 cm in diameter, oriented with its axis along the [100] direction. The crystal, grown from 99.9999%-pure raw material, was obtained from Research Crystals, Inc. A continuous-wave transmission spectrometer[8,9] was used to make the measurements. The use of this spectrometer has been extended during this work to include detection of the dispersion signal. (Previously, NAR dispersion was only observed in magnetic insulators[10,11] and only for very strong spin-phonon interactions was it possible to separate the absorption and dispersion signals.) Quartz piezoelectric transducers were attached to opposite (100) faces of the sample and acoustic standing waves were excited. If one transducer is driven by a unit voltage $\cos\omega t$, the voltage V across the opposite transducer is given by[12]

$$V = V_0 \cos(\omega t - \varphi), \tag{2a}$$

where

$$V_0 = K/[2\cosh 2\alpha l - 2\cos 2ql]^{1/2}, \tag{2b}$$

$$\varphi = ql + \arctan[\sin(2ql)/(e^{2\alpha l} - \cos 2ql)]. \tag{2c}$$

In these equations α is the ordinary acoustic attenuation coefficient, l is the sample length, and K depends on the conversion efficiency of the transducers. Equations (2) have been modified slightly from those given in Ref. 12, because, in the transmission case, we are considering the voltage at the transducer *opposite* the driving transducer. Using Eqs. (2) it is easily shown that, at a standing-wave resonance ($ql = m\pi$, $m = 0, 1, 2, \ldots$), small changes in V_0 reflect only changes in the attenuation while small changes in φ reflect only changes in the acoustic phase velocity. The spectrometer of Ref. 8 used a crystal detector to detect the absorption by detecting changes in V_0. We have extended the technique by using a phase detector (double balanced mixer) to detect the pure dispersion by detecting changes in φ. In practice, the frequency of the spectrometer was adjusted to the center of an acoustic standing-wave resonance and the magnetic field was scanned through the region corresponding to magnetic resonance. Magnetic field modulation and synchronous detection were used, resulting in the usual derivative signal being recorded. In order to obtain good signal strength, magnetic fields in the range of 60 kOe were used for most of this work.

Equations (1) predict that both the absorption and the dispersion signals are admixtures of χ' and χ'', the actual percentages of χ' and χ'' depending on the parameter β. β, in turn, depends on the three quantities ω, v, and σ_0. Although the dependences of the signals on all three quantities were investigated, the most definitive results came from the study of the dependence on σ_0, the electrical conductivity. The electrical conductivity is most conveniently changed by changing the temperature. The present results span the temperature range of 70 to 275 K. The corresponding range of β for 64-MHz shear waves along the [100] direction is 0.050 to 0.707.

Figure 1 shows experimental derivative curves

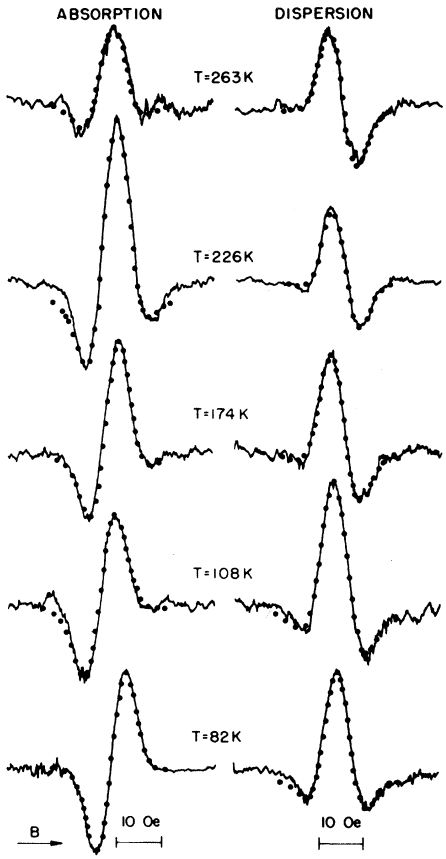

FIG. 1. Derivatives of absorption and dispersion signals at various temperatures. The solid lines are the experimental curves, while the dots represent a theoretical fit to the data. 64-MHz shear waves were propagated along the [100] direction which was also the magnetic field direction.

for absorption and dispersion at various temperatures. These results are for shear waves propagating along the [100] direction, which was also the magnetic field direction. The approximate values of the frequency and field were, respectively, 64 MHz and 57.6 kOe. The exact values were slightly temperature dependent due to the weak temperature dependence of the standing-wave resonant frequency. At $T = 82$ K, β is calculated to be 0.082. Figure 1 shows quite clearly that at this temperature the absorption signal resembles the derivative of χ'' while the dispersion curve resembles the derivative of χ'.[13] As the temperature is increased the absorption and dispersion curves pick up, respectively, a larger component of χ' and χ''. At $T = 263$ K, the shapes of the curves have essentially reversed, as compared to the low-temperature curves.

Attempts were made to fit the curves of Fig. 1 with both Lorentzian and Gaussian line shapes. It was found that the Gaussian gave the better fit. The fitting was done in the following manner. The published value[14] of the electrical conductivity was used to calculate β at a given temperature. With this value of β, Eqs. (1) were used to calculate the percentages of χ' and χ''. Using a Gaussian line shape, a computer was programmed to add the appropriate percentages of $d\chi'/dB$ and $d\chi''/dB$. The points in Fig. 1 represent the results of this computation. The width and height of the computed curve were, of course, adjusted to match the experimental curve, but the shape of the computed curve depends only on the experimental value of β and Eqs. (1). The value of β was computed from independent data and was *not* adjusted to fit the experimental curve. As Fig. 1 shows, Eqs. (1) describe remarkably well the temperature-dependent asymmetries of the absorption and dispersion curves. Our method of analyzing the data permits us to extract the linewidth in terms of the peak-to-peak width of $d\chi''/dB$. We find the width to be 6.8 ± 0.2 Oe over the temperature range investigated. This width is in good agreement with the value of 7.2 Oe reported from NMR measurements,[15] within the skin depth, on single-crystal aluminum with the magnetic field also along the [100] direction.

The experimental curves may be used to determine the value of β, which may then be compared with the value of β computed from the electrical conductivity, frequency, and sound velocity. From the asymmetries of the experimental curves the percentages of χ' and χ'' were determined, assuming a Gaussian line shape. Knowing these percentages, β was computed from Eqs. (1). The results are presented in Fig. 2. The solid line is the value of β computed from the electrical conductivity, frequency, and sound velocity. The circles and triangles represent, respectively, values of β determined from the absorption and dispersion curves. The results again strongly indicate that Eqs. (1) give the correct admixtures of χ' and χ''.

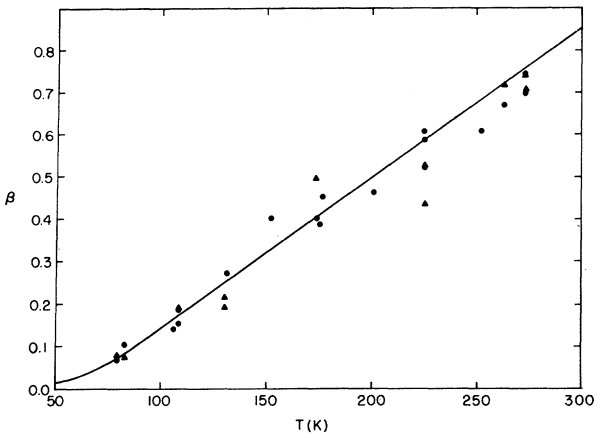

FIG. 2. Temperature dependence of β for the experimental conditions of Fig. 1. The solid line represents the calculated value of β. The circles and triangles represent, respectively, values of β determined from the absorption and dispersion curves.

Shear-wave absorption and dispersion signals were measured at 22.3 MHz, at 79 K, and compared with the results at 63.5 MHz. We find that $\Delta\alpha$ for the latter is a factor of 95 greater than the former. Using the appropriate values of β in Eq. (1a), we find a predicted factor of 70. We also measured the attenuation change for both longitudinal and shear waves at 63 MHz. We found that the shear attenuation was a factor of 50 greater than the longitudinal attenuation. Using the appropriate values of β, v, and $S(\theta)$ in Eq. (1a) we find a predicted increase of 30. In both cases we feel that the agreement between theory and experiment is satisfactory, in view of the difficulty of making reliable intensity measurements.

We have experimentally investigated the nuclear-acoustic-resonance absorption and dispersion signals in aluminum over a temperature range where the theory of Fedders is expected to be valid. We find that the temperature-dependent asymmetry is well accounted for by theory. The dependence of the absorption-signal amplitude on frequency and sound-wave velocity is also in ac-

cord with theory. We are presently extending the work to lower temperatures where the theory used here is no longer valid.

The authors are grateful for helpful discussions with Professor D. A. Krueger and the receipt of a preprint from Professor P. A. Fedders.

*Research supported in part by the U.S. Air Force Office of Scientific Research under Grant No. 71-2042, and in part by Research Corporation.

[1]J. J. Quinn, J. Phys. Chem. Solids 31, 1701 (1970).

[2]J. G. Miller, W. D. Smith, D. I. Bolef, and R. K. Sundfors, Phys. Rev. B 3, 1547 (1971).

[3]J. Buttet, Solid State Commun. 9, 1129 (1971).

[4]J. R. Franz, Phys. Lett. 37A, 197 (1971).

[5]P. A. Fedders, Phys. Rev. B 7, 1739 (1973). Equations (20) of this reference are in error by a factor β. We are indebted to Professor Fedders for this information.

[6]R. A. Alpher and R. J. Rubin, J. Acoust. Soc. Amer. 26, 452 (1954).

[7]J. Buttet, E. H. Gregory, and P. K. Bailey, Phys. Rev. Lett. 23, 1030 (1969).

[8]R. G. Leisure and D. I. Bolef, Rev. Sci. Instrum. 39, 199 (1968).

[9]R. G. Leisure, D. K. Hsu, and B. A. Seiber, Phys. Lett. 42A, 195 (1972).

[10]R. L. Melcher and D. I. Bolef, Phys. Rev. Lett. 20, 1338 (1968).

[11]J. B. Merry and D. I. Bolef, Phys. Rev. Lett. 23, 126 (1969).

[12]J. G. Miller and D. I. Bolef, J. Appl. Phys. 39, 4589 (1968).

[13]G. E. Pake and E. M. Purcell, Phys. Rev. 74, 1184 (1948).

[14]L. A. Hall, *Survey of Electrical Resistivity Measurements on 16 Pure Metals in the Temperature Range 0 to 273°K*, National Bureau of Standards Technical Note 365 (U.S. GPO, Washington, D.C., 1968).

[15]P. L. Sagalyn and J. A. Hofmann, Phys. Rev. 127, 68 (1962).

Magneto-optic Kerr Effect in Ni, Co, and Fe

J. L. Erskine and E. A. Stern
Department of Physics, University of Washington, Seattle, Washington 98195
(Received 30 March 1973)

Magneto-optic Kerr-effect spectroscopy of Ni, Co, and Fe is used to study d-band widths and electron spin polarization (ESP). The d-band widths determined on the basis of magneto-optic Kerr-effect spectra are wider than those established by photoemission, and the sign of ESP of electrons near the Fermi level in Ni is found to be negative, as predicted by band theory. We discuss the relationship between this work and other recent studies of ESP.

Recent experimental studies of Ni, Co, Fe, and Gd have created new interest in the electronic structure of ferromagnets. Photoemission spectroscopy has been used to study the widths and general shapes of occupied d bands in Ni, Co, and Fe[1] and in Gd,[2] and to study $4f$ levels in rare-earth metals.[3,4] Spin-polarized photoemission spectroscopy has been recently introduced and applied to Ni, Co, and Fe[5] and to Gd,[6] and spin-polarized field-emission spectroscopy has been applied to the study of Gd[7] and Ni.[8] In addition, an interesting experimental investigation of the electron spin polarization (ESP) of Fermi level electrons in Ni, Co, Fe, and Gd using spin-dependent tunneling techniques has been recently reported.[9]

These experimental studies have produced some rather interesting results: The spin-dependent tunneling experiments measure a positive ESP of electrons within 0.001 eV of the Fermi level E_F in Ni, Co, Fe, and Gd. This is rather surprising in the case of Ni since the Stoner-Wohlfarth-Slater (SWS) band model of ferromagnetism applied to Ni predicts a net *negative* ESP for electrons near E_F. The spin-polarized photoemission experiments (which probe a depth between 0.4 and 0.8 eV below E_F) also measure a positive ESP in Ni, Co, Fe, and Gd, with a degree of polarization in each case in good agreement with the tunneling results. Several theoretical papers[10-13] have attempted to account for the positive ESP observed in the photoemission experiments, and these papers demonstrate that the spin-polarized photoemission results are not necessarily in disagreement with the SWS theory. However, in Ni, in particular, apparently contradictory experimental results exist which make clear that unanswered questions remain. Spin-