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Discussion of anomalous nuclear-acoustic-resonance signals in aluminum*

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(Received 10 July 1975; revised manuscript received 13 July 1976)

Various explanations of the recent anomalous nuclear-acoustic-resonance (NAR) signals observed in aluminum are discussed. The possible correlation between the phase reversal of the NAR signals and the sign reversal of the Hall coefficient is discussed and shown to be unlikely.

The recent nuclear-acoustic-resonance (NAR) experiments in aluminum reported by Hsu, Leisure, and Seiber¹ (HLS), raise a number of interesting questions. The striking features of their experiments are the following: (i) Upon cooling the sample from ~60 to 30 °K the amplitudes of both the absorption and dispersion signals passed through zero and reemerged with reversed phase; (ii) the temperature at which the signals vanished remained close to 45 °K for all but one of the runs reported, although the magnetic field varied from 46 to 63 kG. The authors of the paper suggest that the vanishing of the signal may be related to the fact that the Hall coefficient of aluminum reverses sign as a function of temperature and magnetic field.²⁻⁴ The connection between the two types of experiments would be expected because the Hall coefficient and the NAR signal are both functions of the off-diagonal elements of the magnetoresistivity tensor.

Measurements of the Hall coefficient in aluminum have been reported over the temperature range from 2 K to room temperature and in magnetic fields up to 30 kG.²⁻⁵ All of the results fit well on a Kohler plot⁶ indicating that the magnetoresistance is a function of B/ρ , where B is the magnetic field and ρ is the electronic resistivity, as predicted by the solution of the Boltzmann transport equation in the relaxation-time approximation. Figure 1 shows a plot of points where the Hall coefficient vanishes as a function of magnetic field and temperature. It is clear from the figure that the zeros of the NAR signals do not fit on the

theoretical extension of the low-field data. HLS¹ suggest two possible reasons why their results do not agree with the Kohler-plot prediction. Their first suggestion is that the Fermi surface of aluminum⁷ is complex, with pieces lying in both the second and third conduction bands; thus a two-band calculation of the NAR interaction might explain their anomalous results. Their second suggestion is that previous experiments⁸ have shown that magnetic breakdown effects may become important in magnetic fields of about 60 kG, thus introducing additional scattering mechanisms, so that a single-relaxation-time solution to the Boltzmann equation might be incorrect. In the following we discuss these ideas and indicate why we believe that the field and temperature at which signals vanish are uncorrelated with those at which the Hall coefficient vanishes.

NAR in aluminum takes place via the interaction of an acoustically generated internal rf magnetic field with the nuclear dipole moments,⁹ the so-called Alpher-Rubin mechanism. Several different theories of NAR¹⁰⁻¹² have been presented covering both the high-, $\omega_c\tau \ll 1$, and low-, $\omega_c\tau \gg 1$, temperature regions. Here ω_c is the cyclotron frequency and τ is the mean electronic relaxation time. In both the high- and low-temperature theories the term in the equation of motion for the ions that contains the coupling of the ions to the electrons and nuclear spins is proportional to

$$(\vec{I} - \vec{R}_0 \cdot \vec{T}) \cdot (\vec{I} - \vec{R} \cdot \vec{T})^{-1} \cdot (\vec{R}_0 - \vec{R}).$$

In this expression \vec{R}_0 is the static resistivity ten-

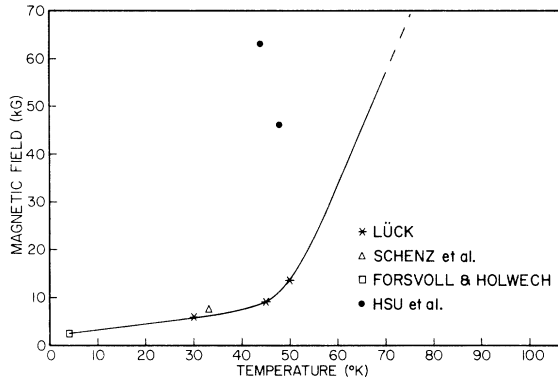


FIG. 1. Magnetic field and temperature dependence of the zeros of the Hall coefficient in aluminum. Points are taken from the data of Refs. 2-4. The curve is a theoretical extension of the low-field data, assuming Kohler's rule. Also plotted are the points at which the NAR signals of HLS pass through zero.

sor, \bar{R} is complete dynamic magnetoresistivity tensor, and $\bar{\Gamma} = -i\beta\sigma_0\bar{\mu}$, where σ_0 is the static conductivity, μ is the magnetic permeability tensor which differs from 1 only because of the slight alignment of the nuclear dipole moments, and $\beta = \omega c^2/4\pi\sigma_0 v^2$, where ω is the frequency of the ultrasonic wave, v is the ultrasonic velocity, and c is the velocity of light. The above expression can be written in terms of the square of the off-diagonal element of the magnetoresistivity tensor, i.e., ρ_{xy}^2 , which is just the square of the Hall resistivity. Since within the theories referred to above the NAR dispersion and absorption signals are proportional to the real and imaginary parts of the coupling term, they would be proportional to the square of the Hall resistivity, and hence might be expected to go to zero when the Hall coefficient changes sign, but would not be expected to show a phase reversal. A straightforward application of neither the high-temperature nor the low-temperature theory, therefore, can explain the anomalous NAR results in aluminum.

The fact that the NAR phase reversals were seen in the region of $\omega_c\tau \sim 1$ raises the question of whether either the high- or the low-temperature theory is applicable in this intermediate-temperature region. We have extended Fedders's¹¹ high-temperature calculation to cover the case of $ql < 1$, $\omega_c\tau < 1$, and find that the NAR signal predicted remains proportional to ρ_{xy}^2 . The extrapolation of Buttet's¹² low-temperature calculation to the region $ql < 1$ also indicates that nothing anomalous happens in this region. Thus other explanations must be sought.

HLS¹ raise the question as to whether a two-band

approach to the theory of NAR might explain their anomalous results. Ashcroft¹³ has proposed that the reversal of the Hall fields in aluminum may occur because of a delicate balance between contributions to the conductivity from the third-band electrons and from the second-band holes. According to this theory the electrons in the third band would dominate the conductivity at low magnetic fields. The third-band carriers, however, have a very small cyclotron mass and thus would be expected to pass into the $\omega_c\tau > 1$ region at a much smaller field than the second-band carriers. Since in the high-field regime the contribution of carriers to the conductivity is proportional to the volume enclosed by the Fermi surface in the band they occupy,¹⁴ the third-band carriers make negligible contribution in this region. These ideas have been substantiated by Douglas and Datars¹⁵ who have calculated the components of the magnetoresistivity tensor using the path-integral method. It is not difficult to incorporate this model into Fedders's calculation of the NAR signals. In his paper Fedders states¹¹ that his calculation was intended to include "anisotropy and non-free-electron-like effects in the conductivity in a self-consistent manner"; it is only necessary to use the correct magnetoresistivity tensor. One can easily show that such a straightforward inclusion of the two-band model into Fedders's calculation results once again in the diagonal coupling terms from which the NAR absorption and dispersion signals are derived, being proportional to the square of the Hall resistivity. Thus within the framework of current theories, the two-band model does not lead to a simple explanation of the NAR-signal phase reversal.

It is interesting to note, however, that any deviation from free-electron behavior introduced into Fedders's calculation causes very different effects to occur. In the following we show that if the Hall coefficient differs from the free-electron value, the ultrasonic mechanical resonances will be distorted, and for the geometry of the experiments of HLS,¹ that of shear waves propagated parallel to the magnetic field and to a [100] direction, the plane of polarization of the shear waves will rotate. We expand Eq. (13) of Fedders's calculation for the free-electron case, omitting for the moment the small coupling to the nuclear magnetization, and find

$$\omega^2\xi_x - q^2v_t^2\xi_x = \frac{i\omega n_0 z e^2}{\sigma_0 M} \left(-\frac{i\beta}{1+i\beta} \omega_c^2 \tau^2 \xi_x - \omega_c \tau \xi_y \right) + \frac{ize\omega}{Mc} B_0 \xi_y \quad (1)$$

and

$$\omega^2 \xi_y - q^2 v_t^2 \xi_y = \frac{i\omega_0 z e^2}{\sigma_0 M} \left(-\frac{i\beta}{1+i\beta} \omega_c^2 \tau^2 \xi_y + \omega_c \tau \xi_x \right) - \frac{iz e \omega}{Mc} B_0 \xi_x. \quad (2)$$

In these equations ξ is the ion displacement, n_0 is the number of conduction electrons, n_0/z is the number of ions per unit volume, and M is the ion mass. The off-diagonal elements in these expressions vanish because

$$\frac{iz e \omega}{M} \left(\frac{n_0 e \omega_c \tau}{\sigma_0} - \frac{B_0}{c} \right) = \frac{iz e \omega}{M} \left(\frac{n_0 e^2 B_0 \tau}{\sigma_0 m c} - \frac{B_0}{c} \right) = 0,$$

from which Fedders's results follow. The second term in the parentheses comes from the direct effect of the magnetic field on the moving ions, the first from the electron-ion interaction. The fact that they cancel indicates that in the free-electron case, the electrons completely shield the ions from direct interaction with the magnetic field. Within the framework of Fedders's calculation, however, this is only true in the free-electron case. This can be seen by noting that in Eqs. (1) and (2), $\omega_c \tau / \sigma_0$ has been substituted for the off-diagonal element of the magnetoresistivity tensor. In cases where the Hall coefficient differs greatly from its free-electron value, and thus Fermi-surface effects are important, this substitution is not justified. Taking this into account, Eqs. (1) and (2) can be rewritten in the following way:

$$\omega^2 \xi_x - q^2 v_t^2 \xi_x = \frac{\omega n_0 z e^2 \omega_c^2 \tau^2}{\sigma_0 M} \frac{\beta}{1+i\beta} \xi_x + \frac{iz^* e \omega}{Mc} B_0 \xi_x, \quad (3)$$

and

$$\omega^2 \xi_y - q^2 v_t^2 \xi_y = \frac{\omega n_0 z e^2 \omega_c^2 \tau^2}{\sigma_0 M} \frac{\beta}{1+i\beta} \xi_y - \frac{iz^* e \omega}{Mc} B_0 \xi_x, \quad (4)$$

where z^* measures the extent to which the conduction electrons are unable to shield the ions from direct interaction with the magnetic field. These

equations can be diagonalized by the substitution $\xi^+ = \xi_x + i\xi_y$ and $\xi^- = \xi_x - i\xi_y$, corresponding to left- and right-handed circularly polarized waves. The + and - waves would travel through the sample with different velocities.

The results of such a velocity shift would be twofold. First, the shape of the ultrasonic mechanical resonances would change. For small shifts a broadening of the resonances would occur, while for larger shifts each mechanical resonance would split into two. Second, the plane of polarization of the ultrasonic wave would rotate. The magnitude of these two effects would depend on the values of z^* and B_0 . Although rotations of the plane of polarization of ultrasonic shear waves have been predicted¹⁶ and observed in the low-temperature regime,¹⁷ $ql > 1$, neither of the above effects have been reported in the regime and $ql < 1$ for which Fedders's calculation is intended. It is possible that under the particular experimental conditions of the experiments of HLS, $\omega_c \tau \sim 1$ and at temperatures below which the Hall coefficient changes sign, the velocity shift between the left- and right-handed circularly polarized waves becomes significant. If so, it is difficult to predict how this would appear in the HLS experiments. In general, such experiments are performed by locking the spectrometer to a mechanical resonance of the sample by means of a feedback loop. The NAR signals detected in this way depend on the shape of the mechanical resonance. If the shape of the resonance is shifting with magnetic field and temperature, the exact effect on the NAR signals would depend on the details of the spectrometer. It is possible that the above effects might produce the anomalous results reported by HLS. The possibility of such effects indicates that more experiments in this temperature and field range are called for.

ACKNOWLEDGMENTS

The author is grateful for helpful discussions with Professor J. C. Swihart and Professor W. L. Schaich.

*Work partially supported by the National Science Foundation under grant No. GH-39133 and by a grant from the Research Corporation.

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