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PHYSICAL REVIEW B VOLUME 7, NUMBER 5 1 MARCH 1973

Acoustic Magnetic Resonance in Metals via the Alpher–Rubin Mechanism*

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The change in ultrasonic attenuation and velocity of metals due to spins coupled via the Alpher–Rubin mechanism is calculated. In particular the results are applied to nuclear- and electronic-spin resonance in paramagnetic metals where discrepancies with previous calculations are found.

I. THEORY

Acoustic magnetic resonance in metals via the Alpher–Rubin[1] mechanism (or magnetic-dipole coupling) has been treated by several authors[2–6] for special cases. However, some of these treatments contain errors. It is the purpose of this paper to give a general self-consistent derivation of the effect and to obtain specific results in the long-wavelength limit for paramagnetic metals. The derivation will be presented in such a way as to see the physical origin of the terms missing from other treatments and to stress the self-consistency requirements. In the rest of this section a general derivation appropriate to any metal is given where deformation-potential effects are not important. Section II contains detailed results for the Alpher–Rubin-induced acoustic-attenuation and -velocity changes in paramagnetic metals due to nuclear and electronic spins in the long-wavelength limit. For the particular case of nuclear resonance with the external magnetic field parallel to the direction of acoustic propagation, our answer is in agreement with Buttet's.[3]

Our treatment of the coupled systems of acoustic waves and conduction electrons is a generalization of the treatment of Rodriguez[7] to apply to noniso-tropic and non-free-electron metals and to include the effects of a magnetic permeability which is not unity and which can depend on frequency. Our treatment does not include effects due to the de-formation-potential and thus may not be valid for materials and/or circumstances where that potential is important.[8] On the other hand, the equation can still be used to include anisotropy and non-free-electron–like effects in the conductivity in a self-consistent manner. The self-consistency of our treatment is manifest in two ways. In the first place, the collision forces between the lattice and the electrons are treated on the same footing. Thus, whether or not the electron-lattice collision force is characterized by a single-collision time, energy and momentum are conserved. In the second place, an eigenvalue equation for the lattice displacement is obtained before any assumptions as to what is small are made. Thus expansion in terms of small quantities can easily be made, ensuring that all terms to a given order are included. Thus we consider an acoustic wave of frequency ω and wave vector $\vec{q}$ whose displacement $\xi(\vec{r}, t)$ is given by

$$\vec{\xi} = \vec{\xi}_0 e^{-i\omega t + i\vec{q}\cdot\vec{r}} . \tag{1}$$

The equation of motion of an ion of mass M in an electric field E and magnetic field B is given by Newton's second law,

$$M\frac{\partial^2\vec{\xi}}{\partial t^2} = C_l\,\vec{\nabla}(\vec{\nabla}\cdot\vec{\xi}) - C_t\,\vec{\nabla}\times(\vec{\nabla}\times\vec{\xi}) + ze\vec{E} + (ze/c)\vec{u}\times\vec{B} + \vec{F}_c . \tag{2}$$

Here C_l and C_t are the elastic constants, $-e$ is the charge on an electron, $\vec{u} = \partial\vec{\xi}/\partial t = -i\omega\vec{\xi}$ is the ionic velocity, and $\vec{F}_c$ is the force on the ions due to collisions with the electrons. It is further assumed that there are n_0 conduction electrons and n_0/z positive ions per unit volume. Thus the density of ions is $\rho = Mn_0/z$.

Under certain simplifying assumptions, Rodriguez and others relate the collision force $\vec{F}_c$ to a phenomenological collision time τ through the equation

$$\vec{F}_c = (mz/\tau)(\langle\vec{v}\rangle - \vec{u}) = -(zm/n_0 e\tau)\vec{j}_e - (mz/\tau)\vec{u} ,$$

where m is the electron mass, $\langle\vec{v}\rangle$ is the average electron velocity, and $\vec{j}_e = -n_0 e\langle\vec{v}\rangle$ is the electronic current density. This is ultimately related to the dc conductivity $\sigma_0 = n_0 e^2\tau/m$. Any simplifying assumptions or restrictions are actually unnecessary at this point. Since the ions are very massive compared to the electrons and the average electron velocity is small compared to the Fermi velocity, the collision force must be equal to $\overleftrightarrow{A}\cdot(\langle\vec{v}\rangle - \vec{u})$. The tensor constant of proportionality $\overleftrightarrow{A}$ can be obtained by noting that in the static ($\omega = 0$, $\vec{q} = 0$) zero-magnetic-field ($\vec{B} = 0$) limit there can be no force on the ions if they are at rest. Thus the collisional force must cancel the $ze\vec{E}$ part of the Lorentz force. Using the additional equation that

$$\langle\vec{v}\rangle = -\vec{j}_e/n_0 e = -\overleftrightarrow{\sigma}_0\cdot\vec{E}/n_0 e$$

in the static limit, we obtain $\overleftrightarrow{A}\cdot\overleftrightarrow{\sigma}_0 = n_0 ze^2$ or

$$\vec{F}_c = -ez\overleftrightarrow{R}_0\cdot\vec{j} , \tag{3}$$

$$\vec{j} = \vec{j}_e + n_0 e\vec{u} , \tag{4}$$

where $\vec{j}$ is the total current density and $\overleftrightarrow{R}_0 = \overleftrightarrow{\sigma}_0^{-1}$ is the static-resistivity tensor in zero external fields. This formula ensures that the calculation will be self-consistent no matter what approximations are made in calculating conductivity. It also obviates any need of worrying about anisotropy or effective masses at this point.

If the space-time–dependent parts of the magnetic fields are sufficiently small, the total current density $\vec{j}$ can be related to the electric field $\vec{E}$ through Maxwell's equations[9]:

$$\vec{j} = \frac{i\omega}{4\pi}\vec{E} + \frac{ic^2}{4\pi\omega}\vec{q}\times[\overleftrightarrow{\mu}^{-1}\cdot(\vec{q}\times\vec{E})] = \overleftrightarrow{\Gamma}\cdot\vec{E} , \tag{5}$$

where $\overleftrightarrow{\mu}$ is the magnetic permeability tensor which may depend on $\vec{q}$, ω, and a static magnetic field $\vec{B}_0$. The one further equation necessary in order to obtain a complete solution is the constitutive equation relating the electronic current density to the electric field $\vec{E}$ and ionic velocity $\vec{u}$. From standard Boltzmann-equation treatments one obtains[7,10]

$$\vec{j}_e = \overleftrightarrow{\sigma}\cdot\vec{E}' = \overleftrightarrow{R}^{-1}\vec{E}' ,$$

where (6)

$$\vec{E}' = \vec{E} - m\vec{u}/e\tau = \vec{E} - n_0 e\vec{u}/\sigma_0 .$$

The diffusion term in the electronic current proportional to the gradient of the density can be related back to the electronic current density through the continuity equation. In our notation $\overleftrightarrow{\sigma}$ includes this term.

In fact, the electronic current density can be shown to be proportional to E' independent of any detailed transport equation. Since the part of $\vec{F}_c$ due to $\vec{u}$ is $-n_0 ze^2\overleftrightarrow{R}_0\cdot\vec{u}$, an equal and opposite force must act on the electrons. Since there are z times as many electrons as ions, the force $-e\vec{E}$ on an electron is replaced by $-e\vec{E} + n_0 e^2\overleftrightarrow{R}_0\cdot\vec{u}$, or $\vec{E}$ is replaced by $\vec{E}'$, where

$$\vec{E}' = \vec{E} - n_0 e\overleftrightarrow{R}_0\cdot\vec{u} . \tag{7}$$

A complete solution is now obtained if $\overleftrightarrow{\mu}$, $\overleftrightarrow{R}$, and $\overleftrightarrow{R}_0$ are known. Since the time-varying part of $\vec{B}$ is proportional to $\vec{\xi}$, $\vec{B}$ in Eq. (2) can be replaced by $\vec{B}_0$, the static external magnetic field, if only small amplitudes are considered. Equations (2)–(5) are combined to give

$$M\omega^2\vec{\xi} - (C_l - C_t)\vec{q}\vec{q}\cdot\vec{\xi} - C_t q^2\vec{\xi} = -ze(\overleftrightarrow{1} - \overleftrightarrow{R}_0\cdot\overleftrightarrow{\Gamma})\cdot\vec{E} + (zei\omega/c)\vec{\xi}\times\vec{B}_0 . \quad (8)$$

From Eqs. (4)–(7) the expression

$$\vec{E} = -i\omega n_0 e(\overleftrightarrow{1} - \overleftrightarrow{R}\cdot\overleftrightarrow{\Gamma})^{-1}\cdot(\overleftrightarrow{R}_0 - \overleftrightarrow{R})\cdot\vec{\xi} \quad (9)$$

for the electric field is obtained. By combining Eqs. (8) and (9) we obtain

$$\{\omega^2 - [(C_l - C_t)\vec{q}\vec{q} + C_t q^2]/M\}\vec{\xi} = (i\omega n_0 ze^2/M)(\overleftrightarrow{1} - \overleftrightarrow{R}_0\overleftrightarrow{\Gamma}), \quad (10)$$
$$(\overleftrightarrow{1} - \overleftrightarrow{R}\cdot\overleftrightarrow{\Gamma})^{-1}\cdot(\overleftrightarrow{R}_0 - \overleftrightarrow{R})\cdot\vec{\xi} + (zei\omega/Mc)\vec{\xi}\times\vec{B}_0 .$$

The acoustic velocity and attenuation are obtained from the real and imaginary parts of the solution of this eigenvalue equation for ω.

II. DISCUSSION

In this section we shall specialize to free-electron cubic paramagnetic metals in the long-wavelength limit. We shall use the usual parameters

$$\omega_c = eB_0/mc ,$$
$$\beta = \omega c^2/4\pi\sigma_0 v^2 , \quad (11)$$
$$v = \omega/q ,$$

where v is the velocity of sound. Since we are not interested here in geometric resonances, we assume that $\omega_c\tau$, $\omega\tau$, $qv_f\tau$, as well as v/c are much less than 1, where τ is an electron-collision time. Equation (10) can then be expanded in powers of $\omega_c\tau$ and $\delta\mu$, where $\mu = 1 + \delta\mu$. We shall also use Rodriguez's coordinate system where $\vec{B}_0 = B_0(0, \sin\theta, \cos\theta)$ and $q = q\hat{z}$. Then $\Gamma = \Gamma_0 + \delta\Gamma$ and $R = R_0 + \delta R$, where

$$(R_0)_{ij} = \sigma_0^{-1}\delta_{ij} ,$$
$$\sigma_0 = ne^2\tau/m ,$$
$$\delta R_{xy} = -\delta R_{yx} = \omega_c\tau\cos\theta/\sigma_0 ,$$
$$\delta R_{zy} = -\delta R_{yz} = \omega_c\tau\sin\theta/\sigma_0 , \quad (12)$$
$$\delta R_{xz} = \delta R_{zx} = \delta R_{ii} = 0 ,$$
$$(\Gamma_0)_{ij} = -i\beta\sigma_0\delta_{ij}(1 - \delta_{iz}) ,$$
$$(\delta\Gamma)_{ij} = -i\beta\sigma_0(\delta\mu)_{ji}(1 - 2\delta_{ij})(1 - \delta_{iz})(1 - \delta_{jz}) .$$

Equation (10) is expanded to first order in $\delta\mu$ and second order in $\omega_c\tau$, yielding

$$\{\omega^2\vec{\xi} - [v_l\vec{q}\vec{q}\cdot\vec{\xi} - v_t\vec{q}\times(\vec{q}\times\vec{\xi})]\} = (zn_0e^2\omega i/\sigma_0 M)\overleftrightarrow{\Phi}\cdot\vec{\xi} + (zei\omega/Mc)(\vec{\xi}\times\vec{B}_0) , \quad (13)$$

where v_l and v_t are the longitudinal and transverse sound velocities and

$$\overleftrightarrow{\Phi} = [\overleftrightarrow{1} - \delta\overleftrightarrow{R}\,\overleftrightarrow{\Gamma}_0\,\overleftrightarrow{A}^{-1} + \delta\overleftrightarrow{R}\,\overleftrightarrow{\Gamma}_0\,\overleftrightarrow{A}^{-1}\sigma_0^{-1}\delta\overleftrightarrow{\Gamma}\,\overleftrightarrow{A}^{-1}](-\sigma_0\delta\overleftrightarrow{R}) \quad (14)$$

and

$$(A)_{ij} = \delta_{ij}[1 + i\beta(1 - \delta_{ij})] . \quad (15)$$

The first two components of the displacement vector $\vec{\xi}$ correspond to the transverse modes and the last component to the longitudinal mode. The first term in the square brackets of Eq. (14), $\overleftrightarrow{\Phi}_1$, exactly cancels the $\vec{\xi}\times\vec{B}_0$ in term in Eq. (13). The second term in the square brackets is the usual Alpher—Rubin term:

$$(\Phi_2)_{ij} = \frac{-i\beta(\omega_c\tau)^2}{1 + i\beta}\delta_{ij}[\cos^2\theta + (\sin^2\theta - \cos^2\theta)(1 - \delta_{ij})] . \quad (16)$$

There are off-diagonal terms of order $(\omega_c\tau)^2$ in Φ_2 connecting the y and z components of $\vec{\xi}$ which are suppressed because they will contribute only to order $(\omega_c\tau)^4$ in the modes.

The last two terms in the square brackets of Eq. (14) are the resonant parts (the parts that depend on the magnetic susceptiblity). Since the longitudinal and transverse modes have distinct velocities, only Φ_{zz} and $\Phi_{\bar{i}\,\bar{j}}$, where $\bar{i}$ and $\bar{j}$ run over x and y, are relevant. In our approximation these terms are

$$(\Phi_3 + \Phi_4)_{zz} = \left(\frac{i\beta}{1 + i\beta} + \frac{\beta^2}{(1 + i\beta)^2}\right)(\omega_c\tau)^2(\delta\mu_{yy})\sin^2\theta , \quad (17)$$
$$(\Phi_3 + \Phi_4)_{\bar{i}\,\bar{j}} = \left(\frac{i\beta}{1 + i\beta} + \frac{\beta^2}{(1 + i\beta)^2}\right)(\omega_c\tau)^2\cos^2\theta\,\delta\mu_{\bar{j}\,\bar{i}} .$$

For a cubic system in our coordinate system the relevant components of $\delta\overleftrightarrow{\mu}$ are

$$\delta\mu_{xx} = 4\pi(\chi_+ + \chi_-) ,$$
$$\delta\mu_{yy} = 4\pi[(\chi_+ + \chi_-)\cos^2\theta + \chi_0\sin^2\theta] , \quad (18)$$
$$\delta\mu_{xy} = -\delta\mu_{yx} = 4\pi i\cos\theta(\chi_+ - \chi_-) .$$

In these equations, χ_m with $m = +$, $-$, or 0 are the spherical components of the susceptibility. In the low-frequency long-wavelength limit, the nuclear and electronic contribution can be written

$$\chi_m^{(\alpha)}(\vec{q}\,\omega) = \chi^{(\alpha)}(\vec{q}, 0)(2 - \delta_{m,0})(m\omega_0 - i\Gamma)/(m\omega_0 - i\Gamma - \omega) , \quad (19)$$

where $m = +1$, -1, or 0, Γ is a decay rate which

may be $\vec{q}$ dependent if spin diffusion is included, $\chi^{(\alpha)}(\vec{q}, 0)$ is the static susceptibility, and α refers to the nuclear or electronic spins.

Keeping only the diagonal terms in $\overleftrightarrow{\Phi}$ and resonant terms in the susceptibility, we obtain the resonant attenuation and velocity shift,

$$\Delta\alpha_i = [\omega B_0^2 \beta S_i(\theta)/2\rho v_i^3(1+\beta^2)^2][(1-\beta^2)\chi_+'' - 2\beta\chi_+'] ,$$

$$(\Delta v_i/v_i) = [B_0^2 \beta S_i(\theta)/2\rho v_i^2(1+\beta^2)^2] \times[(\beta^2-1)\chi_+' - 2\beta\chi_+''] , \quad (20)$$

where

$$\chi = \chi' + i\chi'' , \qquad S_1(\theta) = \cos^2\theta ,$$
$$S_2(\theta) = \cos^4\theta , \qquad S_3(\theta) = \cos^2\theta \sin^2\theta . \quad (21)$$

Two comments should be made about these equations. First, the neglect of off-diagonal terms is not necessarily permitted. This point shall be taken up later in this paper. Second, the resonant-attenuation coefficient is not positive definite! While this may seem strange it does not violate any physical laws because $\Delta\alpha$ is the resonant part of the absorption, and "background" of the usual Alpher–Rubin term is much larger (under the assumption that $\delta\mu$ is small) and positive.

For nuclear acoustic resonance, Eqs. (20) and (21) are not in agreement with Ref. 2, but are in agreement with Ref. 3 where only the case of $\theta = 0$ is considered. We have calculated the decay rate of an acoustic wave which gives the total absorption of energy from the wave. Several authors[2,11] have instead calculated the amount of power absorbed by a nuclear or electronic spin system in order to find the resonance power lost by the acoustic wave using the formula

$$\alpha = (2/\rho\omega v)|H_1/\xi|^2 \mathrm{Im}\chi , \quad (22)$$

where H_1 is the rf magnetic field. An inspection of Eqs. (8) and (9) shows that this procedure is in general incorrect,[11] although in some limiting cases it leads to correct answers.[12] The reason is that the orbital electrons, electronic and nuclear spins, and the acoustic waves are, in general, not separate entities but are rather strongly coupled. In a sense, the power absorbed by the orbital electrons is affected by the electronic or nuclear susceptibility because the electric field felt by the electrons depends on μ through Γ. If the power change in all subsystems is calculated the correct acoustic absorption is obtained, but we feel that our method of directly calculating the acoustic attenuation is superior.

Equation (22) is valid only if all of the lost acoustic energy is transferred to the spins and only if the spins themselves do not contribute to H_1. In general these conditions are not met. The conduction electrons themselves absorb power by the usual Ohmic losses because of the acoustically induced electromagnetic field and H_1 depends on the spins because the induced electric field depends on the total susceptibility through Γ or Maxwell's equations. If the dimensionless parameter β is small enough, Eq. (22) will yield the correct results. The reason is that in this limit the conductivity is large enough so that the Ohmic losses are negligibly small and that the dependence of H_1 on μ is negligible compared with the large H_1 generated by the orbital motion because of the large conductivity.

Finally, let us consider the effects of the nondiagonal terms in Φ on the two transverse modes in the case of nuclear resonance. As can be seen from Eqs. (17) and (18), the off-diagonal components of the resonant part $\Phi_3 + \Phi_4$ are just as large as the diagonal ones. Thus Eq. (20) is correct only if there are other terms splitting the degeneracy of the two transverse modes which are larger than the terms which we have considered. Besides the possibility of static strains or other imperfections in the crystal, the electronic susceptibility itself will effectively mask off-diagonal terms if

$$\chi^{(e)}(\vec{q}, 0) \gg |\chi_+^{(n)}(\vec{q}, \omega)| .$$

If the degeneracy is not sufficiently lifted by other terms, the 2×2 matrix in Eq. (17) must be diagonalized and the two modes have velocities and attenuation given by Eqs. (20) and (21), with

$$S_1(\theta) = (\cos^2\theta + 1)\cos^2\theta, \quad S_2(\theta) = 0 \quad (23)$$

and with complex polarization vectors

$$\hat{e}_1(\theta) = (\hat{e}_y - i\cos\theta\,\hat{e}_x)/(1+\cos^2\theta)^{1/2} ,$$

$$\hat{e}_2(\theta) = (\hat{e}_x - i\cos\theta\,\hat{e}_y)/(1+\cos^2\theta)^{1/2} .$$

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PHYSICAL REVIEW B VOLUME 7, NUMBER 5 1 MARCH 1973

Scintillation Response of NaI(Tl) and KI(Tl) to Channeled Ions*

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The scintillation pulse-height response of NaI(Tl) and KI(Tl) to ^{4}He and ^{16}O ions in the 2–60-MeV range has been studied with the ion beam aligned along low-index planes and axes and also aligned along a random direction. The scintillation efficiency increases by as much as 50% when the ion beam is channeled along a major symmetry direction. The effect of channeling has been observed by recording the pulse-height spectra for monoenergetic ions oriented along {100}, {110}, and {111} planes, and along ⟨100⟩, ⟨110⟩, and ⟨111⟩ axes. The increase in pulse-height response is in semiquantitative agreement with recent model calculations. Observation of this effect permits study of channeling phenomena in thick crystals that are scintillators. In particular, this paper reports a measurement of the critical angle for channeling of 15-MeV ^{16}O along a {100} plane.

I. INTRODUCTION

It is well known that energetic positive ions experience a reduced stopping power dE/dx when incident along low index axes or planes of a crystalline solid.[1] This effect arises from a correlated scattering of the incident ion by the lattice atoms for incidence along major symmetry directions. This correlated scattering and the associated steering of the incident ion along the open regions of the crystal is known as channeling and has been studied in detail in many solids, especially semiconductors and metals.

Luntz and Bartram[2] (denoted LB hereafter) suggested that channeling should have a pronounced effect on the scintillation response of NaI(Tl) and CsI(Tl) to positive ions. Their calculations predicted that the scintillation pulse height from an energetic positive ion could be enhanced by as much as four times its normal value if the ion experiences a channeled rather than a random trajectory, where "random" refers to an ion incident upon the crystal in a nonaligned direction. The cause of this effect can be seen by an examination of the scintillation efficiency dL/dE as a function of stopping power dE/dx for various positive ions. Scintillation efficiency dL/dE is defined as the slope of a pulse-height–versus–energy curve, where L represents the scintillation pulse height arising from a particle of incident energy E that is completely stopped in the crystal. A survey[3] of dL/dE vs dE/dx for various positive ions in NaI(Tl) and CsI(Tl) shows that the scintillation efficiency decreases with increasing stopping power. The effect of aligning the incident beam along a major symmetry direction is to reduce dE/dx, thus to increase dL/dE along the ion's path and therefore produce a greater pulse height L.

This paper reports the results of a series of experiments on the scintillation response of NaI(Tl) and KI(Tl) to ^{4}He and ^{16}O ions in the range 2–60 MeV for random incidence and for incidence along the {100}, {110}, and {111} planes and along the ⟨100⟩, ⟨110⟩, and ⟨111⟩ axes. The results are in qualitative agreement with the calculations of LB as the channeled ions yield a distinctly greater pulse height. The magnitude of the increase in pulse height is, however, substantially less than that predicted by LB.

Most of the work reported here was on NaI(Tl). Several experiments were performed with KI(Tl), confirming that the effects were substantially the same.

II. EXPERIMENT

Experiments were performed with NaI(Tl) scintillation crystals (nominal 0.1-mole% thallium) obtained from Harshaw Chemical Co. as cylindrical