MAGNETIC DEPENDENCE OF LONG-WAVELENGTH PHONONS IN METALS

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The purpose of this note is to indicate the possibility and potential usefulness of altering the energies of long-wavelength phonons in a metal by the application of a large, but feasible, magnetic field. Specifically, the present calculation shows that the speed of longitudinal sound ought to increase by observable amounts when a magnetic field in the range of ten kilogauss or more is applied to the metal. Furthermore this increase in sound velocity may be related to properties of the Fermi surface. For simplicity we confine ourselves here to spherical energy surfaces, but it is conjectured that the anisotropy of the effect in real metals will provide a gross image of the anisotropy of the Fermi surface.

We begin by adopting a Hamiltonian for the metal in a magnetic field which implies the assumptions that the one-electron Bloch states can be adequately represented by plane waves, and that the electrons interact only with longitudinal phonons[1]:

$$H=\sum_{kk'}c_k^+c_{k'}\mathcal{E}_{kk'}+\sum_{BZ}p_k^+p_k+\Omega_k^2q_k^+q_k+\tfrac{1}{2}\sum_k M_k^2\rho_{-k}\rho_k+\sum_k q_kv_k^i\rho_{-k}. \quad (1)$$

In Eq. (1) $\mathcal{E}_{kk'}$ is the matrix element between plane wave states of the one-electron Hamiltonian in a magnetic field. The notation is otherwise identical to that of reference 1. We have neglected the interaction of the ions with the magnetic field as unimportant. From Eq. (1) we obtain the Heisenberg equations of motion for the phonon coordinate q_k, and its derivative $\dot{q}_k$, and find that neglecting Umklapp terms we obtain the same equation of motion that one has in the absence of a field:

$$\ddot{q}_k+\Omega_k^2q_k+v_k^{i*}\rho_k=0, \quad (2)$$

where we have used the relation $v_{-k}^i=v_k^{i*}$ obeyed by the electric-ion interaction matrix element.[1]

As a result of the positive ion motion described by the phonon coordinate q_k, the electron system experiences a longitudinal electric field $\vec{\mathcal{E}}_k$ given by

$$\vec{\mathcal{E}}_k=(1/e)i\vec{k}q_kv_k^i. \quad (3)$$

In response to $\vec{\mathcal{E}}_k$ the electron system moves so as to screen out the positive charge fluctuations giving rise to $\vec{\mathcal{E}}_k$. Such electronic screening currents produce their own electric field, $\vec{\mathcal{E}}_\rho$, which will not in general be entirely longitudinal, due to the presence of the external magnetic field. The total electric field in the metal, $\vec{\mathcal{E}}_T$, is then given by the sum

$$\vec{\mathcal{E}}_T=\vec{\mathcal{E}}_k+\vec{\mathcal{E}}_\rho, \quad (4)$$

and furthermore is linearly related to $\vec{\mathcal{E}}_k$ through the expression

$$\vec{\mathcal{E}}_T=\epsilon^{-1}\cdot\vec{\mathcal{E}}_k, \quad (5)$$

where ϵ^{-1} is the frequency-, wave vector-, and magnetic field-dependent inverse dielectric tensor of the electron gas discussed below.

From Eqs. (4) and (5) we then obtain

$$\vec{\mathcal{E}}_\rho=(\epsilon^{-1}-I)\cdot\vec{\mathcal{E}}_k. \quad (6)$$

Assuming that the electron density fluctuation ρ_k arises solely as a coherent response to the ion motion, we may relate the longitudinal part of $\vec{\mathcal{E}}_\rho$ to ρ_k by means of Poisson's equation. Using Eq. (6) we then obtain

$$i\vec{k}\cdot(\epsilon^{-1}-I)\cdot\vec{\mathcal{E}}_k=-4\pi e\rho_k. \quad (7)$$

Equations (3) and (7) lead to the result

$$\rho_k=(k^2/4\pi e^2)q_kv_k^i\hat{m}_k\cdot(\epsilon^{-1}-I)\cdot\hat{m}_k, \quad (8)$$

where $\hat{m}_k$ is the unit polarization vector of the longitudinal phonon.

Substituting Eq. (8) into Eq. (2) we obtain an equation of motion describing the renormalized phonons,

$$\ddot{q}_k+\omega_k^2q_k=0, \quad (9)$$

where

$$\omega_k^2=\Omega_k^2+(k^2/4\pi e^2)|v_k^i|^2\hat{m}_k\cdot(\epsilon^{-1}-I)\cdot\hat{m}_k \quad (10)$$

represents the actual frequencies of longitudinal phonons in the metal.

We obtain the inverse dielectric tensor, ϵ^{-1}, by first noting that for given frequency and wave vector the linear relation between $\vec{\mathcal{E}}_\rho$ and its source, the electronic screening current $\vec{j}_e$, is conveniently given by

$$\mathcal{E}_\rho = F\cdot\vec{j}_e, \tag{11}$$

where the tensor F which represents the content of Maxwell's equations is given by

$$F = \begin{pmatrix} 4\pi/i\omega & 0 & 0 \\ 0 & 4\pi i\omega/(k^2c^2-\omega^2) & 0 \\ 0 & 0 & 4\pi i\omega/(k^2c^2-\omega^2) \end{pmatrix} \tag{12}$$

in a coordinate system whose 1 axis coincides with the direction of propagation $\hat{m}_k$. The electronic current $\vec{j}_e$, however, responds to the total electric field in the metal, and it may be linearly related to $\vec{\mathcal{E}}_T$ through the frequency- and wave vector-dependent conductivity tensor σ which characterizes the electrons in the presence of an applied magnetic field,

$$\vec{j}_e = \sigma\cdot\vec{\mathcal{E}}_T. \tag{13}$$

From Eqs. (4), (5), (11), and (13) we obtain the result

$$\epsilon^{-1} = (I - F\cdot\sigma)^{-1}. \tag{14}$$

In the long-wavelength limit and for purely Coulomb forces between electrons and ions, one has[1]

$$v_k^{\,i} = (N/M)^{1/2}(4\pi NZe^2/k)i, \tag{15}$$

where N is the density of ions of mass M and valence Z. Further, in this limit $\Omega_k^{\,2}$ is just the square of the ion plasma frequency,[1]

$$\Omega_k^{\,2} = \Omega_p^{\,2} = (4\pi NZ^2e^2/M). \tag{16}$$

By substituting Eqs. (15) and (16) into Eq. (10), we obtain the expression

$$\omega_k^{\,2} = \Omega_p^{\,2}\hat{m}_k\cdot\epsilon^{-1}\cdot\hat{m}_k = \Omega_p^{\,2}(\epsilon^{-1})_{11}, \tag{17}$$

where $\hat{m}_k$ defines the 1 axis of our coordinate system. Equation (17) is an implicit relation for the phonon frequencies because of the dependence of the inverse dielectric tensor on frequency ω_k as well as on wave vector and magnetic field.

Using methods of semiclassical transport theory employing the Boltzmann equation in a magnetic field, we have obtained σ for a completely collisionless electron gas in the limit of long wavelengths and large transverse magnetic fields,[2] such that $\omega_c > kV_F$, where $\omega_c = eH/mc$ is the cyclotron frequency, and V_F the Fermi velocity of the electrons. In obtaining the components of σ in closed form, we have formally taken the limit $\tau\to\infty$ of appropriate expressions containing finite τ. It may be noted that in this limit σ contains no diffusive contribution associated with finite relaxation times. The defining relation Eq. (14) together with Eq. (12) and the components of σ in the domain $\omega_c > kV_F$ enable us to calculate ϵ^{-1} in the corresponding limit. With the transverse magnetic-field direction taken as the 3 axis of a right-handed coordinate system, we have obtained the result

$$(\epsilon^{-1})_{11} = \frac{[\omega_c^{\,2}(k^2c^2-\omega_k^{\,2}) + \omega_p^{\,2}(\tfrac{3}{5}k^2V_F^{\,2}-\omega_k^{\,2})]}{\{(1+\omega_p^{\,2}/\omega_c^{\,2})[\omega_c^{\,2}(k^2c^2-\omega_k^{\,2})+\omega_p^{\,2}(\tfrac{3}{5}k^2V_F^{\,2}-\omega_k^{\,2})]+\omega_p^{\,4}\}}, \tag{18}$$

where $\omega_p = (4\pi NZe^2/m)^{1/2}$ is the electron-plasma frequency.

For phonons $\omega_k < kc$, and for any attainable magnetic field $\omega_c \ll \omega_p$. Employing these inequalities as a basis for further approximation, we combine Eqs. (17) and (18) to obtain a quadratic equation for $\omega_k^{\,2}$:

$$\omega_k^{\,4} + B\omega_k^{\,2} + C = 0, \tag{19}$$

where

$$B = -\{\omega_c^{\,2} + [1+\tfrac{5}{3}(\omega_c/\omega_p)^2(c/V_F)^2]\tfrac{3}{5}V_F^{\,2}k^2\},$$

$$C = [k^2V_F^{\,2}\Omega_p^{\,2}/\omega_p^{\,2}]\omega_c^{\,2}[(\omega_c/\omega_p)^2(c/V_F)^2+\tfrac{3}{5}], \tag{20}$$

and we have made use of the inequality $\omega_p/\Omega_p \gg 1$ in arriving at Eq. (20).

From Eq. (19) we obtain the result,

$$\omega_k^{\,2} = -\tfrac{1}{2}B \pm \tfrac{1}{2}B(1-4C/B^2)^{1/2}. \tag{21}$$

For the values of wavelength and magnetic field with which we are concerned, it is apparent that $4C/B^2 \ll 1$, and the radical may be expanded. Observing that $B<0$, we choose the positive sign in Eq. (21) in order to obtain an expression for $\omega_k^{\,2}$ which goes to zero as the wavelength becomes large, in accord with the properties of acoustic branch phonons. One then obtains

$$\omega_k^2 = -(C/B)(1 + C/B^2 + \cdots), \tag{22}$$

up to terms of sufficient order to include any significant dispersive effects. We substitute Eqs. (20) into Eq. (22) and develop the result in powers of k^2, retaining only the first two terms so that our result is correct up to terms of order k^4. Making additional use of the inequality $\omega_p/\Omega_p \gg 1$ in reducing the coefficient of the dispersive term, we obtain the following explicit expression for the frequencies of longitudinal phonons in the limit of long wavelengths and high transverse fields, $\omega_c > kV_F$:

$$\omega_k^2 = \tfrac{2}{5}(V_F\Omega_p/\omega_p)^2[1+\tfrac{5}{2}(\omega_c/\omega_p)^2(C/V_F)^2] \times k^2\{1-\tfrac{2}{5}(kV_F/\omega_c)^2[1+\tfrac{5}{2}(\omega_c/\omega_p)^2(C/V_F)^2]\}. \tag{23}$$

In the limit of long wavelengths the dispersive term may be neglected and the phonon frequency becomes proportional to wave number,

$$\omega_k = C_s k, \tag{24}$$

where from Eq. (23) the phase velocity of longitudinal sound is given by

$$C_s^2 = \tfrac{2}{5}(V_F\Omega_p/\omega_p)^2[1+\tfrac{5}{2}(\omega_c/\omega_p)^2(C/V_F)^2]. \tag{25}$$

For indefinitely weak magnetic fields $(\omega_c/\omega_p) \times (C/V_F) \ll 1$, and one has

$$C_s^2 = \tfrac{2}{5}(V_F\Omega_p/\omega_p)^2, \tag{26}$$

which differs by a factor of $\frac{5}{6}$ from a result obtained elsewhere.[3] On the other hand it is well within the range of current experimental technique to generate magnetic fields of sufficiently high intensity for $(\omega_c/\omega_p)^2(C/V_F)^2$ to possess a value of 10^{-3}. Changes in the speed of sound as small as one part in 10^7 can be and have been detected recently[4] in studies of the elastic moduli at the superconducting transition. Furthermore, magnetically induced changes in the speed of sound of magnitude $\Delta C_s/C_s \simeq (\omega_c/\omega_p)^2(C/V_F)^2$ have indeed already been observed at room temperature in polycrystalline and single-crystal samples.[5] It would be of considerable interest to study the dependence of sound velocity on magnetic field in single crystals as a function of orientation as well as field strength.

We have calculated the effect of a magnetic field on a phonon propagating perpendicular to the field. In general the change in phonon frequency and sound velocity will depend on the angle between $\vec{H}$ and $\vec{k}$. Thus measurements of the velocity of longitudinal ultrasound in a magnetic field of ten to one hundred kilogauss may be expected to exhibit an anisotropic angular dependence in addition to that ordinarily present in the crystal. Furthermore, we note that in the present free-electron model, the electron mass and Fermi velocity appear explicitly in the coefficients containing the magnetic field dependence. The significant conclusion is that the magnetic-field-dependent part of C_s^2 depends only on properties and parameters of the electron distribution evaluated at the Fermi surface. Indeed, the expression Eq. (25) for C_s^2 can be formally cast into a number of alternatives which explicitly contain parameters commonly quoted in describing the Fermi surface.

We now conjecture that in a real metal with an anisotropic Fermi surface, the same quadratic dependence on H will occur but with a coefficient which depends on the magnetic field orientation with respect to the Fermi surface. We therefore anticipate that the angular dependence of the increase in sound velocity with H^2 will reflect the shape of the Fermi surface, at least in its general structure. Thus a polar plot of $\Delta C_s/H^2C_s$ vs angle measured in the transverse plane ought to have the same symmetry pattern as the Fermi surface. In particular, the occurrence of a large number of open orbits for any given direction of magnetic field may be expected to change the properties of phonons propagating in various directions, according to the corresponding change in the ability of the electrons to screen the fluctuations of ionic charge density which accompany the phonons. The theoretical treatment of these magnetic effects in crystals with such complexities in their band structure will be correspondingly more involved.

It should be mentioned here that because the present calculation rests on the use of the Boltzmann equation in a magnetic field, it completely misses any quantum effects of an oscillatory nature which well may be present in the field range under consideration. We anticipate that the present result provides an average value about which such oscillations may take place. The source of such de Haas–van Alphen type oscillations is to be found in the magnetic-field dependence of ϵ^{-1} when $KT < h\omega_c \ll E_F$, where E_F is the Fermi energy, K is Boltzmann's constant, and T is the absolute temperature. Since oscillations in the conductivity have indeed been observed[6-8] at low temperature and high fields, such oscillations in C_s may be observable.

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[3]D. Bohm and T. Staver, Phys. Rev. 84, 836 (1952). Since the Bohm-Staver result is correct for a collisionless electron gas in zero field, the numerically different result obtained here must be a consequence of the limitations imposed on our model by the order in which we let $\tau \rightarrow \infty$ and $\omega_c > kV_F$. The present numerical factor represents a spurious and unobserved large change in sound velocity. A study of the limiting processes involved is currently under investigation and will be reported on later.

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[5]A. A. Galkin and A. P. Koroliuk, J. Exptl. Theoret. Phys. (U.S.S.R.) 34, 1025 (1958) [translation: Soviet Phys.–JETP 34, 708 (1958)]; G. A. Alers (private communication). Since completing this manuscript the author has been informed by Dr. Alers that he has measured the variation with transverse magnetic field of the velocity of ultrasound in single-crystal aluminum at room temperature with sound propagating along the (110) direction. The experimental result indeed indicates a quadratic dependence on field of the change in sound velocity with $\Delta C_s/C_s = 2.9\times10^{-14}\,H^2$ cgs. This is to be compared with the theoretical estimate $\Delta C_s/C_s = 3.6\times10^{-14}\,H^2$ cgs predicted when one employs the parameter values $m^*/m = 1.6$, with three electrons per atom, and a Fermi momentum of $P_F = 1.5\times10^{-19}$ g cm/sec taken from other experiments, and substitutes them into the relation $\Delta C_s/C_s = \frac{5}{4}(\omega_c/\omega_p) \times (C/V_F)^2$ obtained from Eq. (25). It is to be noted that our result is in semiquantitative agreement with experiment though our model contains the limitations of the collisionless electron gas, and the above experiments were performed under conditions for which $\omega\tau < 1$. The author is indebted to Dr. Alers for communicating these results to him and for permission to quote them prior to publication.

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OBSERVATION OF SURFACE AND IMPURITY STATES IN SILICON BY OXIDE LAYER TUNNELING*

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Conductance and differential capacitances have been measured as a function of frequency and of bias on a number of silicon-silicon oxide-aluminum sandwiches. The results indicate that information about the density and energy of states near the silicon-silicon oxide interface may be deduced from these measurements. Conductance is due to electron tunneling transitions between the aluminum and silicon. This is believed to be the first reported evidence of tunneling to a semiconductor through an oxide film.

Samples are prepared by thermally oxidizing the freshly cleaved (111) surface of a single-crystal silicon specimen. Aluminum is evaporated onto the resulting oxide film. The oxide thickness is judged to be 40 to 60 Å based on capacitance measurements. Because of charge trapped in "outer states" in the oxide,[1] the bands of the p-type silicon used in the present experiments are bent downward at the surface forming a depletion layer. This intrinsic region is essential to the success of the experiment, since otherwise the tunnel current would be nearly Ohmic. The extent of band bending is evident from the position of the band edges relative to zero bias in both figures.

The problem with sample preparation is growing an oxide layer thin enough to permit tunnel current to dominate the conductance and thick enough such that nearly all of the applied bias appears across the oxide rather than the depletion region. This is particularly critical with higher resistivity silicon at low temperatures. When the above conditions are met, the existence of other transport processes is easily ruled out. The films are too thin for space-charge limited current. Virtual temperature independence of the qualitative features eliminates Schottky emission. The fact that an estimated 20% increase in oxide thickness reduces the conductance three orders of magnitude without seriously altering the detailed features of the curves removes the possibility of Ohmic conduction. In addition, the results are readily interpreted in terms of es-