GIANT QUANTUM OSCILLATIONS OF THE ATTENUATION OF TRANSVERSE ACOUSTIC WAVES IN A LONGITUDINAL MAGNETIC FIELD IN METALS

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(Received 21 November 1963)

A number of experiments,[1] in which some geometrical property of the Fermi surface is directly determined, have proven extremely important in the study of the electronic structure of metals and semimetals. The de Haas–van Alphen effect,[2] which allows one to determine the extremal cross-sectional areas of the Fermi surface normal to the direction of an applied magnetic field, is a classic example. The purpose of this note is to point out the existence and importance of a new effect, giant quantum oscillations in the attenuation of transverse acoustic waves propagating parallel to a dc magnetic field. This effect differs from the giant oscillations in acoustic attenuation discussed by Gurevich, Skobor, and Firsov[3] and by Quinn and Rodriguez,[4] in that here we consider electronic transitions in which Δn, the change in the Landau level quantum number, is nonzero. This fact is quite significant; it results in the two oscillatory effects having entirely different periods. The importance of the effect discussed in this paper is that the period of the oscillations can be used to measure the cross-sectional area of the Fermi surface not merely at extrema, but at any plane in k space normal to the magnetic field.

In this work we assume that a metal behaves as a free-electron gas embedded in an isotropic lattice of positive ions, the latter being capable of sustaining both longitudinal and shear waves. This model has been used previously to treat the magnetic field dependence of the velocity of sound,[5] and the helicon-phonon interaction in metals.[6] In references 5 and 6 the following equation is derived:

$$\omega^2 = s^2q^2 \mp \Omega_0\omega + \frac{zmi\omega}{M\tau}\,\frac{(1-i\beta)(\sigma_0 R_\pm - 1)}{1-i\beta\sigma_0 R_\pm}. \qquad (1)$$

The solutions ω of Eq. (1) are the frequencies of the circularly polarized transverse modes of vibration of the system. As discussed in reference 6, there are two acoustic modes and one electromagnetic (helicon) mode for each circular polarization. In Eq. (1), M and Ω_0 are the mass and cyclotron frequency, respectively, of the ions; $\beta = (c^2q^2/\omega_p^2\omega\tau)(1-\omega^2/c^2q^2)$, where ω_p is the plasma frequency and τ the relaxation time of the electrons. σ_0 is the dc conductivity, s the velocity of transverse acoustic waves, and

$$R_\pm = (\sigma_{xx} \mp i\sigma_{xy})^{-1}, \qquad (2)$$

where σ is the magnetoconductivity tensor. The quantum mechanical expression for $\sigma_{xx} \mp i\sigma_{xy}$ is derived in reference 4, and it can be written

$$\sigma_{xx} \mp i\sigma_{xy} = \frac{\omega_p^2}{4\pi i\omega}\left\{1 + \frac{\hbar\omega_0}{N}\sum_{nk_yk_z}(n+\tfrac{1}{2}\pm\tfrac{1}{2})[f_0(E_{n\pm1}(k_z+q)) - f_0(E_n(k_z))][E_{n\pm1}(k_z+q) - E_n(k_z) - \hbar\omega]^{-1}\right\}. \qquad (3)$$

In Eq. (3) $f_0(E)$ is the Fermi distribution function, $E_n(k_z) = \hbar\omega_0(n+\frac{1}{2}) + \hbar^2k_z^2/2m$ is the energy of the quantum state $|nk_yk_z\rangle$, and ω_0 is the electron cyclotron frequency. In order to account for collision effects, one normally replaces ω by $\omega - i/\tau$, but in this work we restrict our attention to the situation where collisions are unimportant. In this situation Eq. (1) can be simplified and rewritten

$$\omega^2 = s^2q^2 \mp \Omega_0\omega + \Omega_p^2\left[1 - \frac{4\pi i}{\omega}\sigma_\pm(q,\omega) - \frac{c^2q^2}{\omega^2}\right]^{-1}, \qquad (4)$$

where Ω_p is the ion plasma frequency. Even in the absence of collisions $\sigma_\pm$ has both a real and imaginary part. The real part of $\sigma_\pm$ is due to a Doppler-shifted cyclotron resonance and gives rise to attenuation of the acoustic wave.[7] For sound waves the real part of $\sigma_\pm(q,\omega)$ is zero for frequencies ω smaller than $\omega_0 s/v_0$, where s is the acoustic velocity and v_0 the Fermi velocity. We restrict ourselves in this work to frequencies ω such that the three roots of Eq. (1) are well separated. In this case γ, the coefficient of attenuation of the energy of the sound wave, is given approximately by

$$\gamma \approx \frac{-(\Omega_p{}^2/s\omega)(4\pi/\omega)\,\mathrm{Re}\sigma_\pm}{[1+(4\pi/\omega)\,\mathrm{Im}\sigma_\pm - c^2q^2/\omega^2]^2+[(4\pi/\omega)\,\mathrm{Re}\sigma_\pm]^2}, \quad (5)$$

where $\mathrm{Re}\sigma_\pm$ and $\mathrm{Im}\sigma_\pm$ denote the real and imaginary parts of $\sigma_\pm$. From Eq. (3), one can show that

$$\mathrm{Re}\sigma_\pm = \frac{-3\hbar\omega_p{}^2\omega_0{}^2}{8mv_0{}^4\omega q}\sum_{n=0}^{\infty}(n+\tfrac{1}{2}\pm\tfrac{1}{2}) \times[f_0(E_{n\pm1}(K_\pm+q))-f_0(E_n(K_\pm))], \quad (6)$$

where

$$K_\pm = (m/\hbar q)(\mp\omega_0+\omega)-\tfrac{1}{2}q. \quad (7)$$

The real part of the conductivity displays giant quantum oscillations as a function of the magnetic field strength.[8]

The physical origin of the giant quantum oscillations is quite simple. In absorbing a phonon of wave vector q and energy $\hbar\omega$, an electron makes a transition from the initial state $|nk_yk_z\rangle$ to the final state $|n\pm1,k_y,k_z+q\rangle$. Conservation of energy and wave vector require that $k_z=K_\pm$, where $K_\pm$ is given by Eq. (7). In order to have absorption the initial state must be occupied and the final state empty. Thus at zero temperature absorption occurs only if the Fermi energy lies between these two states. Since these states are separated in energy by $\hbar\omega$, only over ω/ω_0 of the full period can absorption occur. The period of the oscillations is obtained by determining the change in magnetic field necessary to "push" one Landau level through the Fermi surface at the plane $k_z=K_\pm$. Actually the value of $K_\pm$ changes very slightly over one period; this results in the attenuation being a not exactly periodic function of $B_0{}^{-1}$.

For an arbitrarily shaped Fermi surface, the selection rule $\Delta n=\pm1$ is no longer exact; however, we may expect these transitions to be the predominant source of attenuation under the proper experimental conditions. The period of the oscillatory absorption will be

$$\Delta B_0{}^{-1} = 2\pi e/\hbar c S(\zeta,K) \quad (8)$$

where $S(\zeta,K)$ is the cross-sectional area of the energy surface $E=\zeta$ (Fermi surface) at the plane $k_z=K$. In this case K is approximately the value of k_z which satisfies the equation

$$\frac{\partial S(\zeta,k_z)}{\partial k_z} = q^{-1}\left\{\pm\frac{2\pi|e|B_0}{\hbar c}-\hbar\omega\left[\frac{\partial S(E,k_z)}{\partial E}\right]_{E=\zeta}\right\}. \quad (9)$$

If one knows the cyclotron effective mass of the carriers of interest ($m^*=\hbar^2/2\pi[\partial S(E,k_z)/\partial E]_{E=\zeta}$), Eqs. (8) and (9) allow one to determine $S(\zeta,k_z)$ as a function of k_z.

The oscillations discussed here will be observable if $\hbar\omega_0 \gg kT$, $\omega_0\tau \gg 1$, and if the frequency ω satisfies the conditions[9] $\omega > s\omega_0/v_0$ and $\omega \gtrsim (ms^2/\hbar\tau)^{1/2}$. In a pure sample of sodium, the oscillatory attenuation should be observable at 1.6°K in a magnetic field of 40 000 G with ultrasonic frequencies of the order of a few killomegacycles per second.

The authors would like to thank Morrel Cohen, Murray Lampert, and Robert Parmenter for helpful discussions.

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*Alfred P. Sloan Foundation Fellow; supported in part by the U. S. Atomic Energy Commission.

†Supported in part by U. S. Army Research Office.

[1]See, for example, A. B. Pippard, Rept. Progr. Phys. 23, 176 (1960).

[2]See, for example, D. Shoenberg, Progress in Low-Temperature Physics, edited by J. C. Gorter (North-Holland Publishing Company, Amsterdam, 1957), Vol. 2, p. 226.

[3]V. L. Gurevich, V. G. Skobov, and Yu. A. Firsov, Zh. Eksperim. i Teor. Fiz. 40, 786 (1961) [translation: Soviet Phys.—JETP 13, 552 (1961)]. In principle, the effect discussed by these authors could also be used to study nonextremal cross sections of the Fermi surface, but in practice the angle between the direction of propagation and the magnetic field cannot be controlled with sufficient accuracy to obtain useful results.

[4]J. J. Quinn and S. Rodriguez, Phys. Rev. 128, 2487 (1962).

[5]S. Rodriguez, Phys. Rev. 130, 1778 (1963).

[6]J. J. Quinn and S. Rodriguez, Phys. Rev. Letters 11, 552 (1963); D. N. Langenberg and J. Bok, Phys. Rev. Letters 11, 549 (1963).

[7]The Doppler-shifted cyclotron resonance also gives rise to absorption of helicon waves. The quantum mechanical treatment of this effect has been given else-

where. [J. J. Quinn, Phys. Letters 7, 235 (1963); P. B. Miller, Phys. Rev. Letters 11, 537 (1963).]

[8]One can use the Poisson sum formula [see, for example, R. Courant and D. Hilbert, Methods of Mathematical Physics (Interscience Publishers, Inc., New York, 1953), Vol. 1, p. 76] to put Eq. (6) in a form which explicitly displays the oscillatory behavior. The result is

$$\mathrm{Re}\sigma_{\pm}=\frac{-3\hbar\omega_p{}^2\omega_0{}^2}{8mv_0{}^3\omega q}\sum_{r=-\infty}^{\infty}(-1)^r\left[\pm\frac{1}{2}+\frac{1}{2\pi i}\frac{\partial}{\partial r}\right][\Phi_r(\zeta-\epsilon(K_{\pm})-\hbar\omega)-\Phi_r(\zeta-\epsilon(K_{\pm}))],$$

where

$$\Phi_r(x)=\frac{\pi kT}{\hbar\omega_0}\frac{\sin(2\pi rx/\hbar\omega_0)}{\sinh(2\pi^2 rkT/\hbar\omega_0)},$$

ζ is the Fermi energy, and $\epsilon(K)=\hbar^2K^2/2m$.

[9]This second condition on the frequency arises because of the uncertainty principle. If this condition is satisfied, the meaning of the selection rule (Eq. 7) remains effectively unimpaired by the collision broadening of the Landau levels. It should also be pointed out that the use of the word "giant" to describe the oscillations is really only appropriate when $\hbar\omega \gtrsim kT$ and $\omega T > 1$, conditions which are difficult to satisfy experimentally.

OPTICALLY OBSERVED INNER SHELL ELECTRON EXCITATION IN NEUTRAL Kr AND Xe†

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(Received 23 December 1963)

The purpose of this Letter is to report the observation of the excitation spectra of inner shell d electrons in neutral Kr and Xe by optical spectroscopy. The fine-structure splitting of these electrons amounts to 1-2 eV. Analogous fine structure has been previously observed only in transitions to much deeper levels resulting in x-ray photons of the order of 10^4 eV. The present spectra were observed in the 100-200Å region by absorption spectroscopy using as a background source the "synchrotron light" continuum radiated by the NBS 180-MeV electron synchrotron. Inner-shell structural details, well resolved in this wavelength region, are difficult to observe in the ordinary x-ray region.

Resonances in photoionization continuum absorption having the Beutler-Fano profile[1] have recently been reported for He, Ne, and Ar,[2] and for Kr[3,4] and Xe.[4] The observed structure in the case of Kr and Xe occurred in the 380-600Å region and was shown to be due in part to the excitation of inner subshell s electrons. These high-lying neutral-atom energy states interact with the adjacent photoionization continuum, decaying rapidly into this continuum (autoionization).

The present spectra were observed in the region of 130 Å for Kr and 180 Å for Xe. A three-meter grazing incidence spectrograph was used, having available a resolution of approximately 0.06 Å at 150 Å. The absorption spectra were obtained by admitting the gas into the spectrograph at room temperature and a pressure of approximately 0.07 mm of Hg.

The observed spectra are shown in Fig. 1. For both Kr and Xe, two series of resonances occur, superimposed upon the continuous photoionization absorption for these gases. For Kr, this discrete structure occurs 80 eV above the first ionization limit (14.00 eV) and, in fact, about 20 eV above the third ionization limit (75.52 eV). For Xe, the structure occurs more than 53 eV above the first ionization limit (12.13 eV) and in the region of the third ionization limit (65.46 eV). As is expected for autoionizing series, the widths of the resonances decrease with higher series members.

The measured wavelength and the effective quantum number, n^*, determined for each observed series member, are listed in Table I. In addition, the series limits, determined by a quantum defect plot, are given.

In identifying the transitions associated with these series, a consideration of available x-ray data yields useful information. From differences in observed x-ray transitions, the $M_{\mathrm{IV,V}}$ edge of Kr has been estimated to be (87 ± 10) eV.[5] The $M_{\mathrm{IV,V}}$ edge results from the removal of a $3d$ electron which leaves the Kr ion in a $^2D_{3/2,5/2}$ state. The x-ray data do not yield an estimate of the ^2D splitting. For Xe, x-ray data on the $N_{\mathrm{IV,V}}$ edge are not available. However, the position of this edge is known for the elements on either side of Xe in the periodic table, i.e., Cs and I. From these data one may estimate that the $N_{\mathrm{IV,V}}$ edge of Xe will lie close to 65 eV with a $^2D_{3/2,5/2}$ splitting of approximately 2 eV (16 000 cm^{-1}).

In the case of Kr, additional information is available from spectroscopic data[6] on the isoelec-