Quantum Spectroscopy of the Low-Field Oscillations in the Surface Impedance*†

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We present here a detailed theory of electronic surface quantum states in a low magnetic field, as well as of their effects on the microwave surface impedance. A marked oscillatory structure in the microwave absorption as a function of magnetic field has been carefully observed by Khaikin and by Koch *et al.* The quantized magnetic surface levels are bound states of electrons trapped against the surface by the magnetic field. Even though these levels are somewhat analogous to Landau levels, they have considerably different properties. Resonant transitions between these levels give rise to a series of spectral lines in the surface impedance, just as cyclotron resonance is a result of transitions between Landau levels. The present effect is essentially quantum in nature, however. A considerable amount of quantitative information can be extracted from the experimental data. The Fermi velocity, radius of curvature of the Fermi surface, and mean free time at certain points on the Fermi surface can be obtained. Most novel, however, is the fact that one can extract information on the scattering of electrons by the surface, as a function of impact angle.

I. INTRODUCTION

FOR many years curious oscillations in the surface impedance in low magnetic fields have been observed in metals (Sn, In, Cd, Al, W, Bi, Ga, etc.). The surface impedance oscillates as a function of magnetic field for fixed frequency, or as a function of frequency for fixed field. Oscillations are observed in the range of 0–50 Oe at characteristic microwave frequencies. The existence of detailed structure was first noted by Khaikin[1] and has subsequently been explored by Koch and Kip,[2] and Koch and Kuo.[3] The last report is the most detailed to be published up to the present time, although a considerable amount of new experimental detail is now being prepared for publication.[4]

We wish to advance here a simple theory of the effect which seems to account for the experimental results in considerable detail.[5] The theory reveals that a whole new range of parameters characterizing the metal may be determined by analyzing the experimental results. Not only may orientations, curvatures, and Fermi velocities at certain (cylindrical) portions of the Fermi surface be determined, but one may obtain information on surface scattering as well as ordinary scattering at these points.

In this paper, we set forth the theory for such surface quantum states together with a consideration of their effect on the microwave surface impedance. In a subsequent paper we will present a detailed calculation of line shapes and comparison with experiments, with a view toward ascertaining numerical values for parameters characteristic of the low-field oscillation effect.

The first theory put forward to account for the oscillations is that of Koch *et al.*[2,3] This model proposes that the important electrons are those moving in orbits that skim through the surface layer at the top of their cyclotron orbits. (See Fig. 1.) Those electrons exposed for an odd number of half periods to the radio frequency field in the skin layer absorb more power than other electrons. Expressing the time spent in the skin layer in terms of the angular range (2θ) traversed on the cyclotron orbit, we have

$$\Delta T = 2\theta(R_c/v_F), \tag{1}$$

where R_c is the cyclotron radius, and v_F is the Fermi velocity. With θ related to the skin depth δ as

$$\delta = R_c(1-\cos\theta) \simeq \tfrac{1}{2}R_c\theta^2, \tag{2}$$

we find the field values for maximal absorption as

$$H_n = \frac{\omega^2\delta}{8\pi^2(2n+1)^2}\frac{1}{e}\left(\frac{p_F}{v_F^2}\right), \tag{3}$$

where p_F and e represent the Fermi momentum and electronic charge, respectively. A somewhat more involved argument given by Koch and Kuo[3] gives $H_n \propto 1/(2n+1)$, more nearly in line with the experimentally observed periodicity.

The expression for H_n is in disagreement with the experimental results[3] in three ways. (1) The experimentally observed periodicity for the dR/dH peaks is not as

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[1] M. S. Khaikin, Zh. Eksperim. i Teor. Fiz. **39**, 212 (1960) [English transl.: Soviet Phys.—JETP **12**, 152 (1961)].

[2] J. F. Koch and A. F. Kip, in *Low Temperature Physics LT9* (Plenum Press, Inc., New York, 1965), p. B818.

[3] J. F. Koch and C. C. Kuo, Phys. Rev. **143**, 470 (1965).

[4] J. F. Koch (private communication).

[5] Tsu-Wei Nee and R. E. Prange, Phys. Letters **25A**, 582 (1967).

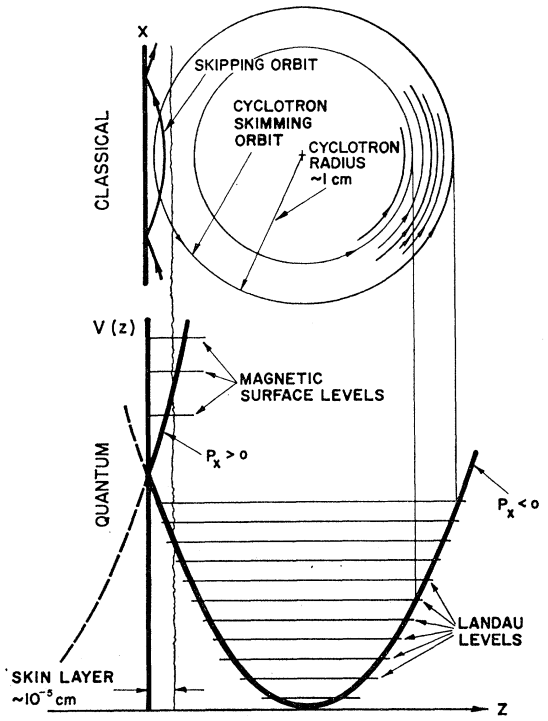

FIG. 1. Electrons traverse the radio-frequency skin layer along both skimming and skipping orbit trajectories. In weak fields ($\approx$10 Oe) the cyclotron radius characteristic of this motion is typically 1 cm. Electrons in the cyclotron orbits are represented by the Landau levels of the lower portion of the figure, whereas the skipping orbits form a system of magnetic surface states. Microwave transitions between surface states account for the impedance oscillations observed in weak magnetic fields as in Fig. 2.

$(2n+1)^{-2}$. The fit to $(2n+1)^{-1}$ is at best approximate. (2) Noting that the skin depth $\delta(\omega) \propto \omega^{-1/3}$ is appropriate for the anomalous skin effect regime, we find $H_n \propto \omega^{5/3}$. The data show unambiguously that the correct scaling law is $H \propto \omega^{3/2}$. (3) The skin depth may be changed appreciably at fixed frequency by changing the polarization of the microwave electric field. The structure does not change experimentally as predicted from Eq. (3). These limitations on the theory were recognized by the original authors, who were merely proposing a model in order to suggest further experiments.

An alternate classical scheme considers electronic orbits which skip along the surface. (See Fig. 1.) This idea has been stressed by Khaikin,[6] although it was brought to our attention by the independent work of Burke and Ferrell.[7] Using arguments essentially as those presented above, one arrives at nearly identical expressions for H_n.[6] We believe that it is a quantum version of the classical skipping orbit idea serves to understand the phenomenon.

The classical arguments as given are incomplete. In any case, it is necessary to sum over all orbits traversing the skin layer. Different orbits can arise because there are different guiding centers. Thus one must ask why the δ on the left-hand side of Eq. (2) should not be replaced by a spread of possible values. Secondly, different parts of the Fermi surface will have different values of the cyclotron radius. This difficulty can be obviated by assuming that the important orbits come from a cylindrical piece of Fermi surface,[3] an assumption which we also will adopt. To some extent, this assumption is justified by the failure to observe oscillations in the alkalis, or in the noble metals. However, there are other possible reasons of an experimental nature which could account for the failure, and it is premature to conclude that the effect will not be seen in these materials.

We have belabored the classical theory at some length because the ideas are not far from correct; one needs to put in only a minimal amount of quantum theory to obtain the correct results. Van Gelder was the first to notice that quantum effects could be important in the surface impedance problem.[8–10] He has made an extensive study of quantum effects in the surface impedance, where he points out that quantum effects could well be important because the wavelength of the electrons at their classical turning point (in the radial direction) is of the order of magnitude 10^{-4}–10^{-5} cm for the low magnetic-field values of interest. These points will be made clearer subsequently.

Van Gelder treats the Koch-Kip skimming orbit considerations quantum mechanically, whereas we quantize the Khaikin skipping orbit. The former is much more difficult both mathematically and numerically, so the results are not clear cut. While Van Gelder was eminently successful in explaining the frequency scaling for the observed microwave effects he fails to properly account for the periodicity and line shapes of the oscillations.

II. MAGNETIC SURFACE LEVELS

In this section, we study the elementary properties of magnetic surface levels in the simplest case. We begin by supposing that the electronic energy levels are given, in the absence of a field, by a function $\epsilon(p_x,p_z)$ which is characteristic of a cylindrical piece of Fermi surface, i.e., we neglect the p_y dependence of the energy states. [For the sake of clarity, one may imagine that $\epsilon(p_x,p_z)=p_x^2/2m_1+p_z^2/2m_3$, although it will shortly develop that no such global knowledge of $\epsilon(p_x,p_z)$ is necessary.] Let the z direction be into the metal. In the presence of a magnetic field H applied in the positive

[6] M. S. Khaikin, Zh. Eksperim i Teor. Fiz. Pis'ma v Redaktsiyu 4, 164 (1966) [English transl.: Soviet Phys.—JETP Letters 4, 113 (1966)].

[7] E. Burke and R. A. Ferrell (1964) (unpublished).

[8] A. P. Van Gelder, Phys. Letters **21**, 18 (1966). (More precisely, D. C. Mattis and Dresselhaus, Phys. Rev. **111**, 403 (1958), had already noted effects of Landau levels in the very high-field case.)

[9] A. P. Van Gelder, Phys. Letters **22**, 7 (1966).

[10] A. P. Van Gelder, Ph.D. thesis (Nijmegen, 1967) (unpublished). We thank Dr. Van Gelder for making available a copy of his work.

y direction we have an effective Hamiltonian:

$$\mathcal{H}=\epsilon(p_x+eHz,p_z), \tag{4}$$

where we are at liberty to choose the gauge such that

$$\mathbf{A}=(Hz,0,0). \tag{5}$$

Since we are going to be interested finally in gauge invariant quantities, we naturally try to choose a convenient gauge. The choice of the Landau gauge here has the advantage that p_x and p_y remain constants of the motion, at least if a perfectly smooth surface at $z=0$ is considered.

We must now treat p_z as an operator which obeys the canonical commutation relations with z. Then the term $(p_x+eHz)^2/2m_1$ represents a parabolic harmonic oscillator potential whose center depends on the value of p_x chosen. Figure 1 illustrates this situation, where, for H along the positive y direction we see that the center of the oscillator is inside the metal for $p_x<0$ and outside the metal for $p_x>0$. The levels of the oscillator are of course the familiar Landau levels. The interesting states are those which have energies near the Fermi energy. Thus, in a field of 10 Oe, as appropriate for the experiments, Landau levels characterized by a quantum number $n\sim10^6$ are important. The great size of this number naturally leads one to believe that quantum effects due to Landau levels are not important. Indeed, cyclotron resonance resulting from microwave transitions between these levels, can be adequately discussed entirely in classical terms. In the low-field limit that we are considering, one has the cyclotron radius $R_c\sim1$ cm, and one usually has insufficient mean free path for the completion of cyclotron orbits. So far, however, we have not taken into account electronic orbits that are incident on the surface. There is, of course, a relatively high-potential barrier which confines the electrons to the interior of the metal. Making the most obvious approximation, we simply assume that there is an infinitely high barrier at the surface. In other words, we demand that the electronic wave function vanish at $z=0$. The reader will be able to think of numerous problems with this approximation. Deferring all discussion of this point, however, we can see at once that quite new types of electronic states can be found. These states correspond to classical trajectories skipping along the surface by periodic reflection. Although the exact solutions are known (Weber functions), nothing is gained by considering them.

Out of the entire spectrum of colliding orbits, we can decide which electronic states are most important by comparison with the classical situation. There are two conditions determining the important states. The first is that the energy of the state, as already mentioned, must be near the Fermi energy (just how near will be noted later.) The second is that the classical orbits must pass through the skin layer, and the orbits should have velocity nearly parallel to the surface at that point. In other words, the component of velocity v_z should be very small when the electron is in the skin layer, if the electric field is to be successful in exciting the state under consideration.

These considerations severely restrict the values of p_x which have to be considered. They mean that the effective Hamiltonian must be such that the classical turning point for an electron of Fermi energy ϵ_F must lie within a few skin depths of the surface. This can be achieved for p_x either positive or negative. The case of positive p_x has a classical correspondence with the skipping orbits. These orbits obviously have a periodic motion in the z direction and therefore must be quantized. The values of p_x of interest occur near p_{x0} where

$$\epsilon(p_{x0},p_{z0})=\epsilon_F,\quad \frac{\partial\epsilon}{\partial p_{z0}}(p_{x0},p_{z0})=v_{z0}=0. \tag{6}$$

We will show later that it is a generally very good approximation to expand

$$\epsilon(p_x,p_z)=\epsilon_F+(p_x-p_{x0})v_{x0}+(p_z-p_{z0})^2/2m_3 +1/2m_{13}\{(p_x-p_{x0})(p_z-p_{z0}) +(p_z-p_{z0})(p_x-p_{x0})\}+\cdots. \tag{7}$$

Putting in this equation $p_x\rightarrow\delta p_x+ezH+p_{x0}$ and letting $p_z-p_{z0}=(\hbar/i)(\partial/\partial z)$ we have

$$\mathcal{H}=\epsilon_F+\frac{1}{2m_3}\left[\frac{\hbar}{i}\frac{\partial}{\partial z}+\frac{m_3}{m_{13}}(\delta p_x+eHz)\right]^2+\delta p_x v_{x0} +eH_y z v_{x0}-\tfrac{1}{2}(m_3/m_{13}^2)(\delta p_x+eHz)^2. \tag{8}$$

Neglecting the final term in (8), as will later be justified, and making a gauge transformation, we have

$$\mathcal{H}-\epsilon=-\frac{\hbar^2}{2m_3}\frac{\partial^2}{\partial z^2}+eHv_{x0}(z-z_{\epsilon,p_x}), \tag{9}$$

where

$$eHv_{x0}z_{\epsilon,p_x}=\epsilon-\epsilon_F-\delta p_x v_{x0} \tag{10}$$

defines the parameter z_{ϵ,p_x}.

The Schrödinger equation with Hamiltonian (9) is mathematically known as Airy's equation. The solutions falling off for large positive z are the ordinary Airy functions Ai(z).[11] The wave function describing an electron in a skipping trajectory is thus

$$\psi(x,y,z)=A^{-1/2}e^{ik_x x}e^{ik_y y}\phi_n(z), \tag{11}$$

where $k_i=p_i/\hbar$ and A is the area of the surface. The wave function $\phi_n(z)$ is

$$\phi_n(z)=C_n\,\mathrm{Ai}(\alpha z-\alpha z_{\epsilon,p_x}), \tag{12}$$

where the characteristic wave number α is

$$\alpha=(2KeH/\hbar)^{1/3}. \tag{13}$$

[11] M. Abromowitz and I. A. Stegun, *Handbook of Mathematical Functions* (National Bureau of Standards, 1955).

In this expression $K=m_3v_{x0}/\hbar$ is the radius of curvature of the Fermi surface at p_{x0}, p_{z0}. The magnitude of α is typically

$$\alpha\simeq.5\times10^5\, H^{1/3}\ \mathrm{cm}^{-1}. \tag{14}$$

The eigenvalue $\epsilon_{n,p}$ is determined by requiring the vanishing of the wave function at $z=0$; thus

$$\alpha z_{\epsilon,px}=a_n, \tag{15}$$

where

$$\mathrm{Ai}(-a_n)=0,\quad n=1,\,2,\,3,\,\cdots. \tag{16}$$

The a_n are quite well determined by the WKB approximation even for low n, which gives

$$a_n=[\tfrac{3}{2}\pi(n-\tfrac{1}{4}]^{2/3};\quad n=1,\,2,\,3,\,\cdots. \tag{17}$$

The normalization constant C_n is exactly equal to $\alpha^{1/2}[\mathrm{Ai}'-(a_n)]^{-1}$. Thus in the WKB approximation, we have

$$\phi_n(z)=\frac{\alpha^{1/2}}{a_n{}^{1/4}(a_n-\alpha z)^{1/4}}\cos[\tfrac{2}{3}(a_n-\alpha z)^{3/2}-\tfrac{1}{4}\pi], \tag{18}$$

when

$$(\alpha z<a_n).$$

Solving for ϵ by means of (10) and (15) we find

$$\epsilon_{n,p_x}=\epsilon_F+\delta p_x v_{x0}+\hbar v_{x0}(eH/\hbar)^{2/3}(1/2K)^{1/3}a_n. \tag{19}$$

We will later discover that the surface impedance can be calculated from the point of view according to which the incident microwave electric field causes transitions between the energy levels ϵ_{n,p_x} of the system. In such transitions p_x is conserved. The Einstein frequency condition then gives

$$\begin{aligned}\omega&=(\epsilon_{n,p_x}-\epsilon_{m,p_x})/\hbar,\\ \omega_{nm}&=v_x(eH/\hbar)^{2/3}(1/2K)^{1/3}(a_n-a_m).\end{aligned} \tag{20}$$

Since experiments are usually performed at fixed frequency, it is convenient to solve for the field values

$$H_{n,m}=-\frac{\hbar}{e}\frac{(2K)^{1/2}}{v_{x0}{}^{3/2}}\frac{\omega^{3/2}}{(a_n-a_m)^{3/2}}. \tag{21}$$

To indicate how successfully this formula predicts the positions of peaks in the surface resistance derivative dR/dH, we examine in Fig. 2 some recent data on the low-field oscillations in In. With the assumption that the pronounced peak at 36.4 Oe represents the resonance condition due to transitions between states characterized by $n=1$, $m=2$, we determine a value of $K^{1/2}/v_{x0}{}^{3/2}$ as 4.5×10^{-9} $\mathrm{sec}^{3/2}$ cm^{-2}. All subsequent dR/dH peaks of the oscillation are then determined from Eq. (21) and have been marked on the figure. The identification of the dR/dH peaks as characterizing the resonance condition will be justified in a subsequent paper devoted to a calculation of line shapes.[12] The value of the Fermi surface parameters derived in this manner is indeed quite reasonable. As pointed out in a previous publication[5] Eq. (21) accounts equally well for the low-field spectra observed in Sn. More recent work in Bi where the Fermi surface parameters are well established allows an even more critical comparison.[13]

In summary, and at the risk of belaboring the obvious, we point out that (1) the field values for the dR/dH peaks predicted agree nicely with experiments, (2) the correct frequency scaling (i.e., $H_{nm}\propto\omega^{3/2}$) is evident from Eq. (21). (3) There is no dependence on the radio frequency skin depth as in the classical considerations. It may be well to emphasize that the data due to a single cylindrical section of the Fermi surface are resolved into a number of spectral series each characterized by a specific lower state m and going to successively higher states n. These series are identified in Fig. 2 by a sequence of multiple arrows.

III. SURFACE IMPEDANCE

In this section, we obtain an expression for the surface impedance derivative, and discuss some of the approximations made in the preceding section. The reader who is not interested in these rather tedious and formal derivations can skip directly to Eq. (39) at the end of the section.

A. Surface Impedance Variations in the Weak Signal Limit

In order to complete the theoretical treatment, we must attempt to predict the actual form of the contribution of the surface quantum states to the microwave impedance, as well as to discuss the approximations made in the preceding simple argument.

Our experimental grounds we note that the oscillatory part of the surface impedance is a small part of the total.[3] This allows us to base an approximation on the weak dependence of the conductivity kernel on magnetic field. Thus we assume that

$$\delta K_{\mu\nu}=K_{\mu\nu}(z,z',H)-K_{\mu\nu}(z,z',0) \tag{22}$$

is small, where the current, in the presence of magnetic field of strength H and a microwave field E, is given by

$$j_\mu(z)=\sum_\nu\int_0^\infty dz' K_{\mu\nu}(z,z';H)E_\nu(z'), \tag{23}$$

and a time dependence $e^{-i\omega t}$ is assumed. It may be verified that $K_{\mu\nu}(z,z')=K_{\mu\nu}(z',z)$. The following variational expression has been used by Marcus[14]:

$$I=\Big[\sum_\nu\int_0^\infty[E_\nu'(z)]^2dz+\sum_{\nu,\mu}\int_0^\infty dz\int_0^\infty dz' E_\nu(z)\times K_{\nu\mu}(z,z')E_\mu(z')\Big]\Big/\sum_\sigma E_\sigma(0)^2. \tag{24}$$

[12] T. W. Nee, J. F. Koch, and R. E. Prange (to be published).

[13] J. Jensen and J. F. Koch (to be published).

[14] P. M. Marcus, Natl. Bur. Std. Circ. No. 519, 265 (1952).

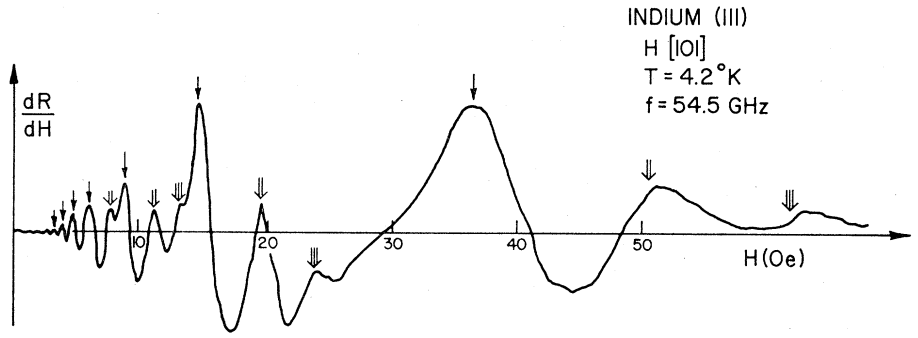

FIG. 2. Variation of the microwave absorption derivative dR/dH with magnetic field applied parallel to the surface of an indium crystals. Identifying the large peak at 36.4 as due to resonant transition between levels characterized by $n=1$, $m=2$, the positions of all other peaks are then determined from the theory and are indicated by the arrows. Single arrows denote the series of transitions starting at ground state $n=1$ and going to successively higher states $m=2$, 3, 4, etc. The double and triple arrowed series represents series where the lower state is $n=2$ and 3, respectively.

The integral I is stationary at the correct electric field value [given $E_\nu(0)$], where it takes the value

$$I=\sum_{\mu,\nu} E_\mu(0)(Z^{-1})_{\mu\nu}E_\nu(0)/\sum_\sigma E_\sigma(0)^2, \qquad (25)$$

in which $(Z^{-1})_{\mu\nu}$ is the inverse surface impedance tensor. Restricting our consideration to the case in which E is in one of the principle directions of the impedance tensor, we may take into account the magnetic field by keeping only those terms linear in δK [Eq. (22)]. The result is

$$Z^{-1}(H)-Z^{-1}(0)=\int_0^\infty dz\int_0^\infty dz'E(z)E(z') \times\delta K(z,z')/E(0)^2. \qquad (26)$$

In (26) the electric field is to be evaluated at zero magnetic field. The usually measured quantities are the real and imaginary parts of the surface impedance derivatives. Thus, with the same accuracy, we find

$$\frac{\partial Z}{\partial H}\simeq -Z(0)^2\frac{\partial}{\partial H}[Z^{-1}(H)]. \qquad (27)$$

[We do not know if the result (26) connecting small changes of surface impedance with small changes of conductivity is in the literature, although somewhat less general results can be found. The closest, and about equivalent for practical purposes, is an equation due to Van Gelder[10] which in our notation specializes (26) to the case in which $Z(0)$ and $E(z)$ are to be calculated on the basis of specular reflection in zero field.]

B. Conductivity Kernel

Thus far, no serious approximation has been made. It is now necessary to obtain an expression for $\delta K(z,z')$ in the course of which some approximations must be introduced. For this purpose, we may use the standard formula[14]

$$j_\alpha(\mathbf{r})=-\langle n(\mathbf{r})\rangle\frac{e^2}{m\omega i}E_\alpha(\mathbf{r})+\int d^3r'\frac{e^3}{\hbar\omega}\int_0^\infty d\tau \times e^{i\omega\tau}\langle[j_\alpha(\mathbf{r},\tau),j_\beta(\mathbf{r}',0)]\rangle E_\beta(\mathbf{r}'). \qquad (28)$$

In (28) the expectation value has the meaning of quantum-mechanical expectation value, averaged over a thermal ensemble, as well as, in principle, an ensemble of states which account for surface roughness and imperfection scattering. Since we are only interested in the oscillatory part of the kernel, we may assume that all the states entering into (28) are constructed of single-particle states of the form as in Eq. (11), and since these lie close to the Fermi surface, we may make the independent quasiparticle approximation. We may thus take the current operators j_x, j_y as defined by the expression

$$\int dxdy\mathbf{j}(\mathbf{r})=-e\sum_{p_x,p_y,m,n}\mathbf{v}_p a_{pm}^\dagger a_{pn}\phi_m(z)\phi_n(z). \qquad (29)$$

In terms of the assumptions regarding the Fermi surface geometry as in the preceding section, only j_x is nonvanishing. Inserting (29) into (28) we make a sort of Hartree-Fock approximation which we do not attempt to justify here. Namely, we put

$$\langle a_{pm}^\dagger(\tau)a_{pn}(\tau)a_{\bar{p}\bar{n}}^\dagger(0)a_{\bar{p}\bar{m}}(0)\rangle =\langle a_{pm}^\dagger(\tau)a_{\bar{p}\bar{m}}(0)\rangle\langle a_{pn}(\tau)a_{\bar{p}\bar{n}}^\dagger(0)\rangle \qquad (30)$$

and further assume

$$\begin{aligned}\langle a_{pn}(\tau)a_{\bar{p}\bar{n}}^\dagger(0)\rangle&=\delta_{p\bar{p}}\delta_{n\bar{n}}G_{pn}^<(\tau),\\ \langle a_{pn}^\dagger(\tau)a_{\bar{p}\bar{n}}(0)\rangle&=\delta_{p\bar{p}}\delta_{n\bar{n}}G_{pn}^>(\tau),\end{aligned} \qquad (31)$$

where as usual $G^\gtrless$ has the temporal Fourier transform[14]

$$G_{pm}^\gtrless(\tau)=\frac{1}{2\pi}\int d\omega\, A_{pm}(\omega)\begin{pmatrix}f(\omega)\\1-f(\omega)\end{pmatrix}e^{-i\omega\tau} \qquad (32)$$

in which $f(\omega)$ is the Fermi distribution function.

The physical content of these approximations is as follows. In (29) we have treated the current in the same effective Hamiltonian approach to band structure as we have treated the Hamiltonian itself. In (30) we have neglected possible correlation effects of the many-particle variety (probably justified), of the impurity scattering variety (also justified in the anomalous skin effect case) and of surface roughness scattering. In (31) the first δ function is justified, since p_x, p_y are conserved, on the average, but we have not found a similar argument in the case of the quantized labels m, n.

Finally, we assume that A takes the form

$$A_{p,m}(\omega)=\Gamma_{p,m}/[(\omega-\epsilon_{p,m}/\hbar)^2+\tfrac{1}{4}\Gamma^2{}_{p,m}], \quad (33)$$

or, in other words, we assume a Lorentzian line shape with characteristic width Γ. We deal with the gauge current [the first term in (28)] by noting that at zero frequency there is no current[15–17] (in the absence of Landau diamagnetism). We thus obtain

$$\frac{\partial}{\partial H}Z^{-1}(H)=\frac{\partial}{\partial H}\frac{ie^2}{\hbar\omega}\int\frac{dk_xdk_y}{(2\pi)^4}\sum_{m,n}\alpha_{mn}{}^2v_\nu{}^2 \times\int d\omega_1d\omega_2A_{n,p}(\omega_1)A_{m,p}(\omega_2)[f(\omega_1)-f(\omega_2)] \times\left[\frac{1}{\omega-\omega_1+\omega_2+i\eta}-\frac{1}{\omega_2-\omega_1}\right], \quad (34)$$

where the electric field is applied in the ν direction, the k_x, k_y are wave numbers corresponding to momenta p_x, p_y, and η appears as an infinitesimally small positive quantity. In (34) the matrix element α_{mn} is given by

$$\alpha_{nm}=\int_0^\infty dz\,\phi_n(z)E(z)\phi_m(z)/E(0). \quad (35)$$

Note that the *ordinary* square of the matrix element appears, which is important, as it is complex.

Leaving the k_y integral to the end, we see that the k_x integral can be performed, if we notice that the only important dependence on this quantity occurs in the functions $\epsilon_{p,m}$. Putting $k_x=k_{x0}$ everywhere but in the energy, and replacing ω_1 by $\omega_1+\epsilon_{p,n}/\hbar$, ω_2 by $\omega_2+\epsilon_{pn}/\hbar$ we see that all the k_x dependence resides in the Fermi functions. Thus, we must perform the integral

$$K=\int dk_x[f(\omega_1+\epsilon_{p,n}/\hbar)-f(\omega_2+\epsilon_{p,n}/\hbar)]. \quad (36)$$

The important values of ω_1, ω_2, and ω_{mn} are very small in comparison with $\epsilon_F/\hbar$. The bracketed function in (36) is large only when $\epsilon_{p,n}$ is very close to the Fermi energy. The width of this region, in frequency, is $|\omega_1-\omega_2|$ or $kT/\hbar$, whichever is larger. The important value of $\omega_1-\omega_2$ is ω, which is generally somewhat smaller than $kT/\hbar$ under experimental conditions.[3] Thus, the k_x integral in (36) may be expanded to $\pm\infty$, and evaluated to give

$$K=(\omega_2-\omega_1)/v_x. \quad (37)$$

We wish to emphasize that this result is independent of temperature, insofar as $kT\ll\epsilon_F$. It is independent of the condition $kT\ll\hbar\omega$, as might be supposed necessary. This situation is familiar in the case of cyclotron resonance and anomalous skin effect calculations, but differs from that of the de Haas–van Alphen effect. Because of the characteristic difference of Fermi functions which appears in the conductivity, the reduction in the probability of transition for a given value of p_x as the temperature is raised is exactly compensated by the increase in the number of values of p_x which can contribute. There will, of course, be a component of the lifetime dependent on temperature, and since both the amplitude and width of the levels depends strongly on the lifetime, there is expected to be an easily observable temperature effect.

This analysis also provides an *a posteriori* justification for the expansion (7). An estimate of the maximum important value of δp_x is provided by Eq. (19) together with the result just obtained; $|\epsilon_{n,p_x}-\epsilon_F|\lesssim kT$. Thus

$$\delta p_x/mv_x\lesssim(kT/mv_x{}^2+(eH/\alpha mv_x)a_n), \quad (38)$$

which is usually of order 10^{-3}.

With the result (37) we are enabled to perform the ω_1, ω_2 integration, to obtain finally

$$\frac{\partial Z(H)}{\partial H}=\frac{\partial}{\partial H}\int\frac{dk_ye^2}{(2\pi)^2i}\frac{v_\nu{}^2}{\hbar v_x}[Z(0)]^2 \times\sum_{m,n}\frac{\alpha_{mn}{}^2}{\omega-\omega_{mn}+i\Gamma_{mn}}, \quad (39)$$

where

$$\Gamma_{mn}=\tfrac{1}{2}(\Gamma_m+\Gamma_n). \quad (40)$$

[For the sake of those who have arrived at this point via the recommended short cut we repeat that v_ν is the component of the Fermi velocity in the direction of the microwave electric field, and α_{mn} is the normalized matrix element of that field, Eq. (35). Γ will be discussed in the next section.]

Formula (39) is plausible enough. It should be noted, however, as both $Z(0)$ and α_{mn} are complex, the absorptive part of Z will not consist entirely of superposition of Lorentzians, but will have contributions from the dispersive part of the energy denominators mixed in. For this reason, the maxima of $Z(H)$ will not be precisely at $\omega_{mn}=\omega$. In a detailed paper devoted to a calculation of line shapes we will elaborate on this point.[12]

IV. LIFETIME OF MAGNETIC SURFACE LEVELS

Aside from the rather technical problem of evaluating the matrix elements α_{mn}, the remaining theoretical task is to find expressions for the widths Γ_m.

[15] A. A. Abrikosov, L. P. Gorkov, and I. E. Dzyaloshinski, *Quantum Statistical Mechanics* (Prentice-Hall, Inc., Englewood Cliffs, N. J., 1965).
[16] L. P. Kadanoff and G. Baym, *Quantum Statistical Mechanics* (W. A. Benjamin, Inc., New York, 1962).
[17] D. C. Mattis and J. Bardeen, Phys. Rev. **111**, 412 (1958).

A. Bulk Scattering

There are three sources of broadening, which we assume are additive. The first is just the ordinary mean free time effect.[18] There is some interest in this quantity however, since it is obviously the mean free time at a particular point of the Fermi surface which is relevant. We shall see that it is possible, in principle, to separate this component of the linewidth from the others, although we are not yet prepared to say with what precision this can be done.

B. Cylinder Approximation

The second source of broadening comes from the breakdown of the cylinder approximation. This can be treated most easily if the magnetic field is precisely in the y direction. In this case, p_x and p_y remain constants of the motion. All the steps up to now can be carried out for each value of p_y, except that it must be understood that p_{x0}, p_{z0} are functions of p_y. In particular, the parameter $v_x/K^{1/3}$ will depend on p_y, thus causing ω_{mn} to be dependent on the y momentum also. As a result of this p_y dependence, there will be a p_y broadening, since for each value of p_y the resonance will be at a slightly field value. However, as is customary, those values of p_y for which ω_{mn} is stationary, will contribute most heavily.

In order to see the general result expected, let us suppose that we may represent $\omega_{mn}(k_y)$ sufficiently well by the formula

$$\omega_{mn}(k_y)\simeq\omega_{mn}(1\mp k_y^2/K^2). \tag{41}$$

Then, neglecting all other dependence on k_y, and extending the limits on the k_y integration in (39) to infinity, we find

$$\frac{\partial Z(H)}{\partial H}=\frac{\partial}{\partial H}\left[e^2K\frac{v_r^2}{v_x}\frac{[Z(0)]^2}{4\pi i\hbar}\times\sum_{m,n}\frac{\alpha_{mn}^2}{[\pm\omega_{mn}(-\omega\omega_{mn}+i\Gamma_{mn})]^{1/2}}\right], \tag{42}$$

where the square root is chosen to have positive imaginary part. Thus, for a bellylike cylinder, the imaginary part of the square root expression is largest for fields such that $\omega_{mn}>\omega$, whereas for a necklike cylinder the opposite is the case.

Thus, it is possible, in principle, to distinguish between necklike and bellylike cylindrical sections by analyzing the symmetry of the line shape.

Experimental evidence to date[3,4,6] shows largely that cylindrical sections of Fermi surface are responsible for the oscillatory effects in the low field impedance. Nevertheless, it appears from (42) that the effect should also be observable for metals with more nearly spherical Fermi surfaces (Na, K, Cu, Ag, and Au), if sufficiently good samples can be prepared.

[18] The mean free time here is the effective free time between any scattering events. Even small angle scattering will effectively remove an electron from its skipping trajectory.

C. Surface Scattering

The most novel aspect of the present effect in the microwave impedance is that it allows an experimental attack to be made on the question of the reflection of electrons from a rough surface. Let us imagine that the roughness is a small effect, i.e., that the probability p that an electron will be reflected specularly is nearly unity.

We can think of the surface roughness as being described by a function $f(x,y)=f(\varrho)$. This function, may be supposed to describe the height of the bumps on the surface, i.e., the surface is at $z=f(\varrho)$. We suppose that $\alpha f\ll 1$. Only the statistical properties of f are of importance (unless one has specially prepared the surface by, for example, growing it against a ruled grating). We thus take, for example,

$$\langle f(\rho'+\rho)f(\rho')\rangle=a^2\exp[-\rho^2/L^2], \tag{43}$$

where the angular brackets signify an average over the surface. Characteristically, one would suppose both a and L to be some few angstroms in magnitude, for a good surface.

If we impose the boundary condition $\psi[\varrho,f(\varrho)]=0$ and regard this as a small perturbation from the previous boundary condition, one obtains for the decay rate of an initial state ψ_i, by the golden rule,

$$\Gamma_i=\frac{2\pi}{\hbar}\sum_f|M_{fi}|^2\delta(\epsilon_f-\epsilon_i). \tag{44}$$

For the case of free electrons, we have

$$M_{fi}=\frac{\hbar^2}{2m}\int d^2\rho f(p)\frac{\partial\psi_f^*}{\partial z}\frac{\partial\psi_i}{\partial z}\bigg|_{z=0}, \tag{45}$$

and one may regard ψ_i and ψ_f as solutions of the problem with $f\equiv 0$. Thus

$$|M_{fi}|^2=A\left(\frac{\hbar^2}{2m}\right)^2\int d^2\rho\, e^{i(k_f-k_i)\rho}\times e^{-\rho^2/L^2}a^2\left(\frac{\partial\phi_f}{\partial z}\right)^2a^2\left|\frac{\partial\phi_i}{\partial z}\right|^2, \tag{46}$$

where the ϕ's are the z-dependent factor in the wave function.

Given our estimate of the magnitude of L, one sees that the final state can range quite fully over the Fermi surface. Such states generally involve appreciable momenta in the z direction and strictly speaking, the accuracy of the boundary condition and the derivation of formula (45) are open to question. We, however, take thc attitude that probably the factor $(\partial\phi_i/\partial z)^2$ is correct, and since essentially all of the magnetic-field dependence and dependence on quantum number resides in this wave function we can obtain a phenomenological formula for the surface scattering component of the lifetime. In view of the normalization of ϕ_n [see Eq.

(18)], we find

$$\Gamma_{\text{surface},m} \propto H, \tag{47}$$

i.e., independent of m and linearly dependent on field.

It is instructive to regard this result from a semiclassical point of view. An electron in a surface state is supposed to skip along the surface. At each bounce, it has the probability $1-p$ of being scattered through a large angle out of the original state. The quantity p was first introduced by Fuchs as a parameter giving the boundary condition to the Boltzmann equation.[19] We adhere to this notation in spite of the fact that Greene[20] has shown that one must distinguish between Fuch's parameter p and the actual specular reflection probability. The total time which an electron can expect to spend in such a state is thus

$$1/\Gamma = T/(1-p) \tag{48}$$

(assuming $1-p \ll 1$). The time T of a single jump may be calculated semiclassically to give

$$T_m = 2\int_0^{a_n/\alpha} \frac{dz}{v_z(z)} = \frac{4Ka_m^{1/2}}{v_x\alpha^2}. \tag{49}$$

The magnitude of T_m is typically 10^{-9} sec. The diffuse reflection probability $(1-p)$ depends on the angle at which the electron strikes the surface. Formula (46) applied to incident plane-wave electrons would seem to indicate that $(1-p) \propto a^2k_z^2$, i.e., $(1-p) \propto \theta^2$, where θ is the angle between the electron trajectory and the surface. However, this formula gives the rate at which a given area of surface scatters an electron; an electrons wave packet will illuminate a greater surface area at small angles. Taking this effect into account, we find

$$1-p \propto a^2k_F^2\theta_m, \tag{50}$$

where

$$\theta_m = \left.\frac{v_z}{v_x}\right|_{z=0} = \frac{\alpha(a_m)^{1/2}}{K}. \tag{51}$$

This evidently yields the same form as (47). The value of θ_m is typically $\theta_m \simeq 10^{-2}$. An approximate examination of the In data suggests that there $1-p \simeq 10^{-2}$. Other laws for the variation of $(1-p)$ can be considered. Greene *et al.*[21,22] have employed the rule $p=1$ for $\theta<\theta_0$, $p=0$ for $\theta>\theta_0$, for which there is no theoretical justification, except that it correctly makes for an increase in specular reflection at glancing angle incidence. Our analysis also bears out this fact.

We add a few remarks on the question of the boundary condition, $\psi(0)=0$, which we have employed. The problem is not too serious if we ignore the crystal lattice. However, it is not so obvious that the vanishing of the actual wave function close to the surface implies that the effective wave function also must vanish, since the actual wave function must contain short wavelength components. If, however, in the spirit of the orthogonalized plane wave (OPW) and pseudopotential approximations, one regards the rapidly varying part of the wave function as arising only in the core of the ions, then it again becomes plausible that the wave function should show the effects of the surface at least as far as a wavelength away.

We do not intend to convince anyone by these remarks. On the contrary, we would welcome alternative proposals for boundary conditions, as we believe the experiments could readily distinguish between various boundary effects.

V. FINAL REMARKS

In a subsequent paper,[12] we will present a calculation of line shapes and detailed numerical aspects showing how well the present theory agrees with the experimental data. Although there is no doubt that the theory is substantially correct, there are some remaining discrepancies, so that one could conceivably include some additional structure from other sources than magnetic surface levels.

The most likely such source is the skimming orbits, which Van Gelder has treated. Van Gelder[10] has pointed out that the matrix elements of the electric field between quantized skimming orbits can have some few oscillations, which could give rise to oscillations in the surface impedance. On the basis of some approximate calculations, we earlier had felt that these oscillations could not be very important; however, Van Gelder has made a careful study and has found that the oscillations do exist, but of unknown line shape and magnitude.

Even if there are no oscillations, one could well expect a relatively slow but observable nonresonant background variation of surface impedance with field,[23] both from the skimming and skipping orbits. We have not attempted to be careful about this variation. In any event, it would be interesting, though difficult, to pick out Van Gelder's oscillations from the evidently more pronounced surface state effects, and to calculate all sources of background variation.

Finally we wish to point out that for the case of superconducting metals there exist qualitatively similar surface states. Such magnetic-field induced surface states were predicted by Pincus[24] and have recently been observed by Koch *et al.*[25,26]

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[19] K. Fuchs, Proc. Cambridge Phil. Soc. **34**, 100 (1938).
[20] R. F. Greene, Phys. Rev. **141**, 687 (1966).
[21] R. F. Greene, Phys. Rev. **141**, 690 (1966).
[22] J. E. Parrott, Proc. Phys. Soc. (London) **85**, 1143 (1965).

[23] There are observed some broad and ill-defined variations in the impedance at fields on the order of 100 Oe in Cu. [See, for example, J. F. Koch, R. A. Stradling, and A. F. Kip, Phys. Rev. **133**, A240 (1964).]
[24] P. Pincus, Phys. Rev. **158**, 346 (1967).
[25] J. F. Koch and P. Pincus, Phys. Rev. Letters **19**, 1044 (1967).
[26] J. F. Koch and C. C. Kuo, Phys. Rev. **164**, 618 (1967).