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Cyclotron resonance revisited

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Résumé. — Les expériences de résonance cyclotron dans les métaux ont été effectuées dans le passé sur des signaux de faible niveau. Dans cet article, le comportement en fort signal est étudié théoriquement à la lumière des derniers développements de la dynamique non linéaire. Les conditions qui pourraient permettre l'observation d'effets spécifiquement non linéaires, y compris le chaos, sont dérivées.

Abstract. — Cyclotron resonance experiments in metals have in the past been performed at very low signal levels. In this paper the large signal behavior is studied theoretically in the light of modern developments in nonlinear dynamics. Conditions that should favor observation of specific nonlinear effects, including chaos, are derived.

An invitation to contribute to a volume paying homage to Jacques Friedel is both an honor and a challenge. Friedel's work has not only been of fundamental importance to solid state physics, but is also marked by exceptional elegance. So, ideally, one would like to contribute something of at least remotely similar profundity and grace. And if one has nothing very deep to say, then what one does have to say should at least have some polish. Hopefully this paper will live up to that modest expectation.

It is widely believed that one should not jeopardize pleasant memories of events by attempting to relive them years later. Quite possibly, cyclotron resonance in metals is a case in point: the visualization of complex features of the Fermi surface by means of the resonance experiments carried out a quarter century ago provided a great deal of aesthetic satisfaction to theorists and experimenters alike, and is still fondly remembered by solid state physicists « d'un certain âge ». My only excuse for tampering with these memories is the realization that some more information might have been squeezed out of this phenomenon at the time, if nonlinear dynamics had then enjoyed its present popularity, and if materials preparation had then been sufficiently advanced to make possible very long mean free paths for the metal electrons.

1. Brief review of cyclotron resonance in very small signal fields.
In the last ten or fifteen years certain universalities have emerged governing the behavior of wide classes of nonlinear resonant systems. Metal electrons in cyclotron resonance form one
such system. The nonlinearity here arises from the fact that the Fermi surface usually differs substantially from a sphere, the more so the closer it comes to the boundary of the first Brillouin zone. For large enough Fermi energy it actually cuts the zone boundaries (at right angles), spilling over into the neighboring zones and, so far as the motion of particles on the Fermi surface (FS) is concerned, the surface must be continued in periodic fashion from one reciprocal cell to the next (repeated zone scheme). Figure 1 shows the portion of FS in the first zone for a simple cubic lattice and for a large (but not too large) Fermi energy. It is clear that continuation of this surface according to the repeated zone scheme yields an open three-dimensional lattice (see, for example, Ref. [1]). If a magnetic field is applied along one of the axes, the resulting electron orbits are relatively simple. Thus two of the six « necks » will have cross sections normal to the field; electrons in these two necks carry out cyclotron motion of the familiar kind, and their orbits are called neck orbits. On the other hand, electrons in a plane normal to the field and at less than a certain critical distance from zone center are constrained by the Fermi surface to rotate around the field in a sense opposite to that of the neck orbits, as though they had a positive charge. These are therefore called hole orbits. Electron and hole orbits are separated by straight orbits which, in the parlance of nonlinear dynamics, are known as separatrices (see Fig. 1).

When the field is tilted at an angle to the crystal axis, other kinds of orbits will arise, not all of which will be closed, and some of which will not even be periodic. In this paper only the case of closed orbits is considered, since they suffice for a demonstration of the important nonlinearities. It should be noted that most of the contribution to the resonant response to a signal will come from so-called extremal orbits. These are orbits in regions of the FS for which the rate of change of frequency with the component of the wavenumber along the magnetic field, $k_H$, is least. For if $\nu$ is some fixed linewidth within which electrons can contribute to the response, the range of $k_H$ values of contributing electrons is given by $\nu = \partial \omega / \partial k_H \cdot \Delta k_H$; thus $\Delta k_H$ is largest where $\partial \omega / \partial k_H$ is smallest. It is easily verified from figure 1 that neck orbits at the zone boundary, and hole orbits lying in a reflection plane normal to the field and passing through the zone center qualify as extremal orbits.

Fig. 1. — Portion of fermi surface in the first B.Z. of a simple cubic lattice, for the case $E_F > 2 E_0$. Magnetic field along z-axis. Solid curves : electron orbits ; dotted curves : hole orbits ; S-S : portions of separatrices. The hole orbits are completed in neighboring zones.
2. The equations of motion.

Most of the advances in nonlinear particle dynamics have made use of action-angle variables. With a view to applying these advances to cyclotron resonance with a minimum of effort, that resonance will likewise be described in terms of these variables. But first, consider the motion in cartesian coordinates in \( k \)-space. In the absence of a radio frequency signal, that motion is integrable. If the FS in the repeated zone scheme is \( E(k) = E_f \), where \( E \) is periodic in the components of the wavevector \( k \), then the equations of motion are

\[
\mathbf{\dot{r}} = \frac{1}{\hbar} \nabla E \times \mathbf{H}.
\]

One integral is \( E = \text{constant} \); the other is \( k \cdot H = \text{constant} \). With the direction of the field \( H \) along the \( z \)-axis, equation (1) may be rewritten

\[
\dot{k}_x = \left( \frac{eH}{\hbar^2 c} \right) \frac{\partial E}{\partial k_y},
\]

\[
\dot{k}_y = -\left( \frac{eH}{\hbar^2 c} \right) \frac{\partial E}{\partial k_x},
\]

and the motion along \( H \) is irrelevant. These equations have Hamiltonian form, with \( k_x \) acting as position and \( k_y \) as momentum, and

\[
h = eHE(k)/\hbar^2 c
\]

acting as « Hamiltonian ». So we have a problem with one degree of freedom, which is always integrable. This is no longer the case when a uniform r.f. electric field, \( F \cos \omega t \), is applied in the \( y \)-direction. The right-hand side of equation (2b) then acquires the additional term \( (eF/\hbar) \cos \omega t \). This can be incorporated into the « Hamiltonian », which becomes

\[
h = -\left( \frac{eF}{\hbar} \right) k_x \cos \omega t + eHE(k)/\hbar^2 c.
\]

Because \( E \) is not a purely bilinear function of the components of \( k \), the solutions of the equations will display stochastic behavior. For arbitrarily small \( F \), this stochasticity is confined to certain correspondingly thin filaments in \( k \) space, but as \( F \) is increased beyond a certain threshold, global chaos sets in. Inasmuch as this scenario is common to nonlinear oscillators in general, it is appropriate to begin with a purely verbal description, prior to a direct attack on the equations. (Refs. [2, 3] should be consulted for more detail.)

3. The driven nonlinear oscillator.

Consider a system with only one degree of freedom. This is integrable in the absence of the drive, the energy being expressible as a function of an action variable, denoted by \( J \). The corresponding conjugate angle variable \( \theta \) increases linearly with time; the proportionality factor, the angular frequency, is the derivative of the energy with respect to the action variable. In the nonlinear case, that frequency is still a function of \( J \), and its derivative is a convenient measure of the strength of the nonlinearity. When a time dependent drive is turned on, the problem is no longer integrable. The drive contributes a term \( V \) to the Hamiltonian where \( V \) in general is a function of \( J \), \( \theta \), and \( t \). For simplicity, let that time dependence be cosinusoidal. Let this extra term in the Hamiltonian be expanded as a Fourier series in \( \theta \), with coefficients still functions of \( J \). Then, writing the time dependence in exponential form, one finds that the Fourier series has terms of the form \( \exp i (n \theta \pm \omega t) \), where \( n \) is any positive or negative integer or zero, and \( \omega \) is the driving frequency. In the absence of the drive, \( J \) is constant in time, equal to \( J_0 \), say. When the drive is turned on \( J \)
acquires a rate of change equal to minus the derivative of the Fourier series with respect to $\theta$. As a first approximation one may write $\theta = \omega (J_0) t$ everywhere in that differentiated Fourier series. Then most of the terms in the series are seen to oscillate more or less rapidly, with frequencies $n \omega (J_0) \pm \omega$, and contribute only a small amount of noise to the new $J$. However, terms with $n$ values such that

$$n \omega (J_0) \pm \omega = 0,$$

cause $J$ to change secularly, and therefore must be treated very carefully. In standard canonical perturbation theory, these so-called resonances appear as vanishing denominators in the perturbation series. It follows that perturbing terms with a potential for causing resonances must, from the outset, be included in the unperturbed part of the Hamiltonian, which then forms the starting point of so-called secular perturbation theory. Consider first the possibility of just one resonance, occurring for some particular value of $n$ (and, of course, also $-n$). Drop all other terms in the Fourier series, retaining only

$$v_n (J) \exp i (n \theta - \omega t) + v_{-n} (J) \exp - i (n \theta - \omega t)$$

which is added to the original unperturbed Hamiltonian $h_0 (J)$ to form a new « unperturbed » Hamiltonian. It is intuitively obvious that to solve the resulting « unperturbed » motion, it is advantageous to go into a rotating frame of reference in which $n \theta - \omega t$ is slowly varying [and exactly equal to zero in the crude approximation $\theta = \omega (J_0) t$]. Technically, this is accomplished by a time-dependent contact transformation to new action-angle variables. The new angle variable is $\hat{\theta} = n \theta - \omega t$; the new action variable is $\hat{J} = J - J_0$. Hopefully, unless the drive is large, $J$ will not stray far from the unperturbed resonant action given by $n \omega (J_0) = \omega$. If that is the case, the transformed Hamiltonian may be expanded in powers of $\hat{J}$, the term linear in $\hat{J}$ cancelling out because of the resonance condition. Also, the $v$'s are evaluated at $J = J_0$, since terms such as $\hat{J} \frac{dv}{dJ_0}$ would be higher order in the drive.

The resulting Hamiltonian, evaluated to second order in $\hat{J}$, has the form (apart from a constant),

$$\hat{h} = \frac{1}{2} \frac{d^2 h_0}{d\hat{J}_0^2} \hat{J}^2 + 2 \Re \{ v_n (J_0) \} \cos \hat{\theta}.$$

This represents a simple pendulum, with momentum variable $\hat{J}$ and angular displacement $\hat{\theta}$. The effective mass of the bobbin is

$$m^* = \left( \frac{d^2 h_0}{d\hat{J}_0^2} \right)^{-1}$$

and the force constant is $2 \Re v_n$. The frequency of infinitely small oscillations is $\nu (J_0) = [2 \Re v_n / m^*]^{1/2}$. Back in the laboratory system this means that the particle carries out oscillations of frequency $\tilde{\omega} = (\omega \pm \nu) / n$. As the amplitude increases, the frequency $\nu$ drops. Finally, at the boundary between librational and rotational motion (the separatrix), the frequency goes to zero. Thus, depending on initial conditions, the librational motion covers a band of frequencies of width $2 \nu (J_0) / n$. For initial conditions sufficiently remote from the stable point $\hat{J} = 0, \hat{\theta} = 0$, « rotation » occurs which in the present case means that $\theta$ steadily deviates from $\omega t$, so that the system has been pulled out of resonance completely. The motion of the representative pendulum may be described in terms of new action-angle variables $J', \theta'$ (not to be confused with, $\hat{J}, \hat{\theta}$, which now play the role of momentum and
displacement). The nonresonant terms of the drive, neglected so far, may now be reconsidered. Among them will usually be a pair that becomes resonant in terms of the new variables, and the process can now be repeated. Around each unperturbed orbit, specified by a fixed value \( J' \), again arises a new pendulum orbit under the influence of the new resonance. Here we simply state the final outcome of this hierarchical procedure: a certain neighborhood of the separatrix arising from inclusion of the primary resonance in the unperturbed Hamiltonian is filled with chaotic orbits. The width of that neighborhood is the larger, the greater the drive, but it vanishes only when the drive is zero. In practice it is much more convenient to study this problem by replacing the differential equations by difference equations relating \( J \) and \( \theta \) at the end of one period of the drive to their values at the beginning of the period. This requires solving the equations of motion over one cycle, and this can be done only approximately. However, injecting the prior knowledge that the motion near the « first level » separatrix is of the most interest makes it possible to solve the motion over one period sufficiently well to obtain reliable information from a consideration of the difference equations. In particular, results can be obtained for the width of the stochastic layer around the separatrix. (For more detail, see Ref. [2] p. 300, and Ref. [3], Sect. 3.2b).

For large enough drive, another kind of stochasticity, so-called global chaos, may come into play. So far we have assumed that there is only one primary resonance and have described its vicinity, treating the primarily nonresonant terms as a perturbation (albeit taking due note of any secular influence they may acquire as the result of the corrected motion). We must now allow for the possibility of several primary resonances in the driving terms: for different values of \( n \) the equation \( n \omega (J) - \omega = 0 \) will in general have different solutions for \( J \). If these resonant \( J \)-values are very different, the motion near any one of them will not be greatly influenced by the motion near another, as long as the drive is small enough. However, as we have seen, the drive produces a finite width of the resonances, the width being of order of the root of the driving field. Considering two resonances with indices \( n \) and \( m \) and action variables \( J_1 \) and \( J_2 \), respectively, these will begin to overlap (their separatrices will touch) when one of the four possible relations

\[
\frac{\omega + \nu}{n} (J_1) \approx \frac{\omega + \nu}{m} (J_2)
\]

is satisfied. (When the signal is periodic but not monochromatic, an even greater variety of overlap conditions will result. Then in the last equation the \( \omega \) on the two sides are replaced by \( p \omega \) and \( q \omega \), respectively, where \( p \) and \( q \) are integers.) Evidently such overlap must occur when the drive is large enough. The resonances then influence each other very strongly; the motion becomes chaotic, and « globally » chaotic if there are many resonance overlaps. Actually, the overlap criterion is usually too stringent: global chaos sets in at lower threshold values of the drive, but the criterion is sufficient for order of magnitude estimates.

In the study of global chaos, it is again convenient to replace the differential equations by difference equations relating action and angle variables from one cycle to the next. Only here we have no a priori reason to favor the region of the separatrix motion and there is no guide to the approximate integration of the motion over one cycle. For this reason, the monochromatic signal is conveniently replaced by a periodic sequence of kicks. The motion between kicks is of course integrable, and thus a so-called map, a pair of difference equations connecting successive \( J, \theta \) values, is obtained. For low enough drive, the « fixed point » of these difference equations is stable against small perturbations. At a threshold value of the drive, the fixed point becomes unstable, and possibly bifurcates into a two-cycle fixed point (i.e. a point for which the \( n \)th and \( n + 2 \)th iterates are equal, but different in value from the equal \( (n - 1) \)th and \( (n + 1) \)th iterates. All this for large \( n \). Eventually, at large enough drive, no order of fixed point is stable any more, and the system is chaotic.
Actually, this device of treating the perturbation as a series of kicks is quite appropriate in the classical cyclotron resonance set-up proposed by Azbel and Kaner. In that set-up the magnetic field is in the surface of the sample, and although the signal is sinusoidal, the electrons see it only when they pass through the skin depth. To achieve resonance, the phase of the field on each passage must be the same, modulo 2 π, and this will be the case if \( [\omega / \omega (J)] = n \), an integer. Thus, there are resonances at all \( J \) values satisfying \( \omega (J) = \omega / n \).

A discussion in terms of a map may be given, the effective drive being proportional to \( \tau \sum \cos [2 \pi n \omega / \omega (J)] \delta [t - 2 \pi n / \omega (J)] \), where \( \tau \) is the sojourn time in the skin depth. This will be almost the usual «kicked oscillator» map, except that the coefficients of successive \( \delta \)-functions are here equal only for resonant \( J \) values.


We are now ready to apply these principles to cyclotron resonance in metals. We shall treat only the simplest case of a simple cubic metal, with magnetic field directed along one of the axes. In tight binding approximation we may then write

\[
E(k) = E_0 (3 - \cos k_x a - \cos k_y a - \cos k_z a),
\]

where \( a \) is the lattice spacing. \( E_0 \) is chosen so that the \( E \) versus \( k \) relation at the very bottom of the band is the usual quadratic one; this requires \( E_0 = \hbar^2/ma^2 \). We concentrate on the Fermi surface \( E(k) = E_f \). Henceforth we set \( k_x a = x, k_y a = y, k_z a = z \). The magnetic field \( H \) is in the \( z \) direction, and we are concerned with the motion in a plane normal to this direction, at fixed \( k_z \), that is at fixed \( z \). Equations (2) in terms of the new variables are

\[
\dot{x} = \partial h / \partial y
\]

\[
\dot{y} = - \partial h / \partial x,
\]

where

\[
h = - (aeFx/\hbar \omega_c) \cos \omega t + E(x, y ; z)/E_0.
\]

Here \( \omega_c = eH/mc \) is the free-electron cyclotron frequency, time is measured in units of \( 1/\omega_c \), and the colon before the \( z \) indicates that \( z \) is to be considered fixed. For the present we pick out only the dominant frequency component from the Azbel-Kaner series of effective pulses. We begin by solving for the motion under the unperturbed Hamiltonian \( h_0 = E/E_0 \). Since \( x \) and \( y \) behave like position and momentum, respectively, the action variable for a periodic orbit at a particular \( z \) value is

\[
J = \oint y \, dx / 2 \pi = \int dx \, dy / 2 \pi
\]

\[
= \frac{1}{2 \pi} \cdot \text{Area of normal section of } (E/E_0) \text{ at } z.
\]

The corresponding natural frequency (in units of \( \omega_c \)) is

\[
\omega (J) = \frac{\partial h / \partial E}{\partial J / \partial E} \left( \frac{E}{E_0} \right)
\]

\[
= 2 \pi / (\text{rate of change of area with energy}).
\]
This is a well-known result [4]. In order to treat the driving field in this framework, we need to express $x$ in terms of $J$ and $\theta$. For this purpose we need to solve the Hamilton-Jacobi equation at Fermi energy:

$$\cos \left( \frac{\partial S}{\partial x} \right) + \cos x = \eta$$

where $\eta = 3 - \cos z - E_f/E_0$ (recall that $z$ is fixed). $S$ is the generating function, in terms of which

$$y = \frac{\partial S}{\partial x}$$

and

$$J = \oint dx \left( \frac{\partial S}{\partial x} \right) / 2 \pi ,$$

and the angle variable is

$$\theta = \frac{\partial S}{\partial J} ,$$

where in the last relation the integration constant in the solution for $S$ has been expressed in terms of $J$.

Fortunately, we never need $S$ itself, only one of its derivatives. We have

$$S = \int_0^x dx \arccos \left( \eta - \cos x \right) ,$$

where we have arbitrarily chosen the lower limit of integration to be zero; which is permissible for electron orbits, but not for hole orbits (see Fig. 1). We have

$$\theta = \frac{\partial S}{\partial J} = \frac{\partial S}{\partial \eta} \frac{\partial \eta}{\partial \theta}$$

and from equation (5), $\partial J/\partial \eta$ is the increment in $\partial S/\partial \eta$ in going around the closed orbit. Now

$$\frac{\partial S}{\partial \eta} = - \int_0^x dx \left[ 1 - (\eta - \cos x)^2 \right]^{-1/2} .$$

This integral can be expressed in terms of elliptic functions. The range of permissible values of $\eta = \cos x + \cos y$ is evidently $-2 < \eta < 2$. In the range $0 < \eta < 2$ we have electron orbits. It that range, the substitutions $z = \tan (x/2)$, $\cos x = (1 - z^2)/(1 + z^2)$ give

$$\frac{\partial S}{\partial \eta} = - \left[ \frac{2}{\sqrt{\eta (2 + \eta)}} \right] \int_0^{\tan (x/2)} dz \left[ \left( \eta^{-1} (2 - \eta) - z^2 \right) \right]^{-1/2}$$

$$= - F \left( \arcsin w ; \frac{1}{2} \sqrt{4 - \eta^2} \right)$$

where $w = 2 \tan (x/2)/\sqrt{(2 - \eta) [\eta + (2 + \eta) \tan^2 (x/2)]}$, and $F(\varphi, k)$ is the incomplete elliptic integral $\int_0^\varphi dx/[1 - k^2 \cos^2 x]^{1/2}$. The maximum permitted value of $x$ is such that $\tan^2 (x/2) = (2 - \eta) / \eta$. When that value is attained, $\arcsin w$ becomes $\pi/2$, and the elliptic integral is complete. By symmetry, four of these complete integrals are added up to give the increment in $\partial S/\partial \eta$ around the circuit. It follows that

$$\frac{\partial J}{\partial \eta} = - \left( 2/\pi \right) F(\pi/2 ; k) = - \left( 2/\pi \right) K(k)$$

(7c)
in the usual notation for elliptic integrals where \( k = \frac{1}{2} (4 - \eta^2)^{1/2} \). Thus we finally get
\[
\theta = (\pi/2 \, K) \, F (\arcsin w, k),
\]
and this is inverted to read
\[
w = sn \left( \frac{2 \, K \theta / \pi}{2 \, k' K} \right) = \left( \frac{\pi}{2 \, k' K} \right) \sum \frac{\sin (2 \, n - 1) \theta}{\sinh \left( (2 \, n - 1) \, \pi K'/2 \, K \right)} \tag{8a}
\]
where \( sn \) is one of the Jacobi elliptic functions and \( K'(k) = K(\sqrt{1 - k^2}) \). From this, and the definition of \( w \), the Fourier series for \( x \) can in principle be constructed and placed into the perturbing Hamiltonian \(-\left( a e F x / \omega_c \right) \cos \omega t\). In practice, the coefficients in the Fourier series must be found numerically. As they stand, they will be functions of \( \eta \), but they can be reexpressed in terms of \( J \) through the equation
\[
J = \frac{2}{\pi} \int_0^\eta d\eta' \, K[k(\eta')] \, K'[k(\eta')].
\]
Using some elliptic function identities, equation (8) may be written somewhat more simply as
\[
\tan \frac{x}{2} = \frac{1}{2} \sqrt{\eta (2 - \eta)} \, sn \left( \frac{2 \, K \theta / \pi}{2 \, k' K} \right) \, \frac{dr}{d \left( 2 \, K \theta / \pi \right)} \tag{9}
\]
The frequency is
\[
\omega(J) = \frac{2 \, \pi}{dJ/d(E_f/E_0)} = \frac{\pi^2}{K[k(\eta)]}.
\]
The separatrix between electron and hole orbits is at \( \eta = 0 \). At that point \( k = 1 \) and \( K \) is infinite. For \( \eta \) slightly larger than zero, the leading term in the expansion of \( K \) is \( -\ln (1 - k^2)^{1/2} \approx -\ln \eta \). So, as is normal for all nonlinear oscillators, the natural frequency goes to zero on the separatrix.

For hole orbits, \(-2 < \eta < 0\). A very similar procedure as for electrons, but allowing for the fact that the smallest value of \( |x| \) for any hole orbit must be greater than zero, gives the result
\[
\frac{\partial S}{\partial \eta} = -F \left[ w_h; \frac{1}{2} (4 - \eta^2)^{1/2} \right]
\]
with
\[
w_h = \frac{\arcsin 2}{(2 - |\eta|)(2 + |\eta| + |\eta| \tan^2(x/2))^{1/2}}
\]
and corresponding results for \( \partial J / \partial \eta \). At \( \eta = 0 \) the expressions for the electron and hole orbits join smoothly.

Finally, we write down the measure of nonlinearity:
\[
\frac{\partial \omega}{\partial J} = -2 \, \pi \, \frac{\partial}{\partial J} \frac{1}{\partial J / \partial \eta} \frac{\partial J}{\partial \eta} = -2 \, \pi \, \frac{\partial^2 J / \partial \eta^2}{(\partial J / \partial \eta)^3}.
\]
When written in the alternative form \(2 \pi \omega \partial \omega / \partial \eta\), this measure is seen to go to infinity as \((\eta \ln^2 \eta)^{-1}\) as the separatrix is approached. This is not surprising, since the orbits change qualitatively from one side of the separatrix to the other.

5. Local chaos in cyclotron resonance.

The perturbing Hamiltonian \(h^1 = -(aeFx/h\omega_c) \cos \omega t\) must now be expanded in a Fourier series in the angle variables. This can be done by solving equation (8) or equation (9) for \(x\), but the necessary integrals must presumably be evaluated numerically. In two limiting cases the leading order term can be written down analytically: when \(\eta = 2 - \delta\), with \(\delta\) small (corresponding to a very small Fermi sphere at the center of the zone), then using the Fourier series

\[ sn(2K\theta/\pi)/dn(2K\theta/\pi) = -\frac{\pi}{kk'} \Sigma(-)^n \sin [(2n-1)\theta]/\cosh [(2n-1)\pi K'/2K] \]

(with \(k' = \sqrt{1-k^2}\) in equation (9), and noting that \(k \approx \sqrt{\delta}\), \(K \approx \pi/2\), and \(k' = K(k') \approx -\ln \sqrt{\delta}\)), we find

\[ \tan(x/2) = -(2\sqrt{2}/\pi) \Sigma(-)^n \delta^{n-(1/2)} \sin(2n-1)\theta, \]

where only the leading orders in \(\delta\) have been kept in each harmonic. From this it is seen that the successive harmonics in the series for \(x\) itself are all odd, and their amplitudes likewise proceed as \(\delta^{n-1/2}\). The first two terms give

\[ \frac{x}{2} = (2\sqrt{2}/\pi) \delta^{1/2} \sin \theta - (2\sqrt{2}/\pi) \left(1 + \frac{1}{48}\right) \delta^{3/2} \sin 3\theta + \ldots \] (10)

The nonlinearity parameter \(d\omega/dJ\), near \(\eta = 2\), is easily found to be 1/2. The other limiting case relatively easy to discuss is \(\eta \approx 0\), in which the orbits are near the separatrix. Then \(k \approx 1\), \(k' \approx \sqrt{\eta}/2\), so that \(K \approx -\ln \sqrt{\eta}/2\), \(K' \approx \pi/2\) and

\[ \tan(x/2) = -\frac{\pi}{\ln \sqrt{\eta}/2} \Sigma(-)^n \sin(2n-1)\theta. \] (11)

This Fourier series is formally divergent, indicating that, as one would expect, all harmonics are present with about equal strength as the separatrix is approached, no matter how small the perturbation. Formally, the series in equation (11) essentially represents a periodic succession of \(\delta\)-functions of alternating sign located at \(\theta = \pm\) odd half-integral multiples of \(\pi/2\). This is confirmed by equation (9). In the limit \(\eta = 0\), the zeroes of the function \(dx\) [the infinities of \(\tan(x/2)\)] are located at these values of \(\theta\), so that \(x = \pm\pi\) there. For intermediate values of \(\theta\), \(x\) is very nearly either zero or \(2\pi\), if only because of the prefactor \(\sqrt{\eta}\). For small but finite \(\sqrt{\eta}\), \(x\) moves rapidly from one hyperbolic point to the next as \(\theta\) passes through odd half-integral multiples of \(\pi\).

In the general case, \(x\), like \(\tan(x/2)\) will be representable as a series of odd sine harmonics. Thus the Hamiltonian can be written

\[ h = h^0(J) + \Sigma \nu_n(J) [\sin(n \theta - \omega t) + \sin(n \theta + \omega t)], \] (12)

where the sum extends over odd positive integers only. (The coefficients are evaluated as functions of \(\eta\), but can be reexpressed in terms of \(J\).) In lowest order, \(\theta = \omega(J) t \approx \omega t\), and for the moment we keep only the resonance at \(n = 1\). Then we transform to a rotating...
coordinate system by means of generating function $G$ expressed in terms of the old action variable $J$, and the new angle variable $\hat{\theta}$:

$$G(J, \hat{\theta}) = -(J - J_0)(-\hat{\theta} + \omega t + \pi/2)$$

where $J_0$ is a constant to be determined. The relations for the new action $\hat{J}$ and the old angle $\theta$ are

$$\hat{J} = -\frac{\partial G}{\partial \hat{\theta}} = J - J_0, \quad \theta = -\frac{\partial G}{\partial J} = \omega t - \hat{\theta} - \frac{\pi}{2}$$

and the new Hamiltonian is

$$\hat{h}(\hat{J}, \hat{\theta}) = h(J, \theta) + \frac{\partial G}{\partial t} = -\omega \hat{J} + h^0(J_0 + \hat{J}) + v_1(J_0 + \hat{J}) \cos \hat{\theta}.$$ 

If $v_1$ is small enough, we expect $\hat{J}$ to differ only slightly from $J_0$, by something of order $v_1$. Therefore, $v_1$ in the foregoing expression may be replaced by its value at $J_0$, and $h^0$ may be expanded in powers of $\hat{J}$. Choosing $J_0$ such that $\hat{\theta}_0 = a_0 \delta_0$, we then have up to second order in $\hat{J}$,

$$\hat{h} \approx \frac{1}{2} \frac{\partial \omega (J_0)}{\partial J_0} \hat{J}^2 + v_1(J_0) \cos \hat{\theta}.$$ \hspace{1cm} (13)

(Actually, this is not quite correct. A more accurate determination of $J_0$ would involve the derivative of $v_1$, but this is normally dismissed as unimportant.) Equation (13) represents a pendulum, if the variables $\hat{J}, \hat{\theta}$ are reinterpreted as ordinary momentum and angular displacement. The effective mass is $m* = [\omega'(J_0)]^{-1}$. Explicitly, for the case of a small FS near the zone center we have, from equation (10),

$$v_1 = - (2 \sqrt{2aeF/\pi \hbar \omega_c}) \delta^{1/2}.$$ 

So in this case the small oscillation frequency around the equilibrium position is

$$\{ [2 \sqrt{2} \cdot a e F / \hbar \pi \omega_c] \cdot \partial \omega (J_0) / \partial J_0 \}^{1/2} \cdot \delta^{1/4},$$

and this is also the total width of the resonance, since the frequency is zero on the pendulum separatrix (no to be confused with the separatrix dividing the unperturbed electron and hole orbits).

Next, the primarily nonresonant terms must be introduced (see Sect. 3), i.e. the sum

$$\Sigma v_n \left\{ \sin [(n - 1) \omega t - n \hat{\theta} - n \pi/2] + \sin [(n + 1) \omega t - n \hat{\theta} - n \pi/2] \right\},$$

where in the summation of the first term the case $n = 1$ is omitted, since that term has already been fully taken into account. For simplicity we retain only the first of the two terms with $n = 3$, that is $v_3 \cos (3 \hat{\theta} - 2 \omega t)$, so we have the perturbed pendulum Hamiltonian

$$\hat{h} = \frac{1}{2} \omega'(J_0) \hat{J}^2 + v_1 \cos \hat{\theta} + v_3 \cos (3 \hat{\theta} - 2 \omega t).$$ \hspace{1cm} (14)

It is convenient to replace this Hamiltonian by an entirely equivalent holonomous one by introducing an extra pair of conjugate variables $-h$ and $t$, or, more conveniently, $\omega I = -h$ and $\Omega = \omega t$, and using the extended Hamiltonian

$$\bar{h} = \omega I + \frac{1}{2} \omega'(J_0) \hat{J}^2 - v_1 \cos \hat{\theta} - v_3 \sin (3 \hat{\theta} + 2 \Omega).$$ \hspace{1cm} (15a)
Since the equation of motion for \( \Omega \) gives \( \Omega = \omega t \), the motion of \( \dot{f} \) and \( \dot{\theta} \) under \( \hat{h} \) is exactly the same as their motion under \( \hat{h} \). Also \( \dot{f} = -\partial \hat{h}/\partial t \). Therefore to study the progress of \( \hat{h} \) under the perturbation we may study the progress of \( I \). Only the secular aspect of the change in \( I \) is of interest. We construct a map connecting \( I \) and the phase of the \( v_3 \) term at the beginning of one period of the slow separatrix motion to the beginning of the next period. (The largest secular changes in these quantities will occur near the separatrix.) We have \( I_{n+1} = I_n + \Delta I \), where

\[
\Delta I = -v_3 \int \cos [3 \dot{\theta}(t) - 2 \omega t + \chi_n] \, dt ,
\]

is the Melnikov-Arnold integral. Here \( \chi_n \) is the phase at the beginning of the \( n \)th period. In this expression the motion of \( \dot{\theta} \) may be approximated by its motion exactly on the separatrix, and there the limits of integration may be chosen to be \( \pm \infty \), because the period there is so long. The well-known result for that motion is (in the present notation)

\[
\dot{\theta} = 4 \arctan (\exp \nu t) ,
\]

where \( \nu \) is the small-oscillation frequency of the pendulum. The maximum contribution to the integral in equation (15) comes from the region in which \( d\dot{\theta}/dt \) comes as close as possible to \( 2 \omega \) (to see this, change variables from \( t \) to \( -t \)). The maximum rate of change of \( \dot{\theta} \) is at \( t = 0 \), where it is \( 2 \nu \). Since \( \dot{\theta} \) is odd in \( t \), we have

\[
\Delta I = -v_3 \cos \chi_n \int \cos [3 \dot{\theta}(t) - 2 \omega t] \, dt .
\]

Of course, \( 2 \nu \) is normally nowhere near as large as \( 2 \omega \), and therefore the integrand always oscillates. However, the region of «closest approach» \( t = 1/(2 \omega - 3 \nu) \approx 1/2 \omega \) produces a shift in the average level of the oscillation, and thus a secular change in \( \Delta I \). For the present case, this shift can be evaluated by the theory of residues; we quote the result from reference [2]. For the most likely case \( \omega \gg 3 \nu \), that result is

\[
\Delta I = I_{n+1} - I_n = - \left( \frac{v_3}{\nu} \right) \cdot [4 \pi (4 \omega/\nu)^5/5!] \cdot \exp \left( -\pi \omega/\nu \right) \cdot \cos \chi_n .
\]

As for the phase, \( \chi_{n+1} \), it is evidently equal to \( \chi_n - 2 \omega T \), where \( T \) is period of an orbit very close to the separatrix. On the pendulum separatrix (just as on the separatrix between hole and electron orbits) that period goes to zero. The standard result is (see Ref. [1], Sect. 1.3a)

\[
T = \nu^{-1} \ln \left| \frac{32}{w} \right| \]

where \( w \) is the difference between the separatrix energy and the potential energy \( v_1 \), relative to \( v_1 \). Because \( h = -\omega I \), that relative difference is

\[
w = -\frac{\omega I - v_1}{v_1} ,
\]

and goes to zero on the separatrix. Thus

\[
\chi_{n+1} = \chi_n + \left( \frac{2 \omega}{\nu} \right) \ln \left| \frac{32}{w_{n+1}} \right| .
\]
Strictly, the subscript of $w$ in this equation should be $n$, but, because of the structure of the difference relation for $w$, the choice $n + 1$ is more convenient. The location of fixed points is not affected by this. Using equation (17) to change variables from $I$ to $w$, we get

$$w_{n+1} = w_n - w_0 \cos \chi_n$$

where

$$w_0 = \frac{v_3}{v_1} \frac{\omega}{\nu} \left[ 4 \pi (4 \omega / \nu)^5 / 5! \right] \cdot \exp \left( - \frac{\pi \omega}{\nu} \right).$$

Stochastic motion results if the fixed points of recursion relations equations (18) and (19) are unstable. We consider here only period 1 fixed points. Their instability does not necessarily indicate chaos, since they may be replaced by stable period 2 fixed points, and so on. However, the instability condition for period 1 fixed points should give an indication of the order of magnitude of the system parameters needed for chaos. Period 1 fixed points are at $(2 \omega / \nu) \ln \left| 32/w_\infty \right| = 2 \pi \cdot \text{integer}, \ m$, giving $w_\infty = \pm 32 \exp \left( - \frac{\pi m \nu}{\omega} \right)$. Then from equation (19), $\chi_\infty = \pm \pi / 2$. A linear stability analysis shows that $\chi_\infty = - \pi / 2$ is always unstable, while $\chi_\infty = + \pi / 2$ is stable, provided $w_\infty > w_0 \omega / 2 \nu$. Always, $\nu \ll \omega$, so that this criterion is essentially

$$32 > \left( \frac{v_3}{v_1} \right) \left( \frac{\omega}{\nu} \right)^6 \left[ \frac{(8 \pi)^5}{120} \right] \cdot \exp \left( - \frac{\pi \omega}{\nu} \right).$$

6. Orders of magnitude.

From equation (10), $v_3/v_1$ is of order $\delta$, and $(8 \pi)^5 \approx 10^7$, so that, for stability

$$32 > 8.3 \times 10^4 \times \delta \times (\omega / \nu)^6 \times \exp \left( - \frac{\pi \omega}{\nu} \right).$$

$aeF/h$ is a characteristic frequency associated with the applied r.f. field amplitude $F$. For $a = 5$ Å, it amounts to about $F \times 10$ MHz/V/cm. $\omega$ is of order $\omega_c$, which for an ordinary electron mass is about $H \times 2.6$ MHz/Oe. Thus $\omega / \nu = 0.25 (H/F)$, with $H$ in oersteds and $F$ is V/cm. For $F$ of order of a V/cm, and $H$ of order 1 kOe, the exponential in equation (20) would be so small that observing the stochastic layer would be out of the question. Even near the hole-electron separatrix, where $\delta \omega I / \delta J_0$ is very large, and where $\delta$ could effectively be replaced by unity, prospects would not be good. If, on the other hand, the metal could support a field of, say, 10 V/cm, and the electron relaxation time were so long ($\gg 4 \times 10^{-9}$ s) that cyclotron resonance at 100 Oe would become feasible, the effect might be observed. Certainly it would be advantageous to look for conditions in which the electron-hole separatrix occurs at $ak_z \approx 0$ or else at $ak_z \approx \pm \pi$, where the density of orbits is high. For the model discussed here, this would require a ratio $E_f/E_0 \approx 2$ or else $E_f/E_0 \approx 4$.

In the presumably less interesting case of semiconductors, conditions for observations of the stochastic layer are more favorable, since much larger r.f. fields can be supported in that case.


As noted in section 3, in the classic geometry for cyclotron resonance, the signal seen by the electrons consists of a series of pulses which reinforce one another if $\omega (I) = \omega / p$, where $p$ is an integer. The signal field seen by the electrons is a function of position that can approximately be written

$$\bar{F}(X) = F \lambda \cos \omega t \delta (X),$$
where $X$ is the distance from the plane surface $X = 0$ of the sample, and $\lambda$ is the skin depth. From a well-known $\delta$-function identity this may be written

$$F(t) = F\lambda \cos \omega t \Sigma \delta (t - t_n)/|dX/dt_n|$$

$$= F\lambda \cos \omega t \Sigma \delta (t - t_n)/|a\delta E/\hbar\delta x|$$

$$= F\lambda \cos \omega t (\hbar/\omega) \Sigma \delta (t - t_n)/|\sin x|$$

for the present model. Here the $t_n \approx 2\pi n/\omega(J)$ are the instances at which the path traverses the skin depth. Writing the $\delta$-functions in the form $\omega(J) \delta[\omega(J) t - 2\pi n]$, gives for the perturbing term in the Hamiltonian

$$h^1 = \lambda \omega(J) \left( \frac{eAF}{\hbar\omega_e} \right) \left( \frac{\hbar}{aE_0} \right) \cos \omega t \left( \frac{x}{|\sin x|} \right) \Sigma \delta[\omega(J) t - 2\pi n] .$$

The sum is a periodic function of $\omega(J) t$ with period $2\pi$, and can be written as a Fourier series

$$\{1 + 2 \sum_{n=1}^{\infty} \cos[n\omega(J) t]\} .$$

Retaining only potentially resonant terms, we find

$$h^1 = B \left( \frac{x}{|\sin x|} \right) \sum_{n=1}^{\infty} \cos[n\omega(J) t - \omega t]$$

(22)

where $B = (\lambda/a)(eAF/\hbar\omega_e)[2\hbar\omega(J)/E_0]$. Furthermore, in equation (22), $\omega(J) t$ may be replaced by $\theta$, since between visits to the skin depth, the motion is unperturbed. We discuss equation (22) for the case of a small FS near zone center. Then to lowest order in $\delta$, we have $x/|\sin x| = \text{sgn}(\sin \theta)$, whose Fourier series is $\Sigma \sin[(2n-1)\theta]/(2n-1)$. If this series is placed into equation (22), and processed using sum and difference formulae, a series with unbounded coefficients results. This can be remedied by noting that the Fourier series for the periodic $\delta$ function should be cut off at an $n$ value equal to $1/\omega(J) \tau$, where $\tau$ is the time an electron spends in the skin depth. The final result is then

$$h^1 = B \sum_{r=1}^{N} \xi(r) \sin(r\theta - \omega t)$$

where

$$\xi(r) = \sum_{j=1}^{r} \frac{1}{j}$$

and $N$ is the largest integer in $1/\omega(J) \tau$.

The distance between the $r$th and $(r+1)$th primary resonance is thus

$$\left| \omega(J_{r+1}) - \omega(J_r) \right| = \left[ \frac{1}{r} - \frac{1}{(r+1)} \right] \omega = \frac{\omega}{r(r+1)}$$

and rapidly decreases with increasing $r$. It is largest for $r = 1$, when it is $\omega/2$. From the previous section we conclude that the widths of the resonances at $r = 1$ are $[B\delta\omega(J_1)/\delta J_1]^{1/2}$ and $[B\delta\omega(J_2)/\delta J_2]^{1/2}$, respectively. The condition for overlap of $r$ and $r + 1$, and therefore for chaos is thus

$$\frac{\omega}{r(r+1)} < B^{1/2}[\sqrt{\xi(r) \omega'(J_1)} + \sqrt{\xi(r+1) \omega'(J_2)}]$$

(23)

where $J_{1,2}$ are the roots of $\omega(J) = \omega$ and $\omega(J) = \omega/2$. 

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Aside from the question of whether $B$ can be large enough so that equation (23) can be satisfied, there is the question of the strength of the resonances at $\omega (J_1) = \omega$ and $\omega (J_2) = \omega / 2$. There is no doubt that at given $E_f$, two $k_z$ values can be found such that the subharmonic resonance occurs if the fundamental one does. It is necessary only to move $k_z$ close enough to the separatrix condition $\eta = 0$, where the frequency is zero. However, the bulk of the resonance response comes from places where $\delta \omega (J) / \delta z = 0$, which in the present model occurs only at the center of the zone, and at the zone boundary (if the Fermi surface extends that far). Therefore, in general, it will be necessary to work with orbits that are more sparse than the ones that are optimally dense. This is probably a more serious limitation on the observability of the overlap than the relation equation (23) as such. Nevertheless, we estimate the coefficient $B$. Taking $\lambda / a = 10^4$, $2 \omega (J) / E_0 \approx 10^{-4}$, and $aeF / \hbar \omega_c \approx 2.5 F / H$ as in section 3, we have $B \approx 1.6 \sqrt{F / H}$, with $F$ and $H$ in c.g.s. units. For $F = 10$ V and $H = 100$ Oe, we have $B \approx 0.02$. The left-hand side of equation (23) is of order $1 / r(r + 1)$. Choosing a value of $r$, such as $r = 3$, known to be experimentally accessible, and setting the $\xi$s equal to 1, we see that we need nonlinearity parameters of order two or three to produce overlap. This condition is not hard to satisfy, even at considerable distance from the separatrix.


Suppose that the sample is weakly inhomogeneous on a scale short compared with the cyclotron radius but long compared with the distance over which the semiclassical use of $E(k)$ as a Hamiltonian function is justified. Then different $k$-values will be coupled, and (in the absence of a signal) the effective Hamiltonian becomes $\Sigma E(k) + \Sigma \Sigma V(k ; k')$. Recall that the magnetic field singles out the z-direction, and that, since $k$ is confined to the FS, different values of $k_z$ give different energy functions labelled by $k_z$ (or $z = ak_z$). So the full effective Hamiltonian can be written $\Sigma E_z(J) + \Sigma \Sigma V_{zz'}(J, \theta ; J', \theta')$, where, for brevity, $J_z$ is replaced by $J$ and $J_{z'}$ by $J'$, etc. As a first approximation, one may restrict consideration to the motion for one particular pair of values $z$, $z'$ at a time, so one has a system with two degrees of freedom which is still relatively simple to discuss. $V(J, \theta ; J', \theta')$, can be written as a Fourier series $\Sigma_{nm} v_{zz'}(J, J', n m) \exp i(n \theta - m \theta')$. So-called primary coupling resonances can now arise at $n \omega (J) = m \omega (J')$. Close to the electron-hole separatrix discussed earlier, these resonances « pile up ». For, suppose that $J'$ is close to the separatrix action, so that $\omega (J')$ is small, equal to $\delta \omega$, say, and consider the case $m = 1$. Then successive values of $J$ that can enter into the resonance relation are given by solving $\omega (J) = \delta \omega / n$, and if $\delta \omega$ is small, the possible $J$ values are closely spaced. In fact, a stochastic layer arises surrounding the electron-hole separatrix. Unless $v$ is very small, that layer should be sufficiently broad to permit experimental investigation. One expects the resonance response under these conditions to become very noisy.

For a system with strictly two degrees of freedom, different resonance surfaces $n \omega (J) = m \omega (J')$ never intersect on a surface of constant unperturbed energy. Therefore motion from one pair of resonant values $J$, $J'$ to another (so-called Arnold diffusion) is difficult. The system just discussed is not strictly a two degree system, and therefore diffusion is possible, though slow. Presumably a detailed discussion of these more complicated aspects of nonlinearity in cyclotron resonance should await some experimental verification of the more obvious effects described above.

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