Landau diamagnetic response in metals as a Fermi surface effect

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It is demonstrated that the Landau diamagnetism of the free electron gas and a monovalent metal can be considered as a Fermi surface effect. Only a relatively small number of electron states close to the Fermi surface are diamagnetically active whereas the majority of the electron states inside the Fermi surface are diamagnetically inert. This partitioning of the occupied electron states is driven by the structure of Landau levels, around which one can introduce magnetic tubes in the reciprocal space. Completely filled magnetic tubes do not change their energy in an applied magnetic field, and only partially occupied magnetic tubes in the neighborhood of the Fermi surface exhibit a diamagnetic response. Using this partitioning of the occupied electron states we derive a general expression for the steady diamagnetic susceptibility, for calculation of which one needs to know the shape of the Fermi surface and the energy gradient on it. The method is applied to alkali metals, whose Fermi surfaces and energy gradients have been obtained from ab initio band structure calculations. It has been found that the Landau diamagnetic susceptibility is anisotropic depending on the direction of the applied magnetic field with respect to the Fermi surface. This effect is more pronounced for Li and Cs, whose Fermi surfaces show a noticeable deformation from the spherical shape. The method opens a new route for ab initio calculations of the Landau diamagnetism of metals or intermetallic compounds. In the case of free electron gas it is shown that this approach also fully describes the oscillatory de Haas - van Alphen part of the diamagnetic susceptibility. Small oscillations of the Fermi energy found in the model are caused by redistribution (inflow or outflow) of electrons from the equatorial region of the Fermi surface.

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I. INTRODUCTION

When an external magnetic field \( H \) is applied to a metallic solid it induces a closed (orbital) motion of the itinerant electrons resulting in a nonvanishing magnetization antiparallel to \( H \), which we call the Landau diamagnetism \([1]\). In general the Landau diamagnetic susceptibility \( \chi_L \) of solids is small and independent of the temperature. In addition to the steady diamagnetism, at very low temperatures and strong magnetic fields there are oscillatory dependencies (\( \chi_{dHvA} \)) due to the well-known de Haas - van Alphen effect \([2, 3]\).

Despite many efforts in the past, an effective method for \textit{ab initio} calculations of the Landau diamagnetism even within the single-electron paradigm has not been established \([4, 5]\). (As summarized in Ref. \([6]\): “The problem of the low field susceptibility of electrons is old, interesting, and, unfortunately, quite complicated”.) This is because the effect is very difficult from the technical point of view. Even the derivation of this effect for the free electron gas formulated by Landau \([1]\) is rather complex.

In his pioneer work Peierls \([8]\) was the first who extended the Landau treatment to the case of electron band energy law \( E(\vec{k}) \) within the tight binding approximation. (Alternative derivations were proposed by Wilson \([9]\), Hebborn and Sondheimer \([10]\), and recently by Briet \textit{et al.} \([11]\).) Peierls obtained an expression for the magnetic susceptibility consisting of three terms,

\[
\chi = \chi_1 + \chi_2 + \chi_3, \tag{1}
\]

where \( \chi_1 \) is the diamagnetic susceptibility analogous to that of isolated metal atoms, \( \chi_2 \) is a term which has no simple physical interpretation and the term \( \chi_3 \) at zero temperature is given by

\[
\chi_3 = -\frac{e^2}{48\pi^2\hbar^2c^2} \int \frac{\partial^2 E}{\partial k_x^2} \frac{\partial^2 E}{\partial k_y^2} \left( \frac{\partial^2 E}{\partial k_x \partial k_y} \right)^2 \frac{dS}{\nabla_k E}. \tag{2}
\]

Here the integration is taken over the Fermi surface. The term \( \chi_3 \) was considered as leading in the diamagnetism of conduction electrons \([8, 9, 12]\). For a simple band it reduces to the Landau-Peierls expression where in the equation for free electron susceptibility the electron mass is replaced by an effective mass \( m^* \). In principle, Eq. \( (2) \) suggests that the diamagnetic effect is due to the Fermi surface electron states. However, this statement can not be proved or considered rigorous because of the other contributions (like \( \chi_1, \chi_2 \) in Eq. \( (1) \)) \([8, 10, 12]\). In Ref. \([12]\) on the basis of the study of the density matrix in the magnetic field Wilson, presenting a refined derivation of Eq. \( (2) \), has found that some terms can not be explicitly evaluated and some are not expressible in terms of derivatives of the band energy. More reservations were added by Adams \([13]\) who claimed that the
Landau-Peierls susceptibility was not always the dominant contribution. Kjeldaa and Kohn [14] further suggested that the Landau-Peierls approximation is valid only in the limit of small electron density. This statement has been proven recently by Briet et al. [11] at the mathematical level of accuracy. They have also worked out all satellite terms which in general accompany Eq. (2) but disappear for small electron density.

The full quantum treatment of band diamagnetism in a real solid is a formidable task for a number of reasons. First, the electron motion in the direction of the magnetic field \( H \) differs from the motion in the perpendicular plane. In a solid on the other hand, we deal with the translational symmetry which equally holds in three dimensions. Therefore, on applying the magnetic field to a solid, its translational symmetry becomes broken, and accurate description of electron bands and even their classification within the former first Brillouin zone is impossible [6, 12]. In general in the magnetic field operators of crystal translation do not commute [6, 13]. An effective magnetic band Hamiltonian can be introduced only when the magnetic flux through any triangle formed by two successive translations is a rational number, when expressed in terms of flux quanta \( R \) [13]. These conditions can be satisfied only for some fields \( H \) called rational by defining a superlattice structure with a restored vector translation symmetry but a changed point group symmetry. Superlattice calculations are more demanding because the number of inequivalent atoms in the unit cell is several times larger and the first Brillouin zone is less symmetric. For an arbitrary magnetic field \( H \) the problem can be sorted out by approximating it with some close rational magnetic field \( H' \approx H \).

Second, although the magnetic coupling is much weaker than the electric interactions including the mean field, it is nevertheless responsible for a complete reconstruction of the energy spectrum. Strictly speaking, we have to calculate new energy values and find new state functions explicitly depending on the value of \( H \) and populate them accordingly starting from a state with lowest energy and finishing with states of highest energy \( (E_F^*) \). In this formulation the task has not been solved. Nevertheless, in the past many bright ideas and nontrivial theoretical studies have been invested in this problem [10, 21]. For example, one can start with a nearly free electron gas model assuming that the crystal periodic potential is weak [13, 20]. The other, more strict approach requires calculated electron bands \( E_n(k) \) in the absence of the magnetic field. The full magnetic Hamiltonian \( \hat{H}(\hat{\Pi}) \) is an operator depending on \( \hat{\Pi} = \hat{p} + e \hat{A}/c \), whose components do not commute. The operator \( \hat{H} \) is a matrix function of \( \hat{\Pi} \) with matrix elements depending on the magnetic field. When written in diagonal form it can be sufficient to approximate its diagonal elements by the effective Hamiltonian \( E_n(\hat{\Pi}) \) obtained by replacing \( h\hat{k} \) by the operator \( \hat{k} \) in the energy band function \( E_n(k) \). Pursuing this approach one can work out the explicit form of the effective Hamiltonian which does not couple different bands and gives the new energy levels. Although this approximation is considered good for isolated (non-degenerate) electron bands \( E_n \), the procedure is not rigorous. In particular, in its derivation some terms of the order of \( \hbar \omega / E_F \approx H^2 \) are omitted [22].

In the effective Hamiltonian treatment, the general effect of the magnetic field on electron bands \( \hat{H} \) is two-fold: 1) a gradual transformation of band parameters, 2) breaking up into a series of discrete states. The latter effect is apparently a manifestation of emerging Landau levels. In Ref. [6] the method reached the stage where in principle all necessary expressions have been derived for the calculation of \( \chi_L \) within a pseudopotential (orthogonalized plane wave) basis set. The core formalism however has turned out to be complicated again and the authors use smallness of pseudopotential to obtain estimates of \( \chi_L \) for real metals.

Diamagnetic response in a metal can be obtained by methods of quantum field theory [23, 24]. In Ref. [24] Fourier components of the induced current are calculated through the evolution of the single particle density matrix. The orbital susceptibility \( \chi_{d\sigma}(q, \omega) \) is found as a prefactor for \( q^2 \) \((q \text{ is the wave vector, } \omega \text{ is the field frequency})\). The Landau susceptibility \( \chi_L \) of free electrons is restored at \( \omega = 0 \) and \( q \to 0 \) as a result of cancellation of two terms which diverge as \( 1/q^2 \). In Ref. [25] and [24] a model of conduction electrons interacting with an electromagnetic field is considered. Of particular interest is the electron interaction via current-current potential arising from the exchange of transverse photons. When Dyson equations for complete two-point correlation functions are solved, the Landau susceptibility appears in the corresponding polarization operator [25]. The diamagnetic response comes from the states at the Fermi surface and in general contain terms with the derivatives of the density of states. For arbitrary dispersion however, such terms cannot be expressed as derivatives of the fermionic energy.

There is another approach to the problem based on the semiclassical treatment of the problem [2, 5, 26]. For example, it has proven to be highly effective in describing the de Haas - van Alphen oscillatory part of the magnetic susceptibility. Onsager [2] and Lifshitz [6] based on the semi-classical description of the movement of an electron in a magnetic field, showed that the change in \( 1/H \) is determined by extremal cross-sections of the Fermi surface in a plane normal to the magnetic field. A good historical and theoretical review of the effect is given in the book of Shoenberg [4]. Although the de Haas - van Alphen effect can be understood within the quantum theory [1, 24], the full quantum consideration is not extended to real crystals and in practice the description of the de Haas - van Alphen oscillations relies on the semiclassical theory [4].

From the semiclassical equations it follows that the component of \( \vec{k} \) (in the reciprocal space) parallel to \( \hat{H} \) and the electron energy \( E(\vec{k}) \) are both constants of the electron motion. Therefore, in \( k \)-space electrons move
along curves defined by the intersection of isoenergetic surfaces \((E(\vec{k}) = E_0)\) with planes perpendicular to the magnetic field, i.e. the scalar potential associated with the electron mean field in solid is unchanged. This is a reasonable assumption especially in the limit of small magnetic fields \(\hbar \omega / E_F \rightarrow 0\), which is the case of our consideration.

Electron movement along a closed trajectory (orbit) is a subject of the quantization conditions, Eqs. \(3\), \(6\) below. Although these quantization conditions are familiar from the semiclassical approach, they also can be derived from an equation-of-motion method \[26, 27\]. In this paper we will use this semiclassical approach to obtain a very simple expression for the Landau diamagnetic susceptibility of real metals at zero temperature (although the method can be extended to the case of a finite temperature). The elementary unit in our approach is a magnetic tube of finite width and length, containing a certain Landau level inside it. The occupied tubes whose electron states are completely filled are diamagnetically inert. Only partially occupied tubes located in the neighborhood of the Fermi surface contribute to the diamagnetic effect.

Two important consequences follow from our approach. First, at zero temperature the Landau diamagnetic susceptibility is caused by electron states near the Fermi energy. It is worth noting that the oscillatory part of the diamagnetic susceptibility giving rise to the de Haas - van Alphen effect has been related to the extremal orbits of the Fermi surface for many years. Here we accomplish the relation by ascribing also the steady diamagnetism to the Fermi surface states. The leading role of the Fermi surface in diamagnetism is in accord with the other properties of conduction electrons (for example, the Pauli paramagnetism) which also depend on the peculiarities of electron states near the Fermi energy \[28\]. Second, the presented method can be applied to \textit{ab initio} calculations of the steady diamagnetic susceptibility of real metals. In this paper we consider the simplest case, which is the case of monovalent alkali metals. We hope that in principle based on our simple expression \[35\] below, the Landau susceptibility can be added to a list of physics quantities available from first principles calculations.

We should also mention the important question of the Fermi energy \((E_F)\) change in the applied magnetic field. This is a very weak effect, considered first in detail by Kaganov et al. \[29\]. Some consequences of the phenomenon including weak oscillations of the density of states at the Fermi level are further discussed by Shoenberg in Ref. \[2\]. Below we will unveil a mechanism of this effect for the free electron gas case. It is caused by peculiarities of electron population in the equatorial region of the Fermi sphere. Depending on the applied magnetic field there can be a small inflow or outflow of electrons from the equatorial region to other states of the Fermi surface.

The paper is organized as follows. In Sec. \[II\] we demonstrate the method for the free electron gas. There, we introduce a central object for our method – a magnetic tube and consider its properties. The most important one is that the tube whose electron states are completely occupied does not contribute to magnetic response. Then we select diamagnetically active tubes near the Fermi surface and calculate their steady diamagnetic susceptibility \(\chi_L\). The oscillatory (de Haas-van Alphen) contribution \((\chi_{AHV})\) is considered in section \[III\]. Here the analytical calculations become more involved although the general physical picture remains clear and transparent. The description of the steady diamagnetism of real metals, Sec. \[III\] follows the same lines. As in the case of the Fermi gas, we first consider magnetic tubes of real systems and then calculate their steady diamagnetic response. The application of the method for calculations of \(\chi_L\) for various magnetic field directions is given in Sec. \[V\]. Finally, in Sec. \[V\] we summarize main conclusions.

II. FREE ELECTRON GAS

A. Magnetic tubes in \(\vec{k}\)-space

In an external magnetic field \(\vec{H}\) directing along the \(z\)-axis, the energy of the free electron is given by

\[
E = \hbar \omega \left( n + \frac{1}{2} \right) + \frac{\hbar^2 k_z^2}{2m},
\]

where \(n\) is integer (numbering the Landau levels), \(k_z\) is the \(z\)-component of the wave vector \(\vec{k}\), and the cyclotron frequency

\[
\omega = \frac{eH}{mc}.
\]

Here \(m\) and \(e\) are the electron mass and charge; \(c\) is the speed of light. In correspondence with Eq. \[3\] the energy of electron is presented by two contributions, the contribution \(E_\parallel\) from the movement in the plane, perpendicular to \(\vec{H}\) (i.e. in the plane \((k_x, k_y)\)) and the contribution \(E_z = \hbar^2 k_z^2/2m\) from the movement parallel to \(\vec{H}\) (i.e. along the \(z\)-axis). In the following we consider only the component \(E_\parallel\), because the parallel component \(E_\parallel = E_z\) is unchanged in the magnetic field.

Although Eq. \[3\] is obtained within the fully quantum treatment, below we will follow the wide used semiclassical representation of electron orbits in the real and momentum \((\hbar \vec{k})\) space \[2, 3, 26\], which gives exactly the same energy spectrum. The electron movement in the magnetic field in the \((x, y)\) plane along the radial direction \(q = r\) or along the \(q = x\) or \(y\)-axis (depending on the choice of the vector potential gauge) is described by a quantized orbit,

\[
\int p_q \, dq = 2\pi \hbar \left( n + \gamma \right),
\]
which corresponds to the \( n \)-th Landau level with the factor \( \gamma \). In Wentzel, Kramers, Brillouin’ (WKB) method for non-singular integrable orbits \( \gamma = \mu/2 \) \textbf{26}, where \( \mu \) is the Maslov index. In our case \( \mu = 2 \) and \( \gamma = 1/2 \) \textbf{26}. In the \((k_x, k_y)\) momentum plane the electron orbit defined by Eq. \textbf{5} is visualized as a circle whose area \( A_n \) is given by

\[
A_n = \frac{2\pi e H}{c \hbar} (n + \frac{1}{2}), \tag{6}
\]

and its energy is

\[
E_{\perp, n} = \hbar \omega \left( n + \frac{1}{2} \right), \tag{7}
\]

It is well known that the average density of electron states in the \( \vec{k} \)-space remains the same as without magnetic field. To understand better the reconstruction of the electron structure in the magnetic field \( H \), we select in the \( \vec{k} \)-space a tube, whose number of electron states and the energy of all states do not change in the presence of \( H \), Fig.\textbf{1}. For that we consider auxiliary electron orbits of the area

\[
A_n^{\text{aux}} = \frac{2\pi e H}{c \hbar} n \tag{8}
\]

with energies

\[
E_n^{\text{aux}} = \hbar \omega n. \tag{9}
\]

Note the the \( n \)-th Landau orbit defined by Eqs. \textbf{6}, \textbf{7} is situated between the auxiliary orbits \( n \) and \( n+1 \), Fig.\textbf{1} left panel, and its energy \( E_n \) lies between \( E_n^{\text{aux}} \) and \( E_{n+1}^{\text{aux}} \).

Below we show that the number of electron states with energies \( E_n^{\text{aux}} \leq E \leq E_{n+1}^{\text{aux}} \) without field equals the number of electron states condensing on the \( n \)-th Landau level in the presence of the field. The same holds for their total energies.

For that we calculate the density of electron states \( N_\perp \) in the \((k_x, k_y)\)-plane,

\[
N_\perp = \frac{dN_\perp}{dE_\perp} = \frac{2m L_x L_y}{\hbar^2} \frac{\omega}{2\pi}, \tag{10}
\]

and notice that \( N_\perp \) is independent of the energy \( E_\perp \).

(Here \( L_x \) and \( L_y \) are distances of the free electron gas box in \( x \) and \( y \) directions, respectively.) Using \textbf{10}, we find the number of electron states in the \( n \)-th tube without field, i.e. in the energy range \( E_n^{\text{aux}} \leq E \leq E_{n+1}^{\text{aux}} \),

\[
\Delta N_n (H = 0) = \int_{E_n^{\text{aux}}}^{E_{n+1}^{\text{aux}}} N_\perp dE_\perp = N_\perp \hbar \omega = 2N_p. \tag{11a}
\]

Here \( N_p \) is the spatial degeneracy of the Landau levels (without spin polarization),

\[
N_p = \frac{L_y m \omega}{2\pi \hbar} L_x = \frac{L_x L_y e H}{2\pi c \hbar}. \tag{11b}
\]

Calculating the total energy of these states without field,

\[
E_n (H = 0) = \int_{E_n^{\text{aux}}}^{E_{n+1}^{\text{aux}}} N_\perp E_\perp dE_\perp = \frac{1}{2} \left[ (E_n^{\text{aux}})^2 - (E_{n+1}^{\text{aux}})^2 \right] N_\perp = E_n E_{n+1} \Delta N_n, \tag{11c}
\]

we find that it coincides with the energy of all electron tube states condensed on the \( n \)-th Landau level in the presence of the field. Thus, we have proven that

\[
\Delta N_n (H = 0) = \Delta N_n (H \neq 0) = 2N_p, \tag{12a}
\]

\[
E_n (H = 0) = E_n (H \neq 0) = \hbar \omega \left( n + \frac{1}{2} \right) 2N_p. \tag{12b}
\]

**FIG. 1:** Magnetic tube and the corresponding Landau level. On the left: the \((k_x, k_y)\) tube section and the \( n \)-th Landau orbit (the solid circle with the in-plane energy \( E_n \)). The dashed circles correspond to the auxiliary orbits with energies \( E_n^{\text{aux}} \) and \( E_{n+1}^{\text{aux}} \). On the right: the tube in the \( \vec{k} \)-space. Without magnetic field electron states are distributed throughout the whole tube, in the presence – only on the Landau orbit in the middle.

So far our consideration has been limited by the \((k_x, k_y)\) plane. However, since Eqs. \textbf{12a} and \textbf{12b} hold for any \( k_z \)-component, they are fulfilled for the whole tube, \( n \), Fig.\textbf{1}. In other words, if without field, a tube contains electron states which satisfy the inequality \( E_n^{\text{aux}} \leq E_\perp \leq E_{n+1}^{\text{aux}} \) for all \( k_z \) in the range \( k_z^{(1)} \leq k_z \leq k_z^{(2)} \), then in the presence of the magnetic field all these states condense on the \( n \)-th Landau level, that is, \( E_\perp (k_z) = E_{n+1} (k_z) \) throughout the \( k_z \) region, Fig.\textbf{1}. Furthermore, the total electron energy of the tube remains unchanged, which bears some resemblance to the Bohr - van Leeuwen theorem in classical physics.

The upper \( k_z^{(2)} \) and lower \( k_z^{(1)} \) boundary of the tube can be taken arbitrary. For practical reason for each tube \( n \) we define the \( k_z \)-boundaries by its intersection with the Fermi surface, Fig.\textbf{2}. We then obtain two \( n \)-th tubes: the first tube (denoted by 1 in the inset of Fig.\textbf{2}) lies entirely inside the Fermi surface and being completely occupied does not exhibit diamagnetism. The second tube (denoted by 2 in the inset of Fig.\textbf{2}) being only partially filled below the Fermi surface, results in a diamagnetic response. We consider this effect in the following sections.

It is also worth noting that the \( \vec{k} \)-space partitioning depends on the value of the magnetic field, since \( \omega \sim H \), and the tube boundaries are defined by \( \omega \), Eq. \textbf{9}.
B. Diamagnetically active electron states

Consider the Fermi surface and define necessary magnetic tubes parallel to the $z$-axis (in the direction of the magnetic field $H$), Fig. 2 as discussed in Sec. II A. Boundary conditions defined by in-plane circular orbits, Eq. (8), specify a set of concentric cylindrical surfaces, which intersect the Fermi surface in circles perpendicular to the $z$-axis. We then draw the planes of the circles and use them to construct a set of tubes, limited by the planes and the cylindrical surfaces, which lie inside the Fermi sphere. The $(k_x, k_z)$ cross-section of these tubes is schematically shown in Fig. 2. The fully occupied tubes are shown as dashed area. The electron states of the completely filled tubes do not change their energy in a magnetic field. Therefore, the whole effect is due to the states lying in the partially occupied tubes. Their cross-sections in the $(k_x, k_z)$-plane look like a chain of triangles, Fig. 2.

Consider a typical partially occupied tube, whose triangle cross-section in the $(k_x, k_z)$-plane is shown in Fig. 3. We denote two legs of the triangle by $\Delta k_{\perp}$ and $\Delta k_z$. Taking into account that the area of the $(k_x, k_y)$ cross-section of the $n$-th tube is $\Delta A = A_{n+1}^{ax} - A_n^{ax} = 2\pi eH/\hbar c$, and that $\hbar \omega \ll E_F$, we obtain

$$\Delta k_{\perp} = \frac{\Delta A}{2\pi k_{\perp}} = \frac{m\omega}{\hbar k_F \sin \Theta}. \tag{13}$$

(Here $\Theta$ is the polar angle, Figs. 2, 3). Therefore, the narrow surface region of the partially occupied tube is defined by the wave vector quantity $\Delta k_F$ shown in Figs. 2 and 3,

$$\Delta k_F = \Delta k_{\perp} \sin \Theta = \frac{m\omega}{\hbar k_F}. \tag{14}$$

It is remarkable that $\Delta k_F$ is independent of $\Theta$. Therefore, the radius $k_F - \Delta k_F$ determines an auxiliary internal sphere in the $\vec{k}$-space, which can be used for drawing the step-wise line shown in Figs. 2 and 3 separating the fully occupied tubes from the partially occupied ones.

For the angle $\Delta \Theta$ shown in Fig. 3 we obtain

$$\Delta \Theta = \frac{\Delta k_F}{k_F} \frac{1}{\cos \Theta \sin \Theta}. \tag{15}$$

Notice, that Eq. (13) has a singularity at $\Theta = 0$, and Eq. (15) at $\Theta = 0$ and $\pi/2$. Therefore, the polar and equatorial region of the Fermi sphere should be considered more attentively, see Sec. III D below.

Now we find the number of active electron states in the partially occupied tubes,

$$N = \sum_{n=1}^{M} \Delta N_n, \tag{16}$$

where $\Delta N_n \equiv \Delta N(k_z, k_x + \Delta k_z)$ is the number of the electron states in the $n$-th partially filled tube, whose $k_z$-component lies between $k_{z,n} \equiv k_z$ and $k_{z,n+1} \equiv k_z + \Delta k_z$. Using the infinitesimal property of the $(k_x, k_y)$ cross-section we find

$$\Delta N = 2v\Delta V_k = 2\pi v \frac{eH}{\hbar c} \Delta k_z, \tag{17}$$

where $\Delta V_k$ is the volume of the partially occupied tube in $k$-space and $v = V/(2\pi)^3$ (In the general case $\Delta N$ is calculated in Appendix A).

Notice that from Eq. (17) it follows that $\Delta N/\Delta k_z = \text{const}$. Furthermore, $\Delta N/\Delta k_z$ is independent of $m$, which will be fully appreciated in the general case considered below in Sec. III B. Since for usual magnetic fields $\Delta k_F, \Delta k_z \ll k_F$, in Eq. (10) we can substitute the summation with the integration,

$$N = \int dN = \int_{k_z}^{k_{z,\text{max}}} \left( \frac{\Delta N}{\Delta k_z} \right) dk_z. \tag{18}$$

For the free electron gas $k_{z,\text{max}}^\text{min} = -k_F, k_{z,\text{max}}^\text{max} = k_F$. Using (17), (18) we arrive at

$$N = 2\pi v \frac{eH}{\hbar c} \ell_z, \tag{19}$$

where $\ell_z \equiv k_{z,\text{max}}^\text{max} - k_{z,\text{min}}^\text{min} = 2k_F$ is defined exclusively by the projection of the Fermi surface along the direction of the applied magnetic field. Therefore, $N \sim H$. Since the perturbation energy for each electron state can be estimated with $\hbar^2 k_F \Delta k_F / m = \hbar \omega \sim H$, the total energy change in the magnetic field $\sim H^2$, which leads to the constant magnetic susceptibility $\chi$. (The rigorous computation of $\chi$ is given in the next section.)

C. Steady diamagnetic susceptibility

A remarkable property of partially occupied tubes near the Fermi surface is that application of a magnetic field does not lead to electron transitions between different
Eq. (21) leads to an important consequence. The energy of the highest occupied electron level coincides with $E_F$ and the wave vector $k_F$ lies on the Fermi surface even in the applied magnetic field $H \neq 0$. Since the conclusion holds for all partially occupied tubes (with the exception of few equatorial tubes), the highest energy of the occupied electron states $-E_F$ is conserved as the Fermi energy for all tubes and there are no electron transitions between tubes. (The exceptional case of the equatorial region is considered later in Sec. [11D]. Therefore, in the following we can calculate the energy change for each tube separately.

Keeping in mind that in the magnetic field there are two energy contributions: $E_{\perp}$ in the $(k_x, k_y)$ plane and $E_z$ at $k_z$, we obtain for the energy change of the partially filled tube $n$,

$$\Delta E = \Delta E_{L=0} + \Delta E_{L=1} - \Delta E_{L=0}. \tag{22}$$

The notation $\Delta$ on the right hand side is an indication that the corresponding energy refers to the partially filled tube $n$, whose $k_z$ values are in the range from $k_{z,n}$ to $k_{z,n+1} = k_{z,n} + \Delta k_z$, Fig. 2, 3. The quantities $\Delta E_{L=0}$, $\Delta E_{L=1}$, and $\Delta E_{L=0}$ therefore refer to energy components of the tube with ($H \neq 0$) and without ($H = 0$) magnetic field.

The detailed simple calculations of all components are performed in Appendix B (with respect to the energy $E_{\text{ref}}$, Fig. 4). As a result, we get

$$\frac{\Delta E_{L=0}}{\Delta N} = \frac{2}{3} \frac{k_F^2}{m} \Delta k_F = \frac{2}{3} \frac{\hbar}{m} \omega \tag{23}$$

for the energy components without magnetic field and

$$\frac{\Delta E_{L=1}}{\Delta N} = \frac{1}{2} \frac{\hbar}{m} \omega, \tag{24a}$$

$$\frac{\Delta E_{L=1}}{\Delta N} = \frac{1}{4} \frac{\hbar}{m} \omega \tag{24b}$$

in the applied magnetic field. In fact, taking into account that $\Delta N$ stands for the number of electrons, the right hand sides of Eqs. (23), (24a) and (24b) represent average energy values independent on the tube under consideration. The substitutions of (23), (24a) and (24b) in (22) yields

$$\Delta E = \frac{1}{12} \hbar \omega \Delta N > 0. \tag{25}$$

Note that Eq. (25) refers to any partially filled tube. Therefore, making the summation over all tubes and using Eq. (19) with $l_z = 2k_F$, we find

$$E = \frac{1}{12} \hbar \omega N = \frac{1}{3} \pi v k_F \frac{e^2 H^2}{m c^2}. \tag{26}$$

For the magnetic susceptibility $\chi$ we finally have:

$$\chi = -\frac{d^2 E(H)}{dH^2} = -\frac{2}{3} \pi v k_F \frac{e^2}{m c^2} = -\frac{e^2 k_F V}{12\pi^2 mc^2}. \tag{27}$$
This is the celebrated expression, obtained by Landau for the diamagnetic susceptibility of the free electron gas.

D. Oscillatory (equatorial) diamagnetic susceptibility

Earlier (Sec. III C) we have obtained the diamagnetic effect based of calculations of the energy of active electrons in the partially occupied tube of general form. Deviations from the general situation are possible for boundary cases, which are the polar region \((\Theta = 0)\) with the Landau level \(n = 0\), and the equatorial region \((\Theta = \pi/2)\). In Appendix C we analyse the polar region and show that it complies with the general case. In the equatorial region however the situation is very different.

The problem is that the step-wise line shown in Fig. 2 can terminate at the equatorial point with \(\Theta = \pi/2\) in any place with \(k_{\perp,e} \leq k_F\), and the equatorial point does not necessarily lie on the internal sphere of the radius \(k_{\perp,e} \leq k_F\), which is the case for all other tubes. This equatorial tube is occupied if \(h > \Delta k_F/2\) \((r > 1/2)\), and empty if \(h < \Delta k_F/2\) \((r < 1/2)\).

For the first equatorial tube we define the quantity \(\Delta k_{\perp,e,1} = k_F - k_{\perp,e}\), Fig. 5, for which we shall also use a short notation \(h = \Delta k_{\perp,e,1}\). The subscripts \(e,1\) and \(e,2\) refer to the first and second equatorial tube, respectively. As discussed above, \(h\) ranges from 0 to \(\Delta k_F\). Consider the important dimensionless parameter

\[
r = \frac{h}{\Delta k_F}.\tag{28}
\]

Clearly, \(0 < r < 1\). Note, that by varying \(H\) we change the structure of all magnetic tubes, and, consecutively the parameter \(r\), defined by the geometry of the last tube. Therefore, \(r\) implicitly depends on \(H\). It can be shown that in a first approximation \(r\) is proportional to \(1/H\).

For \(\Delta k_{\perp,e,1}\) we obtain

\[
\Delta k_{\perp,e,1} = \sqrt{2k_F h} = \sqrt{2r eH/\hbar c}.\tag{29}
\]

Calculating the number of states in the first equatorial tube without magnetic field, we find

\[
\frac{\Delta N_{e,1}^{H=0}}{\Delta \Theta_{e,1}} = \frac{4r}{3} \frac{N}{\Delta \Theta}.\tag{30}
\]

Based on the analysis of the Cornu spiral sum, Pippard estimated that the relative weight of the extremal region should be \(\Delta N_{e,1}/N \sim \sqrt{H}\) (Eq. (33) of Ref. [30]). This conclusion is in agreement with Eq. (30) since \(\Delta N_{e,1}^{H=0}/N \sim \Delta k_{\perp,e,1} \sim \sqrt{\omega} \sim \sqrt{H}\).

Notice that already in obtaining \(\Delta N_{e,1}^{H=0}\) we have a deviation from the general case, Eq. (30), and

\[
\frac{\Delta N_{e,1}^{H=0}}{\Delta \Theta_{e,1}} = \frac{4r}{3} \frac{N}{\Delta \Theta}.\tag{31}
\]

Here \(\Delta \Theta_{e,1}\) defines the angular size of the first equatorial triangle in the \((k_r, k_z)\)-cross-section. Deviations are also present for the transverse and \(z\)-energy contributions.

Now we consider the situation in the magnetic field \(H \neq 0\). We start as in Sec. III C with finding the wave vector \(\delta k_z\) of the highest occupied electron state along the \(z\)-axis under assumption that all electrons belonging to the first equatorial tube do not leave it. By means of Eq. (28) we get

\[
\delta k_{z,e,1} = \frac{2}{3} \frac{r}{r} \Delta k_{z,e,1}.\tag{32}
\]

Now however the energy of the highest occupied state in general differs from \(E_F\), and therefore from the energy of the highest occupied states in other tubes, Eq. (21). Below we consider the situation for two different cases: \(0 \leq r < 1/2\) \((case\ a)\) and \(1/2 \leq r < 1\ \(case\ b)\).

In case a the energy of the Landau level of the first equatorial tube \(E_n - E_{ref} = h\omega/2\), Fig. 4, is higher than \(E_F\) even at \(k_z = 0\). Therefore, all electrons from this tube move to other tubes where they occupy free states above \(E_F\). As a result, a small rise in \(E_{ref}\) should occur, but since \(\Delta N_{e,1} \ll N\), it is of the order of \(h\omega \Delta N_{e,1}/N \ll h\omega\). Since \(E_F - E_{ref} \ll h\omega\), the energy of the promoted electrons is \(\Delta E_{H \neq 0} = r h\omega\) (with respect to \(E_{ref}^{e,1}\)).
in the equatorial region is shown in Fig. 6. The change of the number of electrons from all regular tubes move to the equatorial tube. The dashed line stands for the contribution from the first equatorial tube, solid line – from the first and second equatorial tubes. The same plot (in units of $(eH/hc)^{3/2}h^2/mk_F$) describes a small oscillatory dependence of the Fermi energy (chemical potential).

In case $b$ the Landau level at $k_z = 0$ lies below $E_F$ and in the magnetic field it becomes partially occupied by electrons with $k_z > 0$. The maximal $z$-wave vector $\delta k_F^z$ of the 1D electron state lying on the Fermi sphere can be found by requiring its energy to be equal to $E_F$,

$$\delta k_F^z = \sqrt{2k_F \left( h - \frac{1}{2}\Delta k_F \right)} = \Delta k_{z,e,1} \sqrt{\frac{r - \frac{1}{2}}{r}}. \quad (33)$$

The number of the occupied electron states in the tube, $\Delta N_{e,1}^{H\neq0,b}$, is determined by

$$\frac{\Delta N_{e,1}^{H\neq0,b}}{\Delta N_{e,1}^{H=0}} = \frac{3}{2r^{3/2}} \sqrt{r - \frac{1}{2}}. \quad (34)$$

The condition $\Delta N_{e,1}^{H=0} > \Delta N_{e,1}^{H\neq0,b}$ in terms of $r$ means $1/2 < r < \sqrt{3}\sin\pi/9$, while $\Delta N_{e,1}^{H=0} \leq \Delta N_{e,1}^{H\neq0,b}$ results in $\sqrt{3}\sin\pi/9 \leq r < 1$. Therefore, if $1/2 < r < 0.529$, electrons from the first equatorial tube partially move to other (regular) tubes as happens in case $a$. For $0.529 \leq r < 1$ the opposite happens, that is a small number of electrons from all regular tubes move to the equatorial tube. The change of the number of electrons in the equatorial region is shown in Fig. 6.

To single out the irregular contribution explicitly, we rewrite it in the following form,

$$E = E_L + \Delta E_{eq}. \quad (35)$$

Here $E_L$ is the diamagnetic (regular) contribution, Eq. (26), and $\Delta E_{eq}$ stands for the irregular term from the equatorial region. If only the first equatorial tube is accounted for, then $\Delta E_{eq} = \Delta E_{eq,1}$, where

$$\Delta E_{eq,1} = \Delta E_{\perp,e,1}^{H\neq0} + \Delta E_{\parallel,e,1}^{H\neq0} - \Delta E_{e,1}^{H=0} + \Delta E_{pr,1} - \Delta E_{corr,1}. \quad (36)$$

Here $\Delta E_{pr,1}$ is the energy of the promoted electrons (transferred to or from regular tubes), while $\Delta E_{corr,1}$ stands for the regular diamagnetic contribution of the equatorial region,

$$\frac{\Delta E_{corr,1}}{\Delta N_{e,1}^{H=0}} = \frac{1}{16r} \frac{\hbar \omega}{\omega}. \quad (37)$$

Collecting all energy terms together, we arrive at

$$\Delta E_{eq} = 2 \frac{m \pi v}{15\sqrt{2}} \frac{\omega^2}{\hbar} \sqrt{\frac{m}{\hbar}} f_{eq}(r). \quad (38)$$

(The factor 2 stands for two equivalent contributions from the upper and lower Fermi semicircles.) For the first equatorial tube we have $f_{eq}(r) = f_{eq,1}(r)$, and the function $f_{eq,1}(r)$ has different dependences for cases $a$ and $b$, described earlier. In case $a$ ($0 \leq r < 1/2$)

$$f_{eq,1}(r) = f_{eq,1}^a(r),$$

in case $b$ ($1/2 \leq r < 1$)

$$f_{eq,1}(r) = f_{eq,1}^b(r),$$

in case $r = 0$

$$f_{eq,1}^a(r) = \sqrt{r} (32r^2 - 5), \quad (39a)$$

$$f_{eq,1}^b(r) = \sqrt{r} (32r^2 - 5) - 80 \left( r - \frac{1}{2} \right)^{3/2}. \quad (39b)$$

The dependence of $\Delta E_{eq,1} \sim f_{eq,1}(r)$ from $r$ is shown in Fig. 7. Note that $\Delta E_{eq,1}(r = 0) \neq \Delta E_{eq,1}(r = 1)$, although $r = 0$ and $r = 1$ refer to the same physical situation. Below we shall see that by including two equatorial tubes, the equality of the energy at $r = 0$ and $r = 1$ is restored.

In calculating the magnetic susceptibility $\chi_{eq}$ one has to keep in mind that $\Delta E_{eq}$ depends on $H$ through $\omega$ explicitly and on $r$ implicitly. It can be shown that the contribution from the derivative of $r(H)$ with respect to the magnetic field $H$ is dominant. Finally, we obtain

$$\chi_{eq} = -\frac{\sqrt{2} m \pi v}{15} \frac{\omega^2}{\hbar} \sqrt{\frac{m}{\hbar}} \frac{\partial^2 f_{eq}(r)}{\partial r^2} \left( \frac{\partial r}{\partial H} \right)^2. \quad (40)$$

The plot of $\chi_{eq}(r)$ is reproduced in Fig. 8. It is worth noting that $\chi_{eq,1}(r)$ diverges at $r \to 0^+$ (the divergence disappears when the second equatorial tube is accounted for) and at $r \to (1/2)^+$. The latter persists in a more refined calculation with two or more equatorial tubes, because it is connected with the onset of the occupation of a new Landau level in the $(k_x,k_y)$ equatorial plane.

Notice that if we limit ourselves to the case of only the first equatorial tube, then in correspondence with Eqs. (39a) and (39b), the energy values at $r = 0$ and $r = 1$ are different, namely $\Delta E_{eq,1}(0) = 0$, $\Delta E_{eq,1}(1) \neq 0$, Fig. 6.
expressed in terms of the dimensionless parameter \( r \sim 1/H \).

The dashed line stands for the contribution from the first equatorial tube, the solid line – from the first and second equatorial tubes.

In reality the physical situation is the same, the contribution from the first equatorial tube is absent, while the second equatorial tube plays the role of the first. The inconsistence exists for the other quantities, for example, for the magnetic susceptibility, Fig. 8.

Therefore, to take into account the irregular term from the second equatorial tube, the solid line – from the first and second equatorial tubes.

In an externally applied magnetic field \( \vec{H} \) pointing along the \( z \)-axis, the energy spectrum of the occupied electron states of real metals is completely changed. Here and below we follow the semiclassical representation of electron orbits in the real and momentum \( (\hbar \vec{k}) \) space as briefly described in Sec. II A. It is worth mentioning that the semiclassical picture for itinerant electrons has been supported by the equation of motion method of Ref. [27], provided that in the quantization condition, Eq. (6), \( \gamma = 1/2 + h \Gamma_1 + h^2 \Gamma_2 \approx 1/2 \) (here \( h = eH/c \)). Notice however, that in the limit of small magnetic field \( h \to 0 \) and \( \gamma \to 1/2 \). Therefore, in the following we will still use \( \gamma = 1/2 \), although this is not critical for the method, see details in Appendix D.

In the momentum space the \( n \)-th Landau orbit in the \( (k_x, k_y) \) plane is defined by the area \( A_n \), Eq. (6), and the orbital motion is associated with the energy \( E_{\perp,n} \), Eq. (7). The real space orbit defined by Eq. (4) is no longer a circle, but a closed curve corresponding to the \( \vec{k} \)-orbit in the \( (k_x, k_y) \) plane rotated through \( \pi/2 \) about the field direction and scaled by \( hc/eH \). In real metals, in contrast to the free electron gas, Eq. (43), the cyclotron frequency \( \omega \) is given by

\[
\omega = \frac{eH}{cm^*},
\]

where the effective cyclotron mass \( m^* \) is defined as

\[
m^* = \frac{\hbar^2}{2\pi} \frac{\partial A}{\partial E}. \tag{44}
\]

Here \( A \) is the area of the electron orbit in the \( (k_x, k_y) \)-plane of the \( \vec{k} \)-space. In practice, \( \partial A/\partial E \) is found from the contour integral

\[
\frac{\partial A}{\partial E} = \oint \frac{dk}{\left| \frac{\partial E}{\partial k} \right|}. \tag{45}
\]

From Eq. (45) we conclude that in general for different Landau levels \( n \) and \( m \) with energies \( E_n \neq E_m \) we have \( (\partial A/\partial E)_n \neq (\partial A/\partial E)_m \) and consequently \( m^*_n \neq m^*_m \), \( \omega_n \neq \omega_m \). For real metals for neighboring Landau levels like \( n \) and \( n' = n \pm 1 \) since \( h\omega \ll E_F \) we still have \( m^*_n \approx m^*_m \), \( \omega_n \approx \omega_m \).

So far, it has been implied that \( k_z = 0 \). Since \( k_z \) is a constant of the electron motion \( \vec{B} \), we can easily extend our consideration to the general case with \( k_z \neq 0 \) as has been done in Sec. II A for the free electron gas. The
most important difference with the Fermi gas is that the quantities $A_n$, $\partial A / \partial E$ and hence $m^*$ and $\omega$ depend on $k_z$. Thus, in general the total electron energy is given by

$$E = E_{\perp,n} + E_z(k_z),$$

(46)

where $E_z(k_z)$ is the electron energy associated with the one dimensional (1D) electron band describing the electron motion in the $z$-direction. Notice that at $k_z = 0$ we have $E_z = 0$. Therefore, in general $E_z(k_z) = E(k) - E_{\perp,n}(k)$, provided that $k$ lies on the Landau level $n$. In the free electron case $E_z(k_z) = \hbar^2 k_z^2 / 2m$.

As in the case of the Fermi gas, we select in the $\vec{k}$-space a tube, whose main property will be that its number of electron states and energy of all states do not change in the presence of the external magnetic field. At each component $k_z$ we consider auxiliary quantized electron orbits $O_n$ with areas

$$A_{aux} = \frac{2\pi eH}{ch} (n + \delta)$$

(47)
in the $(k_x, k_y)$ plane, where $\delta \sim H$ is a small parameter ($\delta < 1$) which is discussed in detail in Appendix D. Its choice is determined by imposing the equality of energy (see Eq. (51b) below) to a very high accuracy. At fixed value of $H$, $A_{aux}$ is a constant area throughout $k_z$ although the shape of the orbit can vary with $k_z$. We associate with $A_{aux}$ a certain energy of the electron movement in the $(k_x, k_y)$ plane, which we denote as $E_{aux}$. At $k_z = 0$ this energy coincides with the orbital band energy, i.e. $E_{aux} = E(k)$ where $k$ lies on the $O_n$ orbit. For $k_z \neq 0$ using the proximity to $E_{\perp,n}$ we have $E_{aux} \approx \hbar \omega (n + \delta)$, Appendix D.

Note that at any fixed $k_z$ the $n$-th Landau level $L_n$ whose area is given by Eq. (4), is sandwiched between auxiliary orbits $O_n$ and $O_{n+1}$, Eq. (47), as shown in Fig. 9. Moreover, for a band with dispersion the Landau energy $E_{\perp,n}$, Eqs. (7), lies between $E_{aux}$ and $E_{n+1}$. Since $E_{aux} - E_{aux} \approx \hbar \omega < E_F$, within the interval $(A_{aux}, A_{aux})$ between the auxiliary orbits $n$ and $n + 1$ we deal with an infinitesimal situation. Below we show that in the $(k_x, k_y)$-plane the number of electron states with energies $E_{aux} \leq E \leq E_{aux}^n$ without magnetic field equals the number of electron states condensing on the $n$-th Landau level in the presence of the field. The same holds for their total energy. We start by noting that the density of electron states $N_\perp(E)$ in the $(k_x, k_y)$-plane is

$$N_\perp(E_{\perp}) = \frac{dN_\perp}{dE_{\perp}} = 2L_xL_y \frac{\partial A}{(2\pi)^2 \partial E} = \frac{m^* L_xL_y}{\hbar^2 2\pi},$$

(48)

(Here the double spin degeneracy is taken into account.) Using (45) and (44), we find that the number of electron states in the $n$-th tube without field, i.e. in the energy range $E_{aux} \leq E \leq E_{aux}^n$, is

$$\Delta N_n(H = 0) = \int_{E_{aux}}^{E_{aux}^n} N_\perp(E_{\perp})dE_{\perp} = 2L_xL_y \frac{A_{aux} - A_{aux}^n}{(2\pi)^2} = 2N_p,$$

(49)

where $N_p$ is the spatial degeneracy of the Landau levels, Eq. (11b). Calculating the energy of these states without field to the accuracy of $(\hbar \omega / E_F)^3 \Delta N_n$, Appendix D we obtain

$$\mathcal{E}_n(H = 0) = \int_{E_{aux}}^{E_{aux}^n} \mathcal{E}_\perp(E_{\perp})dE_{\perp} = E_{\perp,n} \Delta N_n.$$ 

(50)

That is, $\mathcal{E}_n(H = 0)$ coincides with the energy of these states on the $n$-th Landau level in the presence of the field. Thus, we have proven that

$$\Delta N_n(H = 0) = \Delta N_n(H \neq 0) = 2N_p,$$

(51a)

$$\mathcal{E}_n(H = 0) = \mathcal{E}_n(H \neq 0) = \hbar \omega \left( n + \frac{1}{2} \right) 2N_p.$$ 

(51b)

Eqs. (51a) and (51b) are valid for any $k_z$-component, and therefore for the whole tube $n$, Fig. 9 defined by its upper ($k_z^{(2)}$) and lower ($k_z^{(1)}$) boundaries, $k_z^{(1)} \leq k_z \leq k_z^{(2)}$. As for the free electron gas, $k_z^{(2)}$ and $k_z^{(1)}$ are conveniently defined by the intersection of a tube with the Fermi surface, Fig. 10. As a result, we have two $n$-th tubes: the first tube (denoted by 1 in the inset of Fig. 10) lies entirely inside the Fermi surface and does not exhibit diamagnetism, while the second tube (denoted by 2 in the inset of Fig. 10) is only partially filled and results in a diamagnetic response. We consider this effect in the following sections.

B. Diamagnetically active electron states

Consider the Fermi surface and define magnetic tubes in an externally applied magnetic field $H$ pointing in the
positive $z$-direction, Fig. 10. As has been discussed in Sec. IIIA, each magnetic tube $n$ at $k_z$ is sandwiched between two boundary electron orbits $O_n$ and $O_{n+1}$ in the $(k_z, k_y)$-plane whose areas $A_{n\text{aux}}$ and $A_{n+1\text{aux}}$ are defined by Eq. (17). As a result we obtain a set of boundary surfaces, which intersect the Fermi surface in orbits perpendicular to the $z$-axis. The intersection orbits unlike simple circles for the Fermi gas, Sec. II, are rather complicated, and below we describe them in detail. These orbits lie in $(k_z, k_y)$-planes and we can use them to define two $k_z$-boundary conditions, $k_z^{n,1}$ and $k_z^{n,2}$, in such a way that the magnetic tube $n$ lies entirely inside the Fermi surface. The fully occupied tubes are schematically shown in Fig. 10. In Sec. IIIA we have seen that the electron states of the completely filled tubes do not change their energy in magnetic field. Therefore, the whole effect is due to the states lying in the partially occupied tubes passing through the Fermi surface (like tube 2 in the inset of Fig. 10).

Consider a boundary orbit $O_{n,F}$ (defined by $A_{n\text{aux}} = \text{const}$, Eq. (17)) in the $(k_z, k_y)$-plane lying on the Fermi surface. It defines a certain value of $k_z$ which we denote by $k_z,n$. The electron energy band gradient, $\partial E(\vec{k})/\partial \vec{k}$, on $O_{n,F}$ is perpendicular to the Fermi surface and hence to the orbit. In addition, we will use the projection of the gradient in the plane of the orbit, $\partial E(\vec{k})/\partial k_z$, which is also perpendicular to $O_{n,F}$, Figs. 9, 11. The normalized vector in the direction of $\partial E(\vec{k})/\partial k_z$ will be denoted by $e_{k_z}$. Notice that plane $(k_z, e_{k_z})$ is normal to $O_{n,F}$. Cross-sections of magnetic tubes passing through the Fermi surface are schematically shown by horizontal dotted lines in Fig. 10. (In general, Fig. 10 should be understood as composed of many different panels – at most one panel for each tube cross-section, because the energy gradient $\partial E/\partial k_z$ and hence $(k_z, e_{k_z})$-planes still can have different orientations.)

Consider a typical partially occupied tube $n$, whose triangle cross-section in the $(k_z, e_{k_z})$-plane is shown in Fig. 11. Points $O_n'$ and $O_{n+1}'$ are defined by the intersection of the $(k_z, e_{k_z})$-plane with the orbits $O_{n,F}$ and $O_{n+1,F}$, respectively. The side $O_{n}', O_{n+1}'$ is the intersection with the Fermi surface, the side $O_n', R$ is the intersection with the $A_{n\text{aux}}$ boundary surface, and the side $O_{n+1}', R$ is the intersection with the $(k_z, k_y)$-plane of the orbit $O_{n+1,F}$. It is convenient to count energy from $E_{ref} = E(R) = E_F - \hbar \omega$. The vector $RR'$ being perpendicular to $O_{n}', O_{n+1}'$ is collinear with the energy gradient $\partial E/\partial \vec{k}$. Finally, the line $L, L'$ is given by the intersection of Landau orbits $L_n$ at various $k_z$, Eq. (9), with the $(k_z, e_{k_z})$-plane. Since we consider the limit of small magnetic fields, i.e. $\hbar \omega/E_F \to 0$, the triangle is assumed infinitesimal allowing for linear dependencies of relevant quantities. In particular, the line $L, L'$ is parallel to $R, O_n'$, etc., and we can take $\delta = 0$ in Eq. (17), $\gamma = 1/2$ in Eq. (3), Appendix D.

In the following we will need $\Delta k_z, \Delta k_z, \Delta k_F$ expressed in terms of $\partial E/\partial k_z, \partial E/\partial \vec{k}_z, \partial A/\partial k_z$ calculated at the Fermi surface. Details of these calculations are given in Appendix A.

Now in accordance with Eq. (16) we can find the number of the active electron states in all partially occupied tubes. We recall that in Eq. (16) $\Delta N \equiv \Delta N(k_z, k_z + \Delta k_z)$ is the number of the electron states in the $n$-th partially filled tube, whose $k_z$-component lies between $k_{z,n} \equiv k_z$ and $k_{z,n+1} \equiv k_z + \Delta k_z$. The calculation of $\Delta N$ carried out in Appendix A results in Eq. (17), that is

$$\frac{\Delta N}{\Delta k_z} = 2\pi v e H \hbar c = \text{const.} \quad (52)$$

Notice that the right hand side of Eq. (52) is independent of the effective mass $m^*$ and $\partial A/\partial E$. It should be emphasized that the polar and equatorial regions of the Fermi surface are special cases, which are discussed in Sec. IIIA. It turns out that these regions fully comply with the general expression (52), while the equatorial regions (where $\partial A(k_z)/\partial k_z = 0$) give rise to the well known...
de Haas-van Alphen oscillations of the magnetic susceptibility, which is not the subject of the present study.

For small magnetic fields when $\Delta k_z \ll k_F$, in Eq. (10) we can substitute the summation with the integration, Eq. (18). In Eq. (18) $k_{z,\text{min}}^{(n)} (k_{z,\text{max}}^{(n)})$ is the minimal (maximal) $k_z$-component of the vectors $\vec{k}$ lying on the Fermi surface. Using (52) [or Eq. (17)] we arrive at Eq. (19), where $\ell_z \equiv k_{z,\text{max}}^{(n)} - k_{z,\text{min}}^{(n)}$ is defined exclusively by the projection of the Fermi surface along the direction of the applied magnetic field. Notice however, that unlike for free electron gas, now in general $k_{z,\text{min}}^{(n)} \neq -k_F$, $k_{z,\text{max}}^{(n)} \neq k_F$. Furthermore, as discussed later in Sec. IV in real metals $\ell_z$ depends on the orientation of the applied magnetic field with respect to the Fermi surface.

Thus, the number of active electron states $N \sim H$, their total energy change in the magnetic field $\sim H^2$, which gives a constant diamagnetic susceptibility $\chi$.

C. Landau diamagnetic susceptibility

The transformation of the electron states of a partially occupied tube is schematically shown in Fig. 12. In the applied magnetic field $H \neq 0$ all electrons of the partially occupied tube are on the $n$-th Landau orbits with the transverse energy $E_n$, and occupy the lowest in energy $k_z$-states, as shown in Fig. 12.

As we have seen in Sec. II C for the free electron gas the highest occupied energy in the presence of the field coincides with the Fermi energy $E_F$ obtained in the absence of the field. For the real metal the $(k_z, c_{k_z})$-cross-section of a partially occupied tube $n$, Figs. 11 and 12 in general is very different from the $(k_z, k_y)$ cross-section of the Fermi gas shown in Figs. 3 and 4. However, the highest occupied energy is still $E_F$. This follows from Eq. (20), written for the partially occupied tube $n$ for real metal. In terms of the $z-$projection $\delta k_z$ of the wave vector $\vec{k}_L$, shown in Fig. 11, it implies $\delta k_z = \Delta k_z / 2$ just as for the Fermi gas, Eq. (21). Therefore, there are no electron transitions between different tubes and we can calculate the energy change for each tube separately and then sum all contribution for the final result.

Keeping in mind that in the magnetic field there are two energy contributions: $E_1$ in the $(k_x, k_y)$ plane and $E_z$ at $k_z$, Eq. (10), we now calculate the energy change for the tube $n$ in the applied magnetic field, Eq. (22).

The calculation of all components is performed in Appendix B (with respect to the energy $E_{\text{ref}}$) and results in Eqs. (23), (24a) and (24b). For the total energy change we get $\Delta E = \hbar \omega \Delta N / 12$. This is the same expression as for the free electron gas, Eq. (25), but now the cyclotron frequency $\omega$ is different for different tubes, Eq. (43). Therefore, by means of Eqs. (43) and (52) we rewrite the expression for $\Delta E$ in terms of $\Delta k_z$:

$$\Delta E = \frac{1}{6} \pi v \left( \frac{eH}{c} \right)^2 \frac{\Delta k_z}{m^*}. \quad (53)$$

Note that Eq. (53) refers to any partially filled tube. Therefore, making the summation over all tubes we find

$$E = \frac{1}{48 \pi^2} \left( \frac{eH}{c} \right)^2 \int_{k_{z,\text{min}}}^{k_{z,\text{max}}} \frac{dk_z}{m^*(k_z)}. \quad (54)$$

Here, as before $k_{z,\text{min}}^{(n)} (k_{z,\text{max}}^{(n)})$ is the minimal (maximal) $k_z$-component of the Fermi surface. Notice, that $m^*$ found from Eqs. (14) and (15) for orbits in the $(k_x, k_y)$ plane lying on the Fermi surface, is a function of $k_z$. Finally, for the magnetic susceptibility $\chi$ we have:

$$\chi = -\frac{d^2 E(H)}{dH^2} = -V \frac{e^2}{24 \pi^2 c^2} \int_{k_{z,\text{min}}}^{k_{z,\text{max}}} \frac{dk_z}{m^*(k_z)}. \quad (55)$$

In the case of the free electron gas the integral on the right hand side of (55) is reduced to $\ell_z / m = 2k_F / m$ and we arrive at Eq. (27) for the diamagnetic susceptibility of the Fermi gas. In general, Eq. (55) differs from the pioneer expression of Peierls [8], Eq. (2), although for the case of parabolic bands in terms of $k_z$ and $k_y$, Eq. (21), they give the identical results, Appendix B.

As a final and important remark we note that the density of electron states, $g(E_F)$, remains unchanged on applying the magnetic field, although energy gradients at the Fermi surface change their directions and values. We prove this remarkable effect in Appendix C.

D. Special cases and extremal cross-sections of the Fermi surface

General expressions for $\Delta k_\bot$ and $\Delta k_z$, Eqs. (A11) and (A22), become indefinite when $(\partial E / \partial k)_{\parallel} = 0$ or $\partial E / \partial k_z = 0$. The condition $(\partial E / \partial k)_{\parallel} = 0$ occurs at polar points of the Fermi surface and the full account of this situation is given in Appendix C. In that case there are no deviations from the general final equations (52) and (53).
If $\partial A(k_z)/\partial k_z = 0$ then from Eq. (A2) it follows that $\Delta k_z \to \infty$, and such an extremal cross-section corresponds to a maximum or a minimum of $A(k_z)$ as a function of $k_z$. It is well known that extremal orbits and extremal regions give rise to the oscillatory behavior for the diamagnetic response (de Haas – van Alphen effect), Ref. [4]. In our approach for the Fermi gas described in detail in Sec. III it has also been demonstrated that the extremal region consisting of a few tubes in the equatorial region of the Fermi surface, results in the oscillatory behaviour of the diamagnetic susceptibility. This should occur in real metals as well, but in this paper we consider only the regular contribution to energy ($\Delta E^L_{eq}$) leaving the irregular part ($\Delta E^{irr}_{eq}$) for future consideration.

IV. APPLICATION TO ALKALI METALS

We have applied the method to calculations of the diamagnetic response of alkali metals: Li, Na, K, Rb and Cs, which are crystallized in the body centered cubic (bcc) lattice and have only one active electron band. As follows from Sec. III for diamagnetic susceptibility one needs two very important characteristics: the Fermi surface and the energy gradient $\nabla_E^z$ on it. Therefore, we first have performed $ab\initio$ density functional calculations (DFT) of the alkali metals using Moscow-FLAPW code[33]. The Perdew-Burke-Ernzerhof (PBE) variant of the generalized gradient approximation has been employed, with the number of $k$-points 2470. For the Fermi surface and gradient calculations we have used the tetrahedron method[35]. In this method the irreducible part of the Brillouin zone is divided into a set of tetrahedra and within each tetrahedron the linear dependence of the band energy $E(\hat{k})$ is assumed. The cross-sections of the constant energy $E = E_F$ in the $\hat{k}$-space from all relevant tetrahedra give the pieces forming the Fermi surface. Also, in each tetrahedron crossing the Fermi surface we find the value of the energy gradient, $\nabla_E^z$, which is unchanged within the tetrahedron.

The obtained Fermi surfaces (FS) are shown in Fig. 13. The important property of the calculated Fermi surfaces is that the topological space inside each of them is simply (or path) connected. In other words, the occupied electron states form a single piece Fermi surface. (The application of the presented method to more complicated Fermi surfaces and to completely filled electron bands requires additional considerations.)

Since the Fermi surface is invariant under all symmetry operations of the crystal (in our case we deal with the $O_h$ cubic symmetry), it can be expanded in terms of symmetry adapted functions (SAFs) belonging to the fully symmetric ($A_{1g}$) representation of $O_h$[31]. Such SAFs known as cubic harmonics[31], are linear combinations of real spherical harmonics $Y_{l}^{m,s}$ ($\tau = c, s$ stands for the azimuthal dependence of the cosine ($c$) or sine ($s$) type with $l = 4, 6, 8, ...$):

$$K_4(\theta, \phi) = \sqrt{\frac{7}{12}} Y_4^0 + \sqrt{\frac{5}{12}} Y_4^4,$$

$$K_6(\theta, \phi) = \sqrt{\frac{7}{8}} Y_6^0 - \sqrt{\frac{7}{8}} Y_6^4,$$

$$K_8(\theta, \phi) = \frac{33}{64} Y_8^0 + \frac{7}{48} Y_8^4 + \sqrt{\frac{65}{192}} Y_8^8.$$

Here $(\theta, \phi) = \hat{k}$ are polar angles in $k$-space and for spherical harmonics we use the phase convention of Bradley and Cracknell[31]. (For compactness here and below we omit $(\theta, \phi)$ in $Y_{l}^{m,\tau}$.) Therefore, the Fermi surface can be very accurately represented by the expansion

$$k_F(\hat{k}) = C_0^{FS} Y_0^0 + C_4^{FS} K_4(\hat{k}) + C_6^{FS} K_6(\hat{k}) + C_8^{FS} K_8(\hat{k}),$$

where $Y_0^0 = 1/\sqrt{4\pi}$ is the zeroth spherical harmonic ($l = m = 0$). In the expansion the Fermi surface is completely defined by the coefficients $C_0^{FS}, C_4^{FS}, C_6^{FS}$ and $C_8^{FS}$. These coefficients can be obtained numerically from the calculated Fermi surfaces, Fig. 13. For all alkalis they are given in Table I.

![FIG. 13: Calculated Fermi surfaces of alkali metals.](image-url)
The other quantity – energy gradient at the Fermi surface $\nabla_k E_F(\hat{k})$ – also can be expanded in terms of the cubic harmonics, Eq. (56). Since the gradient is always normal to the Fermi surface and therefore its direction can be found at any point of the Fermi surface we need only its modulus expansion,

$$\left| \nabla_k E_F(\hat{k}) \right| = C_0^g Y_0^g + C_4^g K_4(\hat{k}) + C_6^g K_6(\hat{k}) + C_8^g K_8(\hat{k}).$$

(57)

The coefficients $C_0^g$, $C_4^g$, $C_6^g$ and $C_8^g$ obtained from the ab initio calculations are also quoted in Table I. The three dimensional picture of $\left| \nabla_k E_F(\hat{k}) \right|$ as a function of the direction $\hat{k}$ in $k$–space is shown in Fig. 14.

From Fig. 13 and the values of the coefficients $C_i^{FS}$ we conclude that the Fermi surfaces of lithium and cesium demonstrate largest deformations from the spherical shape. Sodium and potassium, on the other hand, have FS shapes which are very close to spherical, while deviations for rubidium lie between these two groups. This finding is even more clearly illustrated by the shape of energy gradients, Fig. 14, which are more sensitive to deviations from the spherical form.

We now study the cyclotron mass $m^*$, which depends on the closed orbit on the Fermi surface in $k$-space, Eqs. (44), (45). The orbit is given by the cross section of the Fermi surface with a plane lying perpendicular to the applied magnetic field $\vec{H}$. Such a plane is uniquely defined by its projection $k_H \equiv k_\|$ on the $z'$-axis parallel to $\vec{H}$. Therefore, the cyclotron mass in the applied magnetic field is a function of $k_H$ and $\vec{H}$ pointing in the direction of $\vec{H}$, that is $m^* = m^*(H, k_H)$. In Fig. 15 we plot $m^*$ as a function of $k_H$ for three directions of magnetic field for potassium with nearly spherical Fermi surface and lithium with a deformed Fermi surface.

Notice that for deformed Fermi surfaces (lithium and cesium) $k_H^{max}$ and $k_H^{min}$ are noticeably different for various directions of $H$, Table I. In Table I we also quote average cyclotron masses $(m^*_a)$ for magnetic field directions [001], [101] and [111], defined according to the

| metal | Li | Na | K | Rb | Cs |
|-------|----|----|---|----|----|
| $\alpha$, Å | 3.50 | 4.29 | 5.35 | 5.585 | 6.14 |
| $\chi_L^{av}$, $10^{-7}$ | -1.753 | -2.086 | -1.624 | -1.515 | -1.179 |
| $\chi_L[001]$, $10^{-7}$ | -1.670 | -2.081 | -1.631 | -1.508 | -1.142 |
| $\chi_L[101]$, $10^{-7}$ | -1.871 | -2.090 | -1.631 | -1.560 | -1.308 |
| $\chi_L[111]$, $10^{-7}$ | -1.714 | -2.084 | -1.618 | -1.485 | -1.114 |
| $m^*_{av}$ | 1.517 | 1.036 | 1.067 | 1.097 | 1.300 |
| $m^*_{av}[001]$ | 1.552 | 1.036 | 1.067 | 1.099 | 1.310 |
| $m^*_{av}[101]$ | 1.462 | 1.036 | 1.065 | 1.080 | 1.214 |
| $m^*_{av}[111]$ | 1.527 | 1.037 | 1.067 | 1.107 | 1.317 |
| $K_{H_{max}}^{av}[001]$ | 0.576 | 0.480 | 0.386 | 0.369 | 0.333 |
| $K_{H_{max}}^{av}[101]$ | 0.609 | 0.481 | 0.386 | 0.375 | 0.353 |
| $K_{H_{max}}^{av}[111]$ | 0.582 | 0.481 | 0.385 | 0.366 | 0.326 |
| $N(E_F)$ | 0.486 | 0.494 | 0.792 | 0.888 | 1.302 |
| $\gamma_{el}$ | 1.146 | 1.166 | 1.867 | 2.094 | 3.068 |
| $\gamma_F$, $10^{-7}$ | 12.207 | 6.728 | 5.565 | 5.487 | 6.051 |
| $C_0^{FS}$ | 2.08862 | 1.70433 | 1.36665 | 1.30909 | 1.19032 |
| $C_4^{FS}$ | -0.01529 | -0.00039 | 0.00092 | -0.00498 | -0.00474 |
| $C_6^{FS}$ | -0.02358 | -0.00065 | -0.00101 | -0.00694 | -0.02149 |
| $C_8^{FS}$ | 0.00660 | -0.00001 | 0.00074 | 0.00263 | 0.00914 |
| $C_0^g$ | 1.40005 | 1.64429 | 1.28035 | 1.19546 | 0.94458 |
| $C_4^g$ | 0.13752 | 0.01058 | -0.00908 | 0.00294 | 0.04106 |
| $C_6^g$ | 0.28278 | 0.01325 | 0.00441 | 0.08695 | 0.24620 |
| $C_8^g$ | -0.10748 | -0.00267 | -0.00733 | -0.05090 | -0.12383 |

FIG. 14: Calculated energy gradient modula of alkali metals. (The order of the elements as in Fig. 13.)
calculated averaged values of $\chi$ points [32]. (The set gives the correct coefficients of exceptions [36].

Table I. They are in fair correspondence with estimates $\chi_m$ of the Landau contributions obtained with other approaches [32].

The steady diamagnetic susceptibility $\chi_L$ from its average value $\chi_L^av$ for various directions of the applied magnetic field $H$ for alkali metals.

FIG. 15: Calculated cyclotron mass $m^*$ for Li and K for directions [001], [101] and [111] of the magnetic field $H$.

The anisotropy of $\chi_L$ with respect to the direction of the applied magnetic field is a remarkable property of the Landau diamagnetism, which singles it out from the Pauli paramagnetism and the Langevin diamagnetism of closed electron shells.

V. CONCLUSIONS

The steady diamagnetic response for the Fermi gas and a real metal with a general simply connected Fermi surface is obtained analytically at zero temperature ($T = 0$) within the widely accepted semiclassical approach [2–4] [26, 27]. The diamagnetic effect is caused by electron states in a very narrow region of the Fermi surface. The consideration is based on a structure in $k$-space called a magnetic tube, which sandwiches the Landau level inside it. The completely occupied tubes are diamagnetically inert. Only partially occupied tubes located at the Fermi surface are responsible for the diamagnetic effect. Although energy gradients at the Fermi surface change in the magnetic field, the total density of electron states at the Fermi energy remains constant, Appendix [II].

We have applied the method for calculations of the Landau diamagnetic susceptibility $\chi_L$ of alkali metals (Li, Na, K, Rb, Cs). The crucial quantities for that are the Fermi surface $k_F(k)$ and energy gradients $\nabla_k E_F(k)$ on it, which are obtained from $ab$ initio band structure calculations. For accurate calculations of $\chi_L$, we expand both $k_F(k)$ and $\nabla_k E_F(k)$ in terms of symmetry adapted functions, Eqs. [56] and [57]. We have demonstrated that the diamagnetic effect depends on the direction $\hat{H}$ of the applied magnetic field $H$. The anisotropy of $\chi_L$ is larger for lithium and cesium and smaller for sodium and potassium. It is worth noting that the Langevin diamagnetism of atomic closed core shells and the Pauli
paramagnetism are isotropic with respect to $\hat{H}$.

The present approach can be applied to other metals or intermetallic compounds. The method can also be extended to the case of non-zero temperatures, where the thermal excitations of one dimensional electron gas consisting of electron states on the Landau levels should be taken into account.

While the steady response is due to the region just below the Fermi surface, the oscillatory behavior of energy and magnetic susceptibility arises from a few partially occupied tubes located at its extremal cross-sections. A small oscillatory change of the Fermi energy of the free electron gas in the applied magnetic field is caused by a transfer (inflow or outflow) of electrons from this equatorial region of the Fermi surface, Sec. [11]

Appendix A: Calculation of $\triangle k_\perp, \triangle k_z, \triangle N$

Here we quote some useful relations in a partially occupied tube shown in Fig. [11] taking into account the infinitesimal properties of its triangular $(k_z, e_{k_z})$ cross-section. In particular, we have

$$\triangle k_\perp = \frac{\hbar \omega}{|\partial E/\partial k_\perp|} > 0. \quad (A1)$$

To calculate $\triangle k_z$ we first consider the area $A$ of the $(k_z, e_{k_z})$-cross-section of the Fermi surface as a function of $k_z$, that is, $A(k_z)$. Then $\triangle A = A_{n+1}^{\text{ref}} - A_n^{\text{ref}} = (\partial A(k_z))/\partial k_z ) \triangle k_z$, and we arrive at

$$\triangle k_z = \frac{2\pi e H}{c h} \frac{1}{|\partial A/\partial k_z|} > 0. \quad (A2)$$

For the number of the electron states in the partially occupied tube $n$, Fig. [11] we find

$$\triangle N = 2v \oint_{O_{n+1}} \triangle S dk = v \triangle k_z \oint_{O_{n+1}} \triangle k_\perp dk = v \triangle A \triangle k_z, \quad (A3)$$

where $v = V/(2\pi)^3$ and $\triangle S$ is the area of the $(k_z, e_{k_z})$-cross-section: $\triangle S = \triangle k_\perp \triangle k_z/2$. This gives Eqs. (17) and (22).

Appendix B: Calculation of $\triangle E^{H=0}$, $\triangle E^{H\neq0}_{\perp}$, $\triangle E^{H\neq0}_{z}$

We first consider the energy of a partially occupied tube $n$ without magnetic field ($H = 0$), left panel of Fig. [12] The reference energy at $\hat{k}(R)$ is $E_{\text{ref}} = \tilde{E}(\hat{k}(R)) = E_p - \hbar \omega$, Fig. [11]. We will calculate the energy by integrating over $x$ from $R$ to $R'$ along the energy gradient direction $\partial E/\partial k_F$ , Fig. [11]. Notice that $O_n' O_{n+1}'$ is perpendicular to $RR'$, the energy at $x$ is $(x/\triangle k_F)\hbar \omega$ and the infinitesimal volume element is $dk dx (x/\triangle k_F)|O_n', O_{n+1}'|$. Denoting $\triangle k_F = |O_n', O_{n+1}'|$, we get

$$\triangle E^{H=0} = 2v \oint_{O_{n+1}} \epsilon dk, \quad (B1)$$

where

$$\epsilon = \int_0^{\triangle k_F} \left( \frac{x}{\triangle k_F} \hbar \omega \right) \left( \frac{x}{\triangle k_F} \triangle k_F \right) dx. \quad (B2)$$

Performing integration in (B2) and taking into account that the area of the cross-section shown in Fig. [11] is $\triangle S = \triangle k_F \triangle k_F/2$ whereas $2v \oint \triangle Sdk = \triangle N$, we arrive at Eq. (21).

We now consider the case $H \neq 0$, the right panel of Fig. [12] The transverse component of an electron state condensed on the Landau level is $\epsilon = E_p + \hbar^2/2 m' + \hbar \omega/2$, and for all electron states we obtain Eq. (24).

Populating the Landau levels along the $L, L'$ line shown in Fig. [11] the one dimensional electron energy changes from $E_{\text{ref}}$ to $E_{\text{ref}} + \hbar \omega/2$. Since in the infinitesimal cross-section approximation of Fig. [11] the energy increases linearly with $k_z$ and the density of electron states along the $L, L'$ line is constant, for the energy of all these occupied states we obtain $\triangle E^{H\neq0}_z = (\hbar \omega/4) \triangle N$, Eq. (21).

Appendix C: Polar region

If the energy gradient at a certain point $P$ of the Fermi surface is parallel to the $z$-axis (or to the magnetic field $\hat{H}$), that is $\partial E(P)/\partial k_\perp \parallel \hat{H}$, then $\partial E(P)/\partial k_\perp = 0$, but on the other hand, the contour path for $E = E(P)$ in Eq. (15) reduces to the point $P$. This leads to the ambiguity for $\partial A/\partial E$ in Eq. (15) and consequently for $m'$ and $\omega$ in Eqs. (14) and (13). This situation corresponds to a polar region around the point $P$ and requires a special treatment.

For convenience we put the origin of the coordinate system at the point $P$ and assume that the center of curvature is located below. The Fermi surface in the vicinity of $P$ is given by the function $k_z^{FS}(k_x, k_y) = f(k_x, k_y)$, whose tensor of the second derivatives is diagonalized:

$$\frac{\partial^2 k_z^{FS}}{\partial k_x^2} |_P < 0, \quad \frac{\partial^2 k_z^{FS}}{\partial k_y^2} |_P < 0, \quad \frac{\partial^2 k_z^{FS}}{\partial k_x \partial k_y} |_P = 0. \quad (C1)$$

This polar region of the Fermi surface is described by the function

$$k_z^{FS}(k_x, k_y) = \frac{1}{2} \left( \frac{\partial^2 k_z^{FS}}{\partial k_x^2} |_P \right) k_x^2 + \frac{1}{2} \left( \frac{\partial^2 k_z^{FS}}{\partial k_y^2} |_P \right) k_y^2. \quad (C2)$$

In the polar region we deal with the zeroth magnetic tube, i.e. $n = 0$, and with the zeroth Landau level. Two
tube’s auxiliary boundary conditions are reduced to
\[ A_n^{aux} = 0, \quad A_1^{aux} = \frac{2\pi eH}{\hbar c}. \] (C3a)
\[ \delta_n^{aux} = \frac{\partial^2 \tilde{F}^{aux} \tilde{S}}{\partial k_x^2} \bigg|_p \partial^2 \tilde{F}^{aux} \tilde{S} \bigg|_p. \] (C4)
(Notice that here \( \delta = 0 \).) Thus, the first quantized auxiliary orbit \( O_n^+ = 1 \) at the Fermi surface is an ellipse at \( \Delta k_z = k_{z1} < 0 \), which can be found by equating its area to \( A_1^{aux} \), Eq. (C3b). The result is
\[ \Delta k_z = -\frac{eH}{\hbar c} \sqrt{\frac{\partial^2 \tilde{F}^{aux} \tilde{S}}{\partial k_x^2}} \bigg|_p \partial^2 \tilde{F}^{aux} \tilde{S} \bigg|_p. \] (C5)
Therefore, the whole polar region with \( 0 \geq k_z \geq \Delta k_z \) represents the zeroth partially occupied tube. It can be shown that the volume of the polar region is
\[ V_P = \frac{1}{2} |\Delta k_z| A_1^{aux}. \] (C6)
The same expression holds for a small sphere cap in the case of the Fermi sphere of the free electron gas. The number of active electron states \( \langle H \rangle = 0 \) is then
\[ \Delta N_n = 2\nu V_P = |\Delta k_z| \frac{2\pi eH}{\hbar c} \nu. \] (C7)
The latter equation demonstrates that \( \Delta N_0/|\Delta k_z| \) follows the general relation \( \mu \), although the curvature of the Fermi surface polar region has been explicitly taken into account. Analogously, one can show that the other quantities (energy etc.) also follow the general consideration.

**Appendix D: Tuning auxiliary boundary conditions**

In this section we will give details of derivation of Eqs. (50) and (51b) and define the small parameter \( \delta \sim \hbar \) in Eq. (47). The middle part of Eq. (50) can be integrated
\[ \frac{\partial^2 \tilde{F}^{aux} \tilde{S}}{\partial k_x^2} \bigg|_p \partial^2 \tilde{F}^{aux} \tilde{S} \bigg|_p. \] (C3b)
where \( E_0 = (E_n^{aux} - E_{n+1}^{aux})/2 \) and we use short notations \( A' = \partial A(E_0)/\partial E \), \( A'' = \partial^2 A(E_0)/\partial E^2 \). With Eq. (D1) we obtain
\[ \delta^* = \frac{1}{2} + \delta - \frac{A''(\triangle E)^2}{12\triangle A} + O(H^3), \] (D5)
where \( O(H^3) \sim H^3 \) and Eq. (D2) becomes
\[ \mathcal{E}_n(H = 0) = \left[ E_0 + \frac{A''(\triangle E)^2}{12\triangle A} \right] \triangle N_n. \] (D6)
On the other hand, for the energy of the tube in the magnetic field with respect to \( E_0 \) one has
\[ \mathcal{E}_n(H \neq 0) = \left[ E_0 - \frac{\Delta A}{A} \delta + \frac{1}{8A}(\triangle E)^2 \right] 2N_p. \] (D7)
Equating Eqs. (D6) and (D7), we obtain
\[ \delta = \frac{\pi e}{12c\hbar} \frac{\mathcal{E}_n}{(A')^2}. \] (D8)
If, as in Eq. (50), one uses \( \gamma = 1/2 + \Gamma_{1}\hbar \), then in Eq. (D8) \( \delta \) should be replaced with \( \langle \delta - \gamma \rangle \sim H \). From Eq. (D8) it follows that \( \delta \) is also a function of energy, \( \delta = \delta(E) \). This in turn leads to a more complicated final expression,
\[ \delta(E) = \frac{\pi e}{12c\hbar} \frac{f(E)}{1 + E f'(E)/f(E)} H, \] (D9)
which can be solved iteratively, starting with \( f_0(E) = A''(E)/A'(E)^2 \) as in Eq. (D8).

Quantities \( \gamma \) and \( \delta \) should be taken into account for the definition of the boundaries of the occupied magnetic tubes, because they concern a large number of electron states inside the Fermi surface. For partially occupied tubes at the Fermi surface they give only small corrections of the order of \( \hbar\omega/E_F \) to main results, which can be omitted.

**Appendix E: Connection with Peierls’ expression**

In his early pioneer work [8], Peierls obtained Eq. (2) for the steady diamagnetic susceptibility. Below we will show that the magnetic susceptibilities given by Eqs. (2) and (3) coincide for the case of the parabolic energy dependence in terms of \( k_x \) and \( k_y \).
\[ E(k) = a(k_x)k_x^2 + b(k_x)k_y^2 + c(k_x)k_xk_y + d(k_z). \] (E1)
Notice that \( a(k_z) \), \( b(k_z) \), \( c(k_z) \), \( d(k_z) \) here are arbitrary smooth functions of \( k_z \) and the \( z \)-axis points in the direction of the applied magnetic field \( H \). The expression in the curly bracket of Eq. (2), being \( \lambda = 4ab - c \), is an invariant of the quadratic form (E1). We then transform (E1) to a new coordinate system \((k'_x, k'_y)\) where it takes the diagonal form \( E(\vec{k}) = a'(k_z)k_x'^2 + b'(k_z)k_y'^2 + d(k_z). \)
Now \( \lambda = 4a'b' \) and in the following will work in this representation. For the integral in Eq. (2) we obtain

\[
I = -4 \int_{FS} \frac{a'(k_z)b'(k_z)}{|\nabla_k E(k)|} dS(k) = 4\pi \int_{k_{zm}^{\max}} \frac{a'(k_z)b'(k_z)}{\sqrt{a'(k_z)b'(k_z)}} dk_z, \quad (E2)
\]

where the first integration is taken over the Fermi surface (FS) and \( k_{zm}^{\max} \) is the maximal (minimal) projection of FS on the \( z \)-axis. The final expression for Eq. (2) becomes

\[
\chi = -\frac{e^2}{12\pi^2 \hbar^2 e^2} \int_{k_{zm}^{\max}} \sqrt{a'(k_z)b'(k_z)} dk_z. \quad (E3)
\]

On the other hand, using Eq. (44) we obtain \( m^*(k_z) = \hbar^2 / 2 \sqrt{a'(k_z)b'(k_z)} \). Substitution of \( m^*(k_z) \) in Eq. (55) gives the same result as before, Eq. (E3).

In general however Eq. (2) differs from Eq. (55). We ascribe it to a number of approximations used in the derivation of Eq. (2) \[1, 11, 13, 14\].

**Appendix F: Density of electron states at \( E_F \)**

Consider the partitioning of the Fermi surface region in partially occupied tubes. The density of electron states (DOS) at the Fermi energy then is given by

\[
g(E_F) = \frac{1}{V} \sum \Delta g_n(E_F) = \int_{k_{zm}^{\max}} \Delta g(E_F) \Delta k_z,
\]

where \( \Delta g_n(E_F) \) is the DOS contribution from the part of the Fermi surface \( S_n \) associated with a partially occupied tube \( n \),

\[
\Delta g_n(E_F) = \frac{1}{4\pi^3} \int_{S_n} \frac{dS}{|\nabla S|}. \quad (F2)
\]

In the applied magnetic field \( H \neq 0 \), Fig. 11 electron states occupy the Landau level \( n \) along the \( L, L' \) line. Since the Landau orbits are perpendicular to the \( z \)-axis, the energy gradient should be taken in the \( z \)-direction with the surface element \( dS \) in the \((k_x, k_y)\)-plane. From Fig. 11 we obtain \( \partial E/\partial k_z = h\omega/\Delta k_z \), \( dS_{\perp} = \Delta k_z dk \). We thus arrive at

\[
\Delta g^{H\neq0}(E_F) = \frac{1}{4\pi^3} \int \frac{\Delta k_z dk}{h\omega}. \quad (F3)
\]

Notice that \( \Delta S = \Delta k_z \Delta k_z / 2 \) is the area of the triangular \((O', A, O_{n+1}'\) -cross-section, which can also be written as \( \Delta S = \Delta k_F \Delta l_F / 2 \). Since \( \partial E/\partial \vec{k} = h\omega/\Delta k_F \), we continue Eq. (F3) as

\[
\Delta g^{H\neq0}(E_F) = \frac{1}{4\pi^3} \int \frac{\Delta l_F dk}{|\partial E/\partial \vec{k}|} = \Delta g^{H=0}(E_F). \quad (F4)
\]

Here in establishing the last equality we have taken into account that \( \Delta l_F dk = dS^{H=0} \) is a part of the Fermi surface perpendicular to the energy gradient \( \partial E/\partial \vec{k} \) in the absence of the magnetic field. Eq. (F3) leads to

\[
g^{H\neq0}(E_F) = g^{H=0}(E_F). \quad (F4)
\]

\[\text{References} \]

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