Magnetism - Oscillations

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When dealing with magnetic fields in condensed matter physics, we encounter oscillations in resistivity i.e. Subhnikov de Haas oscillations and also in magnetization i.e. de Haas - van Alphen oscillations. These are crucial to characterize materials and thus a discussion is in order for them. In 1930's, Bohr and Leeuwen showed that magnetism cannot exist within the framework of classical mechanics. Considering the Hamiltonian for N-particles in 3D in presence of a magnetic field (included by minimal coupling)

$$H = \sum_{i=1}^{3N} \frac{(p_i - eA_i/c)^2}{2m} + V(\boldsymbol{q}_1, \boldsymbol{q}_2, ...\boldsymbol{q}_N) \tag{1}$$

The classical partition function can thus be evaluated

$$Z_c = \int e^{-\beta H} \, d\boldsymbol{q}_1 d\boldsymbol{p}_1 d\boldsymbol{q}_2 d\boldsymbol{p}_2 .....d\boldsymbol{q}_N d\boldsymbol{p}_N \tag{2}$$

A shift of the momentum variables $p'_i = p_i - eA_i/c$ removes the effect of the magnetic vector potential, thus any derivative of the partition function will not lead to magnetization i.e. magnetism. Hence we need quantum mechanics to study magnetism.

The Hamiltonian for a single electron in presence of magnetic field is

$$H = \frac{(\boldsymbol{p} - e\boldsymbol{A}/c)^2}{2m} + V(\boldsymbol{x}) + \mu_B \boldsymbol{\sigma} \cdot \boldsymbol{B} + \xi \boldsymbol{l} \cdot \boldsymbol{\sigma} \tag{3}$$

where the Zeeman and Spin-Orbit interaction terms are included.

$$\xi = \frac{\hbar}{4m^2c^2} \frac{1}{r} \frac{dV}{dr} \qquad \text{and} \qquad \mu_B = \frac{|e|\hbar}{2mc} \tag{4}$$

For a constant field $\boldsymbol{B}$, the vector potential $\boldsymbol{A} = \boldsymbol{B} \times \boldsymbol{r}/2$,

$$\frac{(\boldsymbol{p} - e\boldsymbol{A}/c)^2}{2m} = \frac{\boldsymbol{p}^2}{2m} - \frac{e}{2mc}(\boldsymbol{r} \times \boldsymbol{p}) \cdot \boldsymbol{B} + \frac{e^2}{8mc^2}(\boldsymbol{B} \times \boldsymbol{r})^2 \tag{5}$$

Where if the field is in the z-direction

$$\frac{(\boldsymbol{p} - e\boldsymbol{A}/c)^2}{2m} = \frac{\boldsymbol{p}^2}{2m} - \frac{e}{2mc}(\boldsymbol{r} \times \boldsymbol{p}) \cdot \boldsymbol{B} + \frac{e^2B^2}{8mc^2}(x^2 + y^2) \tag{6}$$

Thus the complete Hamiltonian is

$$H = \frac{\boldsymbol{p}^2}{2m} + \mu_B \left(\frac{\boldsymbol{l}}{\hbar} + \boldsymbol{\sigma} \right) \cdot \boldsymbol{B} + \frac{e^2B^2}{8mc^2}(x^2 + y^2) + V(\boldsymbol{x}) + \xi \boldsymbol{l} \cdot \boldsymbol{\sigma} \tag{7}$$

For a system with Z-electrons

$$H = \sum_{i=1}^{Z} \left[\frac{\boldsymbol{p}_i^2}{2m} + \mu_B \left(\frac{\boldsymbol{l}_i}{\hbar} + \boldsymbol{\sigma}_i \right) \cdot \boldsymbol{B} + \frac{e^2B^2}{8mc^2}(x_i^2 + y_i^2) + V(\boldsymbol{x}_i) + \xi \boldsymbol{l}_i \cdot \boldsymbol{\sigma}_i \right] + \frac{1}{2} \sum_{i \neq j} \frac{e^2}{|\boldsymbol{x}_i - \boldsymbol{x}_j|} \tag{8}$$

We can simply compute the magnetization and susceptibility from the derivatives of the average energy in the T=0 limit.

$$M = -\frac{\partial \langle H \rangle}{\partial B} \qquad \chi = N_0 \frac{\partial M}{\partial B} \tag{9}$$

I. DIAGMAGNETISM

Consider the case of system with no spin-orbit interaction and no permanent magnetic moment. (N_0 Avagadro's Number for molar quantities)

$$\left\langle \frac{l_{zi}}{\hbar} + \sigma_{zi} \right\rangle = 0 \tag{10}$$

Therefore

$$\begin{aligned} M &= -\frac{\partial \langle H \rangle}{\partial B} = -\frac{e^2 B}{4mc^2} \sum_{i=1}^{Z} \langle x_i^2 + y_i^2 \rangle \\ \chi_d &= N_0 \frac{\partial M}{\partial B} = -\frac{e^2 N_0}{4mc^2} \sum_{i=1}^{Z} \langle x_i^2 + y_i^2 \rangle \end{aligned} \tag{11}$$

On symmetry grounds

$$\sum_{i=1}^{Z} \langle x_i^2 \rangle = \sum_{i=1}^{Z} \langle y_i^2 \rangle = \frac{1}{3} \sum_{i=1}^{Z} \langle r_i^2 \rangle = \frac{1}{3} Z \langle r^2 \rangle \tag{12}$$

$$\chi_d = N_0 \frac{\partial M}{\partial B} = -\frac{Z e^2 N_0}{6mc^2} \langle r^2 \rangle \tag{13}$$

This is the atomic or diamagnetic susceptibility.

II. DE HAAS - VAN ALPHEN EFFECT

To understand this we need require the non-interacting Hamiltonian in presence of a magnetic field.

$$H = \sum_{i=1}^{N} \frac{(\boldsymbol{p} - e\boldsymbol{A}/c)^2}{2m} + \sum_{i=1}^{N} \mu_B \boldsymbol{\sigma} \cdot \boldsymbol{B} \tag{14}$$

where the first terms describes Landau levels, diamagnetism, and de Haas - van Alphen effect. The second term describes the Pauli paramagnetism.

A. Landau Levels

Considering the magnetic field in the z-direction, the vector potential can be chosen in the Landau gauge $\boldsymbol{A} = (0, Bx, 0)$. Thus the Schrodinger equation for the single electron Hamiltonian is

$$\left[\frac{(\boldsymbol{p} - e\boldsymbol{A}/c)^2}{2m} + \mu_B \boldsymbol{\sigma} \cdot \boldsymbol{B} \right] \psi = \varepsilon \psi \tag{15}$$

where

$$\begin{aligned} &\varepsilon_\sigma(n, k_z) = \left(n + \frac{1}{2} \right) \hbar\omega_c + \frac{\hbar^2 k_z^2}{2m} + \sigma \mu_B B \quad n = 0, 1, 2, ... \quad \sigma = \pm 1 \\ &\psi_{n,k_y,k_z,\sigma}(x, y, z) \propto \exp\left(-k_y y + i k_z z\right) H_n \left(\frac{x - k_y l_B^2}{l_B} \right) \exp\left(-\frac{(x - k_y l_B^2)^2}{2 l_B^2} \right) \end{aligned} \tag{16}$$

where

$$\omega_c = \frac{|e| B}{mc} \quad l_B = \sqrt{\frac{\hbar}{m\omega_c}} \tag{17}$$

The system is clearly periodic in y- and z-directions and thus k_y and k_z are integer multiples of $2\pi/L_y$ and $2\pi/L_z$. However, we can see from the wavefunction that we have a shifted Harmonic oscillator with the center as $k_y l_B^2$. The center needs to lie within the sample

$$-\frac{L_x}{2} \le k_y l_B^2 \le \frac{L_x}{2} \tag{18}$$

This implies that

$$-\frac{eBL_x}{2\hbar c} \le k_y \le \frac{|e|BL_x}{2\hbar c} \tag{19}$$

Thus the degeneracy of each LL is

$$\frac{k_y^{max} - k_y^{min}}{2\pi/L_y} = \frac{|e|BL_xL_y}{2\pi\hbar c} \tag{20}$$

Let us define $N_\sigma(\varepsilon)$ as the number of states with energy $\varepsilon_\sigma \le \varepsilon$. To obtain this quantity, we need to multiply the number of possible values of k_z by the LL degeneracy for given values of n and σ and energy $\varepsilon \le \varepsilon_\sigma(n, k_z)$.

$$k_z^{max/min} = \pm\frac{\sqrt{2m}}{\hbar}\sqrt{\varepsilon - \hbar\omega_c\left(n + \frac{1}{2}\right) - \sigma\mu_B B} \tag{21}$$

Therefore the number of states with energy $\varepsilon \le \varepsilon_n$ are

$$\frac{k_z^{max} - k_z^{min}}{2\pi/L_z} = \frac{L_z\sqrt{2m}}{\pi\hbar}\sqrt{\varepsilon - \hbar\omega_c\left(n + \frac{1}{2}\right) - \sigma\mu_B B} \tag{22}$$

Therefore all possible state with energy ε is

$$N_\sigma(\varepsilon) = \frac{|e|B\sqrt{2m}L_xL_yL_z}{2\pi^2\hbar^2 c}\sum_{n=0}^{n_{max}}\sqrt{\varepsilon - \hbar\omega_c\left(n + \frac{1}{2}\right) - \sigma\mu_B B} \tag{23}$$

where n_{max} is the maximum allowed n for which the square root is real.

B. Free Energy

For a given Hamiltonian of the kind

$$H = \sum_\alpha (\varepsilon_\alpha - \mu)\, n_\alpha \tag{24}$$

The thermodynamic potential in the grand canonical ensemble is

$$\Omega = -k_B T \log Z \tag{25}$$

where the partition function is given by

$$\begin{aligned} Z &= \sum_{\{n_\alpha = 0,1\}} \exp\left[-\beta H\right] = \sum_{\{n_\alpha=0,1\}} \exp\left[-\beta\sum_\alpha (\varepsilon_\alpha - \mu)\, n_\alpha\right] \\ &= \prod_\alpha \sum_{\{n_\alpha=0,1\}} \exp\left[-\beta\left(\varepsilon_\alpha - \mu\right) n_\alpha\right] \\ &= \prod_\alpha \left[1 + e^{-\beta(\varepsilon_\alpha - \mu)}\right] \end{aligned} \tag{26}$$

Therefore

$$\Omega = -k_B T \log Z = -k_B T \sum_\alpha \log\left[1 + e^{-\beta(\varepsilon_\alpha - \mu)}\right] \tag{27}$$

The Helmholtz Free Energy is thus

$$\begin{aligned} F &= \Omega + \mu N = N\mu - k_B T \log Z = N\mu - k_B T \sum_\alpha \log\left[1 + e^{-\beta(\varepsilon_\alpha - \mu)}\right] \\ &= N\mu - k_B T \sum_{\sigma=\pm 1} \int d\varepsilon \log\left[1 + e^{-\beta(\varepsilon-\mu)}\right] D_\sigma(\varepsilon) \end{aligned} \tag{28}$$

where $D_\sigma(\varepsilon)$ is the density of states with energy ε which can be written as

$$D_\sigma(\varepsilon) = \frac{dN_\sigma(\varepsilon)}{d\varepsilon} \tag{29}$$

where $N_\sigma(\varepsilon)$ is the number of states upto energy ε.

$$\begin{aligned} F &= N\mu - k_B T \sum_{\sigma=\pm 1} \int d\varepsilon \log\left[1 + e^{-\beta(\varepsilon-\mu)}\right] \frac{dN_\sigma(\varepsilon)}{d\varepsilon} \\ &= N\mu - \sum_{\sigma=\pm 1} \int d\varepsilon f(\varepsilon) N_\sigma(\varepsilon) \end{aligned} \tag{30}$$

where

$$f(\varepsilon) = \frac{1}{1 + e^{\beta(\varepsilon-\mu)}} \tag{31}$$

Therefore

$$F = N\mu - \frac{|e|B\sqrt{2m}L_x L_y L_z}{2\pi^2\hbar^2 c} \sum_{\sigma=\pm 1} \int d\varepsilon \frac{1}{1 + e^{\beta(\varepsilon-\mu)}} \sum_{n=0}^{n_{max}} \sqrt{\varepsilon - \hbar\omega_c\left(n + \frac{1}{2}\right) - \sigma\mu_B B} \tag{32}$$

Defining dimensionless variables

$$\mu_B = \frac{|e|\hbar}{2mc} \quad E_0 = \frac{\mu}{2\mu_B B} \quad E = \frac{\varepsilon}{2\mu_B B} \quad \Theta = \frac{k_B T}{2\mu_B B} \quad \Omega = L_x L_y L_z \tag{33}$$

In terms of which

$$\begin{aligned} F &= N\mu - \frac{|e|B\sqrt{2m}\Omega(2\mu_B B)^{3/2}}{2\pi^2\hbar^2 c} \sum_{\sigma=\pm 1} \int dE \frac{1}{1 + e^{(E-E_0)/\Theta}} \sum_{n=0}^{n_{max}} \sqrt{E - n - \frac{1}{2} - \frac{\sigma}{2}} \\ &= N\mu - \frac{4m^{3/2}\Omega(\mu_B B)^{5/2}}{\pi^2\hbar^3} \sum_{\sigma=\pm 1} \int dE \frac{1}{1 + e^{(E-E_0)/\Theta}} \sum_{n=0}^{n_{max}} \sqrt{E - n - \frac{1}{2} - \frac{\sigma}{2}} \\ &= N\mu - \frac{4m^{3/2}\Omega(\mu_B B)^{5/2}}{\pi^2\hbar^3} \sum_{\sigma=\pm 1} \int dE f(E) \sum_{n=0}^{n_{max}} \frac{2}{3}\frac{d}{dE}\left(E - n - \frac{1}{2} - \frac{\sigma}{2}\right)^{3/2} \\ &= N\mu + \frac{8m^{3/2}\Omega(\mu_B B)^{5/2}}{3\pi^2\hbar^3} \sum_{\sigma=\pm 1} \int_{-\infty}^{\infty} dE f'(E) \sum_{n=0}^{n_{max}} \left(E - n - \frac{1}{2} - \frac{\sigma}{2}\right)^{3/2} \\ &= N\mu + \alpha \sum_{\sigma=\pm 1} \int_{-\infty}^{\infty} dE f'(E) \sum_{n=0}^{n_{max}} \left(E - n - \frac{1}{2} - \frac{\sigma}{2}\right)^{3/2} \\ &= N\mu + \alpha \int_{-\infty}^{\infty} dE \left[f'(E+1/2) + f'(E-1/2)\right] \sum_{n=0}^{n_{max}} \left(E - n - \frac{1}{2}\right)^{3/2} \end{aligned} \tag{34}$$

To evaluate the above, we use the identity

$$\sum_{n=0}^{n_{max}} \left(E - n - \frac{1}{2}\right)^{3/2} = \sum_{l=-\infty}^{\infty} (-1)^l \int_0^E dx\, (E-x)^{3/2} e^{2\pi i l x} \tag{35}$$

Proof: Poisson Formula
A periodic function with period T can be Fourier expanded.

$$x(t) = \sum_{l=-\infty}^{\infty} x(l) e^{2\pi i l t/T} \tag{36}$$

The Fourier coefficients of the expansion are

$$x(l) = \frac{1}{T} \int_{-T/2}^{T/2} dt\, x(t) e^{-2\pi i l t/T} \tag{37}$$

Considering the periodic function to be a Dirac comb

$$x(t) = \sum_{n=-\infty}^{\infty} \delta(t - nT) \tag{38}$$

the corresponding coefficients are

$$\begin{aligned} x(l) &= \frac{1}{T} \int_{-T/2}^{T/2} dt \sum_{n=-\infty}^{\infty} \delta(t - nT) e^{-2\pi i l t/T} \\ &= \frac{1}{T} \int_{-T/2}^{T/2} dt\, \delta(t) e^{-2\pi i l t/T} \\ &= \frac{1}{T} \end{aligned} \tag{39}$$

Therefore

$$x(t) = \sum_{l=-\infty}^{\infty} x(l) e^{2\pi i l t/T} \Rightarrow \sum_{n=-\infty}^{\infty} \delta(t - nT) = \frac{1}{T} \sum_{l=-\infty}^{\infty} e^{2\pi i l t/T} \tag{40}$$

Choosing the period to be $T = 1$,

$$\sum_{n=-\infty}^{\infty} \delta(t - n) = \sum_{l=-\infty}^{\infty} e^{2\pi i l t} \tag{41}$$

Thus for a Dirac comb of the form $\sum_{n=-\infty}^{\infty} \delta(t - n - 1/2)$

$$\sum_{n=-\infty}^{\infty} \delta(t - n - 1/2) = \sum_{l=-\infty}^{\infty} e^{2\pi i l (t-1/2)} \tag{42}$$

If we now multiply the Poisson formula by a function and integrate as shown

$$\begin{aligned} \int_0^{\infty} dt\, f(t) \sum_{n=-\infty}^{\infty} \delta(t - n - 1/2) &= \int_0^{\infty} dt\, f(t) \sum_{l=-\infty}^{\infty} e^{2\pi i l (t-1/2)} \\ \Rightarrow \quad \sum_{n=0}^{\infty} f(n + 1/2) &= \int_0^{\infty} dt\, f(t) \sum_{l=-\infty}^{\infty} e^{2\pi i l (t-1/2)} \\ &= \sum_{l=-\infty}^{\infty} \int_0^{\infty} dt\, f(t) e^{2\pi i l t} e^{i\pi l} \\ &= \sum_{l=-\infty}^{\infty} \int_0^{\infty} dt\, f(t) e^{2\pi i l t} (-1)^l \end{aligned} \tag{43}$$

Now to consider a finite sum, we can construct a composite function $f(t)$ which is non-zero for $n < n_{max}$ and 0 for $n > n_{max}$. For $n = n_{max}$, $t = E$,

$$\sum_{n=0}^{n_{max}} f(n+1/2) = \sum_{l=-\infty}^{\infty} \int_0^E dt\, f(t) e^{2\pi i l t} (-1)^l \tag{44}$$

If $f(t) = (E-t)^{3/2}$,

$$\sum_{n=0}^{n_{max}} (E-n-1/2)^{3/2} = \sum_{l=-\infty}^{\infty} (-1)^l \int_0^E dt\, (E-t)^{3/2} e^{2\pi i l t} \tag{45}$$

To continue with the evaluation of the free energy

$$\begin{aligned}
\sum_{n=0}^{n_{max}} \left(E-n-\frac{1}{2}\right)^{3/2} &= \sum_{l=-\infty}^{\infty} (-1)^l \int_0^E dx\, (E-x)^{3/2} e^{2\pi i l x} \\
&= \int_0^E dx\, (E-x)^{3/2} + \sum_{l=1}^{\infty} \int_0^E dx\, (E-x)^{3/2} \left[(-1)^l e^{2\pi i l x} + (-1)^{-l} e^{-2\pi i l x}\right] \\
&= \int_0^E dx\, (E-x)^{3/2} + 2\sum_{l=1}^{\infty} \int_0^E dx\, (E-x)^{3/2} (-1)^l \mathrm{Re}\left[e^{2\pi i l x}\right] \\
&= \frac{2}{5}E^{5/2} + 2\sum_{l=1}^{\infty} \int_0^E dx\, (E-x)^{3/2} (-1)^l \mathrm{Re}\left[e^{2\pi i l x}\right] \\
&= \frac{2}{5}E^{5/2} + 2\sum_{l=1}^{\infty} (-1)^l \mathrm{Re}\left[\int_0^E dx\, (E-x)^{3/2} e^{2\pi i l x}\right] \\
&= \frac{2}{5}E^{5/2} + 2\sum_{l=1}^{\infty} (-1)^l \mathrm{Re}\left[\int_0^E dx\, (E-x)^{3/2} \frac{d}{dx}\frac{e^{2\pi i l x}}{2\pi i l}\right] \\
&= \frac{2}{5}E^{5/2} + 2\sum_{l=1}^{\infty} (-1)^l \mathrm{Re}\left[(E-x)^{3/2} \frac{e^{2\pi i l x}}{2\pi i l}\bigg|_0^E - \frac{3}{2}\int_0^E dx\, (E-x)^{1/2} \frac{e^{2\pi i l x}}{2\pi i l}\right] \\
&= \frac{2}{5}E^{5/2} + 2\sum_{l=1}^{\infty} (-1)^l \mathrm{Re}\left[-\frac{E^{3/2}}{2\pi i l} - \frac{3}{2}\frac{1}{2\pi i l}\int_0^E dx\, (E-x)^{1/2} \frac{d}{dx}\frac{e^{2\pi i l x}}{2\pi i l}\right]
\end{aligned} \tag{46}$$

the first term after the integration by parts is purely imaginary and thus can be dropped,

$$\begin{aligned}
\sum_{n=0}^{n_{max}} \left(E-n-\frac{1}{2}\right)^{3/2} &= \frac{2}{5}E^{5/2} + 2\sum_{l=1}^{\infty} (-1)^l \mathrm{Re}\left[-\frac{3}{2}\frac{1}{2\pi i l}\int_0^E dx\, (E-x)^{1/2} \frac{d}{dx}\frac{e^{2\pi i l x}}{2\pi i l}\right] \\
&= \frac{2}{5}E^{5/2} + 2\sum_{l=1}^{\infty} (-1)^l \mathrm{Re}\left[\frac{3}{2}\frac{E^{1/2}}{(2\pi i l)^2} + \frac{3}{4}\frac{1}{2\pi i l}\int_0^E dx\, (E-x)^{-1/2} \frac{e^{2\pi i l x}}{2\pi i l}\right] \\
&= \frac{2}{5}E^{5/2} + 2\sum_{l=1}^{\infty} (-1)^l \mathrm{Re}\left[-\frac{3E^{1/2}}{8\pi^2 l^2} - \frac{3}{16\pi^2 l^2}\int_0^E dx\, (E-x)^{-1/2} e^{2\pi i l x}\right] \\
&= \frac{2}{5}E^{5/2} - \sum_{l=1}^{\infty} (-1)^l \frac{3E^{1/2}}{4\pi^2 l^2} - \sum_{l=1}^{\infty} (-1)^l \frac{3}{8\pi^2 l^2} \mathrm{Re}\left[\int_0^E dx\, (E-x)^{-1/2} e^{2\pi i l x}\right]
\end{aligned} \tag{47}$$

Using

$$\sum_{l=1}^{\infty} \frac{(-1)^l}{l^2} = -\frac{\pi^2}{12} \tag{48}$$

$$\sum_{n=0}^{n_{max}} \left(E-n-\frac{1}{2}\right)^{3/2} = \frac{2}{5}E^{5/2} + \frac{E^{1/2}}{16} - \sum_{l=1}^{\infty} (-1)^l \frac{3}{8\pi^2 l^2} \mathrm{Re}\left[\int_0^E dx\, (E-x)^{-1/2} e^{2\pi i l x}\right] \tag{49}$$

substituting $x = E - u^2$,

$$\sum_{n=0}^{n_{max}} \left(E - n - \frac{1}{2}\right)^{3/2} = \frac{2}{5}E^{5/2} + \frac{E^{1/2}}{16} - \sum_{l=1}^{\infty}(-1)^l \frac{3}{4\pi^2 l^2}\text{Re}\left[\int_0^{\sqrt{E}} du\, e^{2\pi i l(E-u^2)}\right] \tag{50}$$

This can now be substituted in the expression for the free energy

$$\begin{aligned} F &= N\mu + \alpha\int_{-\infty}^{\infty} dE\,[f'(E+1/2) + f'(E-1/2)]\left(\frac{2}{5}E^{5/2} + \frac{E^{1/2}}{16} - \sum_{l=1}^{\infty}(-1)^l \frac{3}{4\pi^2 l^2}\text{Re}\left[\int_0^{\sqrt{E}} du\, e^{2\pi i l(E-u^2)}\right]\right) \\ &= F_o + F_{osc} \end{aligned} \tag{51}$$

where F_o contains the non-oscillatory terms whereas F_{osc} has the oscillatory terms.

$$F_{osc} = -\alpha\int_{-\infty}^{\infty} dE\,[f'(E+1/2) + f'(E-1/2)]\left(\sum_{l=1}^{\infty}(-1)^l \frac{3}{4\pi^2 l^2}\text{Re}\left[e^{2\pi i l E}\int_0^{\sqrt{E}} du\, e^{-2\pi i l u^2}\right]\right) \tag{52}$$

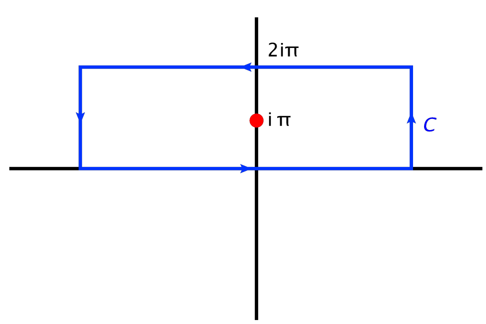
The u-integral is an error function. Since the Fermi function derivative is peaked about E_0 i.e. dimensionless chemical potential, we can replace the upper limit of u-integral by ∞

$$\begin{aligned} F_{osc} &= -\alpha\int_{-\infty}^{\infty} dE\,[f'(E+1/2) + f'(E-1/2)]\left(\sum_{l=1}^{\infty}(-1)^l \frac{3}{4\pi^2 l^2}\text{Re}\left[e^{2\pi i l E}\int_0^{\infty} du\, e^{-2\pi i l u^2}\right]\right) \\ &= -\alpha\int_{-\infty}^{\infty} dE f'(E)\left(\sum_{l=1}^{\infty}(-1)^l \frac{3}{4\pi^2 l^2}\text{Re}\left[(e^{2\pi i l(E+1/2)} + e^{2\pi i l(E-1/2)})\frac{e^{-i\pi/4}}{2\sqrt{2l}}\right]\right) \\ &= -\alpha\int_{-\infty}^{\infty} dE f'(E)\left(\sum_{l=1}^{\infty}(-1)^l \frac{3}{4\pi^2 l^2}\text{Re}\left[2\cos(\pi l)e^{2\pi i l E}\frac{e^{-i\pi/4}}{2\sqrt{2l}}\right]\right) \\ &= -\alpha\sum_{l=1}^{\infty}(-1)^l \frac{3}{4\sqrt{2}\pi^2 l^{5/2}}\cos(\pi l)\text{Re}\left[\int_{-\infty}^{\infty} dE f'(E) e^{2\pi i l E - i\pi/4}\right] \end{aligned} \tag{53}$$

Thus we need to evaluate the integral

$$\begin{aligned} \int_{-\infty}^{\infty} dE f'(E) e^{2\pi i l E - i\pi/4} &= -2\pi i l\int_{-\infty}^{\infty} dE f(E) e^{2\pi i l E - i\pi/4} \quad \text{ignoring the surface term in integration by parts} \\ &= -2\pi i l\int_{-\infty}^{\infty} dE \frac{1}{1+e^{(E-E_0)/\Theta}} e^{2\pi i l E - i\pi/4} \\ &= -2\pi i l\Theta\int_{-\infty}^{\infty} dy \frac{1}{1+e^{y}} e^{2\pi i l(\Theta y + E_0) - i\pi/4} \\ &= -2\pi i l\Theta e^{2\pi i l E_0 - i\pi/4}\int_{-\infty}^{\infty} dy \frac{1}{1+e^{y}} e^{2\pi i l\Theta y} \end{aligned} \tag{54}$$

To solve this consider the contour integral



$$\oint_C dz \frac{1}{1+e^z} e^{2\pi i l\Theta z} \tag{55}$$

where the function has poles at $z = \pm(2n+1)i\pi$. Choosing the contour of the form shown which includes a pole at $z = +i\pi$. Therefore

$$
\begin{aligned}
&\oint_C dz \frac{1}{1+e^z} e^{2\pi i l \Theta z} = 2\pi i \mathrm{Res}|_{i\pi} \\
&\int_{-\infty}^{\infty} dy \frac{1}{1+e^y} e^{2\pi i l \Theta y} + \int_{\infty}^{-\infty} dy \frac{1}{1+e^{y+2\pi i}} e^{2\pi i l \Theta (y+2\pi i)} = 2\pi i e^{-2\pi^2 l \Theta} \\
&\int_{-\infty}^{\infty} dy \frac{1}{1+e^y} e^{2\pi i l \Theta y} (1 - e^{-4\pi^2 l \Theta}) = 2\pi i e^{-2\pi^2 l \Theta} \\
&\int_{-\infty}^{\infty} dy \frac{1}{1+e^y} e^{2\pi i l \Theta y} = 2\pi i \frac{e^{-2\pi^2 l \Theta}}{(1 - e^{-4\pi^2 l \Theta})} = \frac{\pi i}{\sinh(2\pi^2 l \Theta)}
\end{aligned}
\tag{56}
$$

Therefore

$$
\begin{aligned}
\int_{-\infty}^{\infty} dE f'(E) e^{2\pi i l E - i\pi/4} &= -2\pi i l \Theta e^{2\pi i l E_0 - i\pi/4} \int_{-\infty}^{\infty} dy \frac{1}{1+e^y} e^{2\pi i l \Theta y} \\
&= e^{2\pi i l E_0 - i\pi/4} \frac{2\pi^2 l \Theta}{\sinh(2\pi^2 l \Theta)}
\end{aligned}
\tag{57}
$$

and thus the free energy

$$
\begin{aligned}
F_{osc} &= -\alpha \sum_{l=1}^{\infty} (-1)^l \frac{3}{4\sqrt{2}\pi^2 l^{5/2}} \cos(\pi l) \mathrm{Re}\left[\int_{-\infty}^{\infty} dE f'(E) e^{2\pi i l E - i\pi/4}\right] \\
&= -\alpha \sum_{l=1}^{\infty} (-1)^l \frac{3}{4\sqrt{2}\pi^2 l^{5/2}} \cos(\pi l) \mathrm{Re}\left[e^{2\pi i l E_0 - i\pi/4} \frac{2\pi^2 l \Theta}{\sinh(2\pi^2 l \Theta)}\right] \\
&= -\alpha \sum_{l=1}^{\infty} (-1)^l \frac{3}{4\sqrt{2}\pi^2 l^{5/2}} \frac{2\pi^2 l \Theta}{\sinh(2\pi^2 l \Theta)} \cos(\pi l) \cos(2\pi l E_0 - \pi/4) \\
&= -\alpha \sum_{l=1}^{\infty} (-1)^l \frac{3}{2\sqrt{2} l^{3/2}} \Theta \cos(\pi l) \frac{\cos(2\pi l E_0 - \pi/4)}{\sinh(2\pi^2 l \Theta)} \\
&= -\alpha \sum_{l=1}^{\infty} (-1)^l \frac{3}{2\sqrt{2} l^{3/2}} \frac{k_B T}{2\mu_B B} \cos(\pi l) \frac{\cos\left(\frac{\pi l \mu}{\mu_B B} - \frac{\pi}{4}\right)}{\sinh\left(\frac{\pi^2 l k_B T}{\mu_B B}\right)}
\end{aligned}
\tag{58}
$$

Given that the free energy has an oscillatory dependence on the magnetic field, the resultant measurements of magnetization also has oscillatory dependence.