HAIT Journal of Science and Engineering, Volume 1, Issue 2, pp. 235-240

Magnetic free energy of a two-dimensional metal I

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Received 15 September 2003, accepted 15 March 2004

Abstract

In this and a following note we present an essentially exact expression for the the steady and oscillatory (de Haas-van Alphen) free energy of a (noninteracting) spin-split ($g = 2$) two dimensional electron gas subject to a weak periodic potential and a normal uniform magnetic field. The sole restriction is to oscillations corresponding to electron orbits entailing at most one Bragg reflection from a reciprocal lattice line.

PACS: 73.20.Dx, 71.25.Hc

1 Introduction

Since oscillatory effects were first seen in the thermodynamic properties of two-dimensional materials [1, 2], activity in this area has grown significantly. There is particular interest in the class of charge transfer salts based on the bis(ethylenedithio)tetrafulvalene molecule (BEDT-TTF salts) [3] which form nearly ideal two-dimensional metals. A good deal of theoretical interest stems from the observation in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ and related compounds [4 - 6] of oscillations inconsistent with the highly successful semi-classical theory [7] developed for three-dimensional materials. A number of *ad hoc* explanations have been advanced to clarify this, which are by and large semi-classical in nature. These include the use of the Peierls substitution in empirical band models [8] and frequency modulation via oscillations in the chemical potential [9]. The need for a fully quantum mechanical treatment has been emphasized by Fortin and Ziman [10] who indicated how proper tunneling corrections could be grafted into the semi-classical network theory [11].

The aim of this note is to present a fully quantum mechanical expression for the thermodynamic potential for an ideal two-dimensional metal in a perpendicular uniform magnetic field. The sole limitation is in retaining only terms to second order in the ratio of the energy gaps at zone lines to the chemical potential. This is tantamount to using the usual nearly-free electron approximation and is complementary to the tight binding model introduced by Kim and Vagner [12], and presently extensively invoked. An excellent review of this work is contained in [13].

To this end, consider a two-dimensional noninteracting electron gas occupying an area L^2 in the xy plane subject to a weak lattice potential $V(\vec{r})$ and a normal uniform magnetic field $\vec{B}$. The system is described by the Hamiltonian

$$\mathcal{H} = \frac{1}{2m^*}(\vec{p} - \frac{e}{c}\vec{A})^2 + 2\mu_0 B S_z + V(\vec{r}) = \mathcal{H}_0 + V \tag{1}$$

with $\vec{A} = (-By, 0, 0)$. The electron g-factor is assumed to be 2, but it can easily be modified in the following formulas. The eigenstates of $\mathcal{H}_0$ are

$$\frac{1}{\sqrt{2^n n! L}}(\frac{eB}{\pi\hbar c})^{1/4} e^{-\frac{1}{2}(\eta-\eta_0)^2} H_n(\eta - \eta_0) e^{ikx} \chi_\sigma,$$

$$E(k, n, \sigma) = \mu_0^* B(2n + 1) + \mu_0 B\sigma, \tag{2}$$

$$\eta = (eB/\hbar c)^{1/2} y, \quad \eta_0 = -(\hbar c/eB)^{1/2} k, \quad \sigma = \pm 1.$$

The partition function to second order in V, where it is assumed that the average potential vanishes, is

$$Z(s) = Tr[e^{-s\mathcal{H}_0}] + \frac{1}{2}s^2 Tr[\int_0^1 du V(\vec{r}) e^{-s(1-u)\mathcal{H}_0} V(\vec{r}) e^{-su\mathcal{H}_0}]. \tag{3}$$

After carrying out the spin trace this has the form

$$Z(s) = 2\cosh(\mu_0 Bs)[z_0(s) + z_2(s)],$$

$$z_0(s) = L^2(m^*/2\pi\hbar)^2 \mu_0^* B)\text{csch}(\mu_0 * Bs). \tag{4}$$

Next the potential is expressed in terms of the reciprocal lattice vectors $\vec{K}$

$$V(\vec{r}) = \sum_{K\neq 0} V_{\vec{K}} e^{-i\vec{K}\cdot\vec{r}}. \tag{5}$$

To evaluate $z_2(s)$ we require the matrix element

$$M =< n, k|V(\vec{r})|n', k' >=$$

$$\frac{2\pi^{1/2}}{L} \sum_{K\neq 0} V_K \delta(k' - k - K_x) \int_{-\infty}^{\infty} d\eta e^{-\frac{1}{2}[(\eta-\eta_0)^2+(\eta-\eta_0')^2]}$$

$$e^{-i(\hbar c/eB)^{1/2}K_y\eta} \frac{H_n(\eta - \eta_0)}{\sqrt{2^n n!}} \frac{H_{n'}(\eta - \eta_0')}{\sqrt{2^{n'} n'!}}. \tag{6}$$

This gives

$$z_2(s) = \frac{s^2}{2\pi} \sum_{K,K'} V_K^* V_{K'} \int_{-\infty}^{\infty} dk dk' \delta(k' - k - K_x)\delta(k' - k - K_x')\cdot$$

$$\int_0^1 du \int_{-\infty}^{\infty} d\eta d\eta' e^{i(\hbar c/eB)^{1/2}(K_y'\eta' - K_y\eta)} S S' \tag{7}$$

with

$$S = \sum_{n=0}^{\infty} e^{-\frac{1}{2}[(\eta-\eta_0)^2+(\eta'-\eta_0)^2]} \frac{H_n(\eta - \eta_0)H_n(\eta' - \eta_0)}{2^n n!} e^{-su\mu_0^* B(2n+1)}. \tag{8}$$

Since

$$\sum_{n=0}^{\infty} e^{-\frac{1}{2}[X^2+Y^2]} \frac{H_n(X)H_n(Y)}{2^n n!} e^{-an} =$$

$$\frac{e^{a/2}}{\sqrt{2\sinh\ a}} \exp\{-\frac{1}{4}[(X+Y)^2 \tanh\frac{a}{2} + (X-Y)^2 \coth\frac{a}{2}]\}, \tag{9}$$

we have for $z_2(s)$

$$\frac{Ls^2}{8\pi^2} \sum_{K,K'} V_K^* V_K \delta_{K_x,K_x'} \int_{-\infty}^{\infty} dk dk' \int_0^1 \frac{du\delta(k - k' + K_x)}{\sqrt{\sinh(2\mu_0^* Bsu)\sinh(2\mu^* Bs(1-u))}}$$

$$\int_{-\infty}^{\infty} d\eta d\eta' e^{i(\hbar c/eB)^{1/2}(K_y'\eta' - K_y\eta)}$$

$$\exp\{-\frac{1}{4}[(\eta + \eta' - 2\eta_0)^2 \tanh(su\mu_0^* B) + (\eta + \eta' - 2\eta_0')^2 \tanh(s(1-u)\mu_0^* B)]-$$

$$\frac{1}{4}(\eta-\eta')^2[\coth(su\mu_0^*B)+\coth(s(1-u)\mu_0^*B)]\}. \quad (10)$$

The η integrations are essentially Gaussian and are easily worked out, following which we have

$$z_2(s)=\frac{Ls^2}{8\pi}(\frac{eB}{\hbar c})^{1/2}\sum_{K,K'}V_K^*V_{K'}\delta_{K_x,K_x'}\int_{-\infty}^{\infty}d\eta_0 d\eta_0'\cdot$$

$$\int_0^1\frac{du}{\sqrt{\sinh(\mu_0^*Bsu)\sinh(\mu_0^*Bs(1-u))}}\frac{1}{\sqrt{(T_1+T_1')(T_2+T_2')}}\cdot$$

$$\exp[i(eB/\hbar c)^{1/2}K_-\frac{\eta_0T_1+\eta_0'T_1'}{T_1+T_1'}]\exp[-\frac{T_1T_1'}{T_1+T_1'}(\eta_0-\eta_0')^2]\cdot$$

$$\exp[-\frac{1}{4}(\hbar c/eB)\left(\frac{K_-^2}{T_1+T_1'}+\frac{K_+^2}{T_2+T_2'}\right)]\delta(\eta_0'-\eta_0+(\hbar c/eB)^{1/2}K_x), \quad (11)$$

where $T_1=\tanh(\mu_0^*Bsu)$,$T_2=1/T_1$ and the prime on T denotes that u is replaced by $1-u$. $K_\pm=K_y'\pm K_y$. The η_0 integrations simply give delta functions and the result simplifies to

$$z_2(S)=\frac{L^2s^2}{8\pi}\left(\frac{eB}{\hbar c}\right)\mathrm{csch}(\mu_0^*Bs)\sum_{K\neq 0}|V_K|^2\cdot$$

$$\int_0^1\exp\left(-\frac{eB}{\hbar c}K^2\frac{\sinh(\mu_0^*Bsu)\sinh(\mu_0^*su'))}{\sinh(\mu_0^*Bs)}\right)du. \quad (12)$$

Finally, the partition function (per unit area) to second order in the lattice potential is

$$Z(s)=Z_0(s)+Z_2(s),$$

$$Z_0(s)=\frac{m^*}{2\pi\hbar^2}\mu_0^*B\frac{\cosh(\mu_0Bs)}{\sinh(\mu_0^*Bs)}, \quad (13)$$

$$Z_2(s)=\frac{m^*}{4\pi\hbar^2}(\mu^*Bs^2)\frac{\cosh(\mu_0Bs)}{\sinh(\mu_0^*Bs)}\sum_{\vec{K}\neq 0}|V_K|^2G(\vec{K},s),$$

$$G(\vec{K},s)=\int_0^1\exp\left[-\epsilon_K\frac{\sinh(\mu_0^*Bsu)\sinh(\mu_0^*Bs(1-u))}{(\mu_0^*B)\sinh(\mu_0^*Bs)}\right]du.$$

The free energy and magnetization corresponding to the term $Z_0(s)$ have been treated in detail in [14]. We have for the thermodynamic potential $\Phi=F-n\zeta$, where F is the Helmholtz free energy,

$$\Phi_0=-\frac{\pi m^*}{2\pi\hbar^2\beta}(\mu_0^*B)\int_{c-i\infty}^{c+i\infty}\frac{e^{\beta\zeta s}\cosh(\mu_0B\beta s)}{s\ \sin(\pi s)\sinh(\mu_0^*B\beta s)}\frac{ds}{2\pi i}. \quad (14)$$

For future reference we just consider the oscillatory part due to the imaginary poles $s_k = i\pi k/\mu_0^* B\beta$ $k = \pm 1, \pm 2, \ldots$

$$\Phi_0^{osc} = \frac{m^*\mu_0^* B}{\pi\hbar^2\beta}\sum_{k=1}^{\infty}\frac{(-1)^{k+1}}{k}\frac{\cos(m^*\pi k/m)\cos(\pi k\zeta/\mu_0^* B)}{\sinh(\pi^2 k/\mu_0^* B\beta)}. \qquad (15)$$

At zero temperature this can be summed exactly to give

$$\Phi_0 = \frac{m^*(\mu_0^* B)^2}{24\pi^3\hbar^2}([\pi^2 - 3(\lambda+\alpha)]_p + [\pi^2 - 3(\lambda-\alpha)]_p), \qquad (16)$$

where $\lambda = \zeta/\mu_0^* B$, $\alpha = m^*/m$ and $[]_p$ denotes the periodic extension from $[-\pi, \pi]$.

2 Effect of the lattice in a weak magnetic field

The lattice contribution to the free energy at $T = 0$ is

$$F_2 = \left(\frac{m^*}{4\pi\hbar^2}\right)(\mu_0^* B)\sum_{K\neq 0}|V_K|^2\int_{c-i\infty}^{c+i\infty} e^{\zeta s}\frac{\cosh(\mu_0 Bs)}{\sinh(\mu_0^* Bs)}G(K,s)\frac{ds}{2\pi s}. \qquad (17)$$

After expanding in ascending powers of $\mu_0^* B$, we have to second order,

$$F_2^{no} = \frac{m}{8\pi\hbar^2}\sum_{K\neq 0}|V_K|^2\int_{c-i\infty}^{c+i\infty}\frac{ds}{2\pi i}\frac{e^{\zeta s}}{s}\{\,{}_1F_1(1;3/2;-\frac{1}{4}\epsilon_K s)$$

$$+\frac{1}{2}(\mu_0^* B)^2[(\alpha^2 - \frac{1}{3})s^2\,{}_1F_1(1;3/2;-\frac{1}{4}\epsilon_K s) + \frac{\epsilon_K}{45}s^3\,{}_1F_1(3;7/2;-\frac{1}{4}\epsilon_K s)]\}, \qquad (18)$$

where ${}_1F_1$ is the confluent hypergeometric function. Finally, the inverse Laplace transforms of ${}_1F_1$ can be evaluated to yield

$$F_2^{no} = \left(\frac{m}{8\pi\hbar^2}\right)\sum_{K\neq 0}|V_K|^2\{[1 - \sqrt{1 - A_K}\Theta(1 - A_K)] +$$

$$\frac{1}{2}\left(\frac{\mu_0^* B}{\zeta}\right)^2[\frac{1}{4}(\alpha^2 - \frac{1}{3})A_K^2\frac{\Theta(1-A_K)}{(1-A_K)^{3/2}} + \frac{1}{6}A_K^2\frac{(\frac{3}{8}A_K^2 - A_K + 1)}{(1-4A_K)^{5/2}}\Theta(1-A_K)]\}, \qquad (19)$$

where $A_K = 4\zeta/\epsilon_K$ and Θ denotes the unit step function. Were the field dependence of ζ to be of no importance, the corresponding contribution to the susceptibility would be

$$\chi_0^{(2)} = \left(\frac{m}{8\pi\hbar^2}\right)\frac{(\mu_0^*)^2}{4\zeta^2}\frac{A_K^2\Theta(1-A_K)}{(1-A_K)^{5/2}}[(\alpha^2 - \frac{1}{3}) + \frac{2}{3}\frac{(\frac{3}{8}A_K^2 - A_K + 1)}{(1-4A_K)}]. \qquad (20)$$

The next note in this series will deal with the effect of the lattice potential on the free energy and magnetization. In particular, it should bear on various problems in interpreting the de Haas - van Alphen oscillations for various quasi 2D systems.

This work was supported under the NSF Grant DMR-0121146.

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