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Landau-level width: Magnetic-field and temperature dependences

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We calculate the effect of level broadening on the steady part of the magnetic moment and specific heat by deriving an analytic expression for the free energy of a two-dimensional electron gas in a uniform magnetic field, with an arbitrary Landau-level broadening and at a finite temperature. Measurements of these effects may provide a new way for gaining more information about the magnetic-field and temperature dependences of the Landau-level width. This in turn may lead to information about the density of states. In the particular case of Gaussian broadening, the broadening correction to the steady magnetic moment is as large as the steady magnetic moment itself.

I. INTRODUCTION

Two-dimensional electron systems, which can be created in semiconductor inversion layers and GaAs-GaAlAs superlattices, have been the subject of intensive investigations, both theoretically and experimentally.¹ The model of a two-dimensional electron gas (2D EG) was first employed by Peierls² in 1933 to explain the oscillatory magnetic susceptibility (the de Haas-van Alphen effect) of a three-dimensional metal. Because of the experimental realizations of a 2D EG, the development of the general theory for a 2D EG has been accelerated. Despite the increasing theoretical discussions on the subject, some problems remain to be considered.

The problem that we are concerned with here is the inconsistency between recent measurements of the density of states (DOS) of the 2D EG.³⁻⁹ Four different experimental approaches have been used to extract information on the DOS: cyclotron resonance,^{3,4} magnetic susceptibility,⁵⁻⁷ specific heat,⁸ and electric capacitance.⁹ Large discrepancies can be found among the DOS's derived from these experimental results. Determination of the DOS is obviously crucial in both theoretical and experimental studies on the 2D EG. We note that the experiments mentioned above are performed with very strong magnetic fields (~ 10 kG), which emphasize oscillatory behaviors. In this paper we shall propose measurements of broadening effects on the steady magnetic moment and specific heat, e.g., using weak magnetic fields, which we believe may generate a new way to obtain more information about the DOS in experimental studies.

In Sec. II, we formulate the theory of the free energy of a 2D EG, at finite temperature, with an arbitrary form of Landau-level broadening and an arbitrary g factor, and in a uniform magnetic field of arbitrary strength. The steady magnetic moment and the specific heat are then calculated. Some numerical aspects are also discussed. Conclusions are presented in Sec. III.

II. THE LEVEL-BROADENING DEPENDENCES FOR BOTH THE STATIC AND OSCILLATORY FREE ENERGY

For electrons in a magnetic field H the energy spectrum is given by

$$E_{l\sigma} = 2\mu_B H \left[\left(l + \frac{1}{2} \right) + g\sigma/4 \right], \quad l = 0, 1, 2, \dots, \quad \sigma = \pm 1, \quad (1)$$

where $\mu_B = e\hbar/2mc$ is the effective Bohr magneton, m is the effective mass of the electron, g is the Landé factor multiplied by m/m_0 , and m_0 is the electron mass in vacuum.

Suppose each Landau level is broadened by some mechanism, e.g., impurity scattering.^{10,11} Then instead of sharp δ functions, the density of states is the summation of a series of broadened functions $R_\Gamma(E) \equiv (D/\Gamma)R(E/\Gamma)$ say, at each Landau level:

$$D(E) = \sum_{l=0}^{\infty} \sum_{\sigma=\pm 1} \frac{D}{\Gamma} R \left[\frac{E - E_{l\sigma}}{\Gamma} \right], \quad (2)$$

where Γ is the full width at half maximum and $D = eH/2\pi\hbar c = 2.4 \times 10^{10} H \text{ cm}^{-2}$ (H measured in T) is the degeneracy per unit area, such that

$$\int_{-\infty}^{+\infty} R_\Gamma(E) dE \equiv D \int_{-\infty}^{+\infty} R(x) dx = D. \quad (3)$$

Henceforth, we shall always consider a unit area; a generalization to an arbitrary area is trivial.

The free energy is

$$\begin{aligned} F &= n\mu - kT \int_0^{+\infty} \ln \{ 1 + \exp[(\mu - E)/kT] \} D(E) dE \\ &= n\mu + \int_0^{+\infty} \phi(E) \frac{\partial f}{\partial E} dE, \end{aligned} \quad (4)$$

where μ is the Fermi energy, f is the Fermi-Dirac distribution function,

$$f(E) = \frac{1}{\exp[\beta(E - \mu)] + 1}, \quad (5)$$

and

$$\phi(E) = \int_0^E dE' \int_0^{E'} dE'' D(E''). \quad (6)$$

At low temperatures, $\mu/kT \gg 1$, $\partial f/\partial E$ is very close to a δ function at the Fermi energy. As long as the broadening is not too large, in other words, $\mu \gg \Gamma$, inclusions of negative l in Eq. (2) will not greatly influence

the free energy. This is equivalent to the assumption that the density of states given by Eq. (2) vanishes for negative energy. This approximation enables us to em-

ploy the Poisson summation formula.¹² The result is (see the Appendix for details)

$$F = n\mu - \frac{m}{\pi\hbar^2} \left[\frac{\mu^2}{2} + \left(\frac{g^2}{8} - \frac{1}{6} + A(\Gamma) \right) (\mu_B H)^2 + \frac{\pi^2}{6} (kT)^2 + G(\Gamma) (\mu_B H) \mu + 2(\mu_B H)^2 \sum_{l=1}^{\infty} \frac{(-1)^{l-1}}{l} \frac{kT}{\mu_B H} \operatorname{Re}[C_l(\Gamma)] \frac{\cos(gl\pi/2)\cos(\pi l\mu/\mu_B H)}{\sinh(\pi^2 l kT/\mu_B H)} \right], \quad (7)$$

where

$$A(\Gamma) = 2 \sum_{l=1}^{\infty} \frac{(-1)^l}{(\pi l)^2} \cos \left[\frac{gl\pi}{2} \right] \operatorname{Re}[C_l(\Gamma)] - \frac{g^2}{8} + \frac{1}{6}, \quad (8)$$

$$C_l(\Gamma) = \int_{-\infty}^{+\infty} dx R(x) \exp \left[\frac{\pi l \Gamma x i}{\mu_B H} \right], \quad (9)$$

and

$$G(\Gamma) = 2 \sum_{l=1}^{\infty} \frac{(-1)^l}{\pi l} \cos \left[\frac{gl\pi}{2} \right] \operatorname{Im}[C_l(\Gamma)]. \quad (10)$$

Here Re and Im denote the real and the imaginary parts, respectively.

After the free energy is obtained, various thermodynamical quantities can be calculated in a standard fashion. For instance, the Fermi energy is defined in terms of the number of electrons:

$$n = \int_0^{+\infty} dE D(E) \{ \exp[(E - \mu)/kT] + 1 \}^{-1} = 0 = \left[\frac{\partial F}{\partial \mu} \right]_{n,H,T}. \quad (11)$$

In a real 2D EG, the number density is mainly controlled by the gate voltage. If we ignore complications due to localized states, which are important to transport problems, the number density n does not vary with the magnetic field. When one evaluates physical properties, such as the magnetic moment, one should assume that n is a constant. In other words,

$$M = - \left[\frac{\partial F}{\partial H} \right]_{n,T,\mu}. \quad (12)$$

The magnetic moment thus calculated is also a function of μ , which varies with the magnetic field. It is understood that μ should be solved from Eq. (11) and substituted into Eq. (12), in order to obtain the explicit magnetic field dependence for the magnetic moment.

But the task of solving Eq. (11) for μ is often not easy. In 3D cases, especially when electrons are not free, i.e., when the band structure is important, the corresponding situation is worse. Blackman¹³ suggested that the Fermi energy, instead of the number density, can be taken to be

a constant when the magnetic field is not too strong. In other words, the magnetic moment is calculated through^{13,14}

$$M_\mu = - \left[\frac{\partial \Omega}{\partial H} \right]_{T,\mu} \Big|_{\mu=\mu_0}, \quad (13)$$

where μ_0 is the Fermi energy in the absence of magnetic field, and

$$\begin{aligned} \Omega(\mu) &= F - n\mu \\ &= -kT \int_0^{+\infty} \ln \{ 1 + \exp[(\mu - E)/kT] \} D(E) dE \end{aligned} \quad (14)$$

is the grand-canonical potential.

The above constant- μ calculation is based on the following arguments.^{13,14} It is transparent from Eq. (7) that magnetic oscillations are important when

$$H \gtrsim kT/\mu_B \sim T \frac{m}{m_0} \quad (T \text{ in K and } H \text{ in T}). \quad (15)$$

In most of the 3D experiments,¹⁵ magnetic fields used are such that Eq. (15) can be satisfied by only a small number of electrons with very small effective masses. Hence magnetic oscillations in 3D cases are usually very small. The oscillation of the Fermi energy will create a second-order correction to the magnetic moment, which explains why and when the Fermi energy can be taken as a constant. In a 2D EG, Eq. (15) is easily satisfied because the electron effective mass is very small (m/m_0 is 0.0665 for the GaAs-AlGaAs system). Hence we shall take the number density n , instead of the Fermi energy μ , as a constant to evaluate thermodynamical quantities throughout this paper. As we shall see, the discussion on the steady part of the magnetic moment is independent of whether n or μ is taken to be constant if the level broadening is symmetric.

Before calculating the steady magnetic moment and specific heat, we wish to compare our results with those in the literature. The pioneering discussions on Landau-level broadening effects have been given by Dingle.¹⁶ Through a semiclassical argument, he showed that sharp Landau levels are broadened into Lorentzian peaks. However, Dingle's formulation is three dimensional. Here, we concentrate on the 2D case. As it turns out, Dingle's result for the reduction factor of the

de Haas-van Alphen oscillation due to broadening is the same as our result [Eq. (9)].

Different expressions for the free energy of a 2D EG with symmetrically broadened Landau levels have been derived by Shoenberg¹⁷ for the cases where μ or n is constant, respectively. In the case of zero temperature and zero broadening (ideal case), and μ is a constant, the free energy was taken to be

$$F^0(\mu) = D \sum_{n=0}^r E_n \quad (16)$$

where E_r is the highest Landau level below the Fermi level ($E_r < \mu$). Then Shoenberg calculated the free energy (still at absolute zero) with broadening from the zero-broadening case, by using the relation

$$F(\mu) = \int_{-\infty}^{+\infty} F^0(\mu') R[(\mu - \mu')/\Gamma] d(\mu'/\Gamma). \quad (17)$$

The temperature effect can be included by considering a special form of the broadening function.

When n is a constant, the assumption made in Eq. (16) that all the Landau levels below the Fermi energy are fully occupied is removed in Shoenberg's calculation of the free energy in the ideal case. But his discussion in the case of broadening is again based on his constant- μ result [Eqs. (16) and (17) in this paper, or Eq. (16) in Ref. 17].

Since the highest occupied Landau level can be only partially occupied, no matter whether μ or n is a constant, Eq. (16) is not entirely adequate.¹⁸ When the Landau levels are broadened, the levels above (especially the one immediately above) the Fermi level can also contribute to the free energy. Since the summation in Eq. (16) excludes the $(r+1)$ th level and above, this contribution is also ignored in Eq. (17). As a result, in Shoenberg's formulation [see Eq. (16) in Ref. 17] the contribution due to broadening and temperature effects are absent in the steady part of the free energy.

The oscillatory part appears, at first sight, to be different from that obtained in Ref. 17. This is because the broadening function $R(x)$ is assumed to be an even function in Ref. 17, while this restriction has been dropped in our present formulation. In general, the broadening of the Landau levels is asymmetric with respect to the level centers. We present two reasons to support this argument. First of all, the problem of a charged harmonically bound electron in a blackbody heat bath has been solved exactly,¹⁹ and it was found that the energy levels are broadened asymmetrically. Secondly, if we consider the strong magnetic limit and impurity scattering only, the density of states can be obtained explicitly and the asymmetry of the broadening is shown to decrease as the magnetic field increases.²⁰

In the case of symmetric broadening, i.e., $R(x)$ is an even function, we have

$$C_l(\Gamma) = 2 \int_0^{+\infty} dx R(x) \cos \left[\frac{\pi l \Gamma x}{\mu_B H} \right]. \quad (18)$$

Hence the oscillatory part of the free energy in our result [Eq. (7)] reduces to that of Shoenberg [see Eq. (16) in Ref. 17]. There will be more discussions in the Appendix on the free energy in the case where the broadening is asymmetric.

An explicit form of symmetric broadening, i.e., the Lorentzian profile, which has been discussed by Dingle¹⁶ in the 3D case, has recently been considered in the 2D case by Isihara and Shiwa.¹²

In the particular case of symmetric broadening and where there is no spin splitting, i.e., $g=0$, it is easy to check that our result reduces to that in Ref. 12. But when $g \neq 0$, the steady part of our result differs from that in Ref. 12. In what follows we shall explain this discrepancy. In Ref. 21 where Landau levels are not considered to be broadened, the grand canonical potential with nonzero g , $\Omega_1(\mu)$, say, is obtained from that with $g=0$, $\Omega_0(\mu)$, say, as follows:

$$\Omega_1(\mu) = \frac{1}{2} \left[\Omega_0 \left[\mu + \frac{g}{2} \mu_B H \right] + \Omega_0 \left[\mu - \frac{g}{2} \mu_B H \right] \right]. \quad (19)$$

Here

$$\Omega_0(\mu) = -kT \int_0^{+\infty} \ln \{ 1 + \exp[(\mu - E)/kT] \} D_0(E) dE, \quad (20)$$

and $D_0(E)$ is the density of states without the spin splitting, which can be obtained by setting $g=0$ in Eqs. (1) and (2):

$$D_0(E) = 2 \sum_{l=0}^{\infty} \frac{D}{\Gamma} R \left[\frac{E - E_l}{\Gamma} \right], \quad (21)$$

and

$$E_l = 2\mu_B H \left(l + \frac{1}{2} \right). \quad (22)$$

But this way of calculating the grand canonical potential was employed by Isihara and Shiwa¹² in the case where Landau levels are broadened, which we shall show to be incorrect. By definition [Eqs. (21) and (22)], we have

$$D(E) = \frac{1}{2} \left[D_0 \left[E + \frac{g}{2} \mu_B H \right] + D_0 \left[E - \frac{g}{2} \mu_B H \right] \right]. \quad (23)$$

Substituting Eq. (23) into Eq. (14) we have

$$\begin{aligned} \Omega(E) &= -\frac{kT}{2} \int_{(g/2)\mu_B H}^{+\infty} \ln \{ 1 + \exp\{[\mu + (g/2)\mu_B H - E']/kT\} \} D_0(E') dE' \\ &\quad - \frac{kT}{2} \int_{-(g/2)\mu_B H}^{+\infty} \ln \{ 1 + \exp\{[\mu - (g/2)\mu_B H - E']/kT\} \} D_0(E') dE' \\ &\neq \Omega_1(E), \end{aligned} \quad (24)$$

which is the reason for the disagreement.

We now consider the steady part of the free energy, which dominates in the weak magnetic field limit, namely,

$$\mu_B H \lesssim kT. \quad (25)$$

Besides the steady part of the free energy without broadening,²¹

$$F^{(st)} = -\frac{m}{\pi\hbar^2} \left[\frac{\mu^2}{2} + \frac{\pi^2}{6} (kT)^2 + \left[\frac{g^2}{8} - \frac{1}{6} \right] (\mu_B H)^2 \right], \quad (26)$$

we have found the correction to the steady part of the free energy due to Landau-level broadening:

$$\Delta F^{(st)} = -\frac{m}{\pi\hbar^2} [A(\Gamma)(\mu_B H)^2 + G(\Gamma)(\mu_B H)\mu]. \quad (27)$$

As we mentioned, the temperature term in Eq. (26) and the broadening correction given by Eq. (27) have been ignored by Shoenberg.¹⁷

We shall first review the familiar results originated from the steady free energy without broadening. The third term in the bracket of Eq. (26) gives rise to the steady part of the specific heat of the 2D EG:

$$C_v^{(st)} = \frac{m\pi k^2 T}{3\hbar^2}. \quad (28)$$

Similarly, the second term gives the steady magnetic moment

$$M^{(st)} = \left[\frac{g^2}{4} - \frac{1}{3} \right] \frac{m\mu_B^2 H}{\pi\hbar^2}, \quad (29)$$

which consists of the following two parts: the spin paramagnetic moment and the Landau diamagnetic moment, their ratio being the same as in the 3D case.²¹

We next turn to an analysis of the corrections due to the Landau level broadening [Eq. (27)]. In addition to Eq. (29), a new term in the steady magnetic moment arises because of the level broadening, which is the following:

$$\Delta M^{(st)} = \frac{m}{\pi\hbar^2} \left[2A(\Gamma)\mu_B^2 H + G(\Gamma)\mu_B \mu \right. \\ \left. + [A'(\Gamma)(\mu_B H)^2 + G'(\Gamma)(\mu_B H)\mu] \frac{\partial\Gamma}{\partial H} \right], \quad (30)$$

where the primes mean derivatives with respect to Γ . Similarly, in addition to Eq. (28), the contribution to the steady part of the specific heat from the level broadening can be readily calculated:

$$\Delta C_v^{(st)} = -T \frac{\partial^2}{\partial T^2} (\Delta F^{(st)}) \\ = \frac{mT}{\pi\hbar^2} \left[[A''(\Gamma)(\mu_B H)^2 + G''(\Gamma)(\mu_B H)\mu] \frac{\partial\Gamma}{\partial T} \right. \\ \left. + [A'(\Gamma)(\mu_B H)^2 + G'(\Gamma)(\mu_B H)\mu] \frac{\partial^2\Gamma}{\partial T^2} \right]. \quad (31)$$

Now we consider some numbers. The first question relates to the size of the steady magnetic moment in comparison to the saturated magnetic moment. We obtain, by using Eq. (29),

$$\frac{M^{(st)}}{M_0} = \left[\frac{g^2}{4} - \frac{1}{3} \right] \frac{m\mu_B H}{\pi\hbar^2 n} \\ = \left[\frac{g^2}{4} - \frac{1}{3} \right] (n/D)^{-1} \\ = \left[\frac{g^2}{4} - \frac{1}{3} \right] \left[\frac{10^{11} H}{n} \right], \quad (32)$$

where $M_0 = n\mu_B$ is the saturated magnetic moment, n is the number of electrons per unit area (in cm^{-2}), D is the degeneracy per Landau level, and H is measured in T.

We now turn to a discussion of the contribution of broadening to the steady magnetic moment. Needless to say, $\Delta F^{(st)}$ given by Eq. (27) vanishes when $\Gamma = 0$, which is because $A(0) = G(0) = 0$, for any kind of broadening. Next, we consider an explicit form of broadening, Gaussian broadening, where¹¹

$$R_\Gamma(E) = \frac{D}{\pi^{1/2}\Gamma} \exp[-(E/\Gamma)^2], \quad (33)$$

with

$$\Gamma = 2 \left[\frac{\hbar\mu_B H}{\pi\tau} \right]^{1/2}, \quad (34)$$

and τ is the relaxation time. According to Eqs. (8)–(10), we have

$$G(\Gamma) = 0, \quad (35)$$

and

$$A(\Gamma) = 2 \sum_{l=1}^{\infty} \frac{(-1)^l}{(\pi l)^2} \exp \left[- \left[\frac{\pi l \Gamma}{2\mu_B H} \right]^2 \right] \\ \times \cos \left[\frac{g\pi l}{2} \right] - \frac{g^2}{8} + \frac{1}{6}. \quad (36)$$

The series in Eq. (36) converges very rapidly. In fact, it is a good approximation to keep only the first term. By using Eq. (30), we obtain

$$\Delta M^{(st)} \approx \frac{m\mu_B^2 H}{\pi\hbar^2} \left[\frac{1}{3} - \frac{g^2}{4} - \frac{2}{\pi^2} \exp \left[- \frac{\pi^2}{4} \left[\frac{\Gamma}{\mu_B H} \right]^2 \right] \right. \\ \left. \times \cos \left[\frac{g\pi}{2} \right] \right. \\ \left. \times \left[2 + \left[\frac{\Gamma}{\mu_B H} \right]^2 \right] \right]. \quad (37)$$

If we set⁷ $g \sim 0$ and $\Gamma \sim \mu_B H$ in Eq. (37), we know that $\Delta M^{(st)}$ can be as large as $M^{(st)}$ given by Eq. (29).

Next we consider the specific heat. It is straightforward to evaluate the specific heat in the strong magnetic limit. One finds that the maximum specific heat for the Gaussian broadening is the following:

$$C'_v = \frac{m\pi k^2 T}{3\hbar^2} \frac{2}{\pi^{1/2}} \frac{\mu_B H}{\Gamma}, \quad (38)$$

which will be of the same order of magnitude as the zero-broadening specific heat given by Eq. (28), since $\mu_B H \sim \Gamma$ in the strong-field limit. From Eq. (31) we know that the broadening corrections to the steady specific heat will be due to the temperature dependence of the Landau-level width.

Suppose one would like to measure $M^{(st)}$ in the weak-field limit, where $M^{(st)}$ dominates; how weak is the weak magnetic field? At a temperature of 4.2 K and in the case of GaAs, for which $m/m_0 = 0.0665$, Eq. (25) implies that $H \leq 0.5$ T, which is about an order of magnitude smaller than the "strong field" used in the experiments described in Ref. 7. We then go back to Eq. (32). The quantity n/D is the number of Landau levels occupied. For $H = 0.5$ T, $n = 5.4 \times 10^{11} \text{ cm}^{-2}$, $m/m_0 = 0.0665$, and $g \approx 0$, the ratio in Eq. (32) is 0.01. Additionally, the magnetic moment measured in a strong field is⁷ typically on the order of $0.1M_0$. We therefore know that the magnetic moments measured in the weak-field limit are about 1 order of magnitude smaller than those in strong field.

III. CONCLUSIONS

We have considered a two-dimensional electron gas in a uniform magnetic field with an arbitrary Landau level broadening. We have found that thermodynamical properties, such as the free energy, magnetic moment and the specific heat generally consist of steady and oscillatory parts. A nonzero Landau-level broadening contributes additional terms to the steady parts and multiplicative reduction factors to the oscillatory parts. In the particular case of Gaussian broadening, the broadening correction to the steady magnetic moment can be as large as the steady magnetic moment itself. We conclude that measurements of broadening effects on the steady magnetic moment and specific heat will reveal both the temperature and magnetic field dependences of the Landau-level width.

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APPENDIX: PROOF OF EQ. (7)

First we rewrite the density of states given by Eq. (2) as follows:

$$D(E) = \sum_{l=-\infty}^{+\infty} \frac{2D}{\Gamma} R_g \left[\frac{E - E_l}{\Gamma} \right], \quad (A1)$$

where

$$E_l = 2\mu_B H(l + \frac{1}{2}), \quad (A2)$$

$R_g(E/\Gamma)$

$$= \frac{1}{2} \left[R \left[\frac{E - g\mu_B H/2}{\Gamma} \right] + R \left[\frac{E + g\mu_B H/2}{\Gamma} \right] \right]. \quad (A3)$$

Here the terms with negative l are included in Eq. (A1) as an approximation. With the help of the Poisson summation formula

$$\sum_{l=-\infty}^{+\infty} a(l) = \int_{-\infty}^{+\infty} ds a(s) + 2 \sum_{l=1}^{\infty} \int_{-\infty}^{+\infty} ds a(s) \cos(2\pi ls), \quad (A4)$$

Eq. (A1) now has the following form:

$$D(E) = \frac{D}{\mu_B H} \left\{ 1 + 2 \sum_{l=1}^{\infty} \text{Re} \left[\exp \left[\frac{\pi l E i}{\mu_B H} \right] C_l(\Gamma) \right] \times (-1)^l \cos \left[\frac{g l \pi}{2} \right] \right\}, \quad (A5)$$

where $\text{Re}(\dots)$ means the real part of (\dots) and $C_l(\Gamma)$ is given by Eq. (9).

We shall then calculate $\phi(E)$ defined by Eq. (6). We obtain

$$\begin{aligned} \phi(E) = \frac{D}{\mu_B H} & \left\{ \frac{E^2}{2} + \left[\frac{g^2}{8} - \frac{1}{6} + A(\Gamma) \right] (\mu_B H)^2 + G(\Gamma) (\mu_B H) E \right. \\ & \left. + 2(\mu_B H)^2 \sum_{l=1}^{\infty} \frac{(-1)^{l-1}}{(\pi l)^2} \text{Re} \left[C_l(\Gamma) \exp \left[\frac{\pi l E i}{\mu_B H} \right] \right] \cos \left[\frac{g l \pi}{2} \right] \right\}, \quad (A6) \end{aligned}$$

where $A(\Gamma)$ and $G(\Gamma)$ are given by Eqs. (8) and (10), respectively.

We then substitute Eq. (A6) into Eq. (4) to evaluate the free energy. The integral involving the first three terms in Eq. (A6) can be carried out using the following standard formula:²¹

$$\begin{aligned} \int_0^{\infty} \phi(E) \frac{\partial f}{\partial E} dE = -\phi(\mu) - \frac{\pi^2}{6} (kT)^2 \frac{\partial^2 \phi}{\partial E^2} \Big|_{E=\mu} \\ + O[(kT)^4]. \quad (A7) \end{aligned}$$

Since the last term in Eq. (A6) oscillates very rapidly, we shall deal with it more carefully:

$$2 \int_0^{\infty} \exp \left[\frac{\pi l E i}{\mu_B H} \right] \frac{\partial f}{\partial E} dE$$

$$= \exp \left[\frac{\pi l \mu i}{\mu_B H} \right] \int_{-\mu/kT}^{\infty} \exp \left[\frac{2\pi i l Z k T}{\mu_B H} \right]$$

$$\times (\cosh Z)^{-2} dZ. \quad (\text{A8})$$

If we replace $-\mu/kT$ by $-\infty$, the result of the integral in Eq. (A8) is

$$-(2\pi^2 l k T / \mu_B H) / \sinh(\pi^2 l k T / \mu_B H).$$

Noting

$$\text{Re} \left[C_l(\Gamma) \exp \left[\frac{\pi l E i}{\mu_B H} \right] \right] = \text{Re} [C_l(\Gamma)] \cos \left[\frac{\pi l E}{\mu_B H} \right]$$

$$- \text{Im} [C_l(\Gamma)] \sin \left[\frac{\pi l E}{\mu_B H} \right]. \quad (\text{A9})$$

we have finally proved Eq. (7).

The special case where the broadening is symmetric, i.e., $R(x)$ is an even function, has been discussed in the text. Here we consider the more general case where $R(x)$ is arbitrary. We separate $R(x)$ into two parts, i.e., an even term and an odd term. In other words,

$$R(x) \equiv R^+(x) + R^-(x), \quad (\text{A10})$$

where

$$R^{\pm}(x) = \frac{1}{2} [R(x) \pm R(-x)], \quad (\text{A11})$$

and we have

$$R^{\pm}(x) = \pm R^{\pm}(-x). \quad (\text{A12})$$

Equation (9) reduces to the following:

$$C_l(\Gamma) = 2 \int_0^{+\infty} dx R^+(x) \cos \left[\frac{\pi l \Gamma x}{\mu_B H} \right]$$

$$+ 2i \int_0^{+\infty} dx R^-(x) \sin \left[\frac{\pi l \Gamma x}{\mu_B H} \right]$$

$$\equiv C_l^+(\Gamma) + i C_l^-(\Gamma). \quad (\text{A13})$$

Hence Eqs. (8) and (10) can be rewritten as

$$A(\Gamma) = 2 \sum_{l=1}^{\infty} \frac{(-1)^l}{(\pi l)^2} \cos \left[\frac{g l \pi}{2} \right] C_l^+(\Gamma) - \frac{g^2}{8} + \frac{1}{6}, \quad (\text{A14})$$

and

$$G(\Gamma) = 2 \sum_{l=1}^{\infty} \frac{(-1)^l}{\pi l} \cos \left[\frac{g l \pi}{2} \right] C_l^-(\Gamma). \quad (\text{A15})$$

Needless to say, when $R(x)$ is even,

$$R^-(x) = C_l^-(\Gamma) = G(\Gamma) = 0. \quad (\text{A16})$$

¹For a recent review, see, e. g., T. Ando, A. B. Fowler, and F. Stern, *Rev. Mod. Phys.* **54**, 437 (1982).

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