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## Ginzburg-Landau theory of diamagnetic phase transitions in a quasi-two-dimensional electron gas

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A microscopic derivation of the coefficients of the Ginzburg-Landau equation for the diamagnetic phase transition into a two-dimensional Condon-domain state is presented. Explicit expressions for the critical temperature and for the temperature-dependent coherence length are obtained for the first time. The effect of critical fluctuations on the diamagnetic susceptibility above the critical temperature is calculated and the experimental feasibility of observing this effect is briefly discussed.

It is well known that normal metals under the conditions of strong de Haas-van Alphen effects may undergo a diamagnetic phase transition to Condon domains.<sup>1</sup> A few years ago it was pointed out<sup>2</sup> that in a quasi-two-dimensional dense electron gas this diamagnetic instability may pin the chemical potential between Landau levels. This will drive a quantum Hall effect (QHE) within a single domain—an ideally conducting phase (ICP). In contrast to the conventional QHE, where the chemical potential is pinned in the Landau gap by disorder,<sup>3</sup> in the ICP state each phase plays the role of an electron reservoir for the other phase, keeping the chemical potential fixed in a midgap position. This is clearly an *intrinsic* QHE.

Condon domains can be observed as line splitting in nuclear magnetic resonance.<sup>4</sup> In Ref. 5 it was suggested that the nondissipative nature of the ICP state can be verified by the measurement of the spin-lattice relaxation time  $T_1(B)$  due to the hyperfine interaction between the conduction-electron spins and the nuclear spins. The experimental feasibility of NMR in two-dimensional (2D) electron gases was demonstrated for the first time in Ref. 6. Since within an ideally conducting phase the nuclear spin-lattice relaxation time  $T_1 \rightarrow \infty$ , the relaxation rate of the whole sample is dominated by the relaxation processes within the domain walls.

The interpretation of such an experiment requires the knowledge of the local field *B* within a domain wall. The calculation of this field in the general case, where the temperature is far below the critical temperature, is not an easy problem. The problem can be simplified considerably, however, near the critical temperature by using a Ginzburg-Landau- (GL-) like expansion of the thermodynamic potential. In the existing theory of the Condondomain walls, <sup>7-9</sup> the coefficients of the GL free-energy functional are not known explicitly. In two-dimensional electron gas, however, the free energy, magnetization, and susceptibility for the homogeneous system could be calculated with an exponential  $\propto \exp(-\hbar\omega_c/k_BT)$  accuracy.<sup>2,10</sup> Using these analytical expressions, we construct here a Ginzburg-Landau expansion for diamagnetic phase transitions in 2D electron gases.

Our model system consists of an anisotropic freeelectron gas in which the easy axes (x-y) are perpendicular to a uniform static magnetic field  $H_z$ . Taking into account the effect of magnetic interactions (MI) in a selfconsistent manner, the single-electron energies in this model are

$$\varepsilon_{n,k_z}(B) = (n + \frac{1}{2})\hbar\omega_c + \frac{1}{2}\Delta_z[1 - \cos(k_z d)] + \frac{1}{2}\hbar\omega_e\sigma,$$
(1)

where  $\omega_c \equiv eB/cm_{xy}^*$  is the in-plane cyclotron frequency,  $\omega_e = eB/cm_0$  is the electron-spin Larmor frequency,  $k_z$  is the electron wave number perpendicular to the x-y plane, and d is the distance between neighboring planes. The magnetic field B used in this expression is the local, selfconsistent field,  $B = H + 4\pi M(B)$  where M is the magnetization associated with all the other electrons in their cyclotron orbits. The condition  $m_{xy}^* \ll m_0$  permits neglect of the spin term in Eq. (1) and makes it easier to fulfill the condition of the quasi-two-dimensionality:  $\hbar \omega_c \gg \Delta_z$ . Under these circumstances the electron motion is very coherent and highly 2D, with the corresponding energy spectrum consisting of well-separated Landau levels.

Under the conditions of the diamagnetic instability the chemical potential  $\mu$  is pinned in a midgap position in each ideally conducting phase.<sup>2</sup> The corresponding fixed value of  $\mu$  is given by

$$\mu^* = \hbar \omega_c^{>} n_F = \hbar \omega_c^{<} (n_F + 1) = n_0 \frac{2\pi\hbar^2}{m_{xy}^*}$$

where

$$\omega_c^{>} \equiv eB_{>}/cm_{xy}^*, \ \omega_c^{<} \equiv eB_{<}/cm_{xy}^*,$$

 $B_{>} \equiv B_{n_{F}} = n_{0}\phi_{0}/n_{F}$ , and  $B_{<} \equiv B_{n_{F+1}} = n_{0}\phi_{0}/(n_{F}+1)$ , where  $n_{0}$  is the areal density of electrons, which is constant throughout the sample, and  $\phi_{0} \equiv hc/e$  is the flux quantum. Thus each phase plays the role of an electron reservoir for the other phase, keeping the chemical potential fixed in a midgap position.

The criterion for the diamagnetic instability in the systems under study

$$\chi_0 = n_F^2 \frac{\hbar e}{m_{xy}^* c\phi_0 d} > \frac{1}{4\pi} \,. \tag{2}$$

We may introduce a critical value  $n_c$  for  $n_F$  defined by

$$n_c^2 = \frac{1}{2} \left[ \frac{m_{xy}^*}{m_0} \right] \left[ \frac{d}{r_0} \right], \qquad (3)$$

where  $m_0$  is the free-electron mass and  $r_0 \equiv e^2/m_0c^2$  is the

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classical radius of the electron. In terms of  $n_c$  condition (2) takes the form

$$n_F > n_c . \tag{4}$$

In writing Eq. (1) it is implicitly assumed that the selfconsistent field *B* is spatially homogeneous. Below the critical field  $[B_c - n_0(\phi_0/n_c)]$  the translational symmetry in our system is, however, broken and the self-consistent field *B* becomes spatially inhomogeneous. If the field varies slowly over a cyclotron radius  $R_c - a_H(2n_F)^{1/2}$ , where  $a_H \equiv (c\hbar/eB)^{1/2}$  is the magnetic length, one may define local Landau levels by using an average of the inhomogeneous field over a cyclotron orbit in Eq. (1). For example, in the case where the domain structure is one dimensional, say along the x direction, we define the average field  $\mathbf{B}(x_0)$  for an electron orbit centered at  $x_0$  by (see Ref. 7)

$$B(x_0) = \frac{2}{\pi r_c^2} \int_{x_0}^{x_0 + r_c} B(x) y(x - x_0) dx$$
  
=  $\int K(x - x_0) B(x) dx$ , (5)

where

$$K(x-x_0) \equiv \begin{cases} \frac{2}{\pi r_c^2} \left( 1 - \frac{(x-x_0)^2}{r_c^2} \right)^{1/2}; \ |x-x_0| < r_c, \\ 0; \ |x-x_0| > r_c. \end{cases}$$

This definition is meaningful near the critical temperature since the coherence length characterizing the spatial variation of the field *B* is much larger than  $r_c$  (see later). Note that the spatial variation of B(x) removes the degeneracy of the Landau levels with respect to  $k_y$  through the dependence of  $\varepsilon_{n,k_z}(\mathbf{B}(x_0))$  on  $x_0 (-k_y a_H^2)$ . Thus we may write the grand canonical thermodynamical potential for our 2D electron-gas system near the critical temperature

$$\Omega = -k_B T \sum_{k_y} \sum_{n,k_z} \ln\left(\frac{\mu - \varepsilon_{n,k_z}(\mathbf{B}_{k_y})}{k_B T}\right), \qquad (6)$$

where  $\mathbf{B}_{k_y} \equiv \mathbf{B}(x_0)$ , as a spatial integral (over the orbital centers):  $L_z L_y \int dx_0 \omega(\mathbf{B}(x_0))$  (see Ref. 7).

Since B(x) varies between the values  $B \le n_0 \phi_0 / (n_F + 1)$  and  $B \ge n_0 \phi_0 / n_F$  of the field deep inside each domain, one may define a mean field for the domain-wall region by (see Fig. 1)

$$B_0 \equiv \frac{1}{2} \left( B_{<} + B_{>} \right) \simeq \frac{B_{n_F}}{1 - 1/2n_F} , \qquad (7)$$

and a fluctuating field by

$$B_1(x) \equiv B(x) - B_0, \qquad (8)$$

with  $|B_1(x)| < B_0/2n_F \ll B_0$ . We thus choose to expand the appropriate free energy<sup>7-9</sup>

$$G = \Omega + \frac{1}{8\pi} \int B^2 d^3 r \tag{9}$$

about  $B_0$  to second order in the fluctuating field  $B_1$ . In terms of the Fourier transform  $B_1(q)$  of  $B_1(x_0) - B(x_0)$ 



FIG. 1. (a) Schematic illustration of the H vs B diagram for the homogeneous electron gas near the critical temperature  $T_c$ , and (b) the corresponding domain-wall profile obtained by expanding the free energy of the inhomogeneous system about  $B_0$ .

 $-B_0$ , with respect to  $x_0$  this can be written in the form

$$G = G(B_0) + \frac{1}{4\pi} L_z L_y \int dq |B_1(q)|^2 \times \left(\frac{1}{4\pi} - \chi(B_0) |K(q)|^2\right),$$
(10)

with  $\chi(B_0) = -\partial^2 \omega / \partial B_0^2$ , and  $K(q) = 2j_1(qr_c)/qr_c$ , where  $j_1$  is the spherical Bessel function of order 1.<sup>7</sup> The linear term of this expansion vanishes since  $\partial \omega / \partial B_0 = -M(B_0) = 0$  (see Fig. 2).

Since the variation of  $B_1(x)$  is very slow over a length scale  $r_c$ , we can minimize this free energy with respect to



FIG. 2. The variation of the magnetization M as a function of B around  $B_0$  for the homogeneous electron gas. The different graphs correspond to different temperatures with  $\alpha \equiv \hbar \omega_{c,0}/2k_BT$ .

 $B_1^*(q)$  for  $qr_c \ll 1$ . The result is

$$B_1(q)\left(\frac{1}{4\pi}-\chi(B_0)\right)+\chi(B_0)r_c^2\frac{q^2B_1(q)}{4\pi},$$
(11)

or in real space

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$$\left(\frac{1}{4\pi}-\chi(B_0)\right)B_1(x)-\xi_0^2\frac{\partial^2 B_1(x)}{\partial x^2},\qquad(11')$$

with  $\xi_0^2 \equiv \frac{1}{4} r_c^2 \chi(B_0)$ . This expansion may be continued up to third order in  $B_1$  with the result

$$\left[\frac{1}{4\pi}-\chi(B_0)\right]B_1(x)-\xi_0^2\frac{d^2}{dx^2}B_1(x)-\left[\frac{\partial\chi}{\partial B_0}\right]B_1^2(x)-\frac{1}{3}\frac{\partial^2\chi}{\partial B_0^2}B_1^3(x).$$
(12)

The coefficients of this expansion can be directly calculated from the closed analytical expression for M(B) obtained in Ref. 2. A graphical illustration of this function for various temperatures is shown in Fig. 2. It is clearly seen, just from the symmetry of the graphs, that all the odd derivatives vanish at  $B_0$ . The expressions for the susceptibility  $\chi(B_0)$  and its first and second derivatives are thus

$$\chi(B_0) - \chi_0 - \chi_0 \frac{4k_B T}{\hbar \omega_{c,0}}, \qquad (13a)$$

$$\left(\frac{\partial \chi}{\partial B}\right)_{B-B_{0}} = 0, \qquad (13b)$$

$$\left(\frac{\partial^2 \chi}{\partial B^2}\right)_{B=B_0} = -\frac{\chi_0}{b_0^2} \frac{8k_B T}{\hbar \omega_{c,0}},$$
 (13c)

where  $\omega_{c,0} \equiv eB_0/m_{xy}^*c$  and

$$b_0 \equiv \frac{B_0}{2n_F} \,. \tag{14}$$

Thus the resulting equation for  $b_1(x) \equiv B_1(x)/b_0$  has a GL-like form

$$\epsilon b_1(x) - \xi_0^2 \frac{d^2}{dx^2} b_1(x) + b_1^3(x) = 0, \qquad (15)$$

where the coherence length  $\xi_0$  is given by

$$\xi_0^2 \equiv \frac{3}{32} r_c^2 \frac{k_B T}{\hbar \omega_{c,0}} , \qquad (16)$$

and  $\epsilon \equiv \frac{3}{2} [(T - T_c)/T]$ . The critical temperature  $T_c$  can be readily obtained from the requirement  $1/4\pi - \chi(B_0)$ -0 with  $\chi(B_0)$  given by Eq. (13a). The result is

$$T_{c} = \frac{\hbar \omega_{c,0}}{4k_{B}} \left[ 1 - \frac{n_{c}^{2}}{n_{F}^{2}} \right], \qquad (17)$$

where  $n_c$  is the critical value of  $n_F$ , given by Eq. (3), or in terms of the critical field  $B_c$  by  $n_c = n_0\phi_0/B_c$ .

The nonuniform solutions of Eq. (15) below  $T_c$  are

$$b_1(x) = \pm \sqrt{|\epsilon|} \tanh\left[\frac{x}{\xi(T)}\right],$$
 (18)

where  $\xi(T)$  is the temperature-dependent coherence length given by

$$\xi(T) \equiv \frac{\sqrt{2}\xi_0}{\sqrt{|\epsilon|}} \,. \tag{18'}$$

Each of these solutions describes a domain wall with spatial size of order  $\xi(T)$  and field variation between  $B < -B_0 - (\sqrt{|\epsilon|}/2n_F)B_0$  and  $B > -B_0 + (\sqrt{|\epsilon|}/2n_F)B_0$ , the fields deep inside the domains.

Note that for the expansion (12) to converge,  $|b_1|$  should be smaller than one and the coherence length

$$\xi(T) = \frac{\sqrt{3}}{4} \frac{r_c}{\sqrt{|\epsilon|}} \left(\frac{k_B T}{\hbar \omega_{c,0}}\right)^{1/2}$$

should be longer than the cyclotron radius  $r_c$ . Both of these conditions require  $|\epsilon| \ll 1$ , i.e., that T should be sufficiently close to the critical temperature  $T_c$ . Then the field variation between two neighboring domains  $\Delta B(T)$  $\equiv B_>(T) - B_<(T) = (\sqrt{|\epsilon|}/n_F)B_0$  is much smaller than in the low-temperature limit  $T \ll T_c$ , for which  $\Delta B = B_0/n_F$  and a QHE is in action within each domain. In this low-temperature limit our approach is no longer valid since all the derivatives of M(B) with respect to B both at  $B_<$  and  $B_>$  are nearly divergent (see Fig. 2).

Note also the analogy between Eq. (15) and the GL equation describing a domain wall between the superconducting and the normal regions in the intermediate state of a type-I superconductor (see also Refs. 7-9).

A straightforward application of the GL expansion derived above is the calculation of the effect of critical fluctuations on the orbital susceptibility above  $T_c$ . This is done by considering our expression for the GL free-energy functional  $G[B_1]$  to second order in  $B_1$  [Eq. (10)] and by constructing the partition function for all possible configurations of the fluctuating field  $B_1(q)$ :

$$Z = \prod_{\mathbf{q}} \int dB_1(\mathbf{q}) \exp\left[-\left(\frac{1}{4\pi} - \chi(B_0) \,|\, K(q) \,|^2\right) \frac{|B_1(\mathbf{q}) \,|^2}{k_B T}\right] = \prod_{\mathbf{q}} \left(\frac{\pi k_B T}{\left[1/4\pi - \chi(B_0) \,|\, K(q) \,|^2\right]}\right)^{1/2}.$$
(19)

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The susceptibility fluctuations above  $T_c$  is obtained by differentiating twice with respect to  $B_0$  the free energy of the fluctuations  $F = -k_B T \ln Z$ 

$$\chi_{\rm fluc} = -\frac{1}{V} \frac{\partial^2 F}{\partial B_0^2}$$
$$= \frac{1}{2} \left( \frac{\partial^2 \chi}{\partial B_0^2} \right) \int d^3 q \frac{k_B T |K(q)|^2}{(1/4\pi - \chi(B_0) |K(q)|^2)^2}.$$
(20)

Using the long-wavelength expansion applied in deriving Eq. (11) for our 2D system the integral in Eq. (20) can be readily evaluated with the result

$$\frac{\chi_{\text{fluc}}}{\chi(B_0)} \simeq -\left(\frac{3\pi^2}{8}\right) \left(\frac{n_F}{n_c}\right)^3 \left(1 - \frac{n_c^2}{n_F^2}\right)^3 \left(\frac{T_c}{T - T_c}\right) \frac{1}{n_c} ,$$
(21)

where we take  $\chi(B_0) = 1/4\pi$  for  $T \ge T_c$ .

This critical diamagnetic fluctuation effect becomes quite significant by reducing the magnetic field below the critical field  $B_c$ , that is, by increasing  $n_F$  above the critical value  $n_c$ . By doing so one increases both the amplitude  $(n_F/n_c)^3(1-n_c^2/n_F^2)^3$  of the susceptibility fluctuation in

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Eq. (21) and the Ginzburg critical region

$$\Delta T_c = \left(\frac{3\pi^2}{8}\right) \frac{1}{n_c} \left(\frac{n_F}{n_c}\right)^3 \left(1 - \frac{n_c^2}{n_F^2}\right)^3 T_c$$
$$= \left(\frac{3\pi^2}{32}\right) \frac{1}{n_c} \left(\frac{\hbar\omega_{c,0}}{k_B}\right) \left(1 - \frac{n_c^2}{n_F^2}\right)^4 \left(\frac{n_F}{n_c}\right)^3. \tag{22}$$

Let us estimate this fluctuation effect for possible candidates for exhibiting ICP, say, graphite intercalation compounds GIC's.<sup>11</sup> Using typical numbers for AsF<sub>5</sub> GIC's, i.e., d = 10 Å,  $m_{xy}^* = 0.1m_0$ , and  $E_F = 1$  eV, we have  $n_c = 120$  and condition (4) can be satisfied if the applied magnetic field H is smaller than 10 T. For these values of the parameters we get, from Eq. (22), a critical width  $\Delta T_c$ of the order of 2 K, when the applied field is about onehalf (i.e., about 5 T) of the critical field  $B_c$ . At H = 20 T the magnetic gap  $\hbar \omega_c$  is of the order of 100 K, much larger than the Landau-level broadening.<sup>11</sup> The Landaulevel broadening at lower magnetic fields<sup>12</sup> might, however, smear out this critical fluctuation effect.

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