

# Thermodynamics of the Domain Structure and Determination of the Transition Fields in Type-1 Superconductors

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A detailed electromagnetic and thermodynamical theory of the intermediate state in type-1 superconductors is worked out in the Landau laminar geometry. It is shown that the Landau  $s$  domain bounded by a convex wall where the magnetic field very nearly approaches the critical value is likely to be near the most stable configuration. The analysis of the field distribution in the  $n$  laminas provides a qualitative explanation of the experimentally observed small importance of the branching that Landau first suggested. Explicit analytical expressions of the various contributions to the free energy are derived as functions of the applied magnetic field and of the superconductive fraction. This allows the exact formulation of the equations yielding the transition fields at the ends of the intermediate range. At the lowest order, the departure of the field from the ideal value varies predominantly as  $(\Delta/l)^{1/3}$  at the diamagnetic intermediate-phase transition and as  $(\Delta/l)^{1/2}$  at the intermediate normal-phase transition,  $\Delta$  being the surface energy and  $l$  the thickness of the sample.

## I. INTRODUCTION

It is well known that a type-1 superconductor whose geometrical shape permits the setting up of a uniform internal field with a definite demagnetizing coefficient  $\nu$  undergoes a transition into an intermediate state as the external applied field  $H_0$  approaches the value  $(1 - \nu) H_c$ , where  $H_c$  is the critical field.

Landau<sup>1</sup> first proved the thermodynamical possibility of an intermediate state, earlier mentioned by Peierls,<sup>2</sup> consisting of alternating laminas of superconducting ( $s$ ) and normal ( $n$ ) phases parallel to the field, and derived an expression for the period of the structure in terms of a tabulated function.<sup>3</sup>

Direct observations of the domains by various means<sup>3-8</sup> revealed the agreement of Landau's point of view with experiment. Typical values of the measured period were within a few hundred microns.

The first picture of the intermediate state was refined in 1943 by Landau,<sup>9</sup> who suggested a "branched model" consisting of repeated splittings of  $n$  laminas approaching the surface to avoid the difficulty of having a subcritical region in these laminas. However, this model, which predicts the impossibility of direct observation of the domain structure at the surface of the sample, is not consistent with the observations<sup>3-8</sup> which revealed no branching or only an occasional branching with a few splittings.<sup>6</sup>

In the present paper a complete calculation of the free energy of an ellipsoidal sample is carried out, with some simplifying assumptions. The shape of the normal-superconducting ( $n$ - $s$ ) wall in a cross section of the domains is assumed to belong to a particular family, allowing an exact determination of the field distribution by conformal mapping. This

family includes Landau's critical wall, which turns out to be about the best choice given by the minimum of the suitable thermodynamical potential. The detailed expression of the free energy permits a calculation of the equilibrium field in the  $n$  laminas and the study of the transition fields at the ends of the intermediate range (Sec. V).

In this paper both the study of a thermodynamical potential fitting the present physical situation and the buildup of a "simple model" are investigated (Sec. II). The effects due to the ends of the domains are not taken into account so as to make clearer their contributions to the thermodynamical potential, which is analyzed later on (Secs. III-IV).

## II. SIMPLE MODEL

From the magnetostatic point of view the supercurrents around the sample in the diamagnetic state or around the  $s$  domains in the intermediate state can be replaced by a distribution of magnetic polarization intensity  $\vec{J}(\vec{r})$ . It can be shown through elementary thermodynamics that the process of magnetization, whether it is obtained with a permanent magnet or an external current, does not affect the thermodynamical potential  $F$ . Thus the magnetic contribution is given by

$$W_M = -\mu_0 \int \int \vec{J} \cdot \delta \vec{H}_0 d^3r. \quad (1)$$

The integration is extended to magnetized matter and to the magnetization process. Disorder terms may be dropped as insignificant. When the normal state is taken as a reference, the total thermodynamical potential or free energy is

$$F = W_C + W_S + W_M, \quad (2)$$

where

$$W_C = -\frac{1}{2} \mu_0 H_c^2 \int_s d^3r$$

is the condensation energy of the superconductive fraction of the sample and

$$W_S = \frac{1}{2} \Delta \mu_0 H_c^2 \int_s d^2r$$

is the surface energy of the  $n$ - $s$  walls, characterized by the usual parameter  $\Delta$ .

Consider now an ellipsoidal superconducting sample of demagnetizing coefficient  $\nu$  set in a magnetic field, taken parallel to a symmetry axis for convenience (Fig. 1).

In the diamagnetic state a uniform magnetization intensity  $J$  parallel to the field takes place inside the sample and results in a demagnetizing field  $-\nu J$ .  $J$  is given by the Meissner condition

$$B = \mu_0(H_0 - \nu J + J) = 0,$$

$$J = -H_0/(1 - \nu).$$

The resulting free energy, as deduced from (2), is

$$F_D = -\frac{1}{2} \mu_0 H_c^2 V + \frac{1}{2} \mu_0 H_0^2 V/(1 - \nu).$$

$V$  is the volume of the sample. It is convenient to introduce the reduced magnetic field  $h_0 = H_0/H_c$  and the reduced free energy per unit volume

$$f_D = F_D/(\frac{1}{2} \mu_0 H_c^2 V) = -1 + h_0^2/(1 - \nu). \quad (3)$$

In the intermediate state matter undergoes a splitting which, on a small scale, may be considered as periodic with a period  $a$ . Each period consists of an  $s$  lamina with width  $sa$ , and an  $n$  lamina with width  $na = (1 - s)a$ .  $s = 1 - n$  is the superconductive fraction of any section of the sample perpendicular to the field. Since the number of laminas

is very large, the total area of the  $n$ - $s$  walls is  $2V/a$ , whatever the real geometrical disposition of the laminas.

The magnetization  $J$  in the  $s$  laminas is uniform, except perhaps in a region of extension  $\sim a$  near the external surface. Thus the whole sample behaves, on a large scale, as if it had an uniform magnetization  $\bar{J} = sJ$ , giving rise to a demagnetizing field  $-\nu sJ$ .

As a result the field inside the  $n$  laminas is

$$H_n = H_0 - \nu sJ,$$

while, in the  $s$  laminas, the Meissner condition gives

$$H_0 - \nu sJ + J = 0 \quad \text{or} \quad J = -H_0/(1 - \nu s), \quad (4)$$

whence

$$H_n = H_0/(1 - \nu s). \quad (5)$$

As the contribution of an external very thin disturbed layer is negligible, the free energy  $F_I$  of the system can be easily obtained when  $a$  and  $s$  are given arbitrarily set values:

$$F_I = \frac{V}{2} \mu_0 H_c^2 \left( \frac{2\Delta}{a} - s \right) + \mu_0 \int_0^{H_0} \frac{H_0 \delta H_0}{1 - \nu s} d^3r$$

or

$$f_I = \frac{2\Delta}{a} - s + \frac{s}{1 - \nu s} h_0^2. \quad (6)$$

It will be shown later that the equilibrium value of  $a$  is much larger than  $\Delta$ , so that  $2\Delta/a$  may be dropped. The remaining expression has a minimum with respect to  $s$  when

$$1 - \nu s = h_0 \quad \text{or} \quad s = (1 - h_0)/\nu,$$

and, by inserting this value into (4) and (5), we

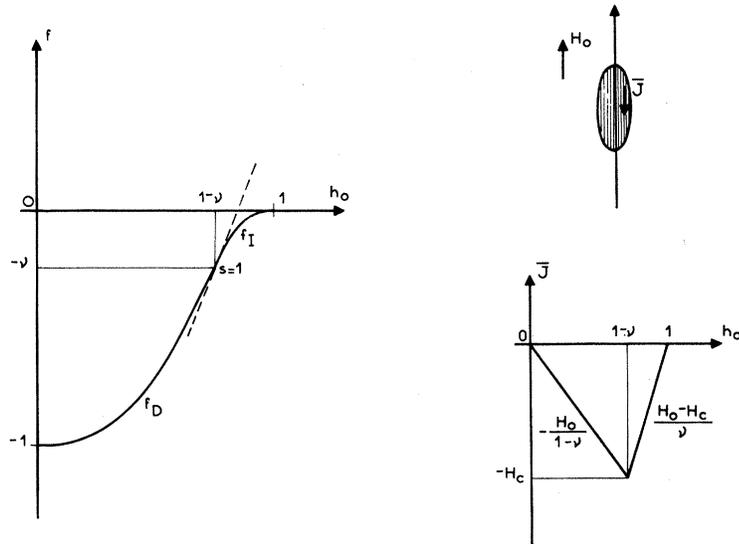


FIG. 1. Free-energy and macroscopic magnetization of an ellipsoidal sample, in the simple model, as functions of the reduced applied field  $h_0$  (demagnetizing factor  $\nu$ ).

have

$$J = -H_c, \quad H_n = H_c.$$

Therefore, in the present model the field takes on exactly the critical value inside the  $n$  laminas. From Eq. (6) the minimum reduced free energy is found to be

$$f_I = -(1 - h_0)^2 / \nu.$$

The transition from the diamagnetic to the intermediate state occurs at a field defined by the equation  $f_D = f_I$ , which yields the usual value

$$h_0 = 1 - \nu.$$

The free energy and the macroscopic magnetization for both states are plotted in Fig. 1 as functions of the applied field  $h_0$ . We have

$$\begin{aligned} \bar{J}_D = J_D &= -\frac{H_0}{1 - \nu}, \\ \bar{J}_I &= -\frac{sH_0}{1 - \nu s} = \frac{H_0 - H_c}{\nu}. \end{aligned} \quad (7)$$

The free-energy curve consists of two arcs of a parabola which have the same tangent with a slope 2 at the transition point.

### III. FIELD DISTRIBUTION

The foregoing model is not able to yield the equilibrium value of the period  $a$ , since the contribution of the domain ends to the free energy has been neglected. We now work out this contribution.

For this purpose we set up a somewhat more ideal geometry by considering a semi-infinite sample bounded by the  $x0y$  plane. The periodic array of  $s$  and  $n$  laminas is parallel to  $y0z$  (Fig. 2). However, the external ellipsoidal shape of the sample is taken into account through the demagnetizing factor  $\nu$ , so that the magnetic flux entering a period per unit of length in the  $y$  dimension is

$$A_0 = na\mu_0 H_0 / (1 - \nu s).$$

The field at a distance greater than  $a$  inside the  $n$

laminas is still equal to  $H_0 / (1 - \nu s)$ . Thus the assumption that all external lines of force are parallel to  $0z$  introduces only slight errors in the values found for the fields very close to the surface. It must obviously be pointed out that the flatter the sample, the smaller the errors.

The shape of the  $n$ - $s$  wall is given by the resolution of the Ginzburg-Landau equations, which determine the distribution of the field and the order parameter. However, in the present situation such a calculation would be of considerable mathematical difficulty and of doubtful interest, since the general features of the result can be inferred from physical arguments.

Far inside the sample, the wall is a plane. The field decreases from the value  $(1 - \nu s)H_0 \approx \lambda j_c$  to zero over a distance called the nonlocal penetration depth  $\lambda$ .  $j_c$  is the critical current density which is related to the fundamental parameters of the superconductor.<sup>10</sup>

In the vicinity of the surface, the wall becomes concave, but its magnetic thickness is still of the order of  $\lambda$ , which is a characteristic length inherent in the Ginzburg-Landau equations. Insofar as the radius of curvature is larger than  $\lambda$ , the detailed structure of the wall can be described with only the surface energy  $\Delta$ , and the shape can be determined by macroscopic electromagnetism and thermodynamics.

To get an insight of what may occur in a real situation, the macroscopic field distribution and the resulting free energy are determined by assuming domain shapes which allow exact calculations.

The magnetic field distribution is conveniently defined by the scalar potential  $\phi$  and the vector potential  $\vec{A}(0, A, 0)$  along the  $y$  axis. Since  $\vec{H} = -\text{grad}\phi = \text{curl}\vec{A} / \mu_0$ , it follows that  $\phi$  and  $A$  obey the Cauchy-Riemann conditions

$$-\phi'_x = -A'_z / \mu_0, \quad -\phi'_z = A'_x / \mu_0.$$

$\phi$  and  $A / \mu_0$  can be regarded as the real and imaginary parts of an analytic function of the com-

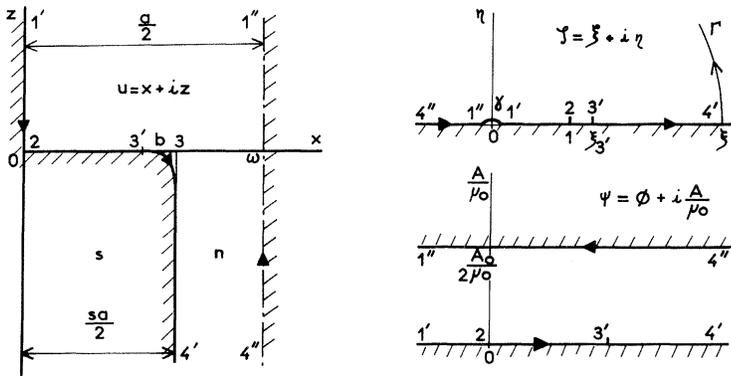


FIG. 2. Geometrical configuration of the cross section of a period, in the laminar model, near the external surface of the sample, and analytical correspondence between the planes  $u(x, y)$ ,  $\xi(\xi, \eta)$ , and  $\psi(\phi, A/\mu_0)$ .

plex variable  $u = x + iz$ , the so-called complex potential

$$\psi = \phi + i(A/\mu_0) .$$

It is readily shown that the complex magnetic field  $H = H_x + iH_z$  is derived from  $\psi$  by

$$-H^* = \frac{d\psi}{du} . \quad (8)$$

The analytic function  $\psi(u)$  defines a conformal transformation of the plane  $u(x, y)$  upon the plane  $\psi(\phi, A/\mu_0)$ . The equipotential lines and the lines of force of the field correspond to the straight lines obtained when  $\phi = Cte$  and  $A = Cte$  and are parallel to the axis in the plane  $\psi(\phi, A/\mu_0)$ .

In Fig. 2 a half-period of the array of the domain is represented. This half-period is bounded by two particular lines of force:  $1'2'3'4'$ , chosen as the line  $A = 0$ , and the straight line  $4''1''$  in the symmetry plane of the  $n$  lamina, where

$$A = \frac{1}{2}A_0 = na\mu_0 H_0 / 2(1 - \nu s) .$$

Hereafter, for convenience, the form of the contour is chosen such that  $3'3' = b$ .

The determination of an analytical function which associates the lines  $1'2'3'4'$  and  $4''1''$  with the straight lines  $A = 0$  and  $A = \frac{1}{2}A_0$  is possible in two steps. In the first step the whole contour is associated with the axis  $\eta = 0$  in a plane  $\zeta(\xi, \eta)$  through a slightly modified Schwartz transformation

$$\frac{du}{d\zeta} = \frac{dx + idz}{d\zeta} = -iB \frac{(\zeta - \xi_{3'})^{1/2} + ik}{\zeta(\zeta - 1)^{1/2}} . \quad (9)$$

$B$  and  $k$  are real constants and the square roots are real as  $\zeta$  is real and large.

In the second step the two halves of the axis  $\eta = 0$  are changed into  $A = 0$  and  $A = \frac{1}{2}A_0$  through the exponential transformation

$$\zeta = \exp(2\pi\mu_0\psi/A_0) . \quad (10)$$

The point 2 at which  $\zeta_2 = 1$  and  $\phi_2 = 0$  are chosen as origin of the potentials, and the various constants  $B$ ,  $k$ , and  $\xi_{3'}$  are determined by the geometrical dimensions of the contour. The analytical correspondence between the planes  $u$ ,  $\zeta$ , and  $\psi$  is represented in Fig. 2. Actually, the space inside the contour corresponds to the upper half-plane  $\eta > 0$  and to the band  $(0, A_0/2\mu_0)$ .

We have through elementary integration

$$4'4'' = \frac{na}{2} = -iB \int_{\Gamma} \frac{(\zeta - \xi_{3'})^{1/2} + ik}{(\zeta - 1)^{1/2}} \frac{d\zeta}{\zeta} = \pi B . \quad (11)$$

$\Gamma$  is the upper half-circle of infinite radius in the plane  $\zeta(\xi, \eta)$ .

In the same way, by integration along the half-circle  $\gamma$  of infinitesimally small radius, we have

$$1''1' = -\frac{1}{2}a = -iB \int_{\gamma} (\xi_{3'}^{1/2} + k) (d\zeta/\zeta)$$

$$= -\pi B(\xi_{3'}^{1/2} + k) . \quad (12)$$

A third relation is obtained by calculating  $3'3'$ :

$$3'3' = b = B \int_{\xi_{3'}}^{\infty} \frac{k d\xi}{(\xi - 1)^{1/2} \xi} = 2Bk \sin^{-1} \frac{1}{\xi_{3'}^{1/2}} . \quad (13)$$

From (11)–(13),  $B$ ,  $k$ , and  $\xi_{3'}$  are determined by the relations

$$B = \frac{na}{2\pi} , \quad \xi_{3'}^{1/2} + k = \frac{1}{n} , \quad k \sin^{-1} \frac{1}{\xi_{3'}^{1/2}} = \frac{\pi b}{na} . \quad (14)$$

We now consider the magnitude of the field along the wall. From (8)–(10) and (14) the complex field is given by

$$H^* = -i \frac{H_0}{1 - \nu s} \frac{(\zeta - 1)^{1/2}}{(\zeta - \xi_{3'})^{1/2} + ik} . \quad (15)$$

Along the wall  $\zeta$  is real and positive, and, as will be shown later,  $H_0/(1 - \nu s)$  tends, at equilibrium, toward the critical field  $H_c$ .  $H(1 - \nu s)/H_0$  can be considered, for the time being, as the reduced field  $h$  the magnitude of which is

$$h = \left( \frac{\xi - 1}{\xi - \xi_{3'} + k^2} \right)^{1/2} .$$

In particular, the critical value  $h = 1$  is obtained all along the wall when

$$\xi_{3'} - k^2 = 1 .$$

Combining with the second equation (14) we get

$$k = (1 - n^2)/2n , \quad \xi_{3'}^{1/2} = (1 + n^2)/2n . \quad (16)$$

These values characterize the critical wall first considered by Landau. As shown below, they happen to approach the best choice for  $b$ . From the third relation (14),  $b$  is then equal to

$$b = \frac{\alpha(1 - n^2)}{2\pi} \sin^{-1} \frac{2n}{1 + n^2} . \quad (17)$$

The distribution of the field in the  $n$  laminas can be deduced from the foregoing relations. The analysis is restricted to the axis  $4''1''$  since the lowest value occurs at the point  $\omega$  (Fig. 2). Along  $4''1''$ ,  $\zeta$  takes on real and negative values. From (9) and (15) the  $z$  coordinate can be calculated as a function of  $h$ . The result for the critical wall is

$$z = (a/\pi) [\tanh^{-1}(n/h) + n^2 \tanh^{-1}(nh) - 2n \tanh^{-1}h] .$$

The equation  $z = 0$  determines the function  $h_{\omega}(n)$  represented in Fig. 3. It is shown that the minimum value which occurs for small  $n$  is 0.648. This curve is in agreement with the values mentioned for  $n = 0$  and  $n = 0.5$  by Lifschitz and Sharvin.<sup>11</sup>

The complete variation of the field along  $4''1''$  is given in Fig. 4 for the typical values  $n = 0.1$  and  $n = 0.5$ . The relative extension  $\Delta z/a$  of the sub-

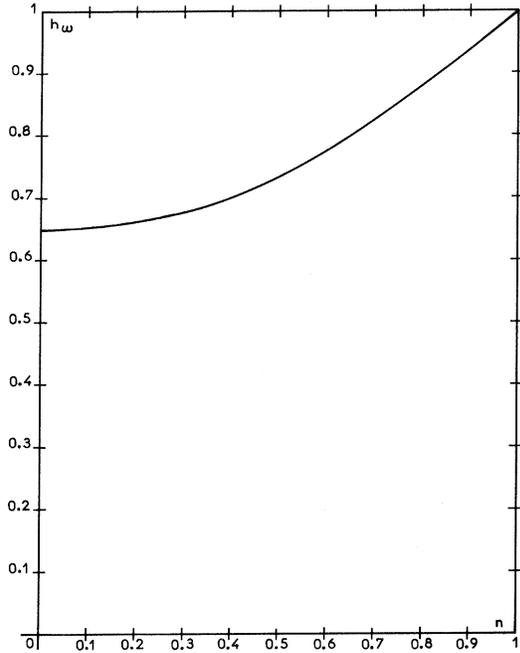


FIG. 3. Plot of the minimum value of the reduced field in the  $n$  domain vs the normal fraction  $n$  for the Landau critical wall.

critical region below  $\omega$ , as defined by

$$\Delta z = (1 - h_\omega) / \left| \frac{dh}{dz} \right|_\omega,$$

is shown to be rather small as long as  $n$  is small ( $dh/dz \rightarrow \infty$  for  $n \rightarrow 0$ ), and more appreciable around  $n \sim 0.5$ . As  $n$  approaches unity the field tends towards the critical value in the whole  $n$  region.

From these results it can be concluded that the branching of the  $n$  laminas is most likely to occur with a few splittings in the medium range, but not near the transition points  $n \approx 0$  and  $n \approx 1$ , where the importance of the subcritical region is negligible.

#### IV. FREE-ENERGY CONTRIBUTION OF DOMAIN ENDS

We now proceed to work out the contribution of the ends of the domains to the magnetic energy (1). Because of the symmetry of the supercurrents with respect to  $yOz$ , the magnetization intensity in the  $s$  domains is along  $Oz$  and depends only on  $z$ . It follows that inside the domain, where  $\vec{J} + \vec{H}^{(i)} = 0$ , the magnetic field is also along  $Oz$  and  $z$  dependent only. Then we have

$$J = -H_z^{(i)},$$

where  $H_z^{(i)}$  is the  $z$  component of the internal field along the wall and is given by

$$H_z^{(i)} = -\frac{d\phi}{dz} = -\frac{n}{1-\nu s} \frac{aH_0}{2\pi} \frac{d\xi}{\xi dz}.$$

As a result the magnetic energy  $W_M$  can be written for a half-period and per unit of length along  $Oy$ :

$$\begin{aligned} W_M &= \mu_0 \int_0^{H_0} \delta H_0 \int_{4'2} x H_z^{(i)} dz \\ &= -\frac{\mu_0 a H_0^2}{4\pi} \frac{n}{1-\nu s} \int_\infty^1 \frac{x d\xi}{\xi}. \end{aligned}$$

Actually, this expression diverges, since the domain is semi-infinite. We separate the contribution of the end of the domain by subtracting the principal part  $W_M^\circ$  at large distance where  $x = \frac{1}{2}sa$  and  $H_z = H_0/(1-\nu s)$ , i. e.,

$$\begin{aligned} W_M^\circ &= \mu_0 \int_0^{H_0} \delta H_0 \int_{-\infty}^0 \frac{sa}{2} \frac{H_0}{1-\nu s} dz \\ &= \frac{\mu_0 H_0^2 sa}{4(1-\nu s)} \int_\infty^{\xi_3'} \frac{dz}{d\xi} d\xi. \end{aligned}$$

This is the contribution of a half-period to the magnetic energy in the simple model (cf. Sec. II). The term of interest can now be written

$$\begin{aligned} \delta W_M &= W_M - W_M^\circ \\ &= -\frac{a\mu_0 H_0^2}{2(1-\nu s)} \left( \int_\infty^1 \frac{n}{2\pi} \frac{x d\xi}{\xi} + \frac{s}{2} \int_\infty^{\xi_3'} \frac{dz}{d\xi} d\xi \right). \end{aligned}$$

$dz/d\xi$  is given by (11). Upon substitution we get

$$\delta W_M = \frac{\mu_0 H_0^2 n a^2}{8\pi(1-\nu s)} \left[ \int_1^\infty \frac{2x}{a} \frac{d\xi}{\xi} - s \int_{\xi_3'}^\infty \left( \frac{\xi - \xi_3'}{\xi - 1} \right)^{1/2} \frac{d\xi}{\xi} \right].$$

Surprisingly, a rigorous expression can be calculated for the terms inside the square bracket. The second integral is elementary. The first one,

$$\int_1^A \frac{2x}{a} \frac{d\xi}{\xi} = s \ln A - \frac{2}{a} \int_1^A \ln \xi \frac{dx}{d\xi} d\xi \quad (A \rightarrow \infty),$$

is obtainable through residue calculation. For a sample of thickness  $l$ , with the restriction  $l \gg a$ , the resulting reduced magnetic energy due to the domain ends is given by

$$\delta w_M = \frac{2\delta W_M}{\frac{1}{2}\mu_0 H_c^2 (al/2)} = \frac{a}{l} g_M(n) h_0^2, \quad (18a)$$

with

$$\begin{aligned} g_M(n) &= \frac{n}{\pi(1-\nu s)} [(1+n)(\alpha+1)\ln(\alpha+1) - (1-n)(\alpha-1) \\ &\quad \times \ln(\alpha-1) - 2n\alpha \ln \alpha - 4 \ln 2], \quad (18b) \end{aligned}$$

where we have used the simpler notation  $\xi_3'^{1/2} = \alpha$ .

In addition, the increment of the  $n$  volume with respect to the simple model, and the increment of the wall length, give rise to two new extra terms in the free energy.

The increment of the  $n$  cross-section area relative to one corner is

$$\delta S = \int_\infty^{\xi_3'} \left( \frac{sa}{2} - x \right) \frac{dz}{d\xi} d\xi.$$

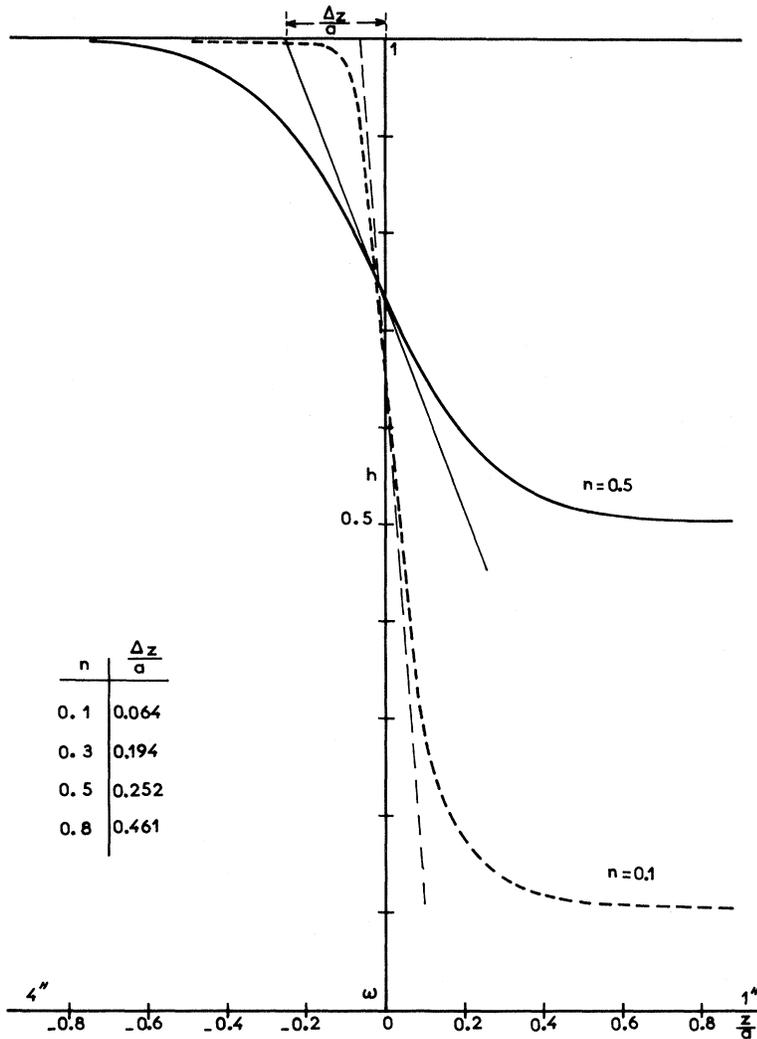


FIG. 4. Variations of the reduced field along the axis 4'' 1'' for the critical wall and relative extension of the subcritical region for typical values of the normal fraction  $n$ .

After integration, the expression of  $x$  for  $\xi > \xi_3$ , is

$$x = \frac{na}{2\pi} [\pi(\alpha - 1) + 2k \tan^{-1}(\xi - 1)^{1/2}],$$

whence  $\delta S$ , by new integration. The reduced increment of the condensation energy is then

$$\delta f_c = 4\delta S/al = (a/l)g_c(n), \quad (19a)$$

with

$$g_c(n) = (kn^2/\pi) [(\alpha + 1)\ln(\alpha + 1) + (\alpha - 1)\ln(\alpha - 1) - 2a\ln\alpha]. \quad (19b)$$

In a similar way the wall-length increment  $\delta l$  for one corner can be exactly calculated and gives rise to a contribution to the free energy

$$\delta f_s = 4\Delta\delta l/al = (\Delta/l)g_s(n),$$

where  $g_s(n)$  is a definite function of  $\alpha$ . This contribution happens to be negligible. Thus the final expression of the reduced free energy is

$$f = \frac{2\Delta}{a} - s + \frac{s}{1-\nu s} h^2 + \frac{a}{l} (g_M h^2 + g_C). \quad (20)$$

The redundant subscript in  $h_0$  is dropped from now onwards.

If, in a more realistic way, we take into account the spatial variation of  $l$  for an ellipsoidal sample,  $l$  is simply replaced by the ratio of the volume to the equatorial area.

Equilibrium values of the independent parameters  $a$ ,  $b$ , and  $s$  or, equivalently,  $a$ ,  $s$ , and  $\alpha$  are given by minimizing this function. We begin with  $\alpha$ , which is found only in the last term. If  $h$  is replaced by  $1/n - \alpha$ , Eqs. (18b) and (19b) can be combined to make a minimum of  $g_M h^2 + g_C$  with respect to  $\alpha$ , to occur for

$$\left(\frac{nh^2}{1-\nu s} + (1-2\alpha n)\right) \ln \frac{\alpha^2 - 1}{\alpha^2} + \left(\frac{h^2}{1-\nu s} - n\right) \ln \frac{\alpha + 1}{\alpha - 1} = 0. \quad (21a)$$

For  $h = 1 - \nu s$ , this gives

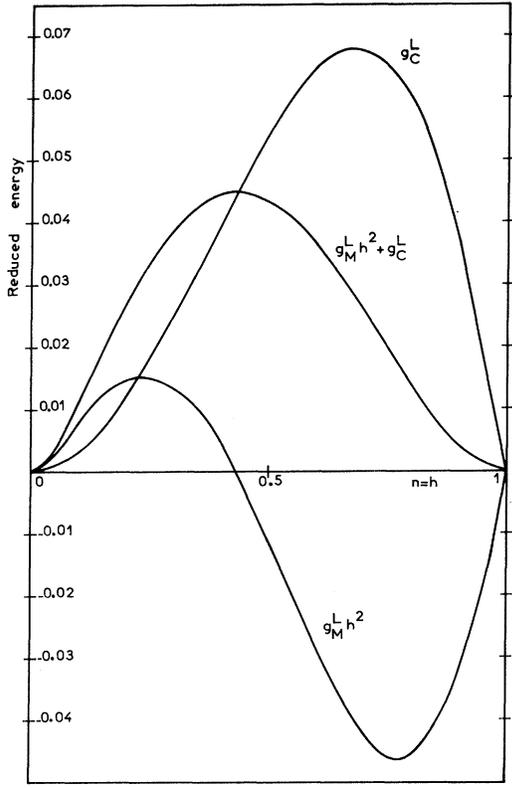


FIG. 5. Representative curves of the magnetic energy ( $g_M h^2$ ), the condensation energy ( $g_C$ ), and the total energy  $g_M h^2 + g_C$  of the domain ends, as functions of  $n=h$ , in the Landau limit  $\nu=1$ .

$$\alpha = \frac{1}{2n} \left( 1 + (1-\nu)n + \nu n^2 + s(1-\nu) \frac{\ln[(\alpha+1)/(\alpha-1)]}{\ln[(\alpha^2-1)/\alpha^2]} \right). \quad (21b)$$

In the limiting case  $\nu=1$ , the root of this equation is  $\alpha = (1+n^2)/2n$ , which is exactly the value of the Landau critical wall defined by the relations (16). At small values of  $1-\nu$  and at intermediate values of  $n$  and  $s$ , the ratio of the logarithms is of the order of 1 and  $\alpha$  does not differ too much from the Landau limit.

Near the diamagnetic intermediate-phase transition where  $n \approx 0$ , it is shown hereafter that the relation  $h = 1 - \nu s$  is still valid. Within the present approximation  $l \gg a$ ,  $1 - \nu$  is larger than the transition values of  $n$  for usual-dimensions samples. The predominant contribution to  $\alpha$  is then

$$\alpha \approx 1/2(1-\nu). \quad (22)$$

At the intermediate normal transition ( $s \approx 0$ ),  $\alpha$  approaches unity, according to the simpler equation derived from (21a):

$$h^2 = (1-\nu s)[1 + \beta(\ln 2\beta/\ln 2) - s + \dots], \quad (23)$$

with  $\beta = \alpha - 1$ .

Explicit calculations of  $g_M$  and  $g_C$  in the Landau case give

$$g_M^L = \frac{n}{(1-\nu s)\pi} \left( \frac{(1+n)^3}{n} \ln(1+n) - \frac{(1-n)^3}{n} \ln(1-n) - (1+n^2) \ln(1+n^2) - 2 \ln 8n \right), \quad (24)$$

$$g_C^L = \frac{1-n^2}{2\pi} [(1+n)^2 \ln(1+n) + (1-n)^2 \ln(1-n) - (1+n^2) \ln(1+n^2)]. \quad (25)$$

The particular value of the function  $g_M h^2 + g_C$  in the limits  $\nu=1$ ,  $h=n$  must be identified with twice the function called  $\psi$  by Landau which has been tabulated by several authors.<sup>3,11</sup> Its exact expression is

$$\begin{aligned} 2\psi^L &= (g_M^L h^2 + g_C^L)_{\nu=1} \\ &= \frac{1}{\pi} \left( \frac{(1+n)^4}{2} \ln(1+n) + \frac{(1-n)^4}{2} \ln(1-n) - \frac{(1+n^2)^2}{2} \ln(1+n^2) - 2n^2 \ln 8n \right). \end{aligned} \quad (26)$$

The representative curves of  $g_M^L h^2 + g_C^L$  along with their sum in these limits, are given in Fig. 5.

The equilibrium value of the period  $a$  is readily obtained from (20):

$$a = \left( \frac{2\Delta l}{g_M h^2 + g_C} \right)^{1/2}. \quad (27)$$

The expression of  $f$  is then

$$f = -s + \frac{s h^2}{1-\nu s} + 2 \left( \frac{2\Delta}{l} (g_M h^2 + g_C) \right)^{1/2}. \quad (28)$$

The equilibrium value of  $s$  is then obtained by minimizing (28) with respect to  $s$ :

$$\frac{\partial f}{\partial s} = -1 + \frac{h^2}{(1-\nu s)^2} + \left( \frac{2\Delta}{l} \right)^{1/2} \frac{g'_M h^2 + g'_C}{(g_M h^2 + g_C)^{1/2}} = 0. \quad (29)$$

Because of the smallness of the last term, the result slightly differs from that of simple model  $s_0 = (1-h)/\nu$ .

Putting  $s = s_0 + \delta s$ , one gets

$$\delta s = -\frac{h}{2\nu} \frac{g'_M h^2 + g'_C}{(g_M h^2 + g_C)^{1/2}} \left( \frac{2\Delta}{l} \right)^{1/2}. \quad (30)$$

The functions of  $s$ ,  $g_M$ ,  $g_C$ ,  $g'_M$ , and  $g'_C$  appearing in the expressions of  $a$  (27) and  $\delta s$  (30) are evaluated at  $s = s_0$ .

The small departure of  $s$  from  $s_0$  gives rise to a slight variation of the field in the  $n$  laminas with respect to the simple-model value (5):

$$\frac{\delta H_n}{H_n} = \frac{\nu \delta s}{1-\nu s_0} \approx -\frac{g'_M h^2 + g'_C}{2(g_M h^2 + g_C)^{1/2}} \left( \frac{2\Delta}{l} \right)^{1/2}.$$

Since in the very interior of the  $s$  domains  $J = -H_n$ , the mean magnetic moment  $\bar{J}_I = sJ$  also undergoes a deviation from the ideal value (7):

$$\delta \bar{J}_I = - \left( \frac{h}{1 - \nu s_0} + \frac{\nu s_0 h}{(1 - \nu s_0)^2} \right) H_c \delta s = - \frac{H_c}{h} \delta s,$$

whence

$$- \bar{J}_I = \frac{H_c - H_0}{\nu} - H_c \frac{g'_M h^2 + g'_C}{2\nu(g_M h^2 + g_C)^{1/2}} \left( \frac{2\Delta}{l} \right)^{1/2}. \quad (31)$$

Thus the dimensional effect resulting from the finite length of the domains is of the order of  $(\Delta/l)^{1/2}$ , as mentioned by de Gennes.<sup>12</sup> This effect is rather small for the usual sample dimensions.

### V. TRANSITION FIELDS

The various expressions obtained above for the thermodynamic quantities in the intermediate state allow us to write the equations governing the equilibrium between the diamagnetic and the intermediate phases (*D-I*), as well as between the intermediate and normal phases (*I-N*). These equations give the values of  $n$  (or  $s$ ) and the values of the field at the transition points.

#### A. *D-I* Transition

The equilibrium condition  $f_D = f_I$  yields the new equation

$$-s + \frac{s}{1 - \nu s} h^2 + 2 \left( \frac{2\Delta}{l} (g_M h^2 + g_C) \right)^{1/2} = -1 + \frac{h^2}{1 - \nu}. \quad (32)$$

At the transition  $n$  and  $h$  are therefore defined by Eqs. (29) and (32). Putting  $2\Delta/l = \epsilon$ , after some arrangements, the following system of two second-degree equations with respect to  $h^2$  is obtained:

$$\frac{h^4}{(1 - \nu)^2 (1 - \nu s)^2} - \left( \frac{2}{(1 - \nu)(1 - \nu s)} + \frac{4\epsilon g_M}{n^2} \right) h^2 + \left( 1 - \frac{4\epsilon g_C}{n^2} \right) = 0, \quad (33)$$

$$\frac{h^4}{(1 - \nu)(1 - \nu s)^3} - \left( \frac{2 - \nu - \nu s}{(1 - \nu)(1 - \nu s)^2} - \frac{2\epsilon g'_M}{n} \right) h^2 + \left( 1 + \frac{2\epsilon g'_C}{n} \right) = 0.$$

By eliminating  $h^4$  we get

$$h^2 = \frac{\nu n^3 + 2\epsilon[2(1 - \nu)g_C + n(1 - \nu s)g'_C]}{\nu n^3/(1 - \nu)(1 - \nu s) - 2\epsilon[2(1 - \nu)g_M + n(1 - \nu s)g'_M]}. \quad (34)$$

The result of the elimination of  $h^2$  is rather complicated, but can be simplified by taking into account the smallness of  $n$  at the transition. From (18b) and (19b), with the value (22) for  $\alpha$ , the fol-

lowing approximate expressions, in the limit  $n \ll 1 - \nu \ll 1$ , can be derived:

$$g_M(n) = - \frac{2n}{\pi(1 - \nu)} [\ln 8(1 - \nu) - 1],$$

$$g_C(n) = \frac{2n}{\pi} (1 - \nu).$$

$g_M$  and  $g_C$  being of first order with respect to  $n$ , the following equation is established:

$$\frac{\nu^2 n^4}{(1 - \nu)^4} \left( g_M + \frac{g_C}{(1 - \nu)^2} \right) = \left( 2g_M + n g'_M \right) + \frac{2g_C + n g'_C}{(1 - \nu)^2} \epsilon + O(\epsilon n^3).$$

Or, since  $n g'_M = -g_M$  and  $n g'_C = -g_C$ , we have

$$n^4 = \frac{(1 - \nu)^4}{\nu^2} \left( g_M + \frac{g_C}{(1 - \nu)^2} \right) \epsilon,$$

whence

$$n = (1 - \nu) \left( (2 - \ln 8(1 - \nu)) \frac{4\Delta}{\pi \nu^2 l} \right)^{1/3}. \quad (35)$$

From (34), the related value of the field is

$$h \simeq (1 - \nu) \left[ 1 + \frac{2(1 - \nu)^3}{\nu n^3} \left( g_M + \frac{g_C}{(1 - \nu)^2} \right) \epsilon \right]^{1/2} \simeq 1 - \nu + \nu n. \quad (36)$$

The field is still related to  $s$  by  $h = 1 - \nu s$ , as supposed in (22).

From (35) and (36) it results that the increment of the transition field above  $1 - \nu$  is predominantly as  $(\Delta/l)^{1/3}$ . For the typical values  $\nu = 0.9$ ,  $\Delta/l = 10^{-4}$  it is found that  $n = 0.71 \times 10^{-2}$  and  $\Delta h/(1 - \nu) = 6.3\%$ .

#### B. *I-N* Transition

The calculations are very similar. The condition  $f_I = f_N$  now gives

$$-s + \frac{s}{1 - \nu s} h^2 + 2\epsilon^{1/2} (g_M h^2 + g_C)^{1/2} = 0, \quad (37)$$

leading with (29) to the system

$$\frac{s^2 h^4}{(1 - \nu s)^2} - \left( \frac{2s^2}{1 - \nu s} + 4\epsilon g_M \right) h^2 + (s^2 - 4\epsilon g_C) = 0, \quad (38)$$

$$\frac{s h^4}{(1 - \nu s)^3} - \left( \frac{s(2 - \nu s)}{(1 - \nu s)^2} + 2\epsilon g'_M \right) h^2 + (s - 2\epsilon g'_C) = 0,$$

whence

$$h^2 = \frac{\nu s^3 - [2g_C - s(1 - \nu s)g'_C] 2\epsilon}{\nu s^3/(1 - \nu s) + [2g_M - s(1 - \nu s)g'_M] 2\epsilon}. \quad (39)$$

$s$  and  $h$  are given by (38) and (23) through (18b) and (19b), but here the derivation of simple approximate expressions is much more involved. However, the elimination of  $h^2$  in (38) and then in Eqs. (23) and (39) leads to the equations

$$\left( \frac{4 \ln 2}{\pi} \right)^2 \left[ \left( \nu + \frac{1 - 2 \ln(\beta/2)}{4 \ln 2} \right) \beta - 2\nu s \right] (2\beta - s) \beta \epsilon^2$$

$$-\frac{2 \ln 2}{\pi} \left[ \left( \frac{1 - 2 \ln 2 \beta}{2 \ln 2} \right)^2 \beta^4 + \frac{1 - 2 \ln 2 \beta}{\ln 2} (1 - 2\nu) \beta^3 s + \left( 1 - 10\nu - \nu^2 + \nu \frac{1 - 2 \ln 2 \beta}{\ln 2} \right) \beta^2 s^2 + 2\nu(2 + \nu) \beta s^3 - \nu^2 s^4 \right] s \epsilon + \left( s + \beta \frac{1 - 2 \ln 2 \beta}{4 \ln 2} \right) \nu^2 \beta s^5 = 0, \quad (40)$$

$$\frac{4 \ln 2}{\pi} \left[ \frac{1 + 2 \ln 2 \beta}{2 \ln 2} \beta^2 - \left( \nu + \frac{\ln 4 \beta}{\ln 2} \right) \beta s + (\nu + 1) s^2 \right] \epsilon - \left( s - \frac{\beta \ln 2 \beta}{\ln 2} \right) \nu s^3 = 0. \quad (41)$$

In the coefficients only the terms which are of the lowest order with respect to  $s$  and  $\beta = \alpha - 1$  have been retained. The analysis shows that a solution exists such that  $s \sim \beta \sim \epsilon^{1/2}$ .

The resulting value of the transition field is then, from (23),

$$h = 1 - \left( \frac{\nu + 1}{2} s - \frac{\beta \ln 2 \beta}{2 \ln 2} \right), \quad (42)$$

which shows that the reduction of the field is of order  $s$ ,  $\beta$ , i. e.,  $(\Delta/l)^{1/2}$ . Numerical computations are required for a comparison with experiment and will be given in a paper to be published later.

With the present assumptions, the equations written above determine completely the transition fields for a flat-ellipsoidal sample, and allow a detailed analysis of the limiting cases corresponding to experimental situations of interest.

The departure of the transition fields from the ideal values  $(1 - \nu)H_c$  and  $H_c$  has been experimentally investigated by different authors on small cylinders<sup>13,14</sup> and thin slabs.<sup>15</sup> Their results generally agree with Landau's branching model. In these experiments the typical parameter  $\Delta/l$  was of a few  $10^{-3}$ . If such values are inserted into the present

unbranched theory, the agreement is just as good. For  $\nu = 0.9$  and  $\Delta/l = 10^{-3}$ , (35) and (36) give  $n \approx 2.7\%$  and  $\Delta h/(1 - \nu) \approx 24.3\%$ . However, it must be borne in mind that the different theories imply the assumption of semi-infinite domains. It is then questionable to apply these results to samples with  $l \lesssim a$  at the transition point.

## VI. CONCLUSION

Through detailed electromagnetic and thermodynamical analysis, analytical expressions have been calculated for the various contributions to the free energy of a superconductive ellipsoidal sample in the intermediate state.

The result permits a precise formulation of the equations describing the phase transitions at the ends of the intermediate range. At the lowest order, the deviation of the field from the ideal value is of the order of  $(\Delta/l)^{1/3}$  for the diamagnetic intermediate-phase transition and of the order of  $(\Delta/l)^{1/2}$  for the intermediate normal-phase transition.

This quantitative theory could be compared with experiments performed on ellipsoidal samples whose thickness is larger than the period of the domain structure.

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