Thermodynamic Behavior of Type-II Superconductors with Small κ near the Lower Critical Field*

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(Received December 1970)

The interaction between well-separated Abrikosov vortices is calculated with the Ginzburg-Landau theory for superconductors with $\kappa<\sqrt{2}$. For $\kappa>1/\sqrt{2}$, vortices repel each other while for $\kappa<1/\sqrt{2}$ there is an attraction. Therefore, in the Ginzburg-Landau regime, all type-II superconductors exhibit a λ transition gradually changing into a first-order transition as $\kappa\rightarrow 1/\sqrt{2}$. Slightly above H_{c1} a transition from the triangular- to the square-flux-line lattice is found to take place. The variational method used admits more general applications.

I. INTRODUCTION

The transition from the Meissner to the mixed state of type-II superconductors which takes place at the lower critical field H_{c1} has some unique features. Although in most cases the transition is of second order in the sense that the first partial derivatives of the Gibb's free energy with respect to the intensive variables are continuous in the thermodynamic limit, a closer look reveals that it has many features of a first-order transition. The reason for this is that above H_{c1} macroscopic quantized flux lines are created, leading locally to jumps in the internal fields. Thus a very sensitive experiment would show that even in a bulk type-II superconductor flux penetrates in small discontinuous steps. The best evidence for the peculiar nature of the transition is probably the observation of substantial superheating at H_{c1}. This phenomenon was discussed previously.[1]

As a consequence of the above-mentioned fact, calculations are usually much harder at H_{c1} than at H_{c2} because there is no mathematically useful order parameter which vanishes at H_{c1}. In this paper, we calculate the thermodynamic behavior of type-II superconductors with $\kappa<\sqrt{2}$ near H_{c1} using the Ginzburg-Landau (GL) theory.[2]

In essence, we derive an expression for the interaction between widely spaced vortices using only the asymptotic form of the magnetic field and order parameter far away from the core of an isolated vortex line. Recently, a number of authors have investigated the structure of an isolated vortex line using more general theories.[3–6] One might be able to adapt our method to these cases. Hopefully, this would lead to a clarification of the nature of the phase transition at H_{c1} outside the GL regime. Such calculations are important also in the light of recent observations of an "intermediate mixed state" which indicates attraction between vortices at certain distances.[7–10]

In Sec. II an expression for the interaction energy is derived valid to second order in the deviations of the order parameter from the isolated vortex solution. The interaction energy ϵ consists of an electrodynamic repulsion ϵ_1 and an attraction ϵ_2 arising from the change in superconducting condensation energy. For type-II superconductors ($\kappa>1/\sqrt{2}$) one finds $\epsilon_1+\epsilon_2>0$ so that the mixed state is stable and there is a λ transition at H_{c1}. For $\kappa=1/\sqrt{2}$, one can show $\epsilon_1+\epsilon_2=0$. In Sec. III some results for $\kappa\gtrsim 1/\sqrt{2}$ are presented. The free energy of the square-flux-line lattice turns out to be lower than that of the triangular one for a penetrated flux B larger than about $0.3H_c$.[2] Square-flux-line lattices have in fact been observed in the region predicted.[11]

II. INTERACTION BETWEEN VORTICES

Writing the GL order parameter as $\psi=fe^{i\phi}$ with real f, and introducing the superfluid velocity $\vec{Q}=\vec{A}-\vec{\nabla}\phi/\kappa$, where $\vec{A}$ is the vector potential, one obtains for the superconducting part of the Helmholtz free energy in the usual reduced units,[12]

$$F=\int d^3r\,\{\tfrac{1}{2}(1-f^2)^2+[(\vec{\nabla}/\kappa)f]^2+\vec{Q}^2f^2+(\mathrm{curl}\vec{Q})^2\}\ . \quad (1)$$

The magnetic field is given by $\vec{h}=\mathrm{curl}\vec{Q}$. Minimizing F leads to the usual GL equations

$$[\vec{Q}^2-(\vec{\nabla}/\kappa)^2]f=f(1-f^2)\ , \quad (2)$$

$$\mathrm{curl}\,\mathrm{curl}\vec{Q}+f^2\vec{Q}=0\ . \quad (3)$$

The isolated vortex carrying one-flux quantum is that solution $f_0, \vec{Q}_0$ of Eqs. (2) and (3) possessing cylindric symmetry and satisfying the boundary conditions

$$f_0\rightarrow 0\ ,\quad Q_0r\rightarrow\kappa^{-1}\quad \text{for } r\rightarrow 0\ ;$$
$$f_0\rightarrow 1\ ,\quad Q_0r\rightarrow 0\quad \text{for } r\rightarrow\infty \quad (4)$$

(r is the distance from the vortex core). Numerical solutions for the isolated vortex have been calculated some time ago by Harden and Arp[13] and by Neumann and Tewordt.[3]

Near H_{c1} the vortices form a widely spaced vortex lattice and their individual structure does not differ much from that of an isolated vortex. Inside the Wigner-Seitz cell centered around a particular vortex we write

$$\vec{Q} = \vec{Q}_0 + \vec{Q}_1 ; \quad f = f_0 + f_1 . \tag{5}$$

The small quantities f_1 and $\vec{Q}_1$ satisfy the perturbational equations corresponding to (2) and (3) which are given by

$$[Q_0^2 + 3f_0^2 - 1 - (\vec{\nabla}/\kappa)^2] f_1 + 2f_0 \vec{Q}_0 \cdot \vec{Q}_1 = 0 , \tag{6}$$

$$\operatorname{curl}\operatorname{curl}\vec{Q}_1 + f_0^2 \vec{Q}_1 + 2f_0\vec{Q}_0 f_1 = 0 . \tag{7}$$

Equations (6) and (7) hold not only if $f_1 \ll f_0$ and $Q_1 \ll Q_0$ are satisfied but also for the case $f_1 \ll f_0$ and $Q_1^2 \ll f_1$. This fact will allow us to retain Eqs. (6) and (7) for superconductors with $\kappa < \sqrt{2}$ even on the boundary of the cell (see below). Now expand the free-energy equation (1) to second order in f_1 and make use of Eqs. (2), (3), and (5)–(7). Taking the integral in (1) over the Wigner-Seitz cell leads to the following expression for the free energy ϵ per vortex line:

$$\epsilon = \epsilon_0^1 + 2\oint d\vec{S} \cdot \left(\vec{Q}_1 \times \operatorname{curl}(\vec{Q}_0 + \tfrac{1}{2}\vec{Q}_1) + \frac{1}{\kappa}\, f_1 \frac{\vec{\nabla}}{\kappa}\,(f_0 + \tfrac{1}{2} f_1) \right), \tag{8}$$

where

$$\epsilon_0^1 = \int d^3 r \left[\tfrac{1}{2}(1 - f_0^2)^2 + \left(\frac{\vec{\nabla}}{\kappa} f_0 \right)^2 + Q_0^2 f_0^2 + (\operatorname{curl}\vec{Q}_0)^2 \right]. \tag{9}$$

The integral in (8) extends over the surface of the cell while integral (9) is taken over its volume. On the boundary of the cell the quantities $g = 1 - f_0$ and $\vec{Q}_0$ are small and Eqs. (6) and (7) reduce to

$$\vec{\nabla}^2 f_1 - 2\kappa^2 f_1 = 0 , \quad \nabla^2 \vec{Q}_1 - \vec{Q}_1 = 0 . \tag{10}$$

For $\kappa < \sqrt{2}$, the asymptotic forms of Eqs. (2) and (3) are identical with Eqs. (10). A solution of (10) appropriate for a vortex lattice is therefore given by[14]

$$f_1(\vec{r}) = \sum_{i \neq 0} g(\vec{r} - \vec{r}_i) , \quad \vec{Q}_1(\vec{r}) = \sum_{i \neq 0} \vec{Q}_0(\vec{r} - \vec{r}_i) \tag{11}$$

(the $\vec{r}_i$ are the lattice points). Also, from the asymptotic solutions of (2) and (3) [see Eqs. (16)] and from Eqs. (11), one sees that $Q_1^2 \ll f_1$ holds inside and on the boundary of the Wigner-Seitz cell. Inserting (11) into (8) yields

$$\epsilon = \epsilon_0 + \epsilon_1 + \epsilon_2 , \tag{12}$$

where ϵ_0 is the energy of an isolated vortex line, i.e., it is given by Eq. (9) with the integral now taken over all space. Furthermore, ϵ_1 and ϵ_2 are given by

$$\epsilon_1 = 2\sum_{i \neq 0} \oint d\vec{S} \cdot \left[\vec{Q}_i \times \operatorname{curl}\left(\vec{Q}_0 + \frac{1}{2} \sum_{j\neq 0;\, j \neq i} Q_j \right) \right], \tag{13}$$

$$\epsilon_2 = 2\sum_{i \neq 0} \oint d\vec{S} \cdot \left[f_i \frac{\vec{\nabla}}{\kappa} \left(f_0 + \frac{1}{2} \sum_{j\neq 0;\, j \neq i} f_j \right) \right], \tag{14}$$

where we have written

$$f_i(\vec{r}) = g(\vec{r} - \vec{r}_i) , \quad Q_i(\vec{r}) = \vec{Q}_0(\vec{r} - \vec{r}_i) . \tag{15}$$

In deriving (12)–(14) use was made of the fact that the terms with $j = i$ which are left out in the sums of Eqs. (13) and (14) just yield the difference between ϵ_0^1 and ϵ_0.

The expressions (13) and (14) can be interpreted in terms of pairwise interactions between vortices. The terms with the double sum then represent the contributions from other Wigner-Seitz cells. This interpretation will become more apparent after further evaluation.

Solving Eqs. (2) and (3) asymptotically for $\kappa < \sqrt{2}$ yields

$$\vec{Q}_0 = c\, K_1(r)\hat{\theta} , \quad g = 1 - f_0 = d K_0(\sqrt{2}\,\kappa r) , \tag{16}$$

where K_0 and K_1 are modified Bessel functions. We determined the constants c and d from the isolated vortex solutions obtained numerically by the authors of Ref. 3. Finally, inserting (15) and (16) into (13) and (14) gives

$$\epsilon_1 = 2\pi c^2 \sum_{i \neq 0} K_0(r_i) , \tag{17}$$

$$\epsilon_2 = -\frac{2\pi}{\kappa^2}\, d^2 \sum_{i \neq 0} K_0(\sqrt{2}\,\kappa r_i) . \tag{18}$$

The evaluation of the integrals is described in Appendix A.

The electrodynamic repulsion between vortices is given by ϵ_1, while ϵ_2 represents an attraction arising from the increase in (negative) superconducting condensation energy as the vortices overlap. The expression for ϵ_1 is valid for all κ. For large κ it holds up to fields near H_{c2} and yields Abrikosov's result.[12] (Note that $c = \kappa^{-1}$ for $\kappa \gg 1$.) The term corresponding to ϵ_2 for large κ is negligible except near H_{c2}.

One can show that for $\kappa = 1/\sqrt{2}$ the relation $d = c/\sqrt{2}$ holds (see Appendix B) and it is then clear that $d \lessgtr c/\sqrt{2}$ for $\kappa \gtrless 1/\sqrt{2}$. Therefore, the following relations hold:

$$\epsilon_1 + \epsilon_2 \gtrless 0 \quad \text{for } \kappa \gtrless 1/\sqrt{2} . \tag{19}$$

Thus for $\kappa < 1/\sqrt{2}$ the vortices attract each other, for $\kappa = 1/\sqrt{2}$ the interaction is zero (in Appendix B it is shown that this result holds for all vortex distances), and for $\kappa > 1/\sqrt{2}$ there is repulsion. In the last case the mixed state is stable and there is a λ point (second-order transition) at[2,15] H_{c1} gradually changing into a first-order transition for

$\kappa \to 1/\sqrt{2}$.

III. RESULTS

From Eqs. (17) and (18) one can calculate the thermodynamic properties near the lower critical field. H_{c1} itself is defined as that applied field H_0 at which the Gibbs's free energies $G = F - 2H_0 B$ for two states with and without a vortex are equal. Since each vortex carries a flux quantum $2\pi/\kappa$, one finds $H_{c1} = \epsilon_0 \kappa / 4\pi$.[12] The relation between B and H_0 is found from the equation

$$2H_0 = \frac{\partial F}{\partial B} = \frac{\partial}{\partial B}\left(\frac{B\kappa}{2\pi}\epsilon\right)$$

leading to

$$H_0 - H_{c1} = +\frac{\kappa}{4\pi}\frac{\partial}{\partial B}\left[B(\epsilon_1 + \epsilon_2)\right] . \qquad (20)$$

Here F is now the spatial average of the free-energy density and B the spatial average of the magnetic field taken over the whole specimen. Since $B\kappa/2\pi$ is equal to the number of vortices per unit area, there exists a simple relation between the lattice constant and the induction depending on the geometry of the lattice. With its help, Eq. (20) can be evaluated. The magnetization curve is found to agree well with experiments by Finnemore *et al.*[16] on niobium which has a κ of 0.78 (see Ref. 2).

We also find the Gibbs's free energy for the square-flux-line lattice to be lower than that of the triangular one in a certain field region. This is rather surprising, since previous calculations valid either near[17,18] H_{c2} or well below H_{c2} and $\kappa \gg 1$ ("London approximation"[19]) have shown the triangular lattice to be stable.[20] To demonstrate our result we have expanded the Gibbs's free energy and Eq. (20) to first order in $\kappa - 1/\sqrt{2}$ in nearest-neighbor approximation. One obtains

$$g \equiv \frac{G}{\kappa - 1/\sqrt{2}} \simeq -B\frac{zb}{2}\left[\alpha K_1(b) + \beta b K_0(b)\right] , \qquad (21)$$

$$h \equiv \frac{H_0 - H_{c1}}{\kappa - 1/\sqrt{2}} \simeq \frac{z}{2}\left[\left(\alpha + \frac{b^2}{2}\beta\right)K_0(b) + \left(\frac{\alpha}{2} + \beta\right) b K_1(b)\right] , \qquad (22)$$

where b is the lattice constant and z the number of nearest neighbors. The numbers α and β are connected with the constants c and d introduced in Eq. (16) by the following relations:

$$\alpha = \frac{d}{d\kappa}\left[c^2 - \left(\frac{d}{\kappa}\right)^2\right]\Bigg|_{\kappa = 1/\sqrt{2}} , \quad \beta = \frac{c^2 + d^2}{2\sqrt{2}} .$$

In Fig. 1, g is plotted as a function of h. For very large vortex distances only the electrodynamic repulsion is of importance. Therefore, very near H_{c1} the triangular lattice is stable as is the case for a London superconductor.[19] However, at $h = 0.13$, a transition to the square lattice takes place. Clearly the transition is of first order and the induction undergoes a jump from $B_\Delta = 0.238$ to $B = 0.247$. Unfortunately, our approximations become inaccurate beyond the transition.

Using the Essmann and Träuble[7,21] technique, Obst[11] was able to observe square lattices in the predicted region on Pb-1.6 wt% Tl ($\kappa = 0.72$). His results also indicate that the flux-line lattice is influenced considerably by the crystal anisotropy. Clearly such effects are not included in our calculation.

IV. CONCLUDING REMARKS

In Sec. II extensive use was made of variational techniques. One convenient feature was the possibility of writing the interaction energy between vortices as a surface integral. It is clear that always when the solution f_0 of a variational problem involving a functional $F[f]$ is perturbed by some function f_1 the change in F appears to first order only in surface terms. If in addition f_1 is a solution of the perturbational equation obtained from Euler's equation of F (i.e., if f_1 is to first order equal to the difference between two neighboring solutions of the variational problem), then also the second-order change of F appears only in surface terms. The reason is that the above-mentioned perturbational equation is identical with the Euler's equation which makes the second variation of F stationary (usually called the Jacobi equation). This is precisely the situation encountered in Sec. II. Therefore, this method is applicable quite generally . We hope

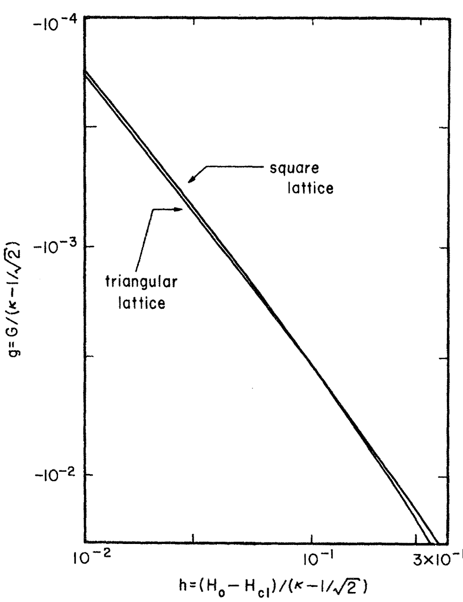

FIG. 1. Gibbs's free energy G of the flux-line lattice in nearest-neighbor approximation as a function of applied field H_0 for κ slightly larger than $1/\sqrt{2}$. Above $h = 0.13$ the square lattice has lower free energy.

to be able to apply it to the generalized GL theory.[3,22,23]

It appears that our theory does not lead to an "intermediate mixed state"[7-10] for specimens with $\kappa > 1/\sqrt{2}$. Owing to the attraction between flux lines however, we see a possibility for such a state when $\kappa \lesssim 1/\sqrt{2}$. The distortion of the magnetic field outside the specimen furnishes the balancing counter force. We hope to investigate this point further.

In Sec. III, we found a transition from the triangular- to the square-flux-line lattice slightly above H_{c1}. Since at H_{c2} the triangular lattice is stable there must be another transition back to triangular symmetry. To push the investigation further, a complete numerical vortex lattice solution of the GL equations seems inevitable. The transition is probably usually broadened or even suppressed owing to the energy barrier separating the two lattices and to interactions with the crystal lattice.

ACKNOWLEDGMENTS

The author wishes to thank Dr. U. Essmann and Professor M. J. Stephen for very helpful discussions.

APPENDIX A

The integrals (13) and (14) are to be evaluated. Inserting (15) and (16) into (13) and (14) yields

$$\epsilon_1 = 2c^2 \sum_{i\neq 0} \oint d\vec{S}\cdot\hat{r}\left\{K_1(|\vec{r}-\vec{r}_i|)\times\left[K_0(r)+\frac{1}{2}\sum_{j\neq 0;\, j\neq i} K_0(|\vec{r}-\vec{r}_j|)\right]\right\}, \quad \text{(A1)}$$

$$\epsilon_2 = -2\sqrt{2}\, d^2 \sum_{i\neq 0}\oint d\vec{S}\cdot\hat{r}\left\{K_0(\sqrt{2}\kappa|\vec{r}-\vec{r}_i|)\times\left[K_1(\sqrt{2}\kappa r)+\frac{1}{2}\sum_{j\neq 0;\, j\neq i}K_1(\sqrt{2}\kappa|\vec{r}-\vec{r}_j|)\right]\right\}. \quad \text{(A2)}$$

Let us introduce the auxiliary functions

$$\vec{h}_i(\vec{r}) = cK_0(|\vec{r}-\vec{r}_i|)\hat{z}\ . \quad \text{(A3)}$$

Then $\vec{h}_i$ satisfies the equation

$$\text{curl curl}\vec{h}_i + \vec{h}_i = 2\pi c\delta^{(2)}(\vec{r}-\vec{r}_i)\hat{z}\ . \quad \text{(A4)}$$

From (A1) or (13) we now obtain

$$\begin{aligned}\epsilon_1 &= 2\sum_{i\neq 0}\oint d\vec{S}\cdot\left[\left(\vec{h}_0+\frac{1}{2}\sum_{j\neq 0;\, j\neq i}\vec{h}_j\right)\times\text{curl}\vec{h}_i\right]\\
&= 2\sum_{i\neq 0}\int d^3r\left[\text{curl}\left(\vec{h}_0+\frac{1}{2}\sum_{j\neq 0;\, j\neq i}\vec{h}_j\right)\cdot\text{curl}\vec{h}_i - \left(\vec{h}_0+\frac{1}{2}\sum_{j\neq 0;\, j\neq i}\vec{h}_j\right)\text{curl curl}\vec{h}_i\right]\\
&= 2\sum_{i\neq 0}\int d^3r\left\{\text{curl}\left(\vec{h}_0+\frac{1}{2}\sum_{j\neq 0;\, j\neq i}\vec{h}_j\right)\cdot\text{curl}\vec{h}_i + \vec{h}_i\cdot\left[2\pi c\delta(r)\hat{z} - \text{curl curl}\left(\vec{h}_0+\frac{1}{2}\sum_{j\neq 0;\, j\neq i}\vec{h}_j\right)\right]\right\}\\
&= 4\pi c\sum_{i\neq 0}\vec{h}_i(0) + 2\sum_{i\neq 0}\oint d\vec{S}\cdot\left[\vec{h}_i\times\text{curl}\left(\vec{h}_0+\frac{1}{2}\sum_{j\neq 0;\, j\neq i}\vec{h}_j\right)\right]\ . \quad \text{(A5)}\end{aligned}$$

One should keep in mind that the integrals are taken either over the surface or the volume of the Wigner-Seitz cell centered around zero. With the help of some more vector analysis one also finds

$$\begin{aligned}&\sum_{i\neq 0}\oint d\vec{S}\cdot\left[\left(\vec{h}_0+\frac{1}{2}\sum_{j\neq 0;\, j\neq i}\vec{h}_j\right)\times\text{curl}\vec{h}_i + \vec{h}_i\times\text{curl}\left(\vec{h}_0+\frac{1}{2}\sum_{j\neq 0;\, j\neq i}\vec{h}_j\right)\right]\\
&\quad= \oint d\vec{S}\cdot\vec{\nabla}\sum_{i\neq 0}\vec{h}_0\cdot\left(\vec{h}_i+\frac{1}{2}\sum_{j\neq 0;\, j\neq i}\vec{h}_j\right)\\
&\quad= \oint d\vec{S}\cdot\vec{\nabla}\left(\vec{h}_k\cdot\vec{h}_0 + (\vec{h}_0+\vec{h}_k)\cdot\sum_{i\neq 0;\, i\neq k}\vec{h}_i + \frac{1}{2}\sum_{i\neq 0;\, i\neq k}\vec{h}_i\cdot\sum_{j\neq 0;\, j\neq k,\, i}\vec{h}_j\right). \quad \text{(A6)}\end{aligned}$$

Here $\vec{h}_k = \vec{h}_0(\vec{r}-\vec{r}_k)$ can refer to any lattice site except the origin. From the symmetry of the vortex lattice it now follows that the expression in (A6) vanishes. To see this, consider one of the plane surface segments, S_1, say, of the Wigner-Seitz cell and let $\vec{r}_k$ be the lattice point obtained by reflection of the origin at S_1. Then the terms appearing in (A6) are symmetric with respect to reflection at S_1. Therefore, their normal derivatives vanish at S_1 and the expression in (A6) is zero. Finally, this result applied to (A5) gives

$$\epsilon_1 = 2\pi c \sum_{i\neq 0} h_i(0) = 2\pi c^2 \sum_{i\neq 0} K_0(r_i) \ . \qquad \text{(A7)}$$

Similarly, one obtains for ϵ_2 the result given in Eq. (18).

APPENDIX B

We show that $d = c/\sqrt{2}$ holds for superconductors with $\kappa = 1/\sqrt{2}$ and furthermore, that in this case the interaction between vortices vanishes exactly. For $\kappa = 1/\sqrt{2}$ the GL equations (2) and (3) admit first integrals which all two-dimensional bulk solutions satisfy. Letting the z axis be the symmetry axis of the superconductor, the first integrals are given by[24]

$$\text{curl}\ \vec{Q} = (1 - f^2)\hat{z}/\sqrt{2} \ , \qquad \text{(B1)}$$

$$\vec{\nabla} f = -f(\vec{Q}\times\hat{z})/\sqrt{2} \ . \qquad \text{(B2)}$$

The asymptotic forms of (B1) and (B2) far away from the core of an isolated vortex are

$$\frac{d}{dr}\left[\frac{1}{r}\frac{d}{dr}(rQ)\right] - Q = 0 \ , \quad \frac{dg}{dr} = -\frac{Q}{\sqrt{2}} \ . \qquad \text{(B3)}$$

The solutions of (B3) are given by

$$Q = cK_1(r) \ , \quad g = cK_0(r)/\sqrt{2} \ ,$$

so that $d = c/\sqrt{2}$ holds.

Next we insert (B1) and (B2) into the free-energy equation (1) integrating over one Wigner-Seitz cell. Making use of Eq. (3) one obtains

$$\epsilon = 2\int d^3r\{Q^2f^2 + (\text{curl}\vec{Q})^2\} = 2\oint d\vec{S}\cdot[\vec{Q}\times\text{curl}\vec{Q}] \ . \qquad \text{(B4)}$$

The integral over the surface of the Wigner-Seitz cell vanishes since the tangential component of $\vec{Q}$ is zero. There is, however, a contribution from the vortex core leading to

$$\epsilon = 2\int_0^{2\pi} d\varphi\, r\frac{1}{r\kappa}\frac{1}{\sqrt{2}} = 2\frac{2\pi}{\kappa}\frac{1}{\sqrt{2}} = 2\phi_0 H_{c1} = \epsilon_0 \ , \qquad \text{(B5)}$$

where ϕ_0 is the flux quantum. This shows that there is no interaction between vortices. At an applied field $H_0 = H_{c1} = H_{c2} = H_c$ the Gibbs's free energy is always zero.

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*Work supported by the National Science Foundation.

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