PHYSICAL REVIEW B VOLUME 24, NUMBER 9 1 NOVEMBER 1981

Critical points of infinite order in charge-density-wave systems and in superconductors

A. E. Jacobs
Department of Physics and Scarborough College, University of Toronto, Toronto, Ontario M5S 1A7 Canada

David Mukamel*
IBM Thomas J. Watson Research Center, Yorktown Heights, New York 10598

M. B. Walker
Department of Physics and Scarborough College, University of Toronto, Toronto, Ontario M5S 1A7 Canada

(Received 2 June 1981)

Two models, one describing charge-density-wave formation and the other (the Ginzburg-Landau model) describing the superconducting state, are studied and shown to exhibit infinite-order critical points. Related models exhibiting a Lifshitz point, a bicritical point, and a tetracritical point are also discussed.

I. INTRODUCTION

Consider a Landau theory of a continuous phase transition for which the free energy relative to the disordered phase is

$$\Delta F = a_2\eta^2 + a_4\eta^4 + a_6\eta^6 + \cdots \quad , \tag{1.1}$$

where η is the order parameter. As discussed in Ref. 1, at an ordinary critical point $a_2=0$ and $a_4>0$, while at a critical point of order $\frac{1}{2}p$, $a_n=0$ for $n<p$ and $a_p>0$.

Suppose that a critical point of order $\frac{1}{2}p$ occurs at a temperature T_c, and that the coefficients $a_n \propto (T-T_c)$ for $n<p$ and temperatures close to T_c. Then, for temperatures T just below T_c,

$$\eta \propto (T_c - T)^{[1/(p-2)]} \quad . \tag{1.2}$$

In this paper we study two models, one describing charge-density-wave formation and the other (the Ginzburg-Landau model) describing the superconducting state, both of which possess critical points of infinite order. At an infinite-order critical point, the coefficients a_p in (1.1) are zero for all p and the free energy is independent of the order parameter. Also, we expect, from taking the limit $p \to \infty$ in (1.2), that the order parameter undergoes a step discontinuity as the temperature is lowered through T_c. In the models studied below, it will be shown that the apparently discontinuous change in the order parameter can be brought about continuously by proceeding via a continuously infinite sequence of states all of which have the same free energy at the infinite-order critical point. Thus, at the infinite-order critical point, a discontinuity in the order parameter (the discontinuity being the difference between the values at temperatures just above and just below T_c) can be brought about continuously.

The behavior at the triple points of these two very different models is highly unusual; the fact that all the coefficients in the Landau expansion vanish at the same point may be due to some underlying symmetry of the models.

II. CHARGE-DENSITY-WAVE MODEL

In what we shall call the charge-density-wave model, the free energy per unit length L relative to that of the normal phase is

$$\Delta F = L^{-1}\int dx\left[v|\psi|^2 + \gamma\left|\left(i\frac{d}{dx}+1\right)\psi\right|^2 - \mathrm{Re}(\psi^3) + \tfrac{1}{2}|\psi|^4\right] \quad . \tag{2.1}$$

This model has been used previously[2–5] to describe a phase transition to a charge-density-wave state, the charge density in the ordered phase being given in terms of the complex order parameter $\psi(x)$. The phase diagram[5] for this model is shown schematically in Fig. 1. The order parameter ψ is zero in the normal state, constant, and nonzero in the commensurate state, and a spatially varying, periodic function in the incommensurate state. In the incommensurate state, $|\psi(x)|$ has a spatial period equal to one-third that of $\psi(x)$ itself. Close to the boundary (in Fig. 1) between commensurate and incommensurate regions, the incommensurate state can be viewed as a periodic domain structure, each domain corresponding to one

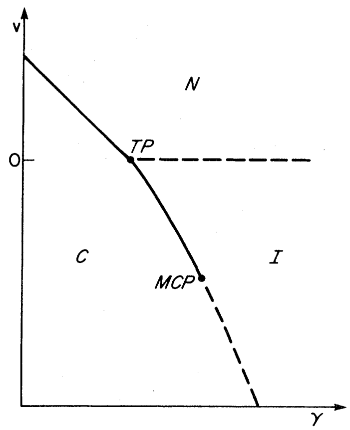

FIG. 1. Phase diagram (schematic only) for the model free energy of Eq. (2.1) showing the regions of stability of the normal (N), commensurate (C), and incommensurate (I) phases. First-order (second-order) transitions are shown by solid (dashed) lines. The triple point (TP) and the multicritical point (MCP) occur at $(\gamma, v) = (\frac{1}{2}, 0)$ and $(6.16, -60.8)$, respectively (Ref. 5). Note that the two first-order lines have different slopes at the triple point, due to the fact that TP is not an ordinary critical point but rather a critical point of infinite order. Numerical calculations (page 4147 of Ref. 5) suggest that the discontinuity in the order parameter ψ at the CI line goes to zero as the triple point is approached; that is, the discommensuration spacing just above the line appears to approach infinity in this limit.

of three distinct commensurate phases differing from one another in the phase of ψ but not its magnitude; at a domain wall (or discommensuration) the phase of ψ changes by $2\pi/3$.

At the triple point $v=0$ and $\gamma=\frac{1}{2}$ in Fig. 1, the normal, commensurate, and incommensurate phases coexist. Variational calculations[3,4] at this point gave the unexpected result that the free energy is independent of the spacing of the discommensurations; an analytical proof of this result has been given in Ref. 5 — a simpler proof is given below. Jackson, Lee, and Rice[4] noticed that, in a Landau expansion of the free energy, the coefficients a_2, a_4, and a_6 as defined in Eq. (1.1) all vanished at the triple point; they concluded that "something unusual is occurring" at the triple point and showed that certain thermodynamic properties are singular as the triple point is approached in the incommensurate phase. We shall show that the coefficients a_p in (1.1) vanish for all p for a Landau expansion appropriate to the model described by Eq. (2.1), and that the triple point is therefore a critical point of infinite order.

All further work will be done at the triple point where the free-energy density [the integrand in Eq. (2.1)] is

$$f=\frac{1}{2}\left|\frac{d\psi}{dx}\right|^2-\mathrm{Re}\left(i\psi\frac{d\psi^*}{dx}\right)+\frac{1}{2}|\psi|^2-\mathrm{Re}(\psi^3)+\frac{1}{2}|\psi|^4 . \tag{2.2}$$

The condition for an extremum of the free energy is that its functional derivative with respect to ψ^* be zero, which gives

$$-\frac{1}{2}\frac{d^2\psi}{dx^2}+i\frac{d\psi}{dx}+\frac{1}{2}\psi-\frac{3}{2}\psi^{*2}+\psi^2\psi^*=0 . \tag{2.3}$$

A first integral of (2.3) is[5]

$$i\frac{d\psi}{dx}=-\psi+\psi^{*2} . \tag{2.4}$$

Equation (2.4) is easily seen to have the solution $\psi=0$ corresponding to the normal phase, and the solutions $\psi=1$, $\psi=\exp(i2\pi/3)$, and $\psi=\exp(i4\pi/3)$ corresponding to distinct commensurate phases. Exact inhomogeneous periodic solutions of (2.4) have been obtained previously[5]; a given solution is characterized by giving its spatial period and a continuous infinity of solutions is obtained by allowing the period to vary continuously from a certain minimum allowed value to infinity; these solutions correspond to incommensurate states.

It will now be shown that all of these solutions of (2.4) must have the same free energy. Any solution of (2.4) must of course satisfy (2.3). By multiplying the left-hand side of (2.3) by ψ^* and subtracting the real part of the result from the right-hand side of (2.2), one finds

$$f=\frac{1}{2}\frac{d}{dx}\left[\mathrm{Re}\left(\psi\frac{d\psi^*}{dx}\right)\right]+\frac{1}{2}\mathrm{Re}(\psi^3)-\frac{1}{2}|\psi|^4 . \tag{2.5}$$

By using (2.4), f can now be rewritten

$$f=\frac{1}{2}\frac{d}{dx}\left[\mathrm{Re}\left(\psi\frac{d\psi^*}{dx}-\frac{1}{3}i\psi^3\right)\right] . \tag{2.6}$$

Such a free-energy density will give only a surface contribution to the free energy, which can be neglected. Thus, for all solutions of (2.4), $\Delta F=0$.

Although exact analytic solutions of (2.4) have been found previously,[5] it is of interest to obtain these solutions in such a way that a Landau expansion of the free energy can be generated in the form of Eq. (1.1) to all orders in η. To this end, we write the order parameter in the periodic incommensurate state in the form

$$\psi(x)=\phi(\delta x)=\phi(t) \tag{2.7}$$

and impose the condition

$$\phi(t)=\phi(t+2\pi) . \tag{2.8}$$

Equation (2.4) is now

$$i\delta\frac{d\phi}{dt}+\phi-\phi^{*2}=0 \ . \tag{2.9}$$

Furthermore, an expansion for both ϕ and δ in powers of the amplitude η of the fundamental wave will be sought in the form

$$\phi(t)=\eta e^{it}+\sum_{n=2}^{\infty}\phi_n\eta^n \ , \tag{2.10}$$

$$\delta=1+\sum_{n=1}^{\infty}\delta_n\eta^n \ . \tag{2.11}$$

Equations (2.10) and (2.11) are now substituted into (2.9) and coefficients of the different powers of η occurring on the left-hand side of (2.9) are set equal to zero. Thus, a sequence of equations is obtained which can be solved in such a way as to obtain the coefficients ϕ_p and δ_p in terms of ϕ_n's and δ_n's having $n<p$. This procedure can be used to obtain ϕ and δ to any desired order, and we find

$$\psi(x)=\eta e^{i\delta x}+(\tfrac{1}{3}\eta^2+\tfrac{4}{27}\eta^4+\tfrac{30}{243}\eta^6)e^{-2i\delta x} -(\tfrac{1}{27}\eta^4+\tfrac{14}{243}\eta^6)e^{4i\delta x}-\tfrac{1}{81}\eta^5e^{-5i\delta x}+O(\eta^7) \tag{2.12}$$

and

$$\delta=1-\tfrac{2}{3}\eta^2-\tfrac{8}{27}\eta^4+O(\eta^6) \ . \tag{2.13}$$

Similar expansions for the amplitude and phase of ψ have been given previously.[5]

The result (2.12), when substituted in the expression (2.1) for the free energy, clearly yields a Landau expansion for the free energy in the form (1.1); the result (2.12) is sufficient to allow the coefficients a_2, a_4, a_6, and a_8 occurring in (1.1) to be found explicitly, and all are found to be zero. In principle, ψ can be found to any desired order in η, and the expansion of ΔF can thus also be found to any desired order in η. Now note that (2.12) represents a continuum of solutions of (2.4), each distinct solution corresponding to a distinct value of η (or δ). Since, as was shown above, all solutions of (2.4) have the free energy $\Delta F=0$, ΔF is independent of η and the coefficients a_p in the expansion (1.1) are zero for all p. Thus, the triple point is an infinite-order critical point.

Notice that, at this infinite-order critical point, an infinite continuum of incommensurate states exists, all having the same free energy. These states are such that one can proceed through them from the normal state with $\psi=0$ to a commensurate state with $|\psi|=1$ while allowing only continuous changes of the order parameter ψ. One does this by starting with the normal state for which $\psi=0$, proceeding to the incommensurate state of Eq. (2.12) for which ψ is infinitesimal and $\delta=1$, and then going through successive incommensurate states with continuously increasing spatial periods until the period becomes infinite, at which point the commensurate state has been reached. In this way, an apparently discontinuous change of the order parameter (from $\psi=0$ to $|\psi|=1$) can be achieved continuously, which is the type of behavior which might be expected at an infinite-order critical point.

It can be shown, by developing an expansion of ψ similar to (2.12) and using it to calculate the free energy correct to terms of order η^6 that if a term proportional to $|\psi|^6$ is added to the free energy density of Eq. (2.2), the infinite-order critical point is no longer present, but that a tricritical point occurs on the normal to incommensurate phase boundary.[4]

We note that in order to perform a scaling analysis in the vicinity of the infinite-order critical point one should first calculate the coefficients $a_{2p}(v,\gamma)$ for small v and $(\gamma-\frac{1}{2})$. This has not been done in the present work.

The model (2.1) corresponds to CDW systems in which the commensurate phase is associated with a reciprocal-lattice vector $q_0=2\pi/m$ with $m=3$. Phase diagrams of systems for which $m\neq 3$ are not given by Fig. 1; in particular they do not exhibit an infinite-order critical point. In the following we analyze the phase diagrams associated with CDW systems with $m\neq 3$. We will show that for $m=2$, the phase diagram exhibits a Lifshitz point while for $m>3$ the three transition lines join via a bicritical or tetracritical point.

Consider first the case $m=2$. The free energy per unit length L relative to that of the normal phase is

$$\Delta F=L^{-1}\int dx\left[v|\psi|^2+\gamma\left|\left(i\frac{d}{dx}+1\right)\psi\right|^2 -\mathrm{Re}(\psi^2)+\tfrac{1}{2}|\psi|^4\right] \ . \tag{2.14}$$

In order to study the N-C and N-I transitions associated with this model we diagonalize the quadratic terms in (2.14) and rewrite the free energy in terms of the Fourier components ϕ_{1q} and ϕ_{2q} of the two eigenvectors ϕ_1 and ϕ_2. The model (2.14) takes the form

$$\Delta F\propto\int dq\,[\lambda_1(q)\phi_{1q}\phi_{1,-q}+\lambda_2(q)\phi_{2q}\phi_{2,-q}+O(\psi^4)] \ , \tag{2.15}$$

where

$$\lambda_{1,2}=(v+\gamma)+\gamma q^2\mp(1+4\gamma^2q^2)^{1/2} \ . \tag{2.16}$$

Since $\lambda_1\le\lambda_2$ the phase transitions from the normal phase are determined by $\lambda_1(q)$. For $\gamma<\frac{1}{2}$, $\lambda_1(q)$ attains its minimum at $q=0$, and therefore $\lambda_1(0)=0$

defines the N-C transition. This transition is located at

$$v+\gamma-1=0 \ . \tag{2.17}$$

For $\gamma > \frac{1}{2}$, however, the minimum of $\lambda_1(q)$ is obtained at $q=q_1$ where

$$q_1^2 = 1-\frac{1}{4\gamma^2} \ . \tag{2.18}$$

The N-I transition is given by $\lambda_1(q_1)=0$, namely

$$v-\frac{1}{4\gamma}=0 \ . \tag{2.19}$$

At the intersection of the two critical lines (2.17) and (2.19) one has $(\partial^2\lambda_1/\partial q^2)_{q=0}=0$, with $(\partial^4\lambda_1/\partial q^4)_{q=0}>0$. This is therefore a Lifshitz point.[6] The (v,γ) phase diagram of the model (2.15) is given in Fig. 2. The N-C line is Ising-like (with $\phi_{1,q=0}$ being the order parameter), while the N-I line is associated with an $n=2$-component order parameter ϕ_{1q_1} and $\phi_{1,-q_1}$. The C-I transition is first order.[7] Detailed scaling and renormalization-group analyses in the vicinity of the Lifshitz point have been carried out in previous studies.[8]

We now consider the case $m>3$. The free energy takes the form:

$$\Delta F = L^{-1}\int dx\left[v|\psi|^2+\gamma\left|\left(i\frac{d}{dx}+1\right)\psi\right|^2 + \tfrac{1}{2}|\psi|^4-\mathrm{Re}(\psi^m)+u|\psi|^{2l}\right] \ , \tag{2.20}$$

where for stability of the free energy we have included a term $u|\psi|^{2l}$ with $2l \geq m$ and $u>0$. Rewriting (2.20) in terms of the Fourier components of ψ we find

$$\Delta F \propto \int dq\,[\lambda(q)\psi_q\psi_q^* + O(\psi_q^4)] \ , \tag{2.21}$$

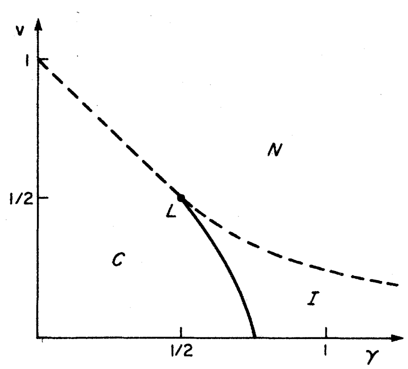

FIG. 2. (v,γ) phase diagram for the case $m=2$. The three phase transition lines meet at a Lifshitz point L. The C-I transition line is schematic.

where

$$\lambda(q)=v+\gamma(1-q)^2 \ . \tag{2.22}$$

This free energy exhibits a N-I transition at $v=0$ for $\gamma \geq 0$. The transition is associated with ψ_{q_1}, with $q_1=1$. However, one has to be careful in applying this result to real CDW systems. The expression (2.22) for $\lambda(q)$ is, in fact, an expansion in powers of $(1-q)$. In the limit of small γ, one has to take into account higher-order terms in $(1-q)$, and consider $\lambda(q)$ of the form

$$\lambda(q)=v+\gamma(1-q)^2+\gamma_3(1-q)^3+O((1-q)^4) \ . \tag{2.23}$$

This amounts to including a term $-\gamma_3\int dx[\psi^*(id/dx+1)^3\psi]$ in the free energy (2.20). The γ_3 term in (2.23) affects the phase diagram quite drastically. For small $\gamma(\gamma>0)$, $\lambda(q)$ will favor ordering of ψ_{q_2} with $q_2 \neq 1$. q_2 is determined by γ, γ_3 and the higher-order terms in (2.23). One therefore expects these CDW systems to exhibit at least two incommensurate phases $I1$ and $I2$, associated with reciprocal-lattice vectors q_1 and q_2, respectively. A possible (v,γ) phase diagram is given schematically in Fig. 3. It exhibits two $n=2$ critical lines, N-$I1$ and N-$I2$ which join at a point M. At this point the two order parameters ψ_{q_1} and ψ_{q_2} become critical simultaneously. This $n=4$ multicritical point is either bicritical [Fig. 3(a)] or tetracritical [Fig. 3(b)] depending upon the various parameters in the free energy.[9] In the bicritical case the $I1$-$I2$ transition is first order. In the tetracritical case, the $I1$ and $I2$ phases are separated by an intermediate phase in which both ψ_{q_1} and ψ_{q_2} are nonzero.[9] Note that this analysis is not dependent on the presence of the term Re (ψ^m) in Eq. (2.20), but rather on the fact that γ becomes small. Mean-field calculations[10,11] and experimental studies[12] suggest that the analysis presented in this section, both for $m=2$ and $m>3$, should be applicable to the phase diagram of TTF-TCNQ under pressure.

III. GINZBURG-LANDAU THEORY OF SUPERCONDUCTORS

We show in this section that this theory[13] has a critical point of infinite order at $\kappa=1/\sqrt{2}$, $H_a=H_c$ where κ is the Ginzburg-Landau parameter, H_a is the applied field, and H_c is the thermodynamic critical field.

In the usual units and notation,[14–17] the Gibbs free

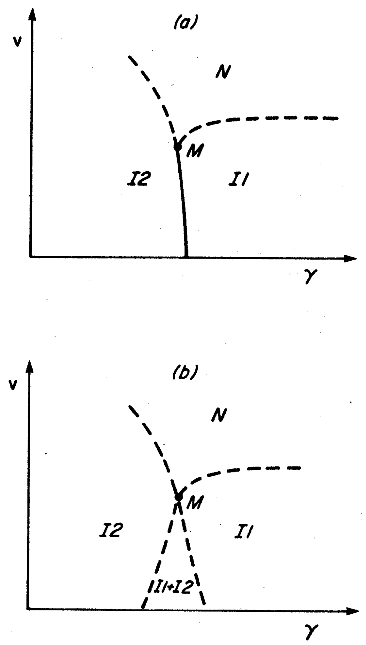

FIG. 3. Schematic (v,γ) phase diagram for the case $m>3$. The phase diagram is either (a) bicritical-like or (b) tetracritical-like, depending upon the various parameters in the free energy. (a) the $I1$-$I2$ transition is first order, and M is a bicritical point; (b) the two incommensurate phases $I1$ and $I2$ are separated by an intermediate phase in which both incommensurate order parameters ψ_{q_1} and ψ_{q_2} are nonzero. M is a tetracritical point. At the multicritical point M all transition lines are parallel to each other. This result is obtained by calculations which go beyond the mean-field approximation (see, for example, Ref. 9).

energy relative to the Meissner state is

$$\Delta G = H_c^2\lambda^2 L(4\pi)^{-1}\int d^2r\,[h^2-2hh_a+\tfrac{1}{2}(1-f^2)^2+\kappa^{-2}(\vec{\nabla} f)^2+f^2v^2] \tag{3.1}$$

for a long cylinder (of length L) parallel to the constant applied field $\vec{H}_a=H_a\hat{z}=\sqrt{2}H_ch_a\hat{z}$. The microscopic magnetic field $\vec{H}(\vec{r})=\sqrt{2}H_ch(\vec{r})\hat{z}$ is also in the z direction. The Ginzburg-Landau order parameter Ψ is written as $\Psi=\Psi_M fe^{i\Phi}$ where Ψ_M is the value of Ψ in the Meissner state and f and Φ are real. Lengths are measured in units of the penetration depth λ. The vector potential is $\vec{A}(\vec{r})=\sqrt{2}H_c\lambda\vec{a}(\vec{r})$ and the superfluid velocity is $\vec{v}=\kappa^{-1}\vec{\nabla}\Phi-\vec{a}$, a vector in the xy plane. The functions h, f, and $\vec{v}$ are independent of the coordinate z. All the temperature dependence of the theory is in H_c and λ.

The states of interest are the normal state ($h=h_a$, $f=0$), the Meissner state ($h=0$, $f=1$, $\vec{v}=0$) and the mixed state, discovered by Abrikosov,[18] in which h, f, and $\vec{v}$ are periodic. The phase diagram is shown in Fig. 4. The Meissner-normal transition is first order; the discontinuity in the order parameter is independent of κ. The Meissner-mixed and mixed-normal transitions are second order; the former is of the λ type. The flux first penetrates, at $H_a=H_{c1}$, in the form of one singly-quantized vortex. The interaction between vortices is repulsive,[19,20] but is exponentially weak at large distances so that many vortices enter with a slight increase of H_a beyond H_{c1}; the magnetization curve is continuous at H_{c1} but has infinite slope at H_{c1}^+. In the intermediate field range $H_{c1}<H_a<H_{c2}$, the vortices form a periodic, two-dimensional array. At $H_a=H_{c2}^-$, the microscopic magnetic field is H_a and the order parameter is infinitesimal; the vortices are singly quantized and form a "triangular" lattice.[14,21]

We show now that the free-energy difference ΔG vanishes at the triple point $\kappa=h_a=1/\sqrt{2}$, independent of the structure of the vortex lattice. The proof is a generalization of that used to show that the normal-superconducting wall energy vanishes[14,15]; it relies on the fact that the Ginzburg-Landau equations

$$\kappa^{-2}\nabla^2 f - f(v^2+f^2-1)=0\ , \tag{3.2}$$

$$\vec{\nabla}\times(\vec{\nabla}\times\vec{v})+f^2\vec{v}=0\ , \tag{3.3}$$

have, for $\kappa=1/\sqrt{2}$, the first integrals

$$\sqrt{2}\vec{h}=(1-f^2)\hat{z}\ , \tag{3.4}$$

$$\sqrt{2}\vec{\nabla} f=f\vec{v}\times\hat{z}\ . \tag{3.5}$$

Multiplication of Eq. (3.2) by f and addition of the resulting left-hand side to the integrand in Eq. (3.1)

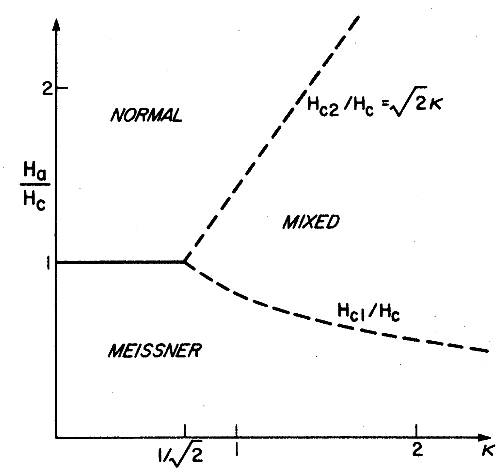

FIG. 4. Phase diagram in the Ginzburg-Landau theory of superconductors.

yields

$$\Delta G \alpha \int d^2r[h^2 - 2hh_a + \tfrac{1}{2}(1-f^4) + \kappa^{-2}\vec{\nabla}\cdot(f\vec{\nabla}f)] \ . \tag{3.6}$$

The integral of the last term vanishes (by virtue of the divergence theorem and the fact that the unit cell can be chosen so that $\vec{\nabla}f$ is parallel to the boundary). Then use of Eq. (3.4) gives $\Delta G = 0$ when $h_a = 1/\sqrt{2}$.

According to the arguments of Sec. II, if a Landau expansion for the order parameter exists, then the fact that $\Delta G = 0$, whatever the periodicity, implies that all the coefficients in the corresponding expansion of the free-energy density vanish; hence the critical point $\kappa = h_a = 1/\sqrt{2}$ is of infinite order. Most of the remainder of this section is devoted to an explicit calculation of the Landau expansion.

We first eliminate the functions h and $\vec{v}$ from Eqs. (3.4) and (3.5) in favor of the function $g = f^2$, obtaining

$$\nabla^2 \ln g = -1 + g \ . \tag{3.7a}$$

The solution of this equation for infinitesimal g is well known[14] to be $g = |\psi_1(\vec{r})|^2$, where

$$\psi_1(\vec{r}) = \sum_{n=-\infty}^{\infty} c_n e^{2\pi i n x/a} e^{-1/4(y+4\pi n/a)^2} \tag{3.7b}$$

and the c_n are infinitesimals. This solution, however, cannot be used as the starting point of an expansion.

A suitable form of Eq. (3.7a) is

$$\nabla^2 \ln g = -\delta + (g - \langle g \rangle) \ , \tag{3.8}$$

where $\langle g \rangle$ is the spatial average of g and

$$\delta = 1 - \langle g \rangle \ . \tag{3.9}$$

The differential equation can be converted into the integral equation

$$\ln g = \ln w + (2\pi)^{-1} \int [g(\vec{r}\,') - \langle g \rangle] \ln|\vec{r}\,' - \vec{r}| d^2r' \ , \tag{3.10}$$

where w satisfies

$$\nabla^2 \ln w = -\delta \ ; \tag{3.11}$$

the divergence of the integral in Eq. (3.10) will be cured momentarily. A solution of Eq. (3.11) which reduces to the proper form when $\delta = 1$ (i.e., when g is infinitesimal) is

$$w = \text{const}|\psi_\delta(\vec{r})|^2 \ , \tag{3.12}$$

$$\psi_\delta(\vec{r}) = \sum_{n=-\infty}^{\infty} C_n e^{2\pi i n \delta x/a} e^{-1/4\delta(y+4\pi n/a)^2} \ . \tag{3.13}$$

Then Eq. (3.10) becomes

$$g(\vec{r}) = \eta^2 |\psi_\delta(\vec{r})|^2 \times \exp\left[\frac{1}{2\pi}\int [g(\vec{r}\,') - \langle g \rangle] \times \ln\left(\frac{|\vec{r}\,' - \vec{r}|}{|\vec{r}\,' - \vec{r}_0|}\right) d^2r'\right] \ , \tag{3.14}$$

where η is a parameter governing the magnitude of g and $\vec{r}_0$ is an arbitrary position vector; note that

$$\eta^2 = g(\vec{r}_0)/|\psi_\delta(\vec{r}_0)|^2 \ . \tag{3.15}$$

Equation (3.14) is our basic result; from it, an expansion of g in powers of η can be obtained by iteration. Clearly the periods of g are those of $|\psi_\delta(\vec{r})|$. From Eq. (3.13), one elementary translation vector is

$$\vec{t}_1 = a\hat{x}/\delta \ ; \tag{3.16}$$

a second is (see Fig. 5)

$$\vec{t}_2 = b(\hat{x}\cos\alpha + \hat{y}\sin\alpha) \tag{3.17}$$

provided that

$$ab\sin\alpha = 4\pi p \ , \tag{3.18}$$

where p is an integer, and

$$C_{n+p} = C_n \exp[2\pi i n \delta(b/a)\cos\alpha] \ . \tag{3.19}$$

A form which preserves the shape of the lattice is obtained by the replacements $a = a'\delta^{1/2}$, $b = b'\delta^{-1/2}$ where a' and b' are independent of δ. One notes that Eqs. (3.14) and (3.16) to (3.19) reduce to the correct forms[14] when $\delta = 1$.

It is an important feature of the above results that the flux-quantization condition

$$\int_{\text{cell}} \hat{z}\cdot\vec{h}\, d^2r = \frac{2\pi p}{\kappa} \tag{3.20}$$

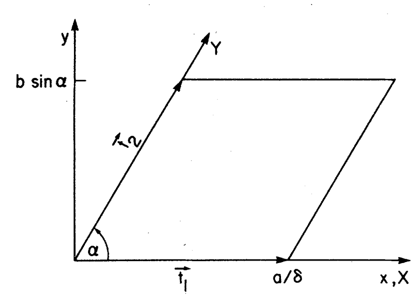

FIG. 5. Elementary translation vectors $\vec{t}_1$ and $\vec{t}_2$ of the vortex lattice. Also shown are the axes of the coordinates $X = x - y/\tan\alpha$ and $Y = y/\sin\alpha$.

is satisifed to all orders in η. From Eq. (3.4),

$$\int_{\text{cell}} \hat{z}\cdot\vec{h}\, d^2r = \frac{A(1-\langle g\rangle)}{\sqrt{2}} \ , \qquad (3.21)$$

where A, the area of the unit cell, is

$$A = ab\delta^{-1}\sin\alpha = 4\pi p/\delta \qquad (3.22)$$

from Eq. (3.18). Hence

$$\int_{\text{cell}} \hat{z}\cdot\vec{h}\, d^2r = 2\sqrt{2}\pi p \ , \qquad (3.23)$$

in agreement with Eq. (3.20).

As in Sec. II, we have two parameters, one (η) governing the amplitude and one (δ) the periodicity. Again, the two are not independent; Eq. (3.9) can be viewed as a self-consistency requirement which determines one, given the other.

It remains to show explicitly that Eq. (3.14) provides an expansion of g in powers of η. To do this, it is convenient to make an explicit choice for the p independent C's. (For $p=1$, each unit cell contains one singly-quantized vortex; for $p=2$, each contains two singly-quantized vortices — i.e., the lattice has a basis — or one doubly-quantized vortex, and so on.) Now, for $\kappa > 1/\sqrt{2}$, it is believed that a "triangular" lattice of singly quantized vortices minimizes the free energy for all values of H_a between H_{c1} and H_{c2}; this choice ($p=1$, $a=b\delta$, $\alpha=\pi/3$) is also simple mathematically. We make it for both these reasons. Taking $C_0=1$, we then have

$$C_n = e^{\pi i n(n-1)/2} \ . \qquad (3.24)$$

Also, $|\psi_\delta(\vec{r})|^2$ has the double Fourier-series expansion

$$|\psi_\delta(\vec{r})|^2 = \sum_{n=-\infty}^{\infty}\sum_{m=-\infty}^{\infty} C_{mn} e^{2\pi i(nX+mY)/b} \ , \qquad (3.25)$$

where $x = X + Y\cos\alpha$, $y = Y\sin\alpha$, $b^2 = 8\pi/(\sqrt{3}\delta)$, and

$$C_{mn} = 3^{-1/4}(-1)^{mn} e^{-\pi(m^2+n^2-mn)/\sqrt{3}} e^{-\pi i n/2} \ ; \qquad (3.26)$$

the asymmetry is removed by the replacement $X \to X + b/4$.

We now use Eq. (3.14) to generate the expansion

$$g(\vec{r}) = \eta^2 g_2(\vec{r}) + \eta^4 g_4(\vec{r}) + \cdots \ ; \qquad (3.27)$$

the only requirement for the validity of this expansion, at least for small η, is that the integral

$$I_k(\vec{r},\vec{r}_0) = \int \frac{d^2r'}{2\pi}[g_k(\vec{r}) - \langle g_k\rangle] \ln\left(\frac{|\vec{r}\,' - \vec{r}|}{|\vec{r}\,' - \vec{r}_0|}\right) \qquad (3.28)$$

exist. Now g_k clearly has the representation

$$g_k = \sum_{n=-\infty}^{\infty}\sum_{m=-\infty}^{\infty} G_{mn}^{(k)} e^{2\pi i(nX+mY)/b} \qquad (3.29)$$

and the average of g_k is

$$\langle g_k\rangle = G_{00}^{(k)} \ . \qquad (3.30)$$

Writing the integral as

$$\int d^2r' \ldots = \lim_{M\to\infty}\int_{-Mb\sin\alpha}^{Mb\sin\alpha} dy' \lim_{N\to\infty}\int_{-Nb}^{Nb} dx' \ldots \ , \qquad (3.31)$$

one finds

$$I_k(\vec{r},\vec{r}_0) = \frac{-3b^2}{16\pi^2}\sum_{mn}{}' \frac{G_{mn}^{(k)}}{m^2+n^2-nm}\left(e^{2\pi i(nX+mY)/b} - e^{2\pi i(nX_0+mY_0)/b}\right) , \qquad (3.32)$$

where the prime means that the term $m=0$, $n=0$ is to be omitted. The sum converges more rapidly than the sum in Eq. (3.29) and therefore I_k is finite for all k. The final step is to calculate δ in terms of η:

$$\delta = 1 - \eta^2\langle g_2\rangle - \eta^4\langle g_4\rangle + \cdots \ , \qquad (3.33)$$

where, from the above results,

$$\langle g_2\rangle = 3^{-1/4} \ , \qquad (3.34)$$

$$\langle g_4\rangle = \frac{-3b^2}{16\pi^2}\sum_{mn}{}' \frac{1}{m^2+n^2-mn}\left(3^{-1/2}e^{-2\pi(m^2+n^2-mn)/\sqrt{3}} - C_{mn}C_{00}e^{2\pi i(nX_0+mY_0)/b}\right) . \qquad (3.35)$$

We have thus proved that the triple point is a critical point of infinite order in the Ginzburg-Landau theory. It is natural to ask what happens when additional terms are added to the free energy. Now the correction terms of order $(1-T/T_c)$ to the free-energy density are known exactly from the work of Tewordt[22] and Neumann and Tewordt.[23] The consequences of the correction terms have been investigated by Tewordt and his group[23,24]; work particularly relevant to that of this article has been done by Jacobs,[25-27] who showed that these "Tewordt" terms lead to major changes in the phase diagram in the vicinity of the triple point. The detailed results are rather complicated because the correction terms depend on the ratio of the BCS coherence length ξ_0 to the mean free path l; the cases $\xi_0/l=0$ and ∞ are representative and are shown schematically in Fig. 6. The reason that the figure is only schematic is that the problem is still partially solved.

For "clean" to relatively "dirty" superconductors ($0 \leq \xi_0/l \lesssim 50$), the critical value of κ for type-II superconductivity is κ_{c1}.[25,27] For $\kappa_{c1} < \kappa < \kappa_{c6}$, the Meissner-mixed transition is first order because of an attractive interaction between vortices.[26] κ_{c1} and κ_{c6} both have the form $(1+\text{const}\Theta)/\sqrt{2}$ where $\Theta = 1 - T/T_c$ and the constants are known[25–27] for all ξ_0/l. Only the end points, not the location, of the first-order line are known. The region κ_{c1} and κ_{c6} for which the lower transition is first order decreases with increasing ξ_0/l and vanishes at $\xi_0/l \approx 50$. For $\kappa > \kappa_{c6}$, the Meissner-mixed transition is second order. The mixed-normal transition is second order for all $\xi_0/l \lesssim 50$. The locations of both second-order lines are known.[25–27]

For $\xi_0/l \gtrsim 50$, the critical value of κ for type-II superconductivity is[25] $\kappa_{c3} = (1+\text{const}\Theta)/\sqrt{2}$ where the constant is known for all ξ_0/l. The lower transition is second order. The upper transition is first order[25] for $\kappa > \kappa_{c3}$ but κ less than some unknown value; the location of the line is also unknown. For sufficiently large κ, the upper transition is second order. Both second-order lines are known.[25]

The feature of these results which is of interest here is that the Tewordt terms split the critical point of infinite order into two points with a first-order line joining them; the length of the first-order line is proportional to the magnitude $(1-T/T_c)$ of the Tewordt terms. This is similar to the behavior found by Jackson, Lee, and Rice[4] in their study of the effect of an additional term $|\psi|^6$ on the charge-density-wave model of Sec. II.

Finally, one can show from the results of Refs. 25 to 27 that there is a value of $\xi_0/l(\approx 50)$ such that both the Meissner-mixed and mixed-normal transitions are second order along their entire lengths (as

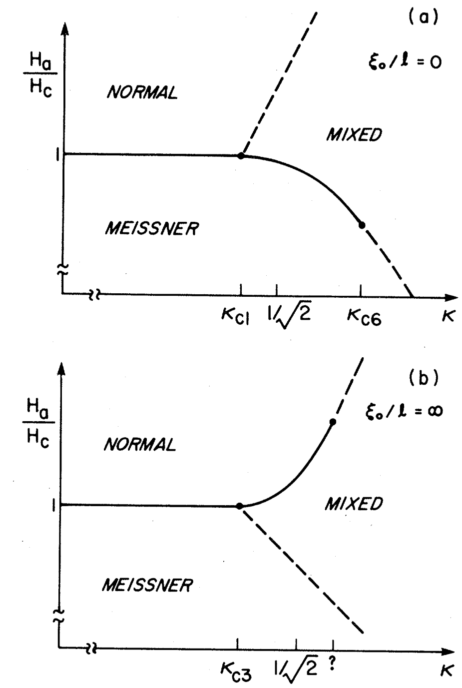

FIG. 6. Phase diagrams (schematic only) obtained in Tewordt's extension of the Ginzburg-Landau theory for the cases $\xi_0/l=0$ and ∞. The locations of the first-order lines on the mixed-phase boundaries are not known; the lines have been drawn to have zero slope at the triple points.

in Fig. 4 for the Ginzburg-Landau theory). One can also show that, at the triple point, the free energy is independent of the structure of the vortex lattice, but only to first order in $1-T/T_c$ and $\kappa - 1/\sqrt{2}$.

ACKNOWLEDGMENTS

This research was supported in part by a grant from the U.S.-Israel Binational Science Foundation (BSF), Jerusalem, Israel, and in part by the Natural Science and Engineering Research Council of Canada.

*Permanent address: Department of Physics, Weizmann Institute of Science, Rehovoth, Israel.

[1]See, e.g., P. Pfeuty and G. Toulouse, *Introduction to the Renormalization Group and to Critical Phenomena* (Wiley, New York, 1977); P. Bastie, J. Lajzerowicz, and J. Schneider, J. Phys. C 11, 1203 (1978); T. S. Chang, G. F. Tuthill, and H. E. Stanley, Phys. Rev. B 9, 4882 (1974); J. C. Slater, J. Chem. Phys. 9, 16 (1941).

[2]W. L. McMillan, Phys. Rev. B 14, 1496 (1976).

[3]K. Nakanishi and H. Shiba, J. Phys. Soc. Jpn. 45, 1147 (1978).

[4]S. A. Jackson, P. A. Lee, and T. M. Rice, Phys. Rev. B 17, 3611 (1978).

[5]A. E. Jacobs and M. B. Walker, Phys. Rev. B 21, 4132 (1980).

[6]R. M. Hornreich, M. Luban, and S. Shtrikman, Phys. Rev. Lett. 35, 1678 (1975).

[7]A. Michelson, Phys. Rev. B 16, 577 (1977).

[8]D. Mukamel and M. Luban, Phys. Rev. B 18, 3631 (1978).

[9]See, for example, K. S. Liu and M. E. Fisher, J. Low Temp. Phys. 10, 655 (1972); J. M. Kosterlitz, D. R. Nelson, and M. E. Fisher, Phys. Rev. B 13, 412 (1976).

[10]E. Abrahams and I. E. Dzyaloshinskii, Solid State Commun. 23, 883 (1977).

[11]C. Hartzstein, V. Zevin, and M. Weger, Solid State Commun. 36, 545 (1980).

[12]R. H. Friend, M. Miljak, and D. Jerome, Phys. Rev. Lett.

40, 1048 (1978); S. Megtert, R. Comès, R. Pynn, C. Vettier, and A. F. Garito, in *The Physics and Chemistry of Low Dimensional Solids,* edited by L. Alcácer (Reidel, London, 1980).
[13]V. L. Ginzburg and L. D. Landau, Zh. Eksp. Teor. Fiz. 20, 1064 (1950). A complete English translation is available in *Men of Physics: L. D. Landau,* edited by D. ter Haar (Pergamon, New York, 1965), Vol. I, pp. 138–167.
[14]D. Saint-James, G. Sarma, and E. J. Thomas, *Type II Superconductivity* (Pergamon, New York, 1969).
[15]A. L. Fetter and P. C. Hohenberg, in *Superconductivity,* edited by R. D. Parks (Marcel Dekker, New York, 1969), Chap. 14.
[16]M. Tinkham, *Introduction to Superconductivity* (McGraw-Hill, New York, 1975).
[17]P. G. de Gennes, *Superconductivity of Metals and Alloys* (Benjamin, New York, 1966).
[18]A. A. Abrikosov, Zh. Eksp. Teor. Fiz. 32, 1442 (1957) [Sov. Phys. JETP 5, 1174 (1957)].
[19]E. Müller-Hartmann, Phys. Lett. 23, 521 (1966).
[20]L. Kramer, Phys. Lett. 23, 619 (1966).
[21]W. M. Kleiner, L. M. Roth, and S. H. Autler, Phys. Rev. 133, A1226 (1964).
[22]L. Tewordt, Z. Phys. 180, 385 (1964); Phys. Rev. 137, A1745 (1965).
[23]L. Neumann and L. Tewordt, Z. Phys. 189, 55 (1966).
[24]L. Tewordt, Z. Phys. 184, 319 (1965); L. Neumann and L. Tewordt, *ibid.* 191, 73 (1966); G. Ebneth and L. Tewordt, *ibid.* 185, 421 (1965).
[25]A. E. Jacobs, Phys. Rev. B 4, 3016 (1971); 4, 3022 (1971).
[26]A. E. Jacobs, Phys. Rev. B 4, 3029 (1971).
[27]A. E. Jacobs, J. Low Temp. Phys. 10, 137 (1973).