Peculiarities of coexistence of phases with different electric conductivities under the influence of an electric current

Yu. Dolinsky* and T. Elperin†

*The Pearlstone Center for Aeronautical Engineering Studies, Department of Mechanical Engineering,
Ben-Gurion University of the Negev, P.O. Box 653, Beer-Sheva 84105, Israel*

In this study, using a Ginzburg-Landau model, we show that in the domain of coexistence of phases under the influence of the electric current, the phase equilibrium curve splits into two different curves for the direct and inverse phase transitions. We derived equations for spinodal and binodal lines in a current-carrying system taking into account the difference of electric conductivities of the phases. It is demonstrated that there exist thermodynamic regions where both phases are metastable simultaneously. The latter effect occurs due to the dependence of the nucleus formation work upon the relation between the conductivities of the phases and to the shift of the phase equilibrium curves for the direct and inverse phase transitions. At small electric currents, the region where both phases are metastable simultaneously corresponds to the coexistence of kink and antikink solutions. [S0163-1829(98)03830-2]

I. INTRODUCTION

In our recent studies,[1–4] we investigated peculiarities of phase transitions in current-carrying systems when such transitions are accompanied by a change of electric conductivity. It was demonstrated that the location of the phase equilibrium curve significantly depends not only upon the relation between the electric conductivities of phases, but also upon the geometry of a system. The case of when a phase transition begins at the surface and propagates into the conductor differs strongly from the case of when a phase transition begins inside the conductor and propagates towards its surface. At the initial stage of the surface phase transition, a phase with a lower conductivity propagates not only without additional work,[3] but ponderomotive forces promote propagation of this phase. Conversely, ponderomotive forces prevent the propagation of the phase with a higher electric conductivity into the conductor from its surface.[3,4] The situation is completely reversed in the case of an internal nucleation. Here, the nucleation of a phase with a lower electric conductivity requires additional energy, while nucleation of a phase with a higher conductivity occurs partially due to the work performed by the ponderomotive forces. These results are pertinent to such cases where a phase transition is described by a zero-dimensional Doering-Volmer-Zeldovich nucleation theory. That is the reason why the above results are expressed through the parameters that are commonly used for the description of these systems, e.g., latent heat of phase transition, specific volume, overheat, etc. On the other hand, several recent studies[5–8] have reported a strong effect of the electric current on phase transitions in alloys when a phase transition is accompanied by a change of an electric conductivity. Such systems like binary alloys can be described using the continual models for the order parameter, whereby one of the most commonly used and simple models is the Ginzburg-Landau model.[9] In this connection it is of interest to construct a model for the description of the above phenomena employing the Ginzburg-Landau model. The latter problem is solved in this study.

II. DESCRIPTION OF THE MODEL

In order to construct a continual model of a phase transition accompanied by a change of electric conductivity in the presence of electric current **j**, we will use the Ginzburg-Landau model for the Gibbs potential Ω. In the regime with a prescribed magnitude of electric current,[10] Ω can be written as

$$\Omega = \Omega_0 - \Omega_m, \quad (1)$$

where Ω_0 is a thermodynamic potential of a current-free system that is determined below and Ω_m is an addition to the thermodynamic potential due to the interaction of electric currents

$$\Omega_m = \frac{1}{2c^2} \int \frac{j(\mathbf{r})j(\mathbf{r}')}{|\mathbf{r}-\mathbf{r}'|} d^3\mathbf{r} \, d^3\mathbf{r}'. \quad (2)$$

Now, let us represent Ω_0 as

$$\Omega_0 = \int [f_0(\eta) - \mu\eta] d^3\mathbf{r} + \Omega_s, \quad (3)$$

where $\Omega_s = (\chi/2)\int(\nabla\eta)^2 d^3\mathbf{r}$ is the surface energy ($\chi > 0$), $f_0(\eta)$ is the specific free energy, and μ is the chemical potential. It was shown in Ref. 11 that equations

$$\dot{\eta} = -\frac{1}{\Gamma_n}\frac{\delta\Omega}{\delta\eta}, \quad \dot{\eta} = \frac{1}{\Gamma_s}\Delta\left(\frac{\delta\Omega}{\delta\eta}\right), \quad (4)$$

where Γ_n and Γ_s are the kinetic coefficients, allow us to describe the relaxation of the order parameter from a metastable phase to a stable phase. The first equation in Eqs. (4) describes a case where the order parameter is not conserved, while the second equation describes a case where the order parameter is conserved. In the latter case, it has the form of a continuity equation.

Thus, the problem is reduced to the problem of determining the explicit dependence of the potential Ω_m upon the order parameter η. In order to solve the latter problem, we use the following model.

Assume that a spherical nucleus with a conductivity σ_1 is formed in a current-carrying host medium with a conductivity σ_0. A change of electric conductivity is related to a change of the order parameter, whereby magnitudes of the order parameter inside the nucleus and far from the nucleus are η_1 and η_0, respectively. Assume for simplicity that the function $\sigma(\mathbf{r})$ is spherically symmetrical with respect to the center of the nucleus $\mathbf{r}_a$.

Configuration of an electric current can be determined from equations of magnetostatics (see Ref. 10):

$$\nabla \cdot \mathbf{j} = 0, \quad \mathbf{j} = \sigma \nabla \varphi, \qquad (5)$$

and their validity at time scales of phase transitions is discussed in Ref. 1. For a given dependence $\sigma(\mathbf{r})$, and boundary conditions for the potential $\varphi(\mathbf{r})$, Eq. (5) determines the density of an electric current $\mathbf{j}(\mathbf{r})$ and, consequently, the potential Ω_m.

Determining the concrete dependence $\sigma(\mathbf{r})$ complicates the analysis of the problem, and it is of little expedience from the physical point of view since the nucleus can be characterized by an electric conductivity only when its size a is much larger than the mean-free-path l, $a \gg l$, and much larger than the width of the skin layer δ, $a \gg \delta$. In the zero-order approximation in parameter δ/a,

$$\nabla \sigma = \mathbf{n}(\sigma_0 - \sigma_1)\delta(a - |\mathbf{r} - \mathbf{r}_a|), \qquad (6)$$

where $\mathbf{n}$ is a unit normal vector to a nucleus surface.

The solution of the magnetostatics problem (5) and (6) in this case is well known (see Ref. 1), and $\mathbf{j} = \mathbf{j}_0 + \delta\mathbf{j}$, where $\mathbf{j}_0$ is the electric current far from the nucleus and

$$\delta\mathbf{j} = -\mathbf{j}_0\xi\left[2\theta(a - |\mathbf{r} - \mathbf{r}_a|) \right.$$
$$\left. + \frac{a^3}{|\mathbf{r} - \mathbf{r}_a|^3}(3\mu^2 - 1)\theta(|\mathbf{r} - \mathbf{r}_a| - a) \right]$$
$$- 3\mathbf{e}_\perp j_0 \xi\mu\sqrt{1 - \mu^2}\frac{a^3}{|\mathbf{r} - \mathbf{r}_a|^3}\theta(|\mathbf{r} - \mathbf{r}_a| - a). \qquad (7)$$

Here,

$$\mu = \frac{(\mathbf{r} - \mathbf{r}_a)\mathbf{j}_0}{|\mathbf{r} - \mathbf{r}_a|j_0}, \quad \theta(x) = \begin{cases} 1, & x \geq 0 \\ 0, & x < 0 \end{cases}, \quad \xi = \frac{\sigma_0 - \sigma_1}{\sigma_1 + 2\sigma_0}.$$

Equation (7) is valid, provided that $\mathbf{j}_0$ is a constant vector of electric current far away from the nucleus, so that the characteristic size of region b where a nucleus is formed, $b \gg a$. When the configuration of electric current is more involved, it can be partitioned into ''linear'' domains and the nucleus is assumed to be formed in one of these domains.

According to Eq. (2), the energy of the magnetic field can be represented as follows:

$$\Omega_m = \Omega_m^0 + \Omega_m^1,$$

where Ω_m^0 is a part of the magnetic energy that does not depend upon the nucleus size,

$$\Omega_m^0 = \frac{\mathbf{j}_0^2}{2c^2} \int \frac{1}{|\mathbf{r} - \mathbf{r}'|} d^3\mathbf{r} \, d^3\mathbf{r}',$$

and

$$\Omega_m^1 = \frac{1}{c^2} \int \frac{\mathbf{j}_0 \delta\mathbf{j}}{|\mathbf{r} - \mathbf{r}'|} d^3\mathbf{r} \, d^3\mathbf{r}'.$$

Here, in the expression for Ω_m, we neglected a contribution of the term

$$\Omega_m^2 = \int \frac{\delta\mathbf{j}(\mathbf{r})\,\delta\mathbf{j}(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d^3\mathbf{r} \, d^3\mathbf{r}'$$

since

$$\Omega_m^2 = \frac{a^3}{b^3}\Omega_m^1 \ll 1.$$

For the case when a nucleus is formed in a cylindrical conductor with radius ρ_0 and length l_0, the detailed calculations of Ω_m can be found in Ref. 3. In this case, in the leading approximation in parameters $a/\rho_0 \ll 1$ and $\rho_0/l_0 \ll 1$, an expression for Ω_m^1 reads:

$$\Omega_m^1 = -2\xi p_m V_a \Phi(\mathbf{r}_a), \quad p_m = \frac{I^2}{c^2\pi\rho_0^2}, \qquad (8)$$

where I is the total electric current, V_a is the volume of the nucleus, and $\Phi(\mathbf{r}_a)$ is the geometrical factor depending upon the length of the conductor and location of the center of the nucleus in the conductor. When configuration of the electric current $\mathbf{j}_0$ is more involved, the magnitude of Ω_m^1 changes only due to the change of the geometrical factor $\Phi(\mathbf{r}_a)$. The geometrical factor can be assumed to be independent of the size of the nucleus and its conductivity, and hereafter it is assumed to be constant.

It is known (see, e.g., Ref. 11) that when a boundary separating phases with different values of the order parameter is sharp, it is possible to transfer from a continual Ginzburg-Landau model to a Doering-Volmer-Zeldovich nucleation model (see, e.g., Ref. 12) using a δ-like variation of the gradient of the order parameter. Similarly, using Eq. (6) one can rewrite the formula (8) for Ω_m^1 as follows:

$$\Omega_m^1 = -\frac{\tilde{p}_m}{3(\sigma_0 - \sigma_1)} \int [(\mathbf{r} - \mathbf{r}_a)\nabla\sigma]d^3\mathbf{r}, \quad \tilde{p}_m = 2\xi\Phi p_m. \qquad (9)$$

Equation (9) is invariant with respect to the choice of the origin of a coordinate system, since the additional energy Ω_m^1 is related only to the gradient of electric conductivity, and substituting Eq. (6) into integral (9) yields Eq. (8).

Hereafter we assume that the electron free path length is the smallest length scale in the system, and the spatial profile of electric conductivity coincides with a spatial profile of the order parameter, i.e.,

$$\nabla\sigma = \frac{\sigma_0 - \sigma_1}{\eta_0 - \eta_1}\nabla\eta,$$

where η_0 and η_1 are the magnitudes of the order parameter far from the boundary between phases. Since, under varia-

tion of the order parameter, its values far from the interface are considered constants, after integration by parts in Eq. (9), and using Eqs. (1) and (3), we find that

$$\frac{\delta\Omega}{\delta\eta} = -\left(\chi\Delta\eta + \mu - \frac{\partial f_0}{\partial\eta} + \frac{\widetilde{p}_m}{\eta_0 - \eta_1}\right). \quad (10)$$

Coexisting values of the order parameter far from the boundary between phases η_0 and η_1 are determined from the system of equations that can be derived from Eqs. (4) and (10), taking into account that far from the front $\Delta\eta \to 0$ and $\dot{\eta} \to 0$.

In the Ginzburg-Landau model, $f_0 = \alpha\eta^2/2 + \beta\eta^4/4$. Substituting the latter expression into Eq. (10), and using the condition $\delta\Omega/\delta\eta = 0$, yields the system of equations for coexisting values of η_0 and η_1:

$$\mu + \frac{\widetilde{p}_m}{\eta_0 - \eta_1} = \alpha\eta_0 + \beta\eta_0^3,$$
$$\qquad\qquad\qquad\qquad\qquad (11)$$
$$\mu + \frac{\widetilde{p}_m}{\eta_0 - \eta_1} = \alpha\eta_1 + \beta\eta_1^3.$$

Subtracting the second equation in Eqs. (11) from the first yields

$$\eta_0^2 + \eta_1\eta_0 + \eta_1^2 = -\frac{\alpha}{\beta}. \quad (12)$$

As generally accepted, assume that $\beta > 0$, and then the domain of coexisting phases is determined by the condition $\alpha < 0$. Introducing the dimensionless variable Ψ, $\eta = (|\alpha|/\beta)^{1/2}\Psi$, we find from Eq. (12) that for a given Ψ_0 there are two possible values for Ψ_1:

$$\Psi_1 = -\frac{\Psi_0}{2} \pm \sqrt{1 - \frac{3}{4}\Psi_0^2}. \quad (13)$$

According to Eqs. (4) and (10), a condition for the thermodynamic stability of a phase is $\partial^2 f/\partial\eta^2 \geqslant 0$ or $|\Psi_{0,1}| > 1/\sqrt{3}$. From Eq. (13) it can be easily seen that there is a unique thermodynamically stable solution Ψ_1 that corresponds to the thermodynamically stable solution Ψ_0 and vice versa. In the range $1/\sqrt{3} < \Psi_0 < 2/\sqrt{3}$, the thermodynamically stable solution reads

$$\Psi_1 = -\frac{\Psi_0}{2} - \sqrt{1 - \frac{3}{4}\Psi_0^2},$$

and in the range $-2/\sqrt{3} < \Psi_0 < -1/\sqrt{3}$ the stable solution is $\Psi_1 = -\Psi_0/2 + \sqrt{1 - \frac{3}{4}\Psi_0^2}$. Using these relations and Eqs. (11), we arrive at the equations for parameter Ψ_0:

$$\frac{\widetilde{\pi}_m}{E_+(\Psi_0)} = \Psi_0^3 - \Psi_0 - h, \quad \frac{1}{\sqrt{3}} < \Psi_0 < \frac{2}{\sqrt{3}},$$

$$\frac{\widetilde{\pi}_m}{E_-(\Psi_0)} = \Psi_0^3 - \Psi_0 - h, \quad -\frac{2}{\sqrt{3}} < \Psi_0 < -\frac{1}{\sqrt{3}}, \quad (14)$$

$$E_\pm(\Psi_0) = \frac{3}{2}\Psi_0 \pm \sqrt{1 - \frac{3}{4}\Psi_0^2},$$

where $\widetilde{\pi}_m = (\beta/|\alpha|^2)\widetilde{p}_m$, $h = (\beta^{1/2}/|\alpha|^{3/2})\mu$.

Similarly, determining Ψ_0 from Eqs. (11)–(12), we arrive at the equations for determining the thermodynamically stable value of the internal phase Ψ_1:

$$\frac{\widetilde{\pi}_m}{\widetilde{E}_+(\Psi_1)} = \Psi_1^3 - \Psi_1 - h, \quad \frac{1}{\sqrt{3}} < \Psi_1 < \frac{2}{\sqrt{3}},$$

$$\frac{\widetilde{\pi}_m}{\widetilde{E}_-(\Psi_1)} = \Psi_1^3 - \Psi_1 - h, \quad -\frac{2}{\sqrt{3}} < \Psi_1 < -\frac{1}{\sqrt{3}}, \quad (15)$$

$$\widetilde{E}_\pm(\Psi_1) = -\left[\frac{3}{2}\Psi_1 \pm \sqrt{1 - \frac{3}{4}\Psi_1^2}\right].$$

In finding the approximate solutions of these equations, one must take into account that the derivatives of functions $E_\pm(\Psi)$ and $\widetilde{E}_\pm(\Psi)$ have singularities at $|\Psi| \to 2/\sqrt{3}$. Far from these points, an approximate expression for Ψ_0 and Ψ_1 can be obtained using Eqs. (14) and Eqs. (15), respectively. For $\widetilde{\pi}_m \ll 1$ we have

$$\Psi_0 = \widetilde{\Psi}_0 + \frac{\widetilde{\pi}_m}{E_\pm(\widetilde{\Psi}_0)(3\widetilde{\Psi}_0^2 - 1)},$$

$$\Psi_1 = \widetilde{\Psi}_1 + \frac{\widetilde{\pi}_m}{E_\pm(\widetilde{\Psi}_1)(3\widetilde{\Psi}_1^2 - 1)},$$

where $\widetilde{\Psi}_{1,0}$ are solutions of the corresponding equations for $\widetilde{\pi}_m = 0$. The values $\widetilde{\Psi}_{1,0}$ can be taken as basic solutions only far away from the boundary of the coexistence of phases.

In order to determine the critical values of the chemical potential h that determine the domain of the coexistence of phases, we take into account that the thermodynamically unstable phase must be considered as an external phase. Then the critical value of h at which the phase loses thermodynamic stability under a given magnitude of electric current can be determined from Eq. (14). Hereafter, the phases with negative and positive values of the order parameter are called the negative and the positive phases, respectively. Then, for a positive phase setting in Eq. (14) $\Psi_0 = 1/\sqrt{3}$, $\sigma_0 = \sigma_+$, $h = h_c^+$, we find that

$$h_c^+ = -h_c^0 + \frac{2}{\sqrt{3}}\frac{\kappa - 1}{\kappa + 2}A, \quad h_c^0 = \frac{2}{3\sqrt{3}}, \quad \kappa = \frac{\sigma_-}{\sigma_+}, \quad (16)$$

where $A = p_m\Phi\beta/|\alpha|^2 \geqslant 0$, σ_+, and σ_-, are conductivities of the positive and negative phases, respectively. The domain of the thermodynamic stability of the positive phase is determined by the relation $h > h_c^+$.

Similarly, for the negative phase $\Psi_0 = -1/\sqrt{3}$, $\sigma_0 = \sigma_-$, $h = h_c^-$ from Eq. (14), we find

$$h_c^- = h_c^0 + \frac{2}{\sqrt{3}}\frac{\kappa - 1}{1 + 2\kappa}A. \quad (17)$$

The domain of stability of the negative phase is determined by the condition $h < h_c^-$, and the domain of the coexistence of phases is determined by the condition $h_c^+ < h$

$<h_c^-$. The critical value of the electric current A_c, whereby at $A>A_c$ phases cannot coexist at any value of h, can be found from the condition $h_c^-=h_c^+$ or

$$A_c=\frac{2}{\sqrt{3}}\frac{(\kappa+2)(1+2\kappa)}{(\kappa-1)^2}\geqslant\frac{4}{\sqrt{3}}.\tag{18}$$

For the electric current $A>A_c$, the phase transition cannot be realized as a phase transition of the first kind.

It is shown below that the equilibrium value of a chemical potential is determined by the following condition:

$$\frac{\tilde{\pi}_m}{\Psi_0-\Psi_1}+h=0.\tag{19}$$

According to Eqs. (14) and (15), the latter condition can be satisfied only at the points $|\Psi_0|=|\Psi_1|=1$.

In order to determine the value of the chemical potential $h=h^-$ corresponding to the equilibrium with respect to the transition from a positive phase to a negative phase, one must take into account that in this case the negative phase is the internal phase, i.e., $\Psi_0=1$, $\Psi_1=-1$. Substituting $\sigma_0=\sigma_+$, $\sigma_1=\sigma_-$ in Eq. (19), we find that

$$h^-=\frac{\kappa-1}{\kappa+2}A.\tag{20}$$

Similarly, one can determine the potential $h=h^+$ corresponding to the equilibrium with respect to the inverse phase transition from the negative phase to the positive phase by setting $\Psi_0=-1$, $\Psi_1=1$, and $\sigma_0=\sigma_-$, $\sigma_1=\sigma_+$:

$$h^+=\frac{\kappa-1}{1+2\kappa}A.\tag{21}$$

It follows from Eqs. (20) and (21) that the phase equilibrium curves for the direct and inverse phase transition are shifted in the same direction, i.e., $\mathrm{sgn}(h^+)=\mathrm{sgn}(h^-)$. Here, one of the phases becomes more stable than in the current-free case, while the other phase destabilizes since part of the domain where this phase is stable in a current-free case becomes metastable. Equations (20) and (21) show that stabilizes the phase with a higher electric conductivity. The physical meaning of the domain $h^+<h<h^-$ is elucidated by the following general considerations. In a current-free system, $h>0$ is a region of stability of the positive phase, while $h<0$ is a domain of the stability of the negative phase. Therefore, in a current-carrying system for sufficiently large but negative h, the negative phase is stable. When h increases and attains the point h^+ where the nuclei of a positive phase can form, the negative phase loses stability and transfers into a metastable state. However, a positive phase is still metastable since, for $h<h^-$, nuclei of a negative phase can form inside a positive phase, and only at $h>h^-$ the positive phase becomes stable. Thus, in the domain $h^+<h<h^-$, both phases are metastable simultaneously. In the following we will show that in this domain the sizes of the critical nuclei for a transition from a negative phase to a positive phase, and for the inverse phase transition, are positive and finite values. Note that we discussed a possibility of the existence of the domain of simultaneous metastability

(DSM) of both phases in Ref. 1 using the zero-dimensional model. However, instead of the coefficient A arising in the present study, the corresponding coefficient in Ref. 1 depends upon the specific volume of the internal phase. The latter provides us with an additional possibility whereby a hysteresis can occur instead of the domain of simultaneous metastability of both phases. In the situation that is considered in this study, $h^+<h^-$ always, i.e., hysteresis does not occur and only DSM occurs. Thus, the points h_c^+,h^+,h^-,h_c^- partition the domain of h into the regions of varying thermodynamic stability of phases. Mutual location of these regions depends upon the parameter $\kappa=\sigma_-/\sigma_+$ and the magnitude of the ''electric current'' A. According to Eqs. (16) and (18), in the domain of the coexistence of phases $h_c^->h_c^+$, and according to Eqs. (20), and (21), $h^->h^+$. Taking into account these conditions, it can be easily seen that there exist six different sequences of the domains:

(a) $h_c^+<h^+<h^-<h_c^-$, (b) $h^+<h_c^+<h^-<h_c^-$,

(c) $h_c^+<h^+<h_c^-<h^-$, (d) $h^+<h_c^+<h_c^-<h^-$, (22)

(e) $h_c^+<h_c^-<h^+<h^-$, (f) $h^+<h^-<h_c^+<h_c^-$.

In order to determine the conditions on electric current where each of the above six cases can be realized, it is convenient to divide the domain of the parameter κ into four regions: $0<\kappa<d$, $d<\kappa<1$, $1<\kappa<1/d$, and $\kappa>1/d$ where $d=2(\sqrt{3}-1)/(4-\sqrt{3})\approx0.65$. However, the region $0<\kappa<d$ is equivalent to the region $\kappa>1/d$, where a positive phase is replaced with a negative one. Thus, all the conclusions concerning the positive phase in the domain $0<\kappa<d$ are valid for the negative phase in the domain $\kappa>1/d$ and vice versa. A similar situation occurs in the domain $d<\kappa<1$, and, correspondingly, in the domain $1/d>\kappa>1$. Therefore, it is sufficient to analyze only the case with $\kappa>1$.

Consider first a domain $1<\kappa<1/d$. The condition $h^-<h_c^-$ and Eqs. (17) and (20) yield

$$\frac{(\kappa-1)(4-\sqrt{3})}{(\kappa+2)(1+2\kappa)}(\kappa d-1)A<\frac{2}{3}.\tag{23}$$

Thus, in this domain, the condition (23) is always valid ($\kappa d<1$), and only three cases (a), (b), (f) can occur in compliance with the inequalities (22). The condition for the occurrence of case (a) is obtained from the condition $h^+>h_c^+$ and Eqs. (16) and (21):

$$A<R=\frac{2}{3(4-\sqrt{3})}\frac{(\kappa+2)(1+2\kappa)}{(\kappa-1)(\kappa-d)}.\tag{24}$$

For the occurrence of case (b), apart from the condition $h^+<h_c^+(A>R)$, the condition $h^->h_c^+$ must be satisfied. The latter condition yields

$$A<P=\frac{2}{3(2-\sqrt{3})}\frac{\kappa+2}{\kappa-1}.$$

For all $\kappa>1$, $P>R$. Therefore, in the range of electric currents $R<A<P$, case (b) occurs. In the range $A_c>A>P$, case (f) occurs.
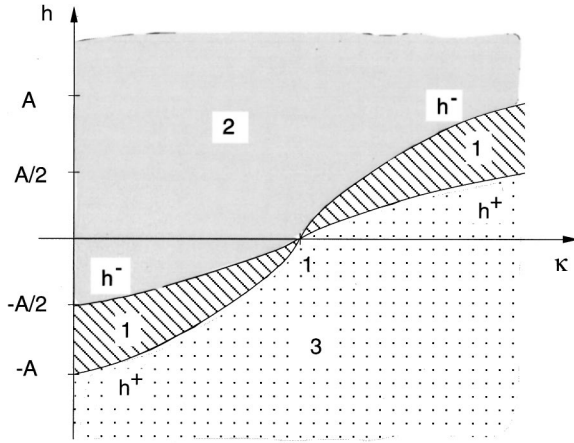
FIG. 1. Diagram of coexistence of phases at small electric currents: (1) DSM; (2) stable positive phase; (3) stable negative phase; h^+ is a binodal line for the $+\Rightarrow-$ phase transition; h^- is a binodal line for the $-\Rightarrow+$ phase transition.

Before the analysis of the domain $\kappa>1/d$, consider thermodynamic situation in each of the domains. When case (a) occurs, DSM is completely embedded in the region of the coexistence of phases. To the left of this domain, $h_c^+<h<h^+$, the negative phase is stable while the positive phase is metastable. At $h_c^->h>h^-$ the negative phase is metastable while the positive phase is stable (see Fig. 1).

When case (b) occurs, the DSM borders from the left with a spinodal line of the positive phase. In this case, in the whole range of the coexistence of phases the negative phase is metastable, and the positive phase is stable in the range $h^-<h<h_c^-$. Case (f) describes a situation where in the whole of the coexistence of phases ($h_c^+<h<h_c^-$) the negative phase is metastable and the positive phase is stable. This situation occurs because, at large electric currents $A_c>A>P$, the coexistence of phases is realized deep inside the region with positive h, such that in a current-free system the negative phase there was thermodynamically unstable. In the presence of an electric current this phase is partially stabilized and it transforms into a metastable state.

Now consider a case with $\kappa>1/d$. In this case the condition (23) yields an additional restriction to Eq. (24):

$$A<R\,\frac{\kappa-d}{\kappa d-1}=R_1. \tag{25}$$

Thus, for $\kappa>1/d$, $R_1>R$, case (b) is realized only when the following condition is satisfied:

$$R_1>A>R. \tag{26}$$

For $A>R_1$, $h^->h_c^-$ and case (f) cannot be realized. Since $h_c^+>h^+$ and $h^->h_c^-$ in the range of electric currents $A>R_1>R$, Eq. (22) shows that in the range $A_c>A>R_1$ case (d) occurs. In the latter case, DSM extends over the whole range of the coexistence of phases. In Fig. 2, we showed, schematically, this situation for $A=1$.

Similarly, we can consider the case $\kappa<1$. In the range $d<\kappa<1$, condition $h_c^+<h^+$ yields
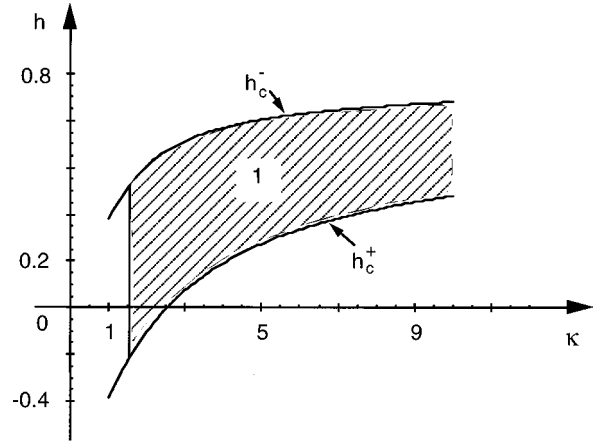

FIG. 2. Diagram of coexistence of phases, case (d): (1) DSM; h_c^- is a spinodal line of the negative phase; h_c^+ is a spinodal line of the positive phase.

$$\frac{(4-\sqrt{3})(1-\kappa)(d-\kappa)}{(1+2\kappa)(\kappa+2)}\,A<\frac{2}{3}. \tag{27}$$

In the range $d<\kappa<1$, inequality (27) is valid without restrictions on the magnitude of the electric current. Thus, in this range of κ, there occur cases (a), (c), (e), and all the conclusions concerning the positive and negative phases in cases (b) and (f) are valid for the negative and positive phases, respectively, in cases (c) and (e). In the range of $\theta<\kappa<d$, instead of cases (a), (b), (d), cases (a), (c), (d), occur, and so on.

Thus, the most significant effect of the electric current is the occurrence of the domain of the simultaneous metastability of two phases. Inside this domain the nuclei of a phase with a higher electric conductivity are formed due to the work of ponderomotive forces, i.e., due to the external source of electric current, while the nuclei of a phase with a smaller electric conductivity are formed due to the surplus of chemical energy.

III. RELAXATION OF THE ORDER PARAMETER

Now, using the approach suggested in Ref. 11, we consider the peculiarities of the relaxation of the order parameter in the presence of an electric current. Using the dimensionless variables $\varphi,h,\tilde{\pi}_m$ defined above, and the dimensionless length x and time τ,

$$r=\sqrt{2}\left(\frac{\chi}{\alpha}\right)^{1/2}x, \quad t=\left(\frac{2}{\Gamma_n\alpha}\right)\tau,$$

we restrict ourselves to the analysis of a nonconserved order parameter in an isotropic case.

Using Eq. (10), we can rewrite Eq. (4) in the new variables as

$$\frac{\partial\Psi}{\partial\tau}=\frac{\partial^2\Psi}{\partial x^2}+\frac{2}{x}\frac{\partial\Psi}{\partial x}+2(\Psi-\Psi^3)+\tilde{h}, \quad \tilde{h}=h+\frac{\tilde{\pi}_m}{\Psi_0-\Psi_1}. \tag{28}$$

Let us look for the solution of Eq. (28) in the form

$$\Psi = \Psi[x - x_0(\tau)],$$

where $x_0(\tau)$ is the location of a front.

Assume that for $x - x_0 \ll x_0$ and $x_0 \gg 1$, solution Ψ is a kink, while for $x - x_0 \sim x_0$ it is close to a constant. Following Ref. 11, the first derivative is taken into account only for $x - x_0 \ll x_0$. Then, Eq. (28) can be rewritten as

$$\frac{\partial \Psi}{\partial \tau} = \frac{\partial^2 \Psi}{\partial x^2} + \frac{2}{x_0} \frac{\partial \Psi}{\partial x} + 2(\Psi - \Psi^3) + \tilde{h}. \qquad (29)$$

For $\tilde{\pi}_m \to 0$, Eq. (29) coincides with Eq. (13) of Ref. 11 if one neglects in the latter the angular dependence of the order parameter Ψ.

In Ref. 11 it was shown that at $\tilde{h} = h$, with the accuracy $o(h^2)$ and $o(e^{-(x - x_0)})$ in the domain $x - x_0 \gg 1$, and with the accuracy $o(h^2)$ in the domain $x - x_0 \ll 1$, the solution of Eq. (29) can be written as follows:

$$\Psi = \pm \tanh[x - x_0(\tau)] + \frac{1}{4} h, \quad -\dot{x}_0 = \frac{2}{x_0} \pm \frac{3h}{2}, \quad (30)$$

where signs $\pm$ in Eq. (30) correspond to the formation of nuclei of the negative and positive phases, respectively. Equations (14) and (15) imply that at $\tilde{h} \to 0$, $\Psi_0 - \Psi_1 \to \pm 2$. Therefore, without losing accuracy, in Eq. (30) we can replace h by $\tilde{h}^\pm = h + \tilde{\pi}_m^\pm / s$, where $s = -2$ in the case of the formation of nuclei of the positive phase, $s = +2$ in the case of the formation of the negative phase, and formulas for $\tilde{\pi}_m^\pm$ are written below. Thus, in the case of the formation of a nucleus of the negative phase, the solution of Eq. (29) reads:

$$\Psi = \tanh[x - x_0^-(\tau)] + \frac{1}{4} \left(h + \frac{\tilde{\pi}_m^-}{2} \right),$$

$$-\dot{x}_0^- = \frac{2}{x_0^-} + \frac{3}{2} \left(h + \frac{\tilde{\pi}_m^-}{2} \right), \qquad (31)$$

where $x_0^-(\tau)$ is the radius of the nucleus of the negative phase and $\tilde{\pi}_m^- = [2(1 - \kappa)/(\kappa + 2)]A$.

In the case of the formation of the nucleus of the positive phase, instead of Eq. (29) we obtain

$$\Psi = - \tanh[x - x_0^+(\tau)] + \frac{1}{4} \left(h - \frac{\tilde{\pi}_m^+}{2} \right),$$

$$-\dot{x}_0^+ = \frac{2}{x_0^+} - \frac{3}{2} \left(h - \frac{\tilde{\pi}_m^+}{2} \right), \qquad (32)$$

where $\tilde{\pi}_m^+ = [2(\kappa - 1)/1 + 2\kappa]A$.

The sizes of the critical nuclei are determined from the conditions $\dot{x}_0^\pm = 0$:

$$x_0^- = - \frac{4}{3} \frac{1}{h + \frac{1 - \kappa}{\kappa + 2} A}, \quad x_0^+ = \frac{4}{3} \frac{1}{h + \frac{1 - \kappa}{1 + 2\kappa} A}. \qquad (33)$$

The conditions $x_0^- > 0$, $x_0^+ > 0$ or $h + [(1 - \kappa)/(\kappa + 2)]A < 0$, $h + [(1 - \kappa)/(1 + 2\kappa)]A > 0$ determine DSM, while conditions $1/x_0^\pm = 0$ determine the phase equilibrium curves (20) and (21).

The conditions (33) yield the following relation between the sizes of the critical nuclei x_0^+ and x_0^- :

$$x_0^+ = \frac{x_0^-}{\gamma x_0^- - 1}, \quad \gamma = \frac{\sqrt{3}}{2} \frac{A}{A_c},$$

where A_c is determined by Eq. (18). Expressing the sizes of the nuclei $x_0^\pm$ in units A_c/A [$x_0^\pm = (A_c/A)\lambda^\pm$], we find that $\lambda^+ = \lambda^- / [(\sqrt{3}/2)\lambda^- - 1]$.

The latter relation shows that when the size of the nucleus of the negative phase increases, $\lambda^- \to \infty$, the size of the nucleus of the positive phase λ^+ decreases, $\lambda^+ \to 2/\sqrt{3}$ and vice versa. If the considered mechanism of the phase transition could have been realized without the influence of other effects, in the DSM the system would transfer periodically from one state into the other since the increase of the critical nucleus for the phase transition $+ \Rightarrow -$ causes a reduction of the critical nucleus for the transition $- \Rightarrow +$. In reality, dispersion of a system into small particles can prevent the occurrence of such a process. In Fig. 1 we plotted a diagram of the coexistence of phases for case (a) and for $|h^-| \ll |h_c^-|$, $|h^+| \ll |h_c^+|$. As can be seen from this figure, DSM occurs without a threshold at arbitrary small values of an electric current A. However, as a magnitude of an electric current decreases the range of values, h, where DSM occurs, shrinks.

Thus, taking into account the change of electric conductivity in current-carrying conductors, there results a more diversified behavior of the system. The most significant difference in comparison with the case of a current-free conductor is the existence of the domain of parameters where both phases are metastable. The existence of such a domain may be characteristic for a wide class of systems subjected to the external forces where the parameters determining the response of the system to the external field differ for both phases. Such a situation may occur due to the effect of the electric field where the dielectric permeabilities of both phases are different (see, e.g., Refs. 13 and 14), due to the external pressure (see Ref. 15), etc. At present, it is not clear whether this effect can occur in systems with a strong magnetic response or segnetoelectrics.

Stochastic behavior of the system in DSM is significantly different from that in the domain where one of the phases is stable, and is not considered here. It is natural to expect that dispersed structures are formed in DSM. The physical reason for the occurrence of DSM is related to the openness of the system with respect to the source of the electric current. As was pointed out above, transition from the phase with a higher electric conductivity to the phase with a lower electric conductivity occurs due to the surplus of chemical energy accumulated in a phase with a high electric conductivity, and ponderomotive forces act to prevent this transition. The inverse phase transition occurs due to the ponderomotive energy, and the work is performed by the source of an electric current. Outside the DSM, the stochastic description of the system may be performed using the traditional approach (see, e.g., Ref. 11).

Due to the strong sensitivity of the behavior of the system with respect to the initial parameters and the physical variety of such systems, comparison of the obtained results with the available experimental data (see Refs. 5–8) is not straightforward. In these studies the following effects of an electric

current on the structure of alloys and upon the crystallization were found: (1) reduction of grain size;[8] (2) strong dependence of the rate of change of Ohmic resistance $\dot{R}$ upon the magnitude of electric current;[7] and (3) high magnitude of the undercooling at which crystallization begins.

Qualitatively, these observations are in accord with the results obtained in Refs. 1, 2, 4, and in the present study. In order to perform a quantitative comparison, the problem must be solved on a more detailed level of description, and the model employed in this study is not sufficient for comparison with the experiments.

As we noted above, comparison of the results obtained with the Ginzburg-Landau model with experiments is quite involved. However, it is expedient to develop this model for the description of phase transitions in current-carrying systems since zero-dimensional nucleation theory (see Refs. 1 and 4) cannot explain the experimentally observed effects of the relatively small electric currents on crystallization. In the following we present some estimates that can be obtained using our previous results obtained with a zero-dimensional model. According to Refs. 1 and 4, the magnitude of the undercooling with respect to the internal crystallization is determined by the following equation:

$$\mu_S(p + \tilde{p}_m + p_s, T) = \mu_L(p, T), \qquad (34)$$

where subscripts S and L correspond to solid and liquid phases, respectively, $\tilde{p}_m = [(1 - \bar{\kappa})/(\bar{\kappa} + 2)]p_m \Phi$, $\bar{\kappa} = \sigma_S/\sigma_L > 1$, and the p_s is the pressure due to surface tension. The magnitude of the $\Delta T/T_0$ can be determined from Eq. (34) in the limit $p_s \to 0$ and

$$\frac{\Delta T}{T_0} = -\frac{v_0 \tilde{p}_m}{\lambda_0},$$

where v_0, $\lambda_0 > 0$, and T_0 are a specific volume, a latent heat of melting, and a crystallization temperature, respectively.

Equation (34) yields the following expression for the size of the critical nucleus a_c:

$$a_c = a_c^0 \frac{1}{1 - \tilde{p}_m/p_s^0}, \qquad (35)$$

where a_c^0 is a size of the critical nucleus without an electric current, $p_s^0 = 2\alpha_s/a_c^0$, and α_s is a coefficient of the surface tension. Since the nucleation rate $J \propto \exp(-4\pi\alpha_s a_c^2/3T)$, while the rate of change of the Ohmic resistance $\dot{R} \propto J$, Eq. (35) yields

$$\dot{R} \propto \exp\left(-\frac{4\pi\alpha_s(a_c^0)^2}{3T}\frac{1}{(1 - I^2\Phi/I_0^2)^2}\right),$$

where I_0 is determined by the following formula:

$$I_0^2 = \pi r_0^2 c^2 \frac{2\alpha_s}{a_c^0}.$$

Usually, the latter quantity is very large, since $\alpha_s \sim 10^3 - 10^4$ dyn/cm, $a_c^0 \sim 10^{-6}$ cm, and for a cylindrical conductor with a radius $r_0 = 1$ cm, $I_0 \sim 10^2$ kA. Experiments show that the characteristic values of the electric current at which the above discussed effects occur are of the order of $I_0 \sim 1$ kA.[7] Therefore, the effect of the electric current with such a magnitude can be explained only by the anomalous behavior of the coefficient of the surface tension that must assume the values $\alpha_s \sim 10^{-1} - 1$ dyn/cm. It is conceivable that such anomalous behavior of the coefficient of the surface tension is caused by the proximity of the region where there occurs a phase transition to the critical temperature (which is the reason why it is expedient to describe this phenomenon using a Ginzburg-Landau model) and because the phase transition occurs in an undercooled state. Anyway, the employed model is the only model that allows us to describe correctly the behavior of the system qualitatively. It must be noted also that, according to our results,[3] the surface crystallization in the presence of an electric current does not occur in an undercooled state but requires an additional overcooling that plays a significant role in the development of the internal phase transition.

---

*Electronic address: yuli@menix.bgu.ac.il

†Electronic address: elperin@menix.bgu.ac.il

[1] Yu. Dolinsky and T. Elperin, Phys. Rev. B **47**, 14 778 (1993).

[2] Yu. Dolinsky and T. Elperin, J. Appl. Phys. **73**, 5283 (1993).

[3] Yu. Dolinsky and T. Elperin, Phys. Rev. B **50**, 52 (1994).

[4] Yu. Dolinsky and T. Elperin, J. Appl. Phys. **80**, 38 (1996).

[5] J. P. Barnak, A. F. Sprecher, and H. Conrad, Scr. Metall. Mater. **32**, 879 (1995).

[6] R. Takemoto and H. Mizubayashi, Acta Metall. Mater. **43**, 1495 (1995).

[7] H. Mizubayashi and S. Okuda, Phys. Rev. B **40**, 8057 (1989).

[8] A. K. Misra, Metall. Trans. A **16**, 1354 (1985).

[9] L. D. Landau and E. M. Lifshitz, *Statistical Physics* (Pergamon, Oxford, 1980).

[10] L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media* (Pergamon, Oxford, 1984).

[11] A. Z. Patashinsky and B. I. Shumilo, Zh. Eksp. Teor. Fiz. **77**, 1417 (1979) [Sov. Phys. JETP **50**, 712 (1979)].

[12] V. P. Scripov, *Metastable Liquids* (Wiley, New York, 1974).

[13] Yu. L. Dolinsky and N. A. Yavorovsky, Sov. Phys. Tech. Phys. **35**, 765 (1990).

[14] P. L. Martson and R. E. Apfel, Phys. Lett. **60A**, 225 (1977).

[15] I. M. Lifshitz and L. S. Gulida, Dokl. Akad. Nauk SSSR **87**, 377 (1952).