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Morphological Instability of Phase Interface between Pyroelectric Domains

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Abstract

The Gibbs-like variational methodology is applied to the heterogeneous systems with rigid pyroelectric or pyromagnetic domains. The processes of depolarization/demagnetization are taken into account by assuming the spatial mobility of the interfaces. The simplest configuration of flat interface separating rigid pyroelectric half-spaces permits explicit analysis of morphological stability.

Keywords

Pyro- and Ferroelectricity, Pyro- and Ferromagnetism, Heterogeneous System, Variational Principles, Cardinal Tensors, Morphological Instabilities

Introduction

We consider the equation of quasi-statics evolution of heterogeneous system consisting of solid nondeformable (rigid) electrically or magnetically polarized domains. By the model of Gibbs [1], heterogeneous systems are conglomerates of macroscopic subdomains, separated by macroscopic interfaces of zero thickness. The Gibbs models and his variational principles allow analyzing both the conditions of equilibrium and the conditions of stability of heterogeneous systems.

When dealing with liquid or gaseous systems the problems of equilibrium and stability mostly to analysis of nonlinear systems of algebraic equations. Gibbs introduced a novel thermodynamic potential playing a crucial role when considering systems with mass exchange between the domains. The mass exchange is of primary importance when dealing with phase transformations between the subdomains. The potential, responsible for equilibrium with respect to mass exchange was coined as chemical potential. Later on, it is occurred that, when dealing with solid deformable phases, the concept of scalar chemical potential of

Gibbs of liquid or gaseous phases should be replaced with tensorial chemical potentials (see, Grinfeld [2]).

When dealing with solid deformable phases, the required calculations become considerably more difficult; in order to suppress difficulties with the calculations the formal techniques of covariant $\delta/\delta t$ -differentiation on moving manifolds have been used (see monographs [2] [3]).

The next step in dealing with heterogeneous systems is to include the effects of electric polarization and magnetization.

Pyroelectric or pyromagnetic substances are those which possess electric or magnetic moments even in the absence of external electric or magnetic macroscopic fields; several approaches to macroscopic modeling of such substances are presented in monographs [4] [5] [6]. Currently dominating scheme of theoretical analysis is presented in the classical monographs of Landau and Lifshitz (1963) which includes various fundamental concepts of thermodynamics. Many brilliant physicists, including Einstein himself, emphasized that thermodynamics is the only discipline fundamentals that will not be overthrown. On the other hand, there are several competing formulations of the thermodynamics fundamentals of macroscopic systems with electric or magnetic polarizations. These formulations are not equivalent to each other, and they lead to different conclusions, sometimes, incompatible with each other. Interested reader can find discussion of different formulation in Rosensweig, R. E. (1985).

Our own analysis, based on the Gibbs variational principles was presented in the series of publications [7] [8] [9] [10] [11].

For the computational simplicity, we assume that the polarization/magnetization densities remain fixed inside each subdomain; such models have been analyzed in [7]. However, they are different in different subdomains. Thus, the polarization/magnetization vectors experience finite jumps across the boundaries (see, **Figure 1**).

Due to the assumption of rigidity, in such models, the material particles cannot move. At the same time, the interfaces between the boundaries are able to migrate. That migration happens not due to the migration of the materials particles

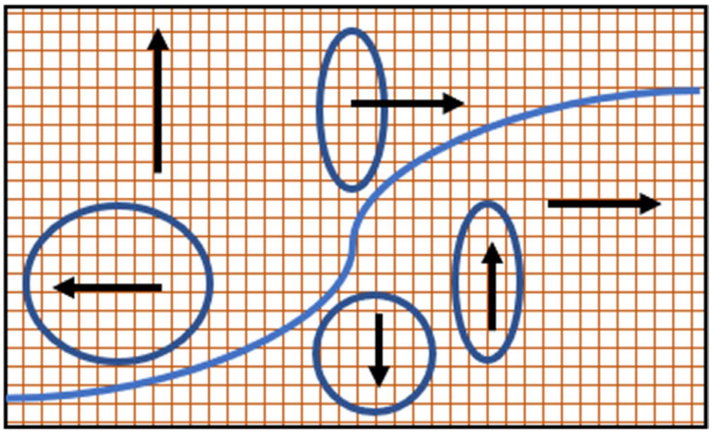

Figure 1. Models of heterogeneous systems with polarized phases.

but due to the processes of depolarization or demagnetization of the substances at the interfaces. In this sense, the boundaries between the polarized domains remind the phase transformations between different phases. Following standard nomenclature, we call these zones of continuity simply different domains (despite some linguistic inconveniences).

For the sake of simplicity, we assume that out of the boundaries the polarization/magnetization vectors are fixed constants. Moreover, we assume, for simplicity, that density of polarization is uniform within each domain of smoothness. For the same reason of computational simplicity, the surface tension is neglected. We distinguish between rigid pyroelectric domains and rigid pyroelectric phases. Pyroelectric domains or substances are characterized as those having fixed polarization vectors per unit volume $\boldsymbol{P}$. The free energy density ψ per unit volume may depend upon the absolute temperature T only. At fixed temperature of the heterogeneous system the function ψ appears to be a constant.

However, generally speaking, the vectors $\boldsymbol{P}$ within different domains differ from each other. The same is true regarding the constant free energy densities ψ within different domains.

Now, what is going on at the interfaces between different domains? This depends upon the nature of the domain. More specifically, two different situations should be distinguished. The first one can be coined as the "chemical" interface. In this situation polarization of the adjacent particles of the bordering domains remains unchanged. In the opposite situation, one of the adjacent particles can change its polarization in favor of the polarization of its neighbor. In this case, the macroscopic boundary between the domains "moves" with respect to the domains. This situation is exactly the same as the situation with different phases. Therefore, the boundary between such domains is called the phase boundary. In other words, we deal with migration of the interface between two domains.

Maybe, the simplest example of such a system is the heterogeneous system consisting of a single domain having a common boundary with vacuum or with the domain having zero polarization. This system can appear as a result of condensation of gas of polarized molecules on the rigid substrate (see, **Figure 2**).

This problem has been analyzed in the publication [7] [8] [9]. The simplest configuration of this sort is the crystalline layer with flat boundary parallel to the substrate. Such a boundary can be morphologically stable or unstable depending on the orientation of the polarization vector $\boldsymbol{P}$ to the boundary normal stable or $\boldsymbol{N}$. Namely, it was demonstrated in Grinfeld [7] that the crystal boundary is

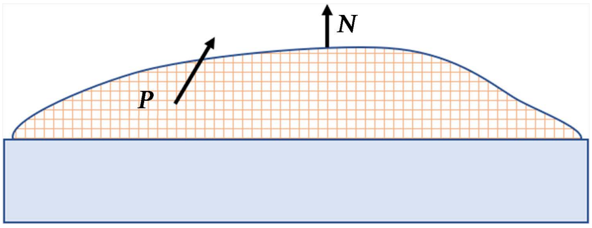

Figure 2. Pyroelectric rigid crystal, growing on substrate.

morphologically unstable if the normal component of the polarization $\left|\boldsymbol{P}_{\perp}\right|$ is bigger than the tangential component of polarization $\left|\boldsymbol{P}_{\|}\right|$:

$$\left|\boldsymbol{P}_{\perp}\right| > \left|\boldsymbol{P}_{\|}\right| \tag{I}$$

The surface tension suppresses the instability for sufficiently short wavelengths $\left|\boldsymbol{k}_{crit}\right|$ such that

$$\frac{\sigma}{2\pi}\left|\boldsymbol{k}_{crit}\right| = \left|\boldsymbol{P}_{\perp}\right|^{2} - \left|\boldsymbol{P}_{\|}\right|^{2} \tag{II}$$

The goal of this paper is to establish the analogy of the relationship (I) for the case of phase interface separating two pyroelectric domains of the shape of half-spaces.

1. Quasi-Static Model of the Heterogeneous System with Pyroelectric Domains

Out of the interfaces equation of electrostatic reads

$$\nabla_{i} D^{i} = 0 \tag{1.1}$$

where D^{i} is the electrostatic displacement vector.

The electrostatic boundary conditions read

$$[\varphi]_{-}^{+} = 0 \tag{1.2}$$

and

$$\left[D^{i}\right]_{-}^{+} N_{i} = 0 \tag{1.3}$$

where N_{i} is the field of unit normal to the interface; φ is the field of electrostatic potential.

We remind the identities for the electric field

$$E_{i} = -\nabla_{i}\varphi \tag{1.4}$$

and the electric displacement D_{i}

$$D_{i} = E_{i} + 4\pi P_{i} = -\nabla_{i}\varphi + 4\pi P_{i} \tag{1.5}$$

Combining Equations (1.1), (1.5), we arrive at the bulk equation

$$\nabla_{i}\left(-\nabla^{i}\varphi + 4\pi P^{i}\right) = 0 \tag{1.6}$$

Since polarization is supposed constant inside each domain, the bulk Equation (1.6) reduces to the Laplace equation

$$\nabla_{i}\nabla^{i}\varphi = 0 \tag{1.7}$$

The interface electrostatics condition (1.3) reduces to the following one:

$$\left[\nabla^{i}\varphi\right]_{-}^{+} N_{i} = 4\pi\left[P^{i}\right]_{-}^{+} N_{i} \tag{1.8}$$

The electrostatics boundary conditions (1.2), (1.3) should be satisfied at any interface—chemical or phase interface. At the same time, one more condition equilibrium should be satisfied at the phase interface. It is the analogy of the condition of equality of the chemical (Gibbs) potentials of the adjacent phases in the classical thermodynamics of liquid/gaseous phases.

In the case of rigid pyroelectric phases, it has appeared that the Gibbs potential should be replaced with the more sophisticated notion and requires the introduction of the so-called Gimel cardinal tensor $\gimel_{ij}$ defined as

$$\gimel_{ij} \equiv \left(\Psi + \frac{1}{8\pi} E^k E_k - \frac{1}{4\pi} E^k D_k \right) z_{ij} + \frac{1}{4\pi} D_i E_j \tag{1.9}$$

The equation of equilibrium across the phase boundary between the rigid domains reads

$$\left[\gimel_{ij} \right]_-^+ N^i N^j = 0 \tag{1.10}$$

If the surface energy is taken into account, Equation (1.10) should be replaced vid the following one:

$$\left[\gimel_{ij} \right]_-^+ N^i N^j = \sigma\left(N^k \right) \xi^{\alpha\beta} b_{\alpha\beta} \tag{1.11}$$

where $\sigma\left(N^k\right)$ is dependence of the surface energy density upon orientation of the normal vector N^k; $\xi^{\alpha\beta}$ and $b_{\alpha\beta}$ are the metrics and the curvature tensors of the interface.

Typically, the process of establishing the "chemical" equilibrium is considerably slower than the process of establishing the electrostatic equilibrium. In other words, the velocity C of migration of the boundaries between the domains is quite slow. In this case, instead of Equation (1.11) of phase equilibrium, it makes sense to use the linear kinetics equation

$$C = -\kappa \left(\left[\gimel_{ij} \right]_-^+ N^i N^j - \sigma B_\alpha^\alpha \right), \tag{1.12}$$

where κ is the kinetics constant, determining the rate of establishing "chemical" equilibrium.

Also, we use the following notation: z^i are the spatial coordinates (the Latin (spatial) indexes run values 1, 2, 3), z_{ij} is the spatial metrics, used for raising and lowering spatial indexes; the spatial covariant differentiation is denoted as ∇_i. Let $z^i = z^i\left(\xi^\alpha, t\right)$ be the equation of the mobile interface between the domains; the Greek (surface) indexes run the values 1, 2. The shift-tensor $z^{i\cdot}_{\cdot\alpha}\left(\xi, t\right)$ is defined as $z^{i\cdot}_{\cdot\alpha}\left(\xi, t\right) \equiv \partial z^i\left(\xi, t\right) / \partial \xi^\alpha$. The surface metrics $\xi_{\alpha\beta}\left(\xi^\alpha, t\right)$ defined as $\xi_{\alpha\beta}\left(\xi^\alpha, t\right) \equiv z_{ij} z^{i\cdot}_{\cdot\alpha} z^{j\cdot}_{\cdot\beta}$ it is for raising and lowering the Greek indexes. We use the notation ∇_α for the surface covariant differentiation based on the tensors z_{ij} and $\xi_{\alpha\beta}$.

2. Linearization in Vicinity of Equilibrium Piecewise Uniform Configuration

Consider the configuration consisting of two domains with the shape of two half-spaces with the common flat phase interface. Assume that all equations of electrostatics and "chemical" (phase) equilibrium are satisfied inside each domain and across the interface. We use degree a mark "∘" for the parameters characterizing this configuration and call it the main configuration. In particular, we use notation $z^{i\circ}_{.}(\xi), z^{i\circ}_{.\alpha}(\xi), N^{i\circ}$ for the equation of the interface, the shift tensor, and the spatial components of the unit normal in this configuration; also we use notation $\varphi^{\circ}_{\pm}, E^{i\circ}_{\pm}$, for the electric potential and electric field in each of the domains in this configuration.

By definition the interface velocity C, vanishes in the main configuration. Assume now that the boundary between the domains slightly deviates from the ideal plane. Then the condition of "chemical" equilibrium (1.11) will not be satisfied anymore, and the process of depolarization will occur at the boundary between the adjacent domains. As a result, the velocity C of the interface will not be equal to zero anymore. Therefore, the electrostatic potential also becomes function of time t. The Laplace bulk Equation (1.7) obviously implies

$$\nabla_i \nabla^i \frac{\partial \varphi}{\partial t} = 0 \tag{2.1}$$

Applying the $\delta/\delta t$-differentiation of the boundary conditions of electrostatics (1.2), (1.3) and using properties of this operation [3] [4], we get, respectively:

$$\left[\frac{\partial \varphi}{\partial t}\right]_-^+ + 4\pi C \left[P_i\right]_-^+ N^i = 0 \tag{2.2}$$

and

$$\left[\nabla_i \frac{\partial \varphi}{\partial t}\right]_-^+ N^i = -4\pi \left[P^\alpha\right]_-^+ \nabla_\alpha C \tag{2.3}$$

Differentiating the kinetics Equation (1.12), we get, neglecting the surface tension

$$\begin{aligned} -K^{-1} \frac{\delta C}{\delta t} = & \left[\frac{1}{4\pi} E^k \frac{\partial E_k}{\partial t} - \frac{1}{4\pi} D^k \frac{\partial E_k}{\partial t} - \frac{1}{4\pi} E^k \frac{\partial E_k}{\partial t}\right]_-^+ + \frac{1}{4\pi}\left[\frac{\partial E_i}{\partial t} E_j + D_i \frac{\partial E_j}{\partial t}\right]_-^+ N^i N^j \\ & - \frac{1}{4\pi}\left[D_i E_j\right]_-^+ \left(z^{i.}_{.\alpha} N^j + z^{j.}_{.\alpha} N^i\right) \nabla^\alpha C \end{aligned} \tag{2.4}$$

Equation (2.4) can be rewritten as follows

$$4\pi K^{-1} \frac{\delta C}{\delta t} = \left[\left\langle E^j - \left(E_i + D_i\right) z^{i\beta}_{..} z^{j.}_{.\beta} \right\rangle \nabla_j \frac{\partial \varphi}{\partial t}\right]_-^+ + \left[D_i E_j\right]_-^+ z^{i.}_{.\alpha} N^j \nabla^\alpha C \tag{2.5}$$

as implied by the following chain:

$$\begin{aligned} & 4\pi K^{-1} \frac{\delta C}{\delta t} = \left[D^k \frac{\partial E_k}{\partial t} - \left(E_i + D_i\right) \frac{\partial E_j}{\partial t} N^i N^j\right]_-^+ + \left[D_i E_j\right]_-^+ z^{i.}_{.\alpha} N^j \nabla^\alpha C \\ & \to 4\pi K^{-1} \frac{\delta C}{\delta t} = \left[\left(D^j - \left(E_i + D_i\right) N^i N^j\right) \frac{\partial E_j}{\partial t}\right]_-^+ + \left[D_i E_j\right]_-^+ z^{i.}_{.\alpha} N^j \nabla^\alpha C \\ & \to 4\pi K^{-1} \frac{\delta C}{\delta t} = \left[\left\langle -D^j + \left(E_i + D_i\right)\left(z^{ij} - z^{i\beta}_{..} z^{j.}_{.\beta}\right)\right\rangle \nabla_j \frac{\partial \varphi}{\partial t}\right]_-^+ + \left[D_i E_j\right]_-^+ z^{i.}_{.\alpha} N^j \nabla^\alpha C \\ & \to 4\pi K^{-1} \frac{\delta C}{\delta t} = \left[\left\langle E^j - \left(E_i + D_i\right) z^{i\beta}_{..} z^{j.}_{.\beta}\right\rangle \nabla_j \frac{\partial \varphi}{\partial t}\right]_-^+ + \left[D_i E_j\right]_-^+ z^{i.}_{.\alpha} N^j \nabla^\alpha C \end{aligned} \tag{2.6}$$

Let us introduce the following notation

$$R_{\pm}^{j} \equiv E_{\pm}^{j} - \left(E_{\pm}^{i} + D_{\pm}^{i}\right) z_{i.}^{.\beta} z_{.\beta}^{j.}, Y_{\pm}^{\alpha} \equiv D_{i\pm} E_{j\pm} z_{..}^{i\alpha} N^{j} \tag{2.7}$$

Using (2.7), we can rewrite Equation (2.5) as follows:

$$4\pi K^{-1} \frac{\delta C}{\delta t} = \left[R^{j} \nabla_{j} \frac{\partial \varphi}{\partial t}\right]_{-}^{+} + \left[Y^{\alpha}\right]_{-}^{+} \nabla_{\alpha} C \tag{2.8}$$

Using the notation $G \equiv \partial\varphi/\partial t$, we can rewrite the bulk Equation (2.1) and the boundary conditions (2.2), (2.3) as follows

$$\nabla_{i} \nabla^{i} G = 0 \tag{2.9}$$

$$[G]_{-}^{+} + 4\pi C [P_{i}]_{-}^{+} N^{i} = 0 \tag{2.10}$$

$$[\nabla_{i} G]_{-}^{+} N^{i} = -4\pi \left[P^{\alpha}\right]_{-}^{+} \nabla_{\alpha} C \tag{2.11}$$

At last, the kinetics Equation (2.5) implies that

$$4\pi K^{-1} \frac{\delta C}{\delta t} = \left[R^{j} \nabla_{j} G\right]_{-}^{+} + \left[Y^{\alpha}\right]_{-}^{+} \nabla_{\alpha} C \tag{2.12}$$

Consider solutions of this system of the following form:

$$\begin{aligned} G\left(z^{a}, z, t\right) &= \mathrm{e}^{\eta t - i k_{a} z^{a}} \begin{cases} \Phi_{+}(k) \mathrm{e}^{-z|k|} & \text{for } z > 0 \\ \Phi_{-}(k) \mathrm{e}^{z|k|} & \text{for } z < 0 \end{cases} \\ C\left(z^{a} t\right) &= \mathrm{e}^{\eta t - i k_{a} z^{a}} S(k) \end{aligned} \tag{2.13}$$

The boundary conditions (2.10), (2.11) imply, respectively:

$$\Phi_{+} - \Phi_{-} = -4\pi [P_{i}]_{-}^{+} N^{i} S \tag{2.14}$$

and

$$\Phi_{+}(k) + \Phi_{-}(k) = 4\pi i \frac{k_{\alpha}}{|k|} \left[P^{\alpha}\right]_{-}^{+} S \tag{2.15}$$

Equations (2.14), (2.15) imply the following values of $\Phi_{\pm}$:

$$\begin{aligned} \Phi_{+} &= \left(-[P_{i}]_{-}^{+} N^{i} + i \frac{k_{\alpha}}{|k|} \left[P^{\alpha}\right]_{-}^{+}\right) 2\pi S \\ \Phi_{-} &= \left([P_{i}]_{-}^{+} N^{i} + i \frac{k_{\alpha}}{|k|} \left[P^{\alpha}\right]_{-}^{+}\right) 2\pi S \end{aligned} \tag{2.16}$$

Using solutions (2.16), we can rewrite (2.13) as follows:

$$\begin{aligned} G\left(z^{a}, z, t\right) &= \mathrm{e}^{\eta t - i k_{a} z^{a}} \begin{cases} \left(-[P_{i}]_{-}^{+} N^{i} + i \frac{k_{\alpha}}{|k|} \left[P^{\alpha}\right]_{-}^{+}\right) 2\pi S \mathrm{e}^{-z|k|} & \text{for } z > 0 \\ \left([P_{i}]_{-}^{+} N^{i} + i \frac{k_{\alpha}}{|k|} \left[P^{\alpha}\right]_{-}^{+}\right) 2\pi S \mathrm{e}^{z|k|} & \text{for } z < 0 \end{cases} \\ C\left(z^{a} t\right) &= \mathrm{e}^{\eta t - i k_{a} z^{a}} S(k) \end{aligned} \tag{2.17}$$

The solution (2.17) obviously implies that

$$G\left(z^a,0,t\right)=\mathrm{e}^{\eta t-ik_a z^a}\begin{cases}\left(-\left[P_i\right]_-^+ N^i+i\dfrac{k_\alpha}{|k|}\left[P^\alpha\right]_-^+\right)2\pi S & \text{for } z>0\\ \left(\left[P_i\right]_-^+ N^i+i\dfrac{k_\alpha}{|k|}\left[P^\alpha\right]_-^+\right)2\pi S & \text{for } z<0\end{cases} \tag{2.18}$$

$$C\left(z^a,t\right)=\mathrm{e}^{\eta t-ik_a z^a}S\left(k\right)$$

Also, we get, using Equations (2.17):

$$\begin{aligned}\nabla_i G&=\nabla_k G\delta_i^k=\nabla_k G\left(N^k N_i+z_{.\beta}^{k.}z_{i.}^{\cdot\beta}\right)=-\frac{\partial G}{\partial z}N_i+\frac{\partial G}{\partial z^\beta}z_{i.}^{\cdot\beta}\\ &=\mathrm{e}^{\eta t-ik_a z^a}\begin{cases}\left(|k|N_i-ik_\beta z_{i.}^{\cdot\beta}\right)\left(-\left[P_k\right]_-^+ N^k+i\dfrac{k_\alpha}{|k|}\left[P^\alpha\right]_-^+\right)2\pi S & \text{for } z>0\\ \left(-|k|N_i-ik_\beta z_{i.}^{\cdot\beta}\right)\left(\left[P_i\right]_-^+ N^i+i\dfrac{k_\alpha}{|k|}\left[P^\alpha\right]_-^+\right)2\pi S & \text{for } z<0\end{cases}\end{aligned} \tag{2.19}$$

We, then, get the following relationships:

$$\begin{aligned}&\left(|k|N_i-ik_\beta z_{i.}^{\cdot\beta}\right)\left(-\left[P_k\right]_-^+ N^k+i\frac{k_\alpha}{|k|}\left[P^\alpha\right]_-^+\right)\\ &=z_{i.}^{\cdot\alpha}z_{k.}^{\cdot\beta}\frac{k_\alpha k_\beta}{|k|}\left[P^k\right]_-^+-|k|N_i\left[P^k\right]_-^+N_k+i\left[P^k\right]_-^+k_\alpha z_{j.}^{\cdot\alpha}\left(\delta_i^j N_k+\delta_k^j N_i\right)\end{aligned} \tag{2.20}$$

and

$$\begin{aligned}&\left(-|k|N_i-ik_\beta z_{i.}^{\cdot\beta}\right)\left(\left[P_k\right]_-^+ N^k+i\frac{k_\alpha}{|k|}\left[P^\alpha\right]_-^+\right)\\ &=z_{i.}^{\cdot\alpha}z_{k.}^{\cdot\beta}\frac{k_\alpha k_\beta}{|k|}\left[P^k\right]_-^+-|k|N_i\left[P^k\right]_-^+N_k-i\left[P^k\right]_-^+k_\alpha z_{j.}^{\cdot\alpha}\left(\delta_i^j N_k+\delta_k^j N_i\right)\end{aligned} \tag{2.21}$$

as implied by the chains:

$$\begin{aligned}&\left(|k|N_i-ik_\beta z_{i.}^{\cdot\beta}\right)\left(-\left[P_k\right]_-^+ N^k+i\frac{k_\alpha}{|k|}\left[P^\alpha\right]_-^+\right)\\ &=-|k|N_i\left[P_k\right]_-^+N^k+\left[P_k\right]_-^+N^k ik_\beta z_{i.}^{\cdot\beta}+|k|N_i i\frac{k_\alpha}{|k|}\left[P^\alpha\right]_-^+-ik_\beta z_{i.}^{\cdot\beta}i\frac{k_\alpha}{|k|}\left[P^\alpha\right]_-^+\\ &=-|k|N_i\left[P_k\right]_-^+N^k+ik_\beta z_{i.}^{\cdot\beta}\left[P_k\right]_-^+N^k+ik_\alpha\left[P^\alpha\right]_-^+N_i+k_\beta z_{i.}^{\cdot\beta}\frac{k_\alpha}{|k|}\left[P^\alpha\right]_-^+\\ &=z_{i.}^{\cdot\beta}\frac{k_\alpha k_\beta}{|k|}\left[P^\alpha\right]_-^+-|k|N_i\left[P_k\right]_-^+N^k+i\left(k_\beta z_{i.}^{\cdot\beta}\left[P_k\right]_-^+N^k+k_\alpha z_{k.}^{\cdot\alpha}\left[P^k\right]_-^+N_i\right)\\ &=z_{i.}^{\cdot\alpha}z_{k.}^{\cdot\beta}\frac{k_\alpha k_\beta}{|k|}\left[P^k\right]_-^+-|k|N_i\left[P^k\right]_-^+N_k+i\left[P^k\right]_-^+k_\alpha z_{j.}^{\cdot\alpha}\left(\delta_i^j N_k+\delta_k^j N_i\right)\end{aligned}$$

and

$$\begin{aligned}&\left(-|k|N_i-ik_\beta z_{i.}^{\cdot\beta}\right)\left(\left[P_k\right]_-^+ N^k+i\frac{k_\alpha}{|k|}\left[P^\alpha\right]_-^+\right)\\ &=-|k|N_i\left[P_k\right]_-^+N^k+z_{i.}^{\cdot\beta}\frac{k_\alpha k_\beta}{|k|}\left[P^\alpha\right]_-^+-i\left(k_\beta z_{i.}^{\cdot\beta}\left[P_k\right]_-^+N^k+N_i k_\alpha\left[P^\alpha\right]_-^+\right)\end{aligned}$$

$$= z_{i.}^{\cdot\beta}\frac{k_\alpha k_\beta}{|k|}\left[P^\alpha\right]_-^+ - |k|N_i\left[P_k\right]_-^+ N^k - i\left(k_\beta z_{i.}^{\cdot\beta}\left[P_k\right]_-^+ N^k + N_i k_\alpha\left[P^\alpha\right]_-^+\right)$$

$$= z_{i.}^{\cdot\alpha} z_{k.}^{\cdot\beta}\frac{k_\alpha k_\beta}{|k|}\left[P^k\right]_-^+ - |k|N_i\left[P^k\right]_-^+ N_k - i\left[P^k\right]_-^+ k_\alpha z_{j.}^{\cdot\alpha}\left(\delta_i^j N_k + \delta_k^j N_i\right),$$

respectively.

Using the relationships (2.20), (2.21), we can rewrite (2.19) as follows

$$\nabla_i G\big|_{z=0\pm} = Se^{\eta t - ik_a z^a}\,2\pi|k|\left[P^k\right]_-^+\left(z_{i.}^{\cdot\alpha} z_{k.}^{\cdot\beta}\frac{k_\alpha k_\beta}{|k|^2} - N_i N_k \pm i\frac{k_\alpha}{|k|}z_{j.}^{\cdot\alpha}\left(\delta_i^j N_k + \delta_k^j N_i\right)\right) \tag{2.22}$$

The kinetics Equation (2.12) implies

$$4\pi K^{-1}\eta S = \left[R^i\nabla_i G\right]_-^+ - ik_\alpha\left[Y^\alpha\right]_-^+ S \tag{2.23}$$

Combining (2.20), (2.21), we get

$$4\pi K^{-1}\eta S = R_+^i\nabla_i G_+ - R_-^i\nabla_i G_- - ik_\alpha\left(Y_+^\alpha - Y_-^\alpha\right)S \tag{2.24}$$

and then

$$\begin{aligned}4\pi K^{-1}\eta S = S2\pi|k|\Bigg\{&\left[P^k\right]_-^+\left(z_{i.}^{\cdot\alpha} z_{k.}^{\cdot\beta}\frac{k_\alpha k_\beta}{|k|^2} - N_i N_k + i\frac{k_\alpha}{|k|}z_{j.}^{\cdot\alpha}\left(\delta_i^j N_k + \delta_k^j N_i\right)\right)R_+^i\\ &-\left[P^k\right]_-^+\left(z_{i.}^{\cdot\alpha} z_{k.}^{\cdot\beta}\frac{k_\alpha k_\beta}{|k|^2} - N_i N_k - i\frac{k_\alpha}{|k|}z_{j.}^{\cdot\alpha}\left(\delta_i^j N_k + \delta_k^j N_i\right)\right)R_-^i\\ &-i\frac{k_\alpha}{2\pi|k|}\left(Y_+^\alpha - Y_-^\alpha\right)\Bigg\}\end{aligned} \tag{2.25}$$

Using (2.25), we proceed as follows

$$\begin{aligned}2K^{-1}\frac{\eta}{K|k|} = &\left[P^k\right]_-^+\left(z_{i.}^{\cdot\alpha} z_{k.}^{\cdot\beta}\frac{k_\alpha k_\beta}{|k|^2} - N_i N_k\right)\left(R_+^i - R_-^i\right)\\ &+i\left(\left[P^k\right]_-^+\frac{k_\alpha}{|k|}z_{j.}^{\cdot\alpha}\left(\delta_i^j N_k + \delta_k^j N_i\right)\right)\left(R_+^i + R_-^i\right) - i\frac{k_\alpha}{2\pi|k|}\left(Y_+^\alpha - Y_-^\alpha\right)\end{aligned} \tag{2.26}$$

Using (2.7), (2.26), we get

$$\begin{aligned}2K^{-1}\operatorname{Im}\eta &= \left(k_\beta z_{i.}^{\cdot\beta}\left[P_k\right]_-^+ N^k + k_\alpha\left[P^\alpha\right]_-^+ N_i\right)\left(R_+^i + R_-^i\right) - k_\alpha\left(Y_+^\alpha - Y_-^\alpha\right)\\ &= \left(k_\alpha z_{i.}^{\cdot\alpha}\left[P_k\right]_-^+ N^k + k_\alpha\left[P^\alpha\right]_-^+ N_i\right)\left(E_+^i + E_-^i - \left(E_+^k + D_+^k + E_-^k + D_-^k\right)z_{k.}^{\cdot\beta} z_{\cdot\beta}^{i.}\right)\\ &\quad -\frac{k_\alpha}{2\pi|k|}\left(D_{i+}E_{j+}z_{..}^{i\alpha}N^j - D_{i-}E_{j-}z_{..}^{i\alpha}N^j\right)\end{aligned} \tag{2.27}$$

and

$$\begin{aligned}2K^{-1}\operatorname{Re}\eta &= \left(z_{i.}^{\cdot\beta}\frac{k_\alpha k_\beta}{|k|}\left[P^\alpha\right]_-^+ - |k|N_i\left[P_k\right]_-^+ N^k\right)\left(R_+^i - R_-^i\right)\\ &= \left(z_{i.}^{\cdot\beta}\frac{k_\alpha k_\beta}{|k|}\left[P^\alpha\right]_-^+ - |k|N_i\left[P_k\right]_-^+ N^k\right)\left[E^i - \left(E^k + D^k\right)z_{k.}^{\cdot\beta} z_{\cdot\beta}^{i.}\right]_-^+\end{aligned} \tag{2.28}$$

We proceed as follows

$$\operatorname{Re}\eta = 2\pi|k|\left(P_{\perp}^{2} - P_{\parallel}^{2}\right)K \tag{2.29}$$

where

$$P_{\perp} \equiv \left[P^{i}\right]_{-}^{+} N_{i}, \quad P_{\parallel} \equiv \frac{k_{\alpha}}{|k|}\left[P^{\alpha}\right]_{-}^{+} \tag{2.30}$$

The relationship (2.29) is implied by the following chain:

$$\begin{aligned}
2K^{-1}\operatorname{Re}\eta &= \left(z_{i.}^{\cdot\beta}\frac{k_{\alpha}k_{\beta}}{|k|}\left[P^{\alpha}\right]_{-}^{+} - |k|N_{i}\left[P_{j}\right]_{-}^{+}N^{j}\right)\left[E^{i} - \left(E^{k} + D^{k}\right)z_{k.}^{\cdot\gamma}z_{\cdot\gamma}^{i.}\right]_{-}^{+} \\
&= z_{i.}^{\cdot\beta}\frac{k_{\alpha}k_{\beta}}{|k|}\left[P^{\alpha}\right]_{-}^{+}\left[E^{i} - \left(E^{k} + D^{k}\right)z_{k.}^{\cdot\gamma}z_{\cdot\gamma}^{i.}\right]_{-}^{+} \\
&\quad - |k|N_{i}\left[P_{j}\right]_{-}^{+}N^{j}\left[E^{i} - \left(E^{k} + D^{k}\right)z_{k.}^{\cdot\gamma}z_{\cdot\gamma}^{i.}\right]_{-}^{+} \\
&= z_{i.}^{\cdot\beta}\frac{k_{\alpha}k_{\beta}}{|k|}\left[P^{\alpha}\right]_{-}^{+}\left[E^{\beta} - \left(E^{k} + D^{k}\right)z_{k.}^{\cdot\beta}\right]_{-}^{+} - |k|\left[P_{j}\right]_{-}^{+}N^{j}\left[E^{i}\right]_{-}^{+}N_{i} \\
&= -4\pi\frac{k_{\alpha}k_{\beta}}{|k|}\left[P^{\alpha}\right]_{-}^{+}\left[P^{\beta}\right]_{-}^{+} + 4\pi|k|\left[P_{j}\right]_{-}^{+}N^{j}\left[P^{i}\right]_{-}^{+}N_{i} = 4\pi|k|\left(P_{\perp}^{2} - P_{\parallel}^{2}\right)
\end{aligned}$$

Next, we will demonstrate that the imaginary part of the decrement η vanishes. First, we get

$$\begin{aligned}
2K^{-1}\operatorname{Im}\eta = &-\left(D_{+}^{i} + D_{-}^{i}\right)z_{i.}^{\cdot\alpha}k_{\alpha}\left[P_{k}\right]_{-}^{+}N^{k} + \left(E_{+}^{i} + E_{-}^{i}\right)k_{\alpha}z_{k..}^{\cdot\alpha}\left[P^{k}\right]_{-}^{+}N_{i} \\
&- k_{\alpha}z_{..}^{i\alpha}\frac{1}{2\pi}\left(D_{i+}E_{j+}N^{j} - D_{i-}E_{j-}N^{j}\right)
\end{aligned} \tag{2.31}$$

as it follows from the chain:

$$\begin{aligned}
2K^{-1}\operatorname{Im}\eta &= \left(E_{+}^{i} + E_{-}^{i}\right)k_{\alpha}z_{i.}^{\cdot\alpha}\left[P_{k}\right]_{-}^{+}N^{k} - \left(E_{+}^{k} + D_{+}^{k} + E_{-}^{k} + D_{-}^{k}\right)z_{k.}^{\cdot\beta}z_{\cdot\beta}^{i.}k_{\alpha}z_{i.}^{\cdot\alpha}\left[P_{j}\right]_{-}^{+}N^{j} \\
&\quad + \left(E_{+}^{i} + E_{-}^{i}\right)k_{\alpha}\left[P^{\alpha}\right]_{-}^{+}N_{i} - \left(E_{+}^{k} + D_{+}^{k} + E_{-}^{k} + D_{-}^{k}\right)z_{k.}^{\cdot\beta}z_{\cdot\beta}^{i.}k_{\alpha}\left[P^{\alpha}\right]_{-}^{+}N_{i} \\
&\quad - k_{\alpha}\frac{1}{2\pi}\left(D_{i+}E_{j+}z_{..}^{i\alpha}N^{j} - D_{i-}E_{j-}z_{..}^{i\alpha}N^{j}\right) \\
&= \left(E_{+}^{i} + E_{-}^{i}\right)k_{\alpha}z_{i.}^{\cdot\alpha}\left[P_{k}\right]_{-}^{+}N^{k} - \left(E_{+}^{k} + D_{+}^{k} + E_{-}^{k} + D_{-}^{k}\right)z_{k.}^{\cdot\alpha}k_{\alpha}\left[P_{j}\right]_{-}^{+}N^{j} \\
&\quad + \left(E_{+}^{i} + E_{-}^{i}\right)k_{\alpha}\left[P^{\alpha}\right]_{-}^{+}N_{i} - k_{\alpha}\left(D_{i+}E_{j+}z_{..}^{i\alpha}N^{j} - D_{i-}E_{j-}z_{..}^{i\alpha}N^{j}\right) \\
&= -\left(D_{+}^{i} + D_{-}^{i}\right)z_{i.}^{\cdot\alpha}k_{\alpha}\left[P_{k}\right]_{-}^{+}N^{k} + \left(E_{+}^{i} + E_{-}^{i}\right)k_{\alpha}z_{k..}^{\cdot\alpha}\left[P^{k}\right]_{-}^{+}N_{i} \\
&\quad - k_{\alpha}z_{..}^{i\alpha}\frac{1}{2\pi}\left(D_{i+}E_{j+}N^{j} - D_{i-}E_{j-}N^{j}\right)
\end{aligned}$$

For further transformation of $\operatorname{Im}\eta$ we have to use the equations of electrostatic equilibrium.

Let E_{+}^{i} be the field in the half-space "+" and the $P_{\pm}^{i}$ polarizations in the half-space. We can express all other fields in terms of E_{+}^{i} and the $P_{\pm}^{i}$. For the field E_{-}^{i} we get

$$E_{-}^{i} = E_{+}^{i} + 4\pi\left[P^{j}\right]_{-}^{+}N_{j}N^{i} \tag{2.32}$$

as implied by the chain:

$$
\begin{aligned}
E_-^i &= E_-^j\delta_j^i = E_-^j\left(N_jN^i + z_{j\cdot}^{\cdot\gamma}z_{\cdot\gamma}^{i\cdot}\right) = E_-^jN_jN^i + E_-^jz_{j\cdot}^{\cdot\gamma}z_{\cdot\gamma}^{i\cdot}\\
&= \left(E_+^j - \left[E^j\right]_-^+\right)N_jN^i + E_+^jz_{j\cdot}^{\cdot\gamma}z_{\cdot\gamma}^{i\cdot}\\
&= E_+^jN_jN^i + 4\pi\left[P^j\right]_-^+N_jN^i + E_+^jz_{j\cdot}^{\cdot\gamma}z_{\cdot\gamma}^{i\cdot}\\
&= E_+^i + 4\pi\left[P^j\right]_-^+N_jN^i
\end{aligned}
$$

Using (2.32), we get

$$
E_+^i + E_-^i = 2E_+^i + 4\pi\left[P^j\right]_-^+N_jN^i \tag{2.33}
$$

Using (2.33), we proceed as follows:

$$
\left(E_+^i + E_-^i\right)N_i = 2E_+^jN_j + 4\pi\left[P^j\right]_-^+N_j, \tag{2.34}
$$

$$
\left(D_+^k + D_-^k\right) = 4\pi\left(P_+^k + P_-^k + \left[P^j\right]_-^+N_jN^k\right) + 2E_+^k, \tag{2.35}
$$

$$
k_\alpha z_{k..}^{\cdot\alpha}\left(D_+^k + D_-^k\right) = k_\alpha z_{k..}^{\cdot\alpha}\left(4\pi\left(P_+^k + P_-^k\right) + 2E_+^k\right) \tag{2.36}
$$

Combining (2.32)-(2.46), we get

$$
\begin{aligned}
&k_\alpha z_{k..}^{\cdot\alpha}\left(D_+^kE_{j+}N^j - D_-^kE_{j-}N^j\right)\\
&= k_\alpha z_{k..}^{\cdot\alpha}\left\{4\pi\left[P^k\right]_-^+E_{j+}N^j - \left(E_+^k + 4\pi P_-^k\right)4\pi\left[P^l\right]_-^+N_l\right\}
\end{aligned} \tag{2.37}
$$

as implied by the following chain:

$$
\begin{aligned}
&k_\alpha z_{k..}^{\cdot\alpha}\left(D_+^kE_{j+}N^j - D_-^kE_{j-}N^j\right)\\
&= k_\alpha z_{k..}^{\cdot\alpha}N^j\left\{\left(E_+^k + 4\pi P_+^k\right)E_{j+} - \left(E_+^k + 4\pi\left[P^l\right]_-^+N_lN^k + 4\pi P_-^k\right)\left(E_{j+} + 4\pi\left[P^l\right]_-^+N_lN_j\right)\right\}\\
&= k_\alpha z_{k..}^{\cdot\alpha}N^j\left\{\left(E_+^k + 4\pi P_+^k\right)E_{j+} - \left(E_+^k + 4\pi P_-^k\right)\left(E_{j+} + 4\pi\left[P^l\right]_-^+N_lN_j\right)\right\}\\
&= k_\alpha z_{k..}^{\cdot\alpha}N^j\left\{\left(E_+^k + 4\pi P_+^k\right)E_{j+} - \left(E_+^k + 4\pi P_-^k\right)E_{j+} - \left(E_+^k + 4\pi P_-^k\right)4\pi\left[P^l\right]_-^+N_lN_j\right\}\\
&= k_\alpha z_{k..}^{\cdot\alpha}\left\{4\pi\left[P^k\right]_-^+E_{j+}N^j - \left(E_+^k + 4\pi P_-^k\right)4\pi\left[P^l\right]_-^+N_l\right\}
\end{aligned}
$$

Combining these relationships, we get

$$
\begin{aligned}
&k_\alpha z_{k..}^{\cdot\alpha}\left(-4\pi\left(P_+^k + P_-^k\right) + \left(2E_+^k + 8\pi P_-^k\right) - 2E_+^k\right)\left[P_j\right]_-^+N^j\\
&+ 2E_+^jN_jk_\alpha z_{k..}^{\cdot\alpha}\left[P^k\right]_-^+ + k_\alpha z_{k..}^{\cdot\alpha}4\pi\left[P^k\right]_-^+\left[P^j\right]_-^+N_j - k_\alpha z_{k..}^{\cdot\alpha}2\left[P^k\right]_-^+E_{j+}N^j = 0
\end{aligned} \tag{2.38}
$$

as implied by the following chain:

$$
\begin{aligned}
&-k_\alpha z_{k..}^{\cdot\alpha}\left(4\pi\left(P_+^k + P_-^k\right) + 2E_+^k\right)\left[P_j\right]_-^+N^j\\
&+k_\alpha z_{k..}^{\cdot\alpha}\left[P^k\right]_-^+\left(2E_+^jN_j + 4\pi\left[P^j\right]_-^+N_j\right)\\
&-k_\alpha z_{k..}^{\cdot\alpha}\left\{2\left[P^k\right]_-^+E_{j+}N^j - \left(E_+^k + 4\pi P_-^k\right)2\left[P^l\right]_-^+N_l\right\}\\
&= k_\alpha z_{k..}^{\cdot\alpha}\left\{-\left(D_+^k + D_-^k\right)\left[P_j\right]_-^+N^j + \left(E_+^i + E_-^i\right)\left[P^k\right]_-^+N_i\right.\\
&\qquad\left.-\frac{1}{2\pi}\left(D_+^kE_{j+}N^j - D_-^kE_{j-}N^j\right)\right\}
\end{aligned}
$$

$$\begin{aligned}
&= -k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left(D_+^k + D_-^k\right)\left[P_j\right]_-^+ N^j + k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left[P^k\right]_-^+\left(E_+^i + E_-^i\right)N_i \\
&\quad -\frac{1}{2\pi}k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left(D_+^k E_{j+}N^j - D_-^k E_{j-}N^j\right) \\
&= -k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left(4\pi\left(P_+^k + P_-^k\right) + 2E_+^k\right)\left[P_j\right]_-^+ N^j \\
&\quad + k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left[P^k\right]_-^+\left(2E_+^j N_j + 4\pi\left[P^j\right]_-^+ N_j\right) \\
&\quad - k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left\{2\left[P^k\right]_-^+ E_{j+}N^j - \left(E_+^k + 4\pi P_-^k\right)2\left[P^l\right]_-^+ N_l\right\} \\
&= k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left\{-\left(D_+^k + D_-^k\right)\left[P_j\right]_-^+ N^j + \left(E_+^i + E_-^i\right)\left[P^k\right]_-^+ N_i\right. \\
&\quad \left. -\frac{1}{2\pi}\left(D_+^k E_{j+}N^j - D_-^k E_{j-}N^j\right)\right\} \\
&= -k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left(D_+^k + D_-^k\right)\left[P_j\right]_-^+ N^j + k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left[P^k\right]_-^+\left(E_+^i + E_-^i\right)N_i \\
&\quad -\frac{1}{2\pi}k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left(D_+^k E_{j+}N^j - D_-^k E_{j-}N^j\right) \\
&= -k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left(4\pi\left(P_+^k + P_-^k\right) + 2E_+^k\right)\left[P_j\right]_-^+ N^j \\
&\quad + k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left[P^k\right]_-^+\left(2E_+^j N_j + 4\pi\left[P^j\right]_-^+ N_j\right) \\
&\quad - k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left\{2\left[P^k\right]_-^+ E_{j+}N^j - \left(E_+^k + 4\pi P_-^k\right)2\left[P^l\right]_-^+ N_l\right\}
\end{aligned}$$

and, at last, we get the required relationship:

$$\begin{aligned}
2K^{-1}\operatorname{Im}\eta &= -\left(D_+^i + D_-^i\right)z_{i\cdot}^{\cdot\alpha}k_\alpha\left[P_k\right]_-^+ N^k + \left(E_+^i + E_-^i\right)k_\alpha z_{k\cdot\cdot}^{\cdot\alpha}\left[P^k\right]_-^+ N_i \\
&\quad - k_\alpha z_{\cdot\cdot}^{i\alpha}\frac{1}{2\pi}\left(D_{i+}E_{j+}N^j - D_{i-}E_{j-}N^j\right) \\
&= 0
\end{aligned} \tag{2.39}$$

Thus, the increment/decrement factor η appears to be real. The relation $\operatorname{Im}\eta = 0$, implied by Equation (2.39), shows that small morphological disturbances can either exponentially decay or grow, but do not include any oscillatory components.

Combining (2.29), (2.39), we arrive at our main result:

$$\frac{\eta}{2\pi|k|K} = P_\perp^2 - P_\parallel^2 \tag{2.40}$$

In terms of the original notation $P_\perp \equiv \left[P^i\right]_-^+ N_i$, $P_\parallel \equiv \left[P^\alpha\right]_-^+ k_\alpha/|k|$, we can rewrite Equation (2.40) as follows:

$$\frac{\eta}{2\pi|k|K} = \left(P_+^i N_i\right)^2 - \left(P_+^\alpha k_\alpha/|k|\right)^2 \tag{2.41}$$

If the phase "minus" is not polarizable, Equation (2.41) reduces to the main result of Grinfeld, 1999 [7]:

$$\frac{\eta}{2\pi|k|K} = \left(P_+^i N_i\right)^2 - \left(P_+^\alpha k_\alpha/|k|\right)^2 \tag{2.42}$$

relating to the pyroelectric crystal in vacuum.

3. Discussion and Conclusions

Based on the variational approach of Gibbs [1], we analyzed the problem of equilibrium and stability of rigid heterogeneous systems with pyroelectric phases (domains). For the case of interface, separating two rigid pyroelectric domains, we establish the rate of growth/decay of small morphological disturbances (Equation (2.40)). The established kinetic relation (2.40) shows that the normal component $P_\perp$ of the polarization jump destabilizes flat interface, whereas the tangential component $P_\parallel$ stabilizes it.

Our analysis, based on linearization of the master system, does not permit exploring big deviations from the initial flat geometry. These deviations can be explored with the help of the full master system (1.1 - 1.9, 1.12). It is natural to expect that spikes will appear in the late stage of evolution of small disturbances (compare with the paper [12] of Banerjee *et al.* (2013)).

Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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