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The Slight Distortions Induced by an Electrostatic Field on Finite Samples of Smectic-A Liquid Crystals

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This paper derives governing equations of the interaction of finite samples of smectic-A liquid crystals with an electrostatic field. Our model takes into account effects of the electric field on the layers distortion and vice-versa. The governing equations are obtained by the principle of virtual work. They are adapted for a particular problem in order to analyze the phase transition of Helfrich–Hurault type induced by an electrostatic field on a finite sample of smectic.

Keywords: continuum models; electro-mechanical coupling; phase transitions; smectic liquid crystals

1. INTRODUCTION

Smectic-A liquid crystals partake some of the essential properties of both solids and fluids. They possess long range orientational order and one-dimensional positional ordering. Their rod-like molecules organize themselves into layers. The average alignment is represented by an unit vector **n**, called the director which is locally perpendicular to the layers. In the presence of an electric field a liquid crystal tends to align its molecules in a direction parallel or orthogonal to the field, depending on the dielectric behaviour of the molecules. In smectic-A systems, the rotation of the molecules drags the distortion of the layers.

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It is known that [6,7] magnetic fields can induce phase transitions in samples of cholesteric liquid crystals. Theoretical results for the Helfrich–Hurault transition in infinite samples of smectic-A liquid crystals can be found in [2]. Stewart in [12] extends the works of Helfrich [6] and Hurault [7] to finite cells of smectic-A liquid crystals subject to an uniform pressure and a magnetic or electric field. However, in all these approaches the electric field is only partially coupled with the mechanical system in the sense that it can induce layer distortion, but it is not affected by this latter.

Deuling [3], Gruler *et al.* [4] and more recently Self *et al.* [11] proposed a model for nematic liquid crystals where the nematic director field distortion is fully coupled with the electrostatic field equations. Similarly, by using continuum mechanics arguments, Napoli [10] proposed a model where the shape equation of the smectic layers is completely coupled with the electrostatic Maxwell equations, allowing the electric field to distort the layers and, at the same time, to be modified by that distortion.

In this paper we propose a variational approach in order to characterize the infinitesimal deformations of flat smectic layers induced by a finite electrostatic field. Equilibrium equations are obtained through a variational principle. The so-obtained equations are specialized to a homeotropic finite cell in order to analyze the phase transition of Helfrich–Hurault type induced by the electrostatic field.

2. EQUILIBRIUM EQUATIONS

Smectics-A liquid crystals can be viewed as a continuum of surfaces $\Sigma(\zeta)$ defined by the equation

$$\omega(\mathbf{x}, \zeta) = 0, \tag{2.1}$$

where $\mathbf{x}$ denotes the position in the space of a point on the surface characterized by the ζ parameter. Moreover, smectics-A are characterized by the property that the rod-likes molecules in each layer described by a vectorial field $\mathbf{n}$ are orthogonal to the respective surfaces. This property is expressed by the relation

$$n_{\propto} = \frac{\omega, \alpha}{\sqrt{\omega_{,\gamma}\omega_{,\gamma}}}, \tag{2.2}$$

so the tilt of the molecules depends on the layer deformation. In (2.2) and all formulae below, we understand sum over all repeated greek indices.

We start from a natural configuration where the layers are flat and hence, the smectic-A can be described by a bunch of parallel planes.

Let $\mathbf{n}^0$ the homogeneous field director for a cell of a smectic in its natural configuration. Then let $O(x,y,z)$ be a Cartesian coordinate system with the third axis parallel to $\mathbf{n}^0$. For slight distortions of flat layers, equation (2.2) becomes

$$\mathbf{n} \simeq (-u_{,x}, -u_{,y}, 1), \quad |u_{,x}|, |u_{,y}| << 1, \tag{2.3}$$

where the scalar field $u(x,y,z)$ represents the layers displacement in $\mathbf{n}^0$ direction. More precisely, the relation (2.3) is obtained by supposing that for a displacement ϵu from the undistorted configuration follows a distortion $\epsilon(\mathbf{n} - \mathbf{n}^0)$ where ϵ is a 'small' parameter; in other words formula (2.3) is the approximation $\mathcal{O}(\epsilon)$ of the relation (2.2). For future developments we assume that the displacement field and its derivatives are $\mathcal{O}(\epsilon)$, hence we neglect the terms $\mathcal{O}(\epsilon^2)$ in equilibrium equations and the terms $\mathcal{O}(\epsilon^3)$ in the energy expression.

2.1. The Variational Principle

By following similar developments pursued in [5] we derive the equilibrium equations by the principle of virtual work. Let us suppose that the smectic-A liquid crystal fills a region Ω of the ordinary Euclidean space, bounded by a closed surface $\partial\Omega$ with an outward normal ν. The stored energy per unit of mass Ψ is a state function and we assume that it depends on ∇u, Δu and π, the electrostatic polarization per unit mass. Then the total stored energy is the sum of two contributions. The first one is the elastic energy (see pp. 343 of [2]), including the nematic energy of distortion (namely the splay and the saddle-splay terms) and the smectic energy of compression of the layer:

$$\mathcal{F}_{elast}[\Omega] = \frac{K_1}{2}\int_\Omega [(u_{,xx} + u_{,yy})^2 + 4(u_{,xy}^2 - u_{,xx}u_{,yy})]dv + \frac{\bar{B}}{2}\int_\Omega u_{,z}^2 dv, \tag{2.4}$$

where K_1 and $\bar{B}$ are positive constants. The quantity $\lambda = \sqrt{K_1/\bar{B}}$ is the characteristic length of the material, of the order of the smectic layer thickness.

The second contribution to the total energy is due to the interaction between the field director and the local electric field. By assuming that the smectic is a perfect insulator, that interaction energy is given by

$$\mathcal{F}_{int}[\Omega] = \frac{1}{2}\int_\Omega \rho\pi_\alpha(\chi^{-1})_{\alpha\beta}\pi_\beta dv, \tag{2.5}$$

where ρ is the mass density and χ^{-1} denotes the inverse of the dielectric susceptibility tensor χ. The last one is connected to the usual

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dielectrics nematic tensor ε (see pp. 133 of [2]) by the relation $\varepsilon_{\alpha\beta} = \delta_{\alpha\beta} + \chi_{\alpha\beta} = \varepsilon_\perp \delta_{\alpha\beta} + (\varepsilon_\parallel - \varepsilon_\perp) n_\alpha n_\beta$, where $\delta_{\alpha\beta}$ is the Kroneker symbol. The constants $\varepsilon_\parallel$, $\varepsilon_\perp > 1$ give the dielectric permectivity of the material along or normal to the molecular axes, respectively. The quantity $\varepsilon_a = \varepsilon_\parallel - \varepsilon_\perp$ measures the crystal dielectric anisotropy; it can assume both positive and negative values.

Let ϕ be the electrostatic potential, which gives rise to the electric field of components $E_\alpha = -\phi_{,\alpha}$; the principle of virtual work is as follows:

$$\begin{aligned}\delta\left[-\int_\Omega \rho\Psi dv + \frac{1}{2}\int_\Omega \phi_{,\alpha}\phi_{,\alpha}\, dv - \int_\Omega \rho\phi_{,\alpha}\pi_\alpha\, dv\right] \\ + \int_{\partial\Omega} t\delta u\, da + \int_{\partial\Omega} m_\alpha \delta(u_{,\alpha})\, da + \int_{\partial\Omega} \sigma^e \delta\phi\, da = 0,\end{aligned} \tag{2.6}$$

for all allowed choices of δu, $\delta(u_{,\alpha})$, $\delta\pi_\alpha$ and $\delta\phi$. The last three integrals in this variational expression represent the virtual work of the boundary pressure t in the displacement direction, the virtual work of the boundary distributed couple $\mathbf{m}$ and the virtual work done by the free electric charge per unit of surface σ^e at the boundary. The second and the third terms in the bracket are the electric field and the matter-field Lagrangian respectively (see for example [5]). Here we have adopted the Lorentz–Heaviside units (see for example [8]).

2.2. Euler–Lagrange Equations and Boundary Conditions

The mass conservation equation and the smectic layers inextensibility restrict the total variation of mass density:

$$\delta\rho + \rho n^0_\alpha \delta(u_{,\alpha}) = 0. \tag{2.7}$$

This means that the smectic can change its mass density only by compressing or expanding the layers. Consequently we have, by applying the transport theorem,

$$\begin{aligned}&\delta\left[-\int_\Omega \rho\Psi dv + \frac{1}{2}\int_\Omega \phi_{,\alpha}\phi_{,\alpha} dv - \int_\Omega \rho\phi_{,\alpha}\pi_\alpha dv\right] \\ &\quad = -\int_\Omega \rho\delta\Psi dv + \frac{1}{2}\int_\Omega [\delta(\phi_{,\alpha}\phi_{,\alpha}) + \phi_{,\alpha}\phi_{,\alpha} n^0_\beta \delta(u_{,\beta})] dv - \int_\Omega \rho\delta(\phi_{,\alpha}\pi_{,\alpha}) dv.\end{aligned} \tag{2.8}$$

Because Ψ depends on ∇u , $\nabla^2 u$ and π we easily obtain

$$\rho\delta\Psi = \rho\left(\frac{\partial\Psi}{\partial u_{,\alpha}}\delta(u_{,\alpha}) + \frac{\partial\Psi}{\partial u_{,\alpha\beta}}\delta(u_{,\alpha\beta}) + \frac{\partial\Psi}{\partial\pi_\alpha}\delta\pi_\alpha\right); \tag{2.9}$$

moreover we notice that

$$\delta(u_{,\alpha}) = (\delta u)_{,\alpha}, \quad \delta(\phi_{,\alpha}) = (\delta\phi)_{,\alpha} - n^0_\beta \phi_{,\beta}(\delta u)_{,\alpha}. \tag{2.10}$$

The last relation takes into account that the electric field can change also because of the layers distortion.

With the aid of equations (2.8), (2.9), (2.10) and of the divergence theorem, after opportune integrations by parts of (2.6), we obtain

$$\begin{aligned} &- \int_{\partial\Omega} \sigma_\alpha \nu_\alpha \delta u \, da + \int_{\partial\Omega} \delta(u_{,\alpha}) M_{\alpha\beta}\nu_\beta \, da - \int_{\partial\Omega} D_\alpha \nu_\alpha \delta\phi \, da \\ &+ \int_\Omega \sigma_{\alpha,\alpha} \delta u \, dv + \int_\Omega D_{\alpha,\alpha}\delta\phi \, dv - \int_\Omega \rho({}^L E_\alpha + \phi_{,\alpha})\delta\pi_\alpha \, dv \\ &+ \int_{\partial\Omega} t\delta u \, da + \int_{\partial\Omega} m_\alpha \delta(u_{,\alpha}) \, da + \int_{\partial\Omega} \sigma^e \delta\phi da = 0, \end{aligned} \tag{2.11}$$

where we have introduced the notations:

$$\sigma_\alpha \equiv \rho \frac{\partial \Psi}{\partial u_{,\alpha}} - M_{\alpha\beta,\beta} - (n^0_\beta \phi_{,\beta}) D_\alpha - \frac{\phi_{,\beta}\phi_{,\beta}}{2} n^0_\alpha, \tag{2.12}$$

$$D_\alpha \equiv -\phi_{,\alpha} + \rho\pi_\alpha, \quad {}^L E_\alpha \equiv \frac{\partial \Psi}{\partial \pi_\alpha}, \quad M_{\alpha\beta} \equiv \rho \frac{\partial \Psi}{\partial u_{,\alpha\beta}}. \tag{2.13}$$

Taking into account the arbitrariness of the fields δu, $\delta(u_{,\alpha})$, $\delta\pi_\alpha$ and $\delta\phi$ we arrive to the Euler–Lagrange equations and boundary conditions:

$$\sigma_{\alpha,\alpha} = 0, \quad D_{\alpha,\alpha} = 0, \quad {}^L E = -\phi_{,\alpha}, \quad \text{in } \Omega, \tag{2.14}$$

$$\sigma_\alpha \nu_\alpha = t, \quad M_{\alpha\beta}\nu_\beta = m_\alpha, \quad D_\alpha \nu_\alpha = \sigma^e, \quad \text{on } \partial\Omega. \tag{2.15}$$

The first and the second equation of the (2.12) represent the momentum equation and the Maxwell electrostatic potential equation, respectively. The third equation expresses the relation between the local field ${}^L\mathbf{E}$, a material property, and the Maxwell electric field. This equation can become more complicated if one takes into account stored energies that depend on polarization gradient [5].

On the boundary, from equation (2.13) it follows that the internal stress and couple stress must be balanced by the external actions t and $\mathbf{m}$; in addition, as it is well known (see for example [8]), since the smectic is supposed to be a perfect insulator, the orthogonal dielectric displacement component is equal to the induced electric charge surface density.

The internal stress σ includes a torque contribution, which is a consequence of the orthogonality constraint of the molecules to the

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layers, and an electric one which derives by the Maxwell stress tensor [5].

By using the expression (2.4) and (2.5), we can write

$$\rho\Psi = \frac{K_1}{2}\{(P^0_{\alpha\beta}u_{\alpha\beta})^2 + 2[P^0_{\alpha\gamma}u_{,\gamma\eta}P^0_{\eta\beta}u_{,\alpha\beta} - (P^0_{\alpha\beta}u_{\alpha\beta})^2]\} + \frac{\bar{B}}{2}(n^0_\alpha u_{,\alpha})^2 + \frac{1}{2}\rho\pi_\alpha(\chi^{-1})_{\alpha\beta}\pi_\beta, \tag{2.16}$$

where

$$\chi_{\alpha\beta} = (\varepsilon_\perp - 1)\delta_{\alpha\beta} + \varepsilon_a(n^0_\alpha - P^0_{\alpha\gamma}u_{,\gamma})(n^0_\beta - P^0_{\beta\eta}u_{,\eta}). \tag{2.17}$$

Here P^0 denotes the projector in the plane orthogonal to $\mathbf{n}^0$, $P^0_{\alpha\beta} = \delta_{\alpha\beta} - n^0_\alpha n^0_\beta$. By applying (2.12) and (2.13) we lead to

$$\sigma_\alpha = -K_1(P^0_{\beta\gamma}u_{,\beta\gamma})_{,\alpha} + \bar{B}n^0_\beta u_{,\beta}n^0_\alpha - \varepsilon_a(n^0_\beta - P^0_{\beta\gamma}u_{,\gamma})\phi_{,\beta}P^0_{\alpha\eta}\phi_{,\eta} - (n^0_\beta\phi_{,\beta})D_\alpha + \frac{\phi_{,\beta}D_{,\beta}}{2}n^0_\alpha, \tag{2.18}$$

$$D_\alpha = -\varepsilon_\perp\phi_{,\alpha} - \varepsilon_a(n^0_\beta - P^0_{\beta\gamma}u_{,\gamma})\phi_{,\beta}n^0_\alpha + \varepsilon_a n^0_\gamma\phi_\gamma P^0_{\alpha\beta}u_\beta, \tag{2.19}$$

$$M_{\alpha\beta} = K_1[P^0_{\gamma\eta}u_{,\gamma\eta}P^0_{\alpha\beta} + 2(P^0_{\alpha\gamma}u_{,\gamma\eta}P^0_{\eta,\beta} - P^0_{\gamma\eta}u_{,\gamma\eta}P^0_{\alpha\beta})], \tag{2.20}$$

where we have used the following identity

$$\frac{\partial\chi^{-1}_{\alpha\beta}}{\partial u_{,\gamma}} = -\chi^{-1}_{\alpha\eta}\frac{\partial\chi_{\eta\xi}}{\partial u_{,\gamma}}\chi^{-1}_{\xi\beta}, \tag{2.21}$$

and the relation

$$\rho(\chi^{-1})_{\alpha\beta}\pi_\beta = -\phi_{,\alpha}. \tag{2.22}$$

It is possible to check that the equation (2.19) expresses the classical relation of proportionality between the dielectric displacement and the electric field through the dielectric tensor ε. This gives us an *a posteriori* justification for the $\mathcal{F}_{int}$ form.

By inserting equations (2.18), (2.19) and (2.20) into (2.12), we obtain a coupled system of partial differential equations for the variables u and ϕ. We remark that the system is linear with respect to the variable u because we have taken into account only slight layer deformations, while it is not linear with respect to ϕ.

The system of differential equations must be supported by the boundary conditions. More precisely, on boundary regions where the fields δu, $\delta(\nabla u)$ and $\delta\phi$ are arbitrary the right hand quantities of

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the conditions (2.13) must be a datum of the problem. On the contrary, when the variation of the fields δu, $\delta(\nabla u)$ and $\delta\phi$ are null on a subregion of the boundary the right hand quantities are given by solving the problem. As we will show in the next section, intermediate situations are obviously possible.

3. MODIFIED HELFRICH-HURAULT CRITICAL FIELD EQUATIONS

Now we adapt our equations for a particular problem. Let us suppose that the smectic liquid crystal fills the region $\Omega \equiv \{0 \leq x \leq a, 0 \leq y \leq b, 0 \leq z \leq d\}$; in an ideal experiment the planes placed at $z = 0$ and $z = d$ can be worked as two electrodes of a capacitor. Let us suppose that the smectic is simply supported on the boundary [9]; so the molecules stick on the boundary but with a free orientation:

$$u = 0, \qquad \text{on} \quad \partial\Omega \tag{3.1}$$

$$\mathbf{m} = 0, \qquad \text{on} \quad \partial\Omega. \tag{3.2}$$

By using equations (3.1), (2.20) and the second of the (2.13), the boundary conditions (3.2) become

$$u_{,xx} = 0 \quad \text{in } x = 0,\ a \qquad u_{,yy} = 0 \quad \text{in } y = 0,\ b. \tag{3.3}$$

We impose electrical boundary conditions, by assigning a voltage difference V between the plates placed at $z = 0$ and $z = d$

$$\phi(x,y,0) = 0, \qquad \phi(x,y,d) = V, \tag{3.4}$$

while on the lateral faces we request that the electric charge vanishes, so we have

$$\sigma^e = 0, \qquad \text{in} \quad x = 0, a \quad \text{and} \quad y = 0, b. \tag{3.5}$$

Then the equilibrium equations (2.12) with the boundary conditions (3.1–3.5) admit the trivial displacement solution

$$u = 0, \quad \text{everywhere}, \qquad \phi = \frac{V}{d} z. \tag{3.6}$$

However, this may not be the only solution. As in the classical Helfrich-Hurault effect, we assume that there is a critical potential V_{cr} at which a solution bifurcation can occur. Then we study the solution of the equilibrium equations in a neighborhood of the critical voltage. Let us suppose, according to perturbation theory, that the

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unknown potential field can be written as

$$\phi = \phi_{cr} + \varphi \tag{3.7}$$

where φ is a small perturbation to the critical potential field ϕ_{cr}; we suppose that φ and its derivatives are $\mathcal{O}(\epsilon)$. Even, let us suppose that the critical potential gives rise to a uniform critical electric field

$$\mathbf{E}_{cr} = -\nabla\phi_{cr} = -\frac{V_{cr}}{d}\mathbf{n}^0, \qquad \phi_{cr} = \frac{V_{cr}}{d}z \tag{3.8}$$

where V_{cr} is the voltage between the two electrodes when the smectic switches from unperturbed to distorted configuration. ϕ_{cr} is a solution of the field equations in case of null deformation and voltage between the electrodes V_{cr}. For future developments we pose $E_{cr} = |\mathbf{E}_{cr}|$.

When the electric voltage between the two electrodes is exactly V_{cr}, we can take

$$\varphi(x,y,0) = 0, \qquad \varphi(x,y,d) = 0. \tag{3.9}$$

Now, we look for solutions of the form:

$$u(x,y,z) = v(x,y)u_0 \sin\Big(\frac{\pi}{d}z\Big), \quad \varphi(x,y,z) = \psi(x,y)\varphi_0 \sin\Big(\frac{\pi}{d}z\Big), \tag{3.10}$$

where u_0 and φ_0 have been introduced in order to work with dimensionless quantities $v(x,y)$, $\psi(x,y)$. Such choice of the fields u and φ satisfy automatically the boundary conditions (3.1), (3.2) and (3.9), and allows us to restrict the study of the problem to the two dimensional region $\bar{\Omega} = \{0 \le x \le a, 0 \le y \le b\}$ bounded by the closed curve $\partial\bar{\Omega}$. The equilibrium equations take the form

$$\Delta_\Sigma^2 v + \Big(\frac{\pi}{\lambda d}\Big)^2 v - \frac{\varepsilon_a}{K_1}E_{cr}^2\Delta_\Sigma v + \frac{\varphi_0}{u_0 K_1}E_{cr}\varepsilon_a\Delta_\Sigma\psi = 0, \tag{3.11a}$$

$$\Delta_\Sigma\psi - \left(\frac{\varepsilon_a}{\varepsilon_\perp} + 1\right)\Big(\frac{\pi}{d}\Big)^2\psi + E_{cr}\frac{\varepsilon_a}{\varepsilon_\perp}\frac{u_0}{\varphi_0}\Delta_\Sigma v = 0, \tag{3.11b}$$

where Δ_Σ and Δ_Σ^2 are the Laplacian and biharmonic operator in the (x,y)-plane.

The boundary conditions for u and ϕ on the surface $\partial\Omega$ lead to the following boundary conditions for v and ψ on the contour $\partial\bar{\Omega}$

$$v = 0 \qquad \text{on } \partial\bar{\Omega}, \tag{3.12}$$

$$v_{,xx} = 0 \quad \text{in } x = 0,\ a, \quad v_{,yy} = 0 \quad \text{in } y = 0,\ b, \tag{3.13}$$

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$$\varepsilon_\perp \varphi_0 \psi_{,x} + \varepsilon_a E_{cr} u_0 v_{,x} = 0, \quad \text{in } x = 0,\ x = a, \tag{3.14a}$$

$$\varepsilon_\perp \varphi_0 \psi_{,y} + \varepsilon_a E_{cr} u_0 v_{,y} = 0, \quad \text{in } y = 0,\ y = b. \tag{3.14b}$$

We remark that if the perturbation potential ψ vanishes, the equation (3.11a) takes the classical form which allows to find the classical Helfrich-Hurault critical electric field. Nevertheless such choice does not satisfy automatically the equation (3.11b); in fact a further hypothesis is required in order to hold the equation (3.11b): its last term must be zero. In nematic literature [13] this hypothesis is called *magnetic approximation* and it consists in putting $\varepsilon_a = 0$ in the Maxwell equation. Consistently, we put $\psi = 0$ and $\varepsilon_a = 0$ in the boundary conditions (3.14); so that are trivially satisfied. With this hypothesis, our problem is identical to the homogeneous version of the problem treated in the section 3 of the Stewart's paper [12].

Within the magnetic approximation one finds that non-trivial solution of equations (3.11) with homogeneous boundary conditions are possible for several values of E_{cr}. The smallest one is the Helfrich-Hurault critical field:

$$E_{cr} = \sqrt{-2\frac{K_1}{\varepsilon_a}\left(\frac{\pi}{\lambda d}\right)} \quad \varepsilon_a < 0; \tag{3.15}$$

for a voltage difference V such that $|V| > |V_{cr}| = E_{cr} d$ the layers undulation is energetically preferred with respect to the undistorted configuration.

The complete coupled case is carefully discussed in [1]. We resume here the principal features:

- As in the classical case, non trivial solutions are possible only for negative values of dielectric anisotropy $\varepsilon_a < 0$.
- The critical field depends also on the ratio between $\varepsilon_\parallel$ and $\varepsilon_\perp$ and not only on their difference as in the classical case.
- When the sample contains a great number of layers it is possible to give an analytical expression for the critical field. Then E_{cr} is greater than the classic one.

4. CONCLUSIONS

We have presented a model describing the interaction of an insulator smectic liquid crystal with an electrostatic field. Our model goes beyond the literature approach [2,12] in which the electrostatic field is not affected by the smectic deformation. We have adapted our

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equations in order to study a Helfrich-Hurault type transition in a finite cell of smectic-A liquid crystal with homeotropic alignment. Thus, we have obtained and discussed a new system of differential equations to determine the critical field of the transition.

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