Regular dendritic patterns induced by non-local time-periodic forcing

T. Börzsönyi\textsuperscript{1,2}, T. Tóth-Katona\textsuperscript{1}, Á. Buka\textsuperscript{1}, and L. Gránásy\textsuperscript{1}
\textsuperscript{1}Research Institute for Solid State Physics and Optics, Hungarian Academy of Sciences, H-1525 Budapest, P.O.B.49, Hungary
\textsuperscript{2}Groupe de Physique des Solides, CNRS UMR 75-88, Universités Paris VI et VII, Tour 23, 2 place Jussieu, 75251 Paris Cedex 05, France

The dynamic response of dendritic solidification to spatially homogeneous time-periodic forcing has been studied. Phase-field calculations performed in two dimensions (2D) and experiments on thin (quasi 2D) liquid crystal layers show that the frequency of dendritic side-branching can be tuned by oscillatory pressure or heating. The sensitivity of this phenomenon to the relevant parameters, the frequency and amplitude of the modulation, the initial undercooling and the anisotropies of the interfacial free energy and molecule attachment kinetics, has been explored. It has been demonstrated that besides the side-branching mode synchronous with external forcing as emerging from the linear Wentzel-Kramers-Brillouin analysis, modes that oscillate with higher harmonic frequencies are also present with perceptible amplitudes.

I. INTRODUCTION

Complex patterns observed in nature attract a considerable interest recently \cite{1}--\cite{3}. The complicated spatiotemporal behavior that leads to the formation of such patterns is usually associated with the instabilities of systems transforming under non-equilibrium conditions. A spectacular example of practical importance is dendritic growth that appears in anisotropic systems where the propagation of the transformation front is coupled with the diffusion of a conserved quantity. The respective diffusional instabilities lead to the formation of a traveling quasi-steady-state tip that emits surface undulations which evolve into side arms in directions determined by the anisotropy of the system \cite{4}. Many of the technologically important materials form by dendritic solidification \cite{1}. Analogous phenomena have been reported in biological systems \cite{5}, anisotropic Hele-Shaw cells \cite{6}, and in cosmology/particle physics \cite{7,8}. Although experiments on the freezing of transparent liquids \cite{9}--\cite{13} clarified many of the essential features of dendrite formation, important questions remained open. For example, the role played by thermal fluctuations in side-branching is the subject of continuing investigations \cite{14}--\cite{19}. It is anticipated that a selective amplification of thermal noise is responsible for the side-branching and for the observed irregularity of dendritic patterns \cite{14}--\cite{18}.

While the steady-state behavior of dendritic growth is understood fairly well in the framework of recent theories (microscopic solvability \cite{19}--\cite{22} and phase-field theories \cite{14}--\cite{28}), less is known of the dynamic response of the dendritic morphology to time-dependent external perturbations. The non-linear systems often develop regular patterns under periodic forcing \cite{20}. Considering the inherent non-linearity of the equations describing dendritic growth, it is reasonable to expect that such periodic perturbations lead to resonance patterns that might be used to influence the growth morphology. Besides its scientific interest, a detailed understanding of such phenomena could open novel routes for designing materials for specific applications.

Time-periodic forcing of dendritic side-branching has already been realized via heating locally the dendrite tip by laser beam \cite{21}--\cite{23}, or by exposing it to an oscillatory flow field \cite{24}. Both methods lead to the formation of fairly regular morphologies in a suitable frequency range. However, these methods cannot be easily used for spatially homogeneous controlling of growth morphologies in large volumes. To circumvent this problem, we demonstrated \cite{25} the possibility for tuning the dendritic morphology by spatially homogeneous time periodic forcing via modulated pressure \cite{26} and heating, that change the undercooling instantaneously and homogeneously in large volumes.

In this paper we present a detailed analysis of the dynamic response of dendritic solidification to oscillating pressure and heating. In identifying the resonance conditions and the most important process parameters, we rely on phase-field modeling, one of the most potent methods for describing morphology evolution. The theoretical predictions will be confronted with experiments on quasi-2D liquid crystal layers, known as suitable model materials \cite{14,23,27}. The rest of this paper is structured as follows. In section II we briefly summarize the phase-field equations that incorporate external forcing, and introduce quantities for characterizing growth morphology. Section III is devoted to the experimental setup and the details of the measurement techniques. In section IV, we present the phase-field predictions and compare them with the experimental results. In section V, our findings are discussed in the light of theory and experiment on tip-localized forcing. Finally, a few concluding remarks will be made in section VI.
II. PHASE-FIELD THEORY WITH EXTERNAL FORCING

We investigate the growth of the crystalline phase into an undercooled single component liquid at a temperature between the melting point and the hypercooling limit (“unit undercooling”) [3]. In this temperature range, the thermal diffusion controls the growth process, as opposed to the (molecule attachment) kinetics controlled mechanism taking place below the hypercooling temperature [3]. Owing to the heat release during solidification, the crystalline phase heats up to its melting point, while the rest of the latent heat is transported into the cold liquid. Under such conditions, the planar front is inherently unstable against thermal perturbations despite the stabilizing effect of the interfacial free energy [10].

The phase-field theory is a powerful tool to study moving boundary problems. It represents a dynamic extension of the Cahn-Hilliard theory of first-order phase transformations, in which the evolution of a non-conserved order-parameter is coupled to thermal or chemical diffusion [12,23,24]. Here we use a thermodynamically consistent version [23] (Model I of [25]) which is extended to incorporate anisotropies of the interfacial free energy and the kinetic coefficient [11]. The local state of the matter is represented by the phase-field \( \phi(r, t) \) [12] that describe the evolution of the phase-field and the temperature field \( T(r, t) \) in 2D

\[
\tau(\theta) \frac{\partial \phi}{\partial t} = Q(T)p' (\phi) - G' (\phi) - \frac{\delta}{\delta \phi} \left\{ \frac{1}{2} [\kappa(\theta)]^2 |\nabla \phi|^2 \right\}, \tag{1}
\]

\[
\{c_p + [p(\phi) - 1]L'(T)) \} \frac{\partial T}{\partial t} + L(T)p' (\phi) \frac{\partial \phi}{\partial t} = k \nabla^2 T, \tag{2}
\]

where \( \tau(\theta) \) is an empirical relaxation coefficient, whose inverse is an intrinsic interfacial mobility, and \( \frac{\delta}{\delta \phi} \) stands for variation with respect to \( \phi \). Other notations are as follows: \( p(\phi) = \phi^3(10 - 15\phi + 6\phi^2) \), \( \kappa^2/2 \) the coefficient of the square-gradient term in the entropy functional, that depends on the orientation represented by the angle \( \theta \) (defined as \( \tan(\theta) = \partial \phi / \partial x \)), \( G(\phi) = \phi^2(1 - \phi)^2 / 4e \), \( e \) is a constant that determines the height of the intermediate maximum of the double well-potential, \( Q(T) = \int_T^\infty \frac{L' (\gamma)}{\gamma^2} d\gamma = -\Delta G'(T) \), \( \Delta G(T) \) the Gibbs free energy difference between the liquid and solid, while \( c_p, L(T) \), and \( k \) are the specific heat of the liquid, the enthalpy difference between the liquid and the solid, and the thermal conductivity, respectively. The primed quantities denote derivatives with respect to the arguments. Note that \( \phi \) and \( p \) are dimensionless. The units of \( k \) and \( \kappa^2 \) are \( \text{W m}^{-1} \text{K}^{-1} \) and \( \text{J m}^{-1} \text{K}^{-1} \), respectively; while \( Q, G, c_p \) and \( 1/e \) are measured in \( \text{J m}^{-3} \text{K}^{-1} \).

The dominant effect of the pressure modulation \( p(t) = p_0 + \Delta p(t) \) is a variation of the melting point described by the Clausius-Clapeyron law \( T_m(p) = T_m(p_0) + \Delta p \Delta V / S_m \), where \( \Delta V \) is the volume change upon solidification, and \( S_m \) is the entropy of fusion. This enters into Eq. (1) via the Gibbs free energy difference which may be approximated as \( \Delta G(T, p) \approx S_m [T_m(p) - T] \) at small undercoolings \( (T_m - T < \Delta T_m) \). The other relevant quantities [specific heat, heat of fusion, and entropy of fusion \( L'(T) \)] are far less sensitive to the variation of pressure. For example, integrating the Clausius-Clapeyron relationship one obtains \( \Delta L = \Delta p \Delta V [1 + \Delta \rho_c / S_m + T(\alpha \Delta V - \alpha_s V_s) / \Delta V] \), where \( \Delta c_p = c_p,l - c_p,s \) is the variation of the specific heat upon melting, \( \alpha_l, s \) are the thermal expansion coefficients, while subscripts \( l \) and \( s \) refer to the liquid and the solid respectively. Under typical conditions of our experiments [pressure amplitude ~ 2 bar, undercooling of 1 K, and physical properties of CCH3 (Appendix A)], the undercooling varies by ~ 6%, while the relative change of \( L \) and \( S_m \) is only \( \sim 10^{-3} \). We found that such pressure effects on \( L \) and \( S_m \) do not influence the results perceptibly; i.e. pressure modulation needs to be taken into account only via \( Q(T) \) in Eq. (1).

Modulated heating, in turn, appears as a time dependent source term in Eq. (2).

At this stage it is advantageous to introduce dimensionless variables. This is done similarly to that in [25]. However, to avoid complications emerging from an oscillating melting point, the reduced temperature has been redefined in terms of a constant reference temperature \( T_r \) as \( u(r, t) = (T - T_r) / (T_r - T_\infty) \), where \( T_\infty \) is the initial temperature of the undercooled liquid. The mathematical problem is invariant to the choice of \( T_r \) so far as \( A = u_m = (T_m - T_r) / (T_r - T_\infty) \) is inserted as in Eq. (3). (With \( T_r = T_m \), the usual form of the phase-field model is recovered.)

The relevant physical properties are combined into dimensionless parameters \( \Delta = c_p(T_r - T_\infty)/L, \alpha = \sqrt{2} \omega S_m L / (12c_\rho \sigma_o), \tau_o = S_m D \beta_0 / \sigma_o, \) and \( \epsilon = \delta / \omega, \sigma' = \sigma / \delta / \delta \theta, \) where \( D \) is the thermal diffusion coefficient, \( \delta \) the interface thickness, and \( \omega \) a reference length comparable to the size of a well developed crystallite. The anisotropies of the interfacial free energy \( \sigma \) and the kinetic coefficient \( \beta \) are represented by the dimensionless functions \( \tilde{\sigma} = 1 + \sigma_o \cos(n \theta) / 2, \beta = 1 + \beta_n \cos(n \theta) / 2 \) of \( n \)-fold symmetry, that are related to the dimensional quantities via relationships \( \sigma(\theta) = \sigma_o \tilde{\sigma}(\theta) \) and \( \beta(\theta) = \beta_o \tilde{\beta}(\theta) \). Here \( \sigma_n \) and \( \beta_n \) are the anisotropy parameters. Lengths and time are scaled by \( \omega \) and \( \omega^2 / D \), respectively. The coefficient \( \kappa \) and the constant \( e \) are related to \( \delta \) and the interfacial free energy [24].

With these notations Eqs. (1) and (2) transform to
\[
e^2\tau_0\partial^2_0 = \phi(1 - \phi)\{(1 - \phi)[\phi - \frac{1}{2} + 30\epsilon x\Delta[u - A(t)]\phi(1 - \phi)]
\]

\[
-\epsilon^2 \frac{\partial}{\partial x}[(\sigma_x^2) \phi(1 - \phi)] + \epsilon^2 \frac{\partial}{\partial y}[(\sigma_y^2) \phi(1 - \phi)] + \epsilon^2 \nabla^2 [\sigma_x^2 \phi(1 - \phi)]
\]

\[
\frac{\partial u}{\partial t} + \frac{1}{\Delta} 30\phi^2 (1 - \phi)^2 \frac{\partial^2 \phi}{\partial t^2} = \nabla^2 u + B(t),
\]

where the modulated pressure and heating are incorporated via terms \(A(t)\) and \(B(t)\), respectively.

In this work Eqs. (3) - (4) have been solved numerically on an \(N \times N\) rectangular grid, \(N = 1000\), that corresponds to an area \(l \times l\) of dimensionless linear size \(l = 5\) and a grid spacing \(\Delta x = 0.005\). An explicit finite difference scheme has been employed in the case of Eq. (3), while Eq. (4) has been handled by the alternating-direction implicit method which is unconditionally stable [41]. At \(t = 0\) a crystalline nucleus (\(\phi = 0\)) is placed at the center of the model area filled by uniformly undercooled liquid (\(\phi = 1, u = -1\)). The reduced temperature and the phase-field were kept constant (\(u = -1\) and \(\phi = 1\)) at the boundaries of the model space. To save CPU time, we used a primitive adaptive mesh technique: far from the solidification front Eq. (4) has been replaced by a difference scheme. The amplitude of regulation with oscillatory heating, see section III.C, was determined.

To characterize the spatiotemporal behavior of the evolving dendritic morphology the following quantities were determined:

1. We measured the width \(w\) of the dendrite behind the tip at a distance of \(\zeta = 0.75\) (150 pixels). To investigate this feature for longer times, we performed separate simulations on a \(1200 \times 600\) grid, oriented so that the larger dimension be parallel with the direction of growth (here we used \(\Delta t = 8 \times 10^{-5}\)). The amplitude \(A\) in the Fourier transform \(W(f)\) of \(w(t)\) characterizes the response synchronous with external forcing.

2. The symmetry of the growth patterns was characterized by the quantities \(a\) and \(b\):

\[
a = \frac{\sum_{i,j=-N/2}^{N/2} (\phi_{i,j} - \phi_{-i,-j})^2}{M}
\]

\[
b = \frac{\sum_{i,j=-N/2}^{0} (\phi_{i,j} - \phi_{j,i})^2}{K}
\]

where \(M\) and \(K\) are the numbers of pixels in which \(0.4 < \phi < 0.6\) for the whole system and for the lower left quarter, respectively. Accordingly, \(a \geq 0\) measures the symmetry of the whole domain with respect to the \((xy)\)-axis, while \(b \geq 0\) quantifies the axisymmetry of a main branch. Both parameters are zero for symmetric patterns.

III. EXPERIMENTAL SETUP AND CONDITIONS

The nematic-smectic B (\(N - S_B\)) phase transition of liquid crystals is recognized as an appropriate model of crystallization in liquids [25, 27]. A specialty of this phase transformation is a large anisotropy of the interfacial free energy [26, a property expected to be advantageous for the regularization of dendritic morphology on theoretical grounds (see section IV.A and [33]).

To test the predictions, experiments have been performed on thin CCH3 liquid crystal layers. Ready-made cells of E.H.C. Co. (Japan) KSRP-10 (of thickness 10 \(\mu\)m) and KSRP-02 (2 \(\mu\)m) have been filled with CCH3 (Merek, Darmstadt). The surface treatment of the bounding glass plates assured the planar alignment of both \(N\) and \(S_B\) phases [the directors \(\mathbf{n}(N)\) and \(\mathbf{n}(S_B)\) that describe the average orientation of the elongated molecules are in the plane of the cell], and the conducting layers on the bounding plates were used as electrodes.

For pressure modulation the liquid crystal cell was placed into a brass box (see Fig. 3) surrounded by a temperature controlled hot-stage of accuracy \(\pm 3\) mK. The gas pressure in the brass box has been regulated by a computer controlled solenoid valve system that switches on and off an excess pressure \(p_\nu\), preset between 0 to 2 bar with an accuracy of \(\pm 0.03\) bar. This allows square wave-like pressure modulations in the frequency range up to \(\nu \sim 2\) Hz.

The modulated heat release in the bulk has been realized by periodically transmitting a high frequency (600 kHz) electric current through the liquid crystal layer produced by gating the signal of a function generator with \(\nu \sim 1\) Hz. The local off-plane heat transport (a precondition of regulation with oscillatory heating, see section IV.A and [33]) is ensured by the quasi-2D sample geometry and by the heat transport through the bounding glass plates.

The growth patterns were monitored in transmitting mode via a polarizing microscope equipped with a CCD camera, a method relying on the different optical properties of the nematic and smectic B phases. The images were stored and processed by a PC. The spatial and time resolutions of the system were \(512 \times 512\) pixels and 0.04 s, respectively. The calibration procedure with 6.3 \(\times\) objective and 3.2 \(\times\) projector combination gave scale factors of \(1.35 \pm 0.01\) \(\mu\)m/pixel in \(x\) direction and \(0.95 \pm 0.01\) \(\mu\)m/pixel in \(y\) direction.
The same setup has been used to measure the pressure coefficient of the temperature of transformation between the smectic-B and nematic phases (see Appendix A).

IV. RESULTS

A. Theoretical predictions

To find resonance patterns we investigated the parameter space defined by the frequency and amplitude of the modulation, the undercooling and the anisotropies of the interfacial free energy and the kinetic coefficient. The calculations were performed with four- and six-fold anisotropies ($n = 4$ and $6$ in expressions for $\tilde{\sigma}$ and $\tilde{\beta}$).

Before presenting our results on non-local periodic forcing we characterize the unmodulated reference state.

1. Free dendritic growth

Independently of the symmetry of the system, a weak uncorrelated (noise-induced) side-branching occurs in the unmodulated reference states shown in Figs. 3(a) and 3(a). The width $w(t)$ of a main dendrite arm measured at a dimensionless distance of $\zeta = 0.75$ behind the tip [Fig. 3(a)] indicates surface undulations that can be decomposed into a spectrum of oscillations which covers the dimensionless frequency range of $\sim 30$ to $\sim 170$, centered around a characteristic frequency of $f_0 \sim 100$ [see insert in Fig. 3(b)]. This behavior accords with that seen in the phase-field simulations of Karma and Rappel [23], and with experimental results [16,30]. In agreement with [23], we find that far from the tip the characteristic wave length $\lambda_0 = \nu/f_0$ depends much more weakly on the distance from the tip than predicted by the microscopic solvability theory [22,44], a behavior interpreted in terms of the “stretching” of the perturbations [23] that travel along the curved perimeter of the dendrite.

Considering that the applied uncorrelated noise probes the dynamic response of the system to a broad variety of frequencies, it is reasonable to seek “resonance” in the vicinity of the characteristic frequency $f_0$ of spontaneous side branching.

2. Pressure modulations

According to the Clausius-Clapeyron law, the modulated pressure translates into a time dependent melting point, and is represented by inserting $A(t) = u_m(t) = a_0g(t)$ and $B(t) = 0$ into Eqs. (3) and (4). Since our experimental setup allows only square-wave modulations (characterized by the filling coefficient $\xi = t_{on}/t_0$, where $t_0$ is the period of oscillations and $t_{on}$ the pulse length), the simulations have been performed for this wave form (unless stated otherwise).

(a) The effect of forcing frequency: The frequency dependence of the growth patterns is shown in Figs. 3(b)–(f) and 3(b)–(f). At low forcing frequencies ($\nu < 15$), the system alternates between two steady-state growth modes yielding uncorrelated side-branching that correspond to the actual undercooling. It is, however, remarkable that the switching transient between the constant pressure stages initiates more pronounced side arms than formed during the constant pressure periods. Regular growth morphologies [Figs. 3(b)–(c) and 3(b)–(e)] are observed in the lower half of dimensionless frequency range of spontaneous thermal side branching shown in the insert of Fig. 3(b)]. At frequencies larger than these, the formation of side branches cannot follow the external forcing, and the uncorrelated thermal side-branching reappears [Figs. 3(f) and 3(f)].

In agreement with our earlier results [33], when regular morphologies are formed, the side-branching and the tip velocity correlate with the pressure modulation.

The parameters $w(t)$, $a$, and $b$ reflect the formation of regular morphologies. For example, a periodic variation of the width $w(t)$ of the dendrite is seen [Fig. 3(a)], which correlates with the external sinusoidal forcing and leads to the formation of regular side branches. Note that besides the forcing frequency (that appears with a far larger amplitude in $W(f)$ than the spontaneous undulations), its second and third harmonics ($2\nu, 3\nu$) are also present albeit with an amplitude that diminishes for higher order harmonics [Fig. 3(b)].

Apparently, periodic forcing with fixed frequency excites several surface modulations; one synchronous with the forcing frequency ($\nu$) and others oscillating with the higher harmonic frequencies ($2\nu, 3\nu, ...$), which indicates a dynamic coupling among these modes associated with the non-linearity of the governing equations.

The relative amplitudes of higher harmonics vary with the wave form of the pressure modulation yielding different growth patterns (see Fig. 3). Fairly similar patterns [cf. Figs. 3(a) and (b)] are observed for sinusoidal and square-wave modulations, provided that for the latter $\xi = 0.5$ [i.e., frequencies $(2n + 1)\nu$, $n = 1, 2, ...$ are present in the forcing spectrum]. When the filling coefficient $\xi$ deviates from 0.5, the frequency/phase content of forcing varies (e.g., the second harmonic $2\nu$ appears) leading to such effects as the formation of side-branches of twice the number corresponding to the base frequency [cf. Fig. 3(a)–(c)]. To understand this behavior we analyze the frequency dependence of the dynamic response of the system to “asymmetric” forcing ($\xi = 0.3$).

The Fourier amplitudes $A_\nu$ and $A_{2\nu}$ (corresponding to modes that oscillate with the forcing frequency and its double, respectively) and the symmetry parameters $a$ and $b$ are presented in Fig. 3 as a function of the forcing frequency $\nu$. $A_\nu$ and $A_{2\nu}$ show maxima if $\nu$ or $2\nu$ are close to
\(f_{\text{exc}} \sim 70\) [Fig. 3(a)], where \(f_{\text{exc}}\) is the frequency where the synchronous response has the maximum amplitude. Note that \(f_{\text{exc}}\) is somewhat below the characteristic frequency \(f_0 \sim 100\) of the unperturbed dendritic growth. A possible explanation of this deviation might be a different “stretching” of perturbations, related to differences seen between shapes of dendrite tips formed in free growth and under external forcing. It is also remarkable, that at low forcing frequencies the amplitude \(A_{2\nu}\) of the second harmonic becomes larger than \(A_\nu\). This inversion of the relative magnitudes of \(A_\nu\) and \(A_{2\nu}\) is responsible for the side-branch “doubling” shown in Fig. 3. A similar phenomenon is seen in Fig. 3(b), where the second harmonic is the dominant mode as indicated by the striking similarity of patterns corresponding to \(\nu = 20\) and 40 [cf. Figs. 3(b) and (c)]. Note the lack of alternating variation of the length of side-branches that appears when the synchronous mode is also present with a significant amplitude [as in Fig. 3(c)].

We find that the amplitude \(A_\nu\) increases approximately exponentially with the distance \(\zeta\) from the tip of the dendrite. In contrast, \(A_{2\nu}\) shows a maximum as a function of \(\zeta\) (Fig. 3). Remarkably, the symmetry parameters \(a\) and \(b\) display minima at lower forcing frequencies [around \(\nu \sim 20\); see Fig. 3(b)] than the maximum of the Fourier-amplitude \(A_\nu\). This difference in the positions of the extrema is a manifestation of the fact that axial symmetry and frequency content are different constituents of regularity. It appears that \(A_\nu\) gives a closer representation of the regularity recognized by the human eye than the symmetry parameters.

The overall frequency dependence of the pattern formation supports our anticipation that regularization via periodic forcing with fixed frequencies is possible when the latter fall in the characteristic frequency range of noise-induced side branching.

(b) The effect of the amplitude of forcing: The Fourier amplitudes \(A_\nu\) and \(A_{2\nu}\) and the symmetry parameters show that with increasing forcing amplitude \(a_0\) the regularity of the growth patterns increases (Fig. 3). This is manifested in monotonously increasing \(A_\nu\) and \(A_{2\nu}\) that saturate for large \(a_0\). At the same time, \(a\) and \(b\) decrease with \(a_0\), reflecting the increasing symmetry of the patterns.

(c) The effect of undercooling: The response to a given pressure modulation correlates with the tendency for “natural” side-branching (Fig. 3). While at large undercoolings (e.g., \(\Delta = 0.65\)), the formation of side-branches is rather intense, leading to well developed side-arms (both with or without forcing), at \(\Delta = 0.5\) or below free growth produces essentially no side-branches, and external forcing is needed to trigger them, also reflected in their smaller size. Note the difference in spatial periodicity, that originates from the temperature dependence of the tip velocity.

The variation of the Fourier amplitudes \(A_\nu\) and \(A_{2\nu}\) with undercooling (\(\Delta\)) is shown in Fig. 3. The maximum of \(A_\nu\) might be understood as follows. We found that with increasing undercooling \(f_{\text{exc}}\) increases. Accordingly, a fixed forcing frequency \(\nu\) produces the maximum amplitude at the undercooling for which \(f_{\text{exc}} \sim \nu\). For the same reason, the maximum amplitude for the second harmonic \(2\nu\) appears at a larger undercooling.

(d) The effect of anisotropy: Theoretical considerations and numerical simulations show that anisotropy plays a central role in the formation of dendritic morphology. It is, therefore, reasonable to expect that it has a similar importance in the formation of regular patterns. This expectation is verified by the dependence of growth forms on the magnitude of anisotropies in the system (Fig. 3). The growth patterns and variations of the Fourier amplitudes \(A_\nu\) and \(A_{2\nu}\), and of the symmetry parameters (Fig. 3) indicate that the regularizing effect of external forcing decreases dramatically with vanishing anisotropy, i.e. without well defined orientational preferences the external perturbations are unable to drive the amoebae-like growth forms into a regular pattern. Note that the tip velocity decreases with decreasing anisotropies (note the difference of simulation times the snapshots correspond to), the excitation with \(\nu = 60\) generates periodic side-arms with increasing spacing.

In these calculations we assumed that both the interfacial free energy and the kinetic coefficient are anisotropic. For the sake of simplicity, their ratio was usually kept constant \(\beta_4/\sigma_4 = -2\). Our further investigations show, however, that resonance patterns form if any of these anisotropies (that of \(\sigma\) or \(\beta\)) is sufficiently large.

3. Modulated heating

(a) Alternating heating and cooling: They lead to essentially the same type of resonance patterns as pressure oscillations (Fig. 3), provided that that the net heat production in a period is negligible. A remarkable difference is, however, that while the average tip velocity \(v_0\) is essentially independent of the amplitude of pressure modulations, it decreases with the amplitude of heat production/extraction (Fig. 3); a phenomenon that might be associated with different efficiencies of heating and cooling.

(b) Oscillatory heating: The introduction of a local off–plane thermal transport described by \(B(t) = b_0g(t) + h[u(r,t) - u_{\infty}]\) was necessary to prevent the melting of the crystal for the heating amplitudes needed to generate regular patterns. Here \(h < 0\) is a dimensionless heat transfer coefficient that imitates heat transfer perpendicular to the plane of the liquid crystal layer towards the surrounding of reduced temperature \(u_{\infty}\). Since this term serves as a local heat sink, the in-plane thermal diffusion
becomes less restrictive. This results in the formation of more compact objects (see Fig. [13]) with thick main arms and less developed side-branches. While with an increasing amplitude of the heat pulses the side-arms become more regular, they become smaller due to the dissipated heat.

Much like pressure modulations, oscillatory heating leads to an oscillating tip velocity. Owing to a decreasing average undercooling accompanied with increasing heating amplitude \( b_0 \), the period averaged velocity \( v_0 \) of the tip decreases roughly linearly with \( b_0 \) (Fig. [19]). At the same time, the amplitude of the oscillatory part of the velocity \( v_o \) increases about linearly.

In summary, our investigations imply that under well defined conditions both types of non-local forcing can be used to control dendritic growth.

**B. Experimental results**

In defining the experimental conditions, we utilized the results of the computer simulations. For CCH3, the time and spatial resolution of our experimental setup is optimal at an undercooling of \( \sim 1 \text{ K} \). Comparable variations of the undercooling \( \sim 0.1 – 0.2 \text{ K} \) are achievable by applying modulation amplitudes as high as 2 bar or \( 3 \times 10^{-4} \text{ W/cm}^2 \), as estimated on the basis of the pressure coefficient of the equilibrium temperature for the nematic–smectic B phases (see Appendix A) and other relevant properties.

The appropriate frequency range of forcing has been determined by Fourier analysis of the width \( w(t) \) of a freely growing dendrite measured at \( 66.5 \mu \text{m} \) behind the tip. Without perturbation, the side-branching is essentially random [Figs. 17(a) and 18(a)]. The characteristic frequency, identified as the peak of the broad spectrum in the insert of Fig. 19, is roughly 1.8 Hz. Accordingly, our experimental setup was designed to cover the frequency range of \( 0 – 2 \text{ Hz} \).

**1. Pressure modulations**

The experimental results for pattern formation in the 2 \( \mu \text{m} \) thick liquid crystal cell are summarized in Fig. 17. Resonance patterns of fairly regular side-branches are observed [Figs. 17(b)–(f)] at forcing frequencies comparable with the characteristic frequency of the unper- turbed state \( \tilde{v} \sim 1.8 \text{ Hz} \). The figure shows germs that nucleated heterogeneously at the same site with essentially the same orientation several times, yielding reproducible patterns. Although the respective dimensionless undercooling \( (\Delta = 0.06) \) is considerably smaller than in the simulations \( (\Delta = 0.4 – 0.68) \), the observed behavior follows closely the predictions of the phase-field theory. This similarity is especially striking when patterns formed at similar reduced frequencies \( (\tilde{v}/f_0) \) are compared [cf. Figs. 2(c) and 17(b) of \( \tilde{v}/f_0 = 0.40 \) and 0.42; or Figs. 2(d) and 17(d) of \( \tilde{v}/f_0 = 0.6 \) and 0.65]. An interesting observation is that far from the dendrite tip irregularities develop in the lengths of the side-branches, despite the even distance of their trunks. We observed an analogous phenomenon driven by the interaction with neighboring side-arms via thermal diffusion fields in long-time phase-field simulations [13].

The effect of the forcing amplitude is shown in Fig. 20. Pressure oscillations of amplitude of 1 bar efficiently regularize the side-branch formation. With increasing amplitude the phenomenon becomes more pronounced as predicted by the phase-field calculations. At the largest pressure amplitudes even the secondary side-branches correlate with the pressure oscillations [see Fig. 20(c)]. Note again that the competition of neighboring side-branches leads to irregularities in their lengths.

**2. Modulated heating**

In analogy to pressure modulations, the periodic heating experiments on the 10 \( \mu \text{m} \) thick liquid crystal cell also reveal regular side-branching [see Figs. 18(a)–(c)]. The formation of side-branches correlates with the external forcing as illustrated in Fig. 18(b), where the black lines denote the position of the tip at the centers of the heating pulses. The correlation is also evident from the power spectrum of the width of the dendrite measured 66.5 \( \mu \text{m} \) behind the tip (Fig. 13). In full accord with the phase-field simulations for asymmetric square waves [Fig. 1(b)], the power spectrum indicates the presence of modes that are either synchronous with the forcing frequency \( \tilde{v} = 0.46 \text{ Hz} \), or oscillate with doubled frequency \( 2\tilde{v} \). Even a peak corresponding to the third harmonic \( 3\tilde{v} \) may be identified, although with an amplitude that is close to the experimental uncertainty.

Remarkably, in the response to “symmetric” forcing \( \xi = 0.5 \), the \( 2\tilde{v} \) mode is also present (see the Fourier-spectrum and the short side-arms of doubled frequency in Fig. 21), although this frequency is absent from the forcing spectrum. This finding confirms the non-linear behavior revealed by phase-field simulations, that higher harmonics missing from the forcing spectrum are also excited [see \( W(f) \) for sinusoidal forcing in Fig. 1].

Increasing the heating amplitude or \( \xi \) so that the period-averaged heating power reaches \( P \approx 10^{-4} \text{ W/cm}^2 \), the formation of the side-branches is suppressed [see Fig. 13(d)], a phenomenon resembling that seen in the numerical simulations (Fig. 13). Further increase of the heating amplitude (and power) melts the dendrites back.

In line with our theoretical predictions (see Fig. 2, of 13), a weak oscillation of the tip velocity has been observed that correlates with the forcing. However, its
amplitude is just above the resolution of the present experimental setup.

A remarkable feature of the "regularized" dendrites, not seen in the simulations, is a shift in the position of the side-branches on the two sides of the main tip [see e.g. Figs. 18(b) and (c)]. As a result, one cannot use the symmetry parameters $a$ and $b$ [defined by eqs. (3) and (4)] for characterization of the pattern's regularity. For the same reason, the Fourier spectra shown in Figs. 19 and 21 have been evaluated from the half-width of the dendrite. Despite the shift of the side-branches, the power spectra on the two sides are fairly similar.

The shift in the position of the side-branches might be attributed to the asymmetry of the dendrite tip (due to the angular dependence $\sigma(\theta)$ [35]), shown by a high resolution snapshot of the tip region of a dendrite grown freely at $\Delta T = 1.0$ °C (Fig. 22). Another remarkable feature is that the left side of the tip is faceted. The first observable surface undulation (marked with arrow in Fig. 22) that evolves later into a side-arm appears on the opposite side. In contrast, the faceted side remains smooth up to the same distance behind the tip.

Finally, one should mention that the electric heating in liquid crystals may have side-effects that are not incorporated into our phase-field model. Switching the electric field on, the orientation of the nematic director $n(N)$ changes from planar to homeotropic (perpendicular to the bounding plates), an effect that influences the magnitude and anisotropy of the interfacial free energy, and may induce local flow in the sample. However, these side-effects are of minor importance, since they are present in full strength much below the electric field needed for regularization.

V. DISCUSSION

In this section we confront our results with those on the dynamic response of dendritic growth to local forcing with fixed frequency as emerging from the microscopic solvability theory (MST) and experiment. It is appropriate to mention, that some features of the MST derivation prevent a quantitative comparison. For example, the MST calculations were performed for a 2D symmetric dendrite, while the anisotropy, the stability coefficient, and the Péclet-number were assumed to be small; conditions that are not met in our simulations and experiments. Therefore, a qualitative comparison is only meaningful. An important further difference between previous work and ours is that we used non-local forcing, as opposed with the tip-localized forcing assumed in the MST.

Let us first recall some of the MST predictions on noise amplification (based on the Wentzel-Kramers-Brillouin approximation) that have been tested by comparison with numerical simulations [44,46] and experiments [30] for fixed frequency perturbations localized at the tip.

(i) Periodic forcing by a fixed frequency $\nu$ leads to surface oscillations of amplitude that increases exponentially with the distance from the dendrite tip up to a critical distance $\zeta_c$ proportional with $1/\nu^2$. For larger distances the amplitude decreases and eventually dies away. If, in turn, the distance from the tip is fixed and $\nu$ is varied, a peak is observed in the amplitude.

(ii) The localized wave packets behave differently; they grow exponentially as they move to arbitrarily large distances from the tip, while the respective characteristic wave length increases.

Our simulations are in line with the previous MST and numerical predictions for tip-localized forcing in the following respects:

(a) In the vicinity of the dendrite tip, the amplitude of the mode synchronous with forcing increases roughly exponentially with the distance $\zeta$ from the tip. Owing to the excessive computation time and memory needed, we were unable to study the decay of this mode in detail. Nevertheless, as expected from the MST, at large frequencies [Fig. 6(f) and 19(f)] the synchronous mode can only be recognized in the vicinity of the dendrite tip. Another sign, that accords with the presence of a critical distance $\zeta_c$ beyond which the excited mode decays, is a maximum observed in the amplitude $A_{2\nu}$ of the second harmonic (Fig. 7) in simulations where the forcing spectrum contains $2\nu$ with a significant amplitude.

(b) The amplitude $A_{\nu}$ measured at a fixed distance behind the dendrite tip shows a maximum as a function of the forcing frequency [Fig. 7(a)].

An interesting feature, revealed by our phase-field simulations, is that besides the synchronous mode, the Fourier spectrum of the dendrite width contains the second and third harmonics with perceptible amplitudes. This finding is confirmed by our experiments. We believe that this is the first direct demonstration of such non-linear effects in connection with diffusional instability induced dendrite formation during a first-order phase transformation. It appears, that this non-linear behavior is not a peculiarity of spatially homogeneous forcing. Although not mentioned in the original works, traces of higher harmonics seem to be present in the respective power spectra for local forcing as well [22,45]. It is worth mentioning, furthermore, that a similar behavior has been observed in the case of anomalous Saffman-Taylor fingering [37], suggesting that the appearance of higher harmonic modes in response to periodic external forcing is a common feature of dendrite formation whether driven by diffusional or mechanical instabilities.

Summarizing, in the present state of affairs it seems that the only specific feature of pattern formation under non-local forcing is a simultaneous triggering of side-branches (of first, second, and higher orders) on independent crystallites throughout the sample.
VI. SUMMARY

Our computer simulations and experiments demonstrated that the dendritic morphology can be regularized by non-local time-periodic forcing realized by modulated pressure and Joule heating. These conditions lead to an oscillatory velocity of the dendrite tip, and yield side-branches at regular distances, provided that the frequency of the modulation is close to the “natural” frequency of free side-branch formation. The dynamical response of the system to such non-local forcing can be understood in general on the basis of previous theoretical and experimental results on tip-localized forcing. However, the non-linear effects, such as the appearance of higher harmonic modes, warrant further theoretical work.

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APPENDIX A: PRESSURE DEPENDENCE OF THE PHASE TRANSITION TEMPERATURE

The pressure dependence of the $N \rightarrow S_B$ phase transition temperature has been measured by a procedure similar to that described in \[36\]. For different $p_c$ a single $S_B$ monodomain has been kept at constant size by controlling the temperature for several hours (until the thermodynamic equilibrium state of the system has approached). Fig. 23 shows the change of the phase transition temperature depending on the pressure. Linear fit on the data gives a slope $\frac{dT_m}{dp} = (0.032 \pm 0.003)$ K/bar for the Clapeyron coefficient.

The Clapeyron coefficient can be also calculated from the relation:

$$\frac{dT_m}{dp} = \frac{T_m^0 \Delta V_{N\rightarrow S_B}}{\Delta H}$$

where $T_m^0$, $\Delta V_{N\rightarrow S_B}$ and $\Delta H$ are the phase transition temperature at atmospheric pressure, the molar volume change on transition and the molar latent heat of fusion, respectively. From the relevant material parameters of CCH3, namely $T_m^0 = 329.45$ K \[23\], $m = 233$ (molar mass), $\rho = 895.4$ kg/m$^3$ (density at 80.6 °C \[18\]), $\Delta H = 6247$ J/mol, $\Delta V_{N\rightarrow S_B}/V = -2.6 \%$, and $\alpha_N = 25 \times 10^{-4}$ 1/K (volumetric expansion coefficient in the nematic phase \[19\]) one obtains $\frac{dT_m}{dp} = 0.033$ K/bar in excellent agreement with the experimental value.

Although this value is much larger than for metals, it is not unusual for liquid crystals. For example, $\sim 0.03$ K/bar has been reported for the nematic – crystal transition in PAA, and $\sim 0.1$ K/bar for the nematic smectic transition in p-methoxybenzoic acid \[50\]. Comparable, or even larger coefficients have been measured on other organic substances such as camphene 0.214 K/atm, picolinic acid 0.0674 K/atm, and succinonitrile 0.0245 K/atm.

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FIG. 1. The experimental setup.
FIG. 2. Dendritic patterns of four-fold symmetry predicted by the phase-field theory for $\Delta = 0.6$, $\sigma_4 = -0.12$, $\beta_4 = 0.24$ and $t = 0.22$: (a) Under constant pressure yielding $A = u_m = 0.048$; and (b)–(f) in the presence of square-wave pressure modulations of amplitude $a_0 = 0.16$ and filling coefficient $\xi = 0.3$. The gray level in the liquid phase indicates the temperature field. The white line marks the position, where the width of the dendrite arm was measured.

FIG. 3. Dendritic patterns of six-fold symmetry predicted by the phase-field theory for $\Delta = 0.6$, $\sigma_6 = -0.04$, $\beta_6 = 0.08$ and $t = 0.22$: (a) Under constant pressure yielding $A = 0.048$; and (b)–(f) in the presence of square-wave pressure modulations of amplitude $a_0 = 0.16$ and filling coefficient $\xi = 0.3$. 
FIG. 4. (a) The width of the dendrite $w(t)$ at a distance of $\zeta = 0.75$ behind the dendrite tip without periodic perturbation [heavy solid line; Fig. 2(a)], in the presence of sinusoidal (light solid line) and square-wave [dotted line; Fig. 2(d)]. (b) The respective Fourier spectra $W(f)$.

FIG. 5. Comparison of growth patterns corresponding to (a) sinusoidal, (b) symmetric ($\xi = 0.5$) and (c) asymmetric ($\xi = 0.3$) square-wave forcing of equal period lengths ($\nu = 30$), and period averaged amplitudes. Note the side-branch “doubling” in panel (c). (Other relevant parameters are as for Fig. 3).

FIG. 6. The amplitude of the Fourier peaks $A_\nu$ and $A_{2\nu}$ measured at $\zeta = 0.75$ behind the tip, and the symmetry parameters $a$ and $b$ as a function of the driving frequency $\nu$. Other relevant parameters are $\Delta = 0.55$, $\xi = 0.3$, $\sigma_4 = -0.12$ and $\beta_4 = 0.24$, while $a_0 = 0.14$ (a), and $a_0 = 0.2$ (b).

FIG. 7. The Fourier amplitudes $A_\nu$ (circles) and $A_{2\nu}$ (triangles) as a function of the distance $\zeta$ from the dendrite tip at two undercoolings $\Delta = 0.55$ (full symbols) and 0.6 (empty symbols). Note the roughly exponential behavior of $A_\nu$ and the saturation/maximum in $A_{2\nu}$. Other relevant parameters are $\nu = 60$, $\xi = 0.3$, $a_0 = 0.16$, $\sigma_4 = -0.12$ and $\beta_4 = 0.24$. 
FIG. 8. The amplitude of the Fourier peaks $A_\nu$ and $A_{2\nu}$, and the symmetry parameters $a$ and $b$ as a function of the forcing amplitude $a_0$ for the system shown in Fig. 2(d).

FIG. 9. The effect of undercooling on pattern formation under periodic forcing. (a) $\Delta = 0.65$, (b) $\Delta = 0.5$. Other parameters as for Fig. 2(d) except that $t = 0.2$ and $t = 0.283$ for panels (a) and (b), respectively.

FIG. 10. The amplitude of the Fourier peaks $A_\nu$, $A_{2\nu}$ as a function of undercooling $\Delta$. Other relevant parameters are $\nu = 60$, $\xi = 0.3$, $a_0 = 0.16$, $\sigma_4 = -0.12$ and $\beta_4 = 0.24$.

FIG. 11. Pattern formation under periodic forcing as a function of anisotropy: $\sigma_4 = -0.08$ (a), $\sigma_4 = -0.04$ (b), $\sigma_4 = -0.02$ (c) and $\sigma_4 = -0.01$ (d). Other relevant parameters are $\Delta = 0.55$, $\nu = 60$, $\xi = 0.3$, $a_0 = 0.16$ and $\beta_4 = -2\sigma_4$. The average growth rate varies with anisotropy, therefore, different times are needed to reach comparable sizes: $t = 0.24$ (a), $t = 0.3$ (b), $t = 0.38$ (c), and $t = 0.44$ (d).

FIG. 12. The amplitude of the Fourier peaks $A_\nu$, $A_{2\nu}$, and the symmetry parameters $a$ and $b$ as a function of anisotropy parameter $|\sigma_4|$ of the interfacial free energy. The kinetic anisotropy has been varied proportionally ($\beta_4 = -2\sigma_4$). Other relevant parameters are $\Delta = 0.55$, $\nu = 60$, $\xi = 0.3$ and $a_0 = 0.16$. 

FIG. 12. The amplitude of the Fourier peaks $A_\nu$, $A_{2\nu}$, and the symmetry parameters $a$ and $b$ as a function of anisotropy parameter $|\sigma_4|$ of the interfacial free energy. The kinetic anisotropy has been varied proportionally ($\beta_4 = -2\sigma_4$). Other relevant parameters are $\Delta = 0.55$, $\nu = 60$, $\xi = 0.3$ and $a_0 = 0.16$. 

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FIG. 13. Pattern formation under periodically alternating heating and cooling of amplitudes $b_0 = 8$ (a) and 64 (b). Other relevant parameters are $\Delta = 0.6$, $\nu = 60$, $\xi = 0.5$, $\sigma_4 = -0.12$, $\beta_4 = 0.24$, while the elapsed time $t = 0.22$.

FIG. 14. The average velocity of the tip under periodic heating and cooling (open circles; parameters as for Fig. [13], and for oscillatory pressure (open triangles; calculated with $\langle p \rangle = 0$, $\xi = 0.5$, other parameters as for Fig. 2(d)).

FIG. 15. Pattern contours formed under periodic heat pulses of amplitudes increasing from left to right ($b_0 = 5.7$, $b_0 = 28.6$, and $b_0 = 51.4$). Other relevant parameters are $\Delta = 0.61$, $\nu = 50$, $\xi = 0.15$, $\sigma_4 = -0.16$, $\beta_4 = 0.32$, $h = -14.3$, $\tau_o = 16$ and $\Delta t = 7 \times 10^{-5}$.

FIG. 16. The average velocity $v_o$ (open triangles) and the amplitude of velocity oscillation $v_\nu$ (full circles) as a function of the heating amplitude $b_0$ for simulations presented in Fig. 15.

FIG. 17. The effect of oscillatory pressure on the smectic-B dendrite growing into undercooled nematic phase ($\Delta T = 1.0 \, ^\circ C$). (a) Without pressure oscillations; (b)–(f) with square wave pressure modulation of parameters $p_e = 2$ bar, $\xi = 0.2$, $\nu = 0.75, 0.91, 1.18, 1.64$, and 1.96 Hz, respectively.
FIG. 18. The effect of the oscillatory heating on smectic-B dendrite growing into undercooled nematic phase ($\Delta T = 1.0^\circ$C). (a) No oscillatory heating; (b) $\tilde{\nu} = 0.2$ Hz, $\mathcal{T} = 6 \times 10^{-5}$ W/cm$^2$, $\xi = 0.14$; (c) $\tilde{\nu} = 0.46$ Hz, $\mathcal{T} = 6 \times 10^{-5}$ W/cm$^2$, $\xi = 0.16$; (d) $\tilde{\nu} = 0.81$ Hz, $\mathcal{T} = 1.1 \times 10^{-4}$ W/cm$^2$, $\xi = 0.61$. The two lines in panels (a) and (c) mark the positions, where the width $w$ of the dendrite has been measured as a function of time (for the Fourier transforms see Fig. 19). Black lines in panel (b) denote the position of the tip at the centers of the heating pulses.

FIG. 19. Fourier spectrum $W(f)$ of the half-width of the dendrite measured 66.5 $\mu$m behind the tip as shown in Fig. 18(c). For comparison, the respective spectrum for free dendritic growth [Fig. 18(a)] is also presented (see insert).

FIG. 20. The effect of the amplitude of oscillatory pressure on the smectic-B dendrite growing into undercooled nematic phase ($\Delta T = 1.0^\circ$C). (a) Without pressure oscillations; and with square wave pressure modulation of parameters $\tilde{\nu} = 1.64$ Hz, $\xi = 0.2$, $p_e = 1$ bar (b) and 2 bar (c), respectively.

FIG. 21. Side-branch formation close to the tip under “symmetric” ($\xi = 0.5$) square-wave heating as displayed by a smectic-B dendrite growing into the undercooled nematic phase. The growth pattern (insert) and the Fourier-spectrum $W(f)$ (main frame) are shown. Black lines in the insert denote the forcing frequency $\tilde{\nu}$. The white line marks the position (53.2 $\mu$m behind the tip), where the width $w$ of the dendrite has been measured as a function of time. Note the presence of the mode oscillating with twice the base frequency of forcing; a mode missing from the forcing spectrum. The relevant parameters are: $\tilde{\nu} = 0.35$ Hz and $\mathcal{T} = 9 \times 10^{-5}$ W/cm$^2$. 
FIG. 22. Shape of the dendrite tip. The arrow points at a surface undulation that later develops into a side-branch.

FIG. 23. Pressure dependence of the temperature of the nematic–smectic B phase transition.