

Elastically buckled film-substrate system as a two-dimensional crystalWenqing Zhu (朱文清) ^{*}*Department of Engineering, University of Cambridge, Cambridge CB2 1PZ, United Kingdom*

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Compressive mechanical stress exceeding a critical value leads to the formation of periodic surface buckling patterns in film-substrate systems. A comprehensive understanding of this buckling phenomenon is essential in applications where the surface topologies are modulated to achieve multifunctionalities. Here, we reformulate the elastic theory of a film-substrate system by treating the compliant substrate as a nonlinear elastic solid. The resulting elastic free energy functional of the deflection field is shown to be equivalent to a minimal density functional of the phase-field crystal theory plus a Gaussian curvature-related term. The proposed elastic model constructs a phase diagram based on free energy minimization, quantitatively agreeing with the buckling transitions observed in previous experiments. The emerging hexagonal buckling system is shown to be equivalent to a two-dimensional crystal with proper scalings. We further conducted simulations of repeated buckling under cyclic stress to demonstrate a dynamically modulated structural adhesive, which resembles the physical process of repeated crystallization and melting near a critical temperature.

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Crystalline systems with periodic structures, whether synthetic or formed in nature, have received intense research interest given their simplicity and representativeness [1]. Understanding their formation and evolution has significant implications in diverse fields such as condensed matter physics, engineering sciences, and life sciences [2–4]. Referring to the phase diagram of condensed matter, a disordered structure generally persists at high temperatures. It corresponds to a uniform atomic density distribution on statistical average. When the matter is cooled down to a critical onset temperature, however, it crystallizes to form a periodically ordered structure, minimizing the system free energy. Molecular dynamics simulations of crystallization with accurate descriptions of interatomic potentials have been able to provide details of the relevant thermodynamics and kinetics [5]. To probe the long-time behavior at a larger length scale, continuum models have also been proposed within the framework of dynamical density functional theory [6–9]. These models have been successfully applied to studies of phase stability and defect dynamics such as the grain boundaries in graphene [10], which is a typical two-dimensional (2D) crystal with a hexagonal lattice.

A relevant macroscopic procedure analogous to crystallization is elastic buckling of soft materials [11]. In a stress-free state, an elastic soft plate has a trivial deflection field. By applying compressive stress exceeding a threshold value, it deforms to a buckled state with periodic out-of-plane

deformation. This type of pattern formation is attributed to the system bifurcation into a minimized strain energy state with characteristic modes of out-of-plane deformation [12]. Particularly, Cai *et al.* reported the emergence of a honeycomb hexagonal buckling pattern, similar to 2D graphene lattice, on an elastic film-substrate system under equi-biaxial compressive stress [13]. We argue that the similarity between crystallization and the elastic buckling of this system lies in the fact that they share a common principle of free energy minimization, while rigorous mathematical proof of the equivalence is yet to be given. The significance of the crystallization-buckling equivalence lies in the following aspects. First, a possible mapping from crystallization to elastic buckling implies that designated observations of elastic instability can be carried out on a film-substrate system to understand the thermodynamics of its mapped crystalline system, given proper parameter scaling. In addition, accurate predictions of surface buckling using a rigorous mathematical framework will enable the design of dynamic pattern control, promoting wide applications in soft robotics with adjustable adhesion [14], sensing [15], measurements of elastic properties [16,17], etc.

In this Letter, we show that a buckled elastic film-substrate system is equivalent to a model 2D crystal according to the classical density functional theory developed by Elder *et al.* [6], except that an extra Gaussian curvature energy term must be considered for the elastic system. Examining the free energy of various periodic lattice phases analytically, we find that the Gaussian curvature-related energy contribution vanishes for a system larger than a single unit cell, rendering the buckling phase diagram identical to that predicted by the density functional theory. Model validation and analyses of buckling characteristics are further presented, regarding the phenomenological crystallization temperature, lattice constants, and crystalline defects.

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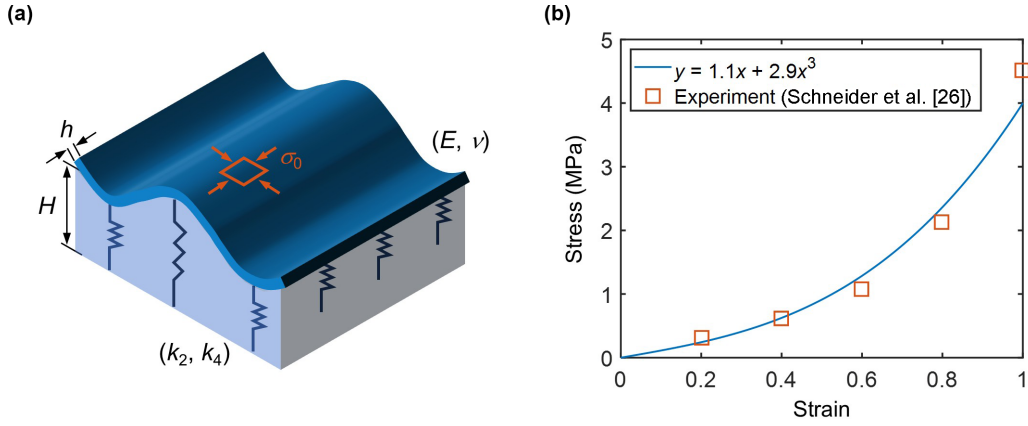


FIG. 1. (a) Schematic of a linear-elastic film bonded on a nonlinear elastic substrate. The film is under equi-biaxial in-plane compression. (b) Spring constants k_2 and k_4 were determined from the experimental stress-strain curve of PDMS (Sylgard 184, Dow Corning) [26].

We start by considering a Föppl-von Kármán film perfectly bonded on a nonlinear elastic substrate. A uniformly distributed equi-biaxial compressive stress σ_0 is applied on the film, as shown in Fig. 1(a). The free energy of this film-substrate system reads $F = F_{\text{stretch}} + F_{\text{bend}} + F_{\text{sub}}$, where the stretching energy of the film [18]

$$F_{\text{stretch}} = - \int d\mathbf{x} [\nabla \cdot \mathbf{u} + |\nabla w|^2] \sigma_0 h / 2, \quad (1)$$

and the bending energy of the film [18]

$$F_{\text{bend}} = \frac{Eh^3}{12(1-\nu^2)} \int d\mathbf{x} [(\nabla^2 w)^2 / 2 - (1-\nu) \det(\nabla \nabla w)]. \quad (2)$$

$\mathbf{u}$ and w are the in-plane and out-of-plane displacement fields. $\nabla = \frac{\partial}{\partial x} \mathbf{e}_x + \frac{\partial}{\partial y} \mathbf{e}_y$ is the in-plane gradient operator. E and ν are Young's modulus and Poisson's ratio of the film; h is the film thickness. Assuming no shear deformation or relative sliding between the film and substrate, the elastic energy stored in the substrate is written as

$$F_{\text{sub}} = \int d\mathbf{x} \left[\frac{k_2}{2} w^2 + \frac{k_4}{4} w^4 \right]. \quad (3)$$

k_2 and k_4 are spring constants. The fourth-order term in F_{sub} is introduced to describe the stiffening of polymeric substrates at large strains (see Fig. 1(b) and the Supplemental Material [19]). Such a form of substrate energy in Eq. (3) has also been adopted by Stoop *et al.* [20], which is a nonlinear version of Winkler's mattress model [21]. In this model, the elastic substrate is represented by a set of parallel springs [22,23]. It is also suitable to describe the nonlinear van der Waals force acting between an elastic film and a rigid substrate [24]. Therefore, the total free energy, scaled by the bending modulus $B = Eh^3/[12(1-\nu^2)]$, reads

$$\tilde{F} = \frac{F}{B} = \int d\tilde{\mathbf{x}} \left[\frac{\lambda}{4} \psi^4 + \frac{1-\varepsilon}{2} \psi^2 - |\tilde{\nabla} \psi|^2 + \frac{1}{2} (\tilde{\nabla}^2 \psi)^2 - (1-\nu) \det(\tilde{\nabla} \tilde{\nabla} \psi) \right], \quad (4)$$

where a characteristic wavenumber $q = \sqrt{6\sigma_0(1-\nu^2)/(Eh^2)}$ is defined, such that $\tilde{\mathbf{x}} = q\mathbf{x}$ and $\psi = qw$. The nondimen-

sional parameters $\lambda = k_4/(Bq^6)$ and $\varepsilon = 1 - k_2/(Bq^4)$. Note that the term involving the $\mathbf{u}$ field is not shown here, since we focus on the variation of the energy functional with respect to w . Also, because a uniform stress σ_0 is prescribed, the variational derivative of the free energy functional without the $\mathbf{u}$ field-related term yields the complete set of equilibrium equations. When stress gradients are present, however, the in-plane deformation needs to be explicitly included. Surprisingly, the expression of the scaled free energy in Eq. (4) is exactly the same as the Swift-Hohenberg (SH) energy functional [25], except that there is an extra Gaussian curvature term $(1-\nu) \det(\tilde{\nabla} \tilde{\nabla} \psi)$. The phase-field crystal (PFC) model adopting the SH functional has been successful in describing crystal nucleation and growth [6,9]. In this PFC model, the scaled free energy functional of atomic density reads

$$F_\rho = \int d\mathbf{x} \left[\frac{\lambda_\rho}{4} \rho^4 + \frac{1-\varepsilon_\rho}{2} \rho^2 - |\nabla \rho|^2 + \frac{1}{2} (\nabla^2 \rho)^2 \right], \quad (5)$$

where λ_ρ and ε_ρ are phenomenological parameters. Note that an increase in ε_ρ reflects a decrease in temperature. The mathematical similarity between Eq. (4) and Eq. (5) suggests that the elastic film-substrate system studied herein can be treated as a model crystal, where the scaled deflection field ψ is equivalent to a time-averaged atomic density field.

To establish the mathematical equivalence, we examine how the Gaussian curvature term in Eq. (4) influences the buckling phase selection. We calculate the scaled free energy densities of four different periodic phases, i.e., the liquid phase (homogeneous phase) $\psi_l = \tilde{\psi}$, the hexagonal lattice phase $\psi_h = \tilde{\psi} + A_h [\cos(q_h \tilde{x}) \cos(q_h \tilde{y} / \sqrt{3}) + \cos(2q_h \tilde{y} / \sqrt{3}) / 2]$, the square lattice phase $\psi_s = \tilde{\psi} + A_s \cos(q_s \tilde{x}) \cos(q_s \tilde{y})$, and the striped phase $\psi_t = \tilde{\psi} + A_t \sin(q_t \tilde{x})$, respectively. In terms of the parameters λ , ε , and $\tilde{\psi}$ (the spatial average of the scaled deflection), we have $f_l = \frac{1-\varepsilon}{2} \tilde{\psi}^2 + \frac{\lambda}{4} \tilde{\psi}^4$, $f_h = \frac{\lambda}{4} \tilde{\psi}^4 + \frac{(1-\varepsilon)}{2} \tilde{\psi}^2 + \frac{3(3\lambda\tilde{\psi}^2-\varepsilon)}{16} A_h^2 + \frac{3\lambda\tilde{\psi}}{16} A_h^3 + \frac{45\lambda}{512} A_h^4$, $f_s = \frac{\lambda}{4} \tilde{\psi}^4 + \frac{(1-\varepsilon)}{2} \tilde{\psi}^2 + \frac{3\lambda\tilde{\psi}^2-\varepsilon}{8} A_s^2 + \frac{9\lambda}{256} A_s^4$, and $f_t = \frac{\lambda}{4} \tilde{\psi}^4 + \frac{(1-\varepsilon)}{2} \tilde{\psi}^2 + \frac{3\lambda\tilde{\psi}^2-\varepsilon}{4} A_t^2 + \frac{3\lambda}{32} A_t^4$, respectively. The amplitudes A_h , A_s , and A_t , are functions of λ , ε , and $\tilde{\psi}$ (see the Supplemental Material for detailed mathematical

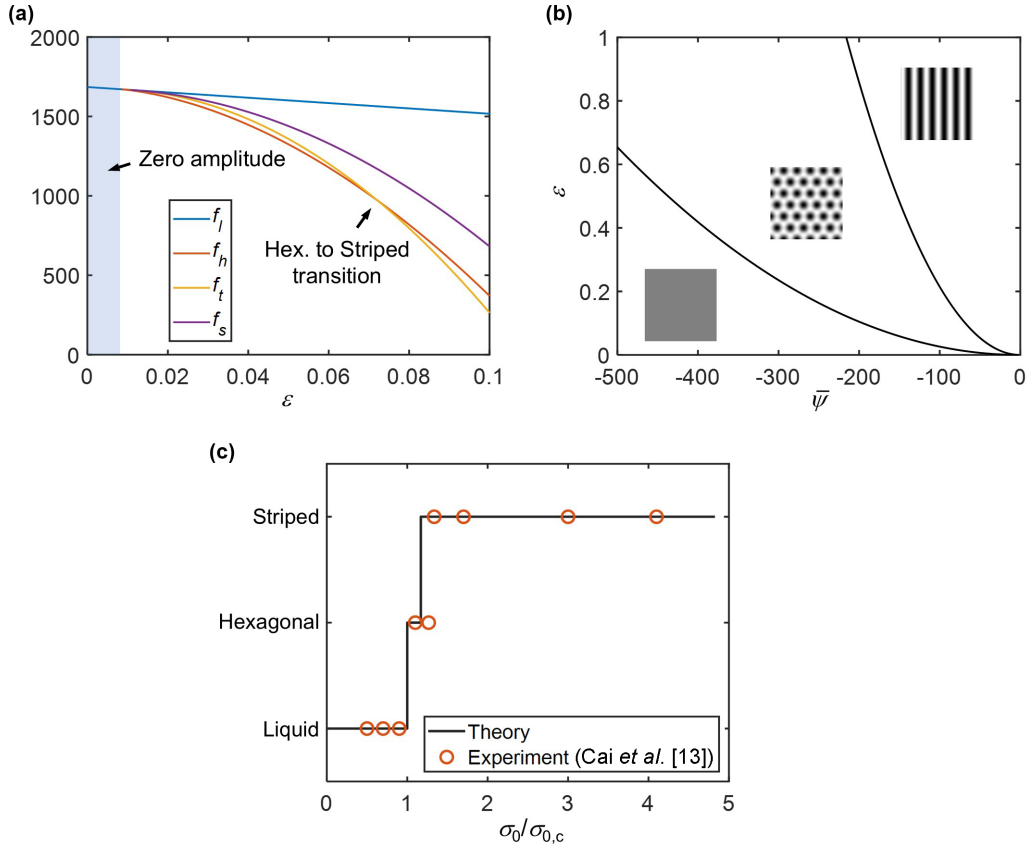


FIG. 2. Phase diagram of the elastic film-substrate system identical to that from the PFC model. (a) Representative free energy densities of different phases with respect to ε . $\bar{\psi} = -58$ and $\lambda = 1.1 \times 10^{-6}$ were used. (b) Phase selection for varying $\bar{\psi}$ and ε at $\lambda = 1.1 \times 10^{-6}$. (c) Phase selection for varying the overstress $\sigma_0/\sigma_{0,c}$ from previous experiment (Ref. [13]) and theoretical prediction.

derivations [19]). The unique third-order term of A_h in f_h is crucial for the stability of the hexagonal phase based on energy minimization. Importantly, it is found that the Gaussian curvature term does not contribute to the total free energy, i.e., $\int \det(\tilde{\nabla} \tilde{\nabla} \psi) d\tilde{\mathbf{x}} = 0$. This ensures that the expressions of free energy densities for all the phases are identical to those derived from the PFC model in 2D [6]. Thus, obtained from the energy minimization principle, the phase diagram of the elastic film-substrate system is the same as that derived from the 2D PFC model with SH functional [Eq. (5)] [6]. It is worth noting that for nonperiodic boundary conditions, such as clamped edges, the Gaussian curvature term does not vanish, and the material is not in an equi-biaxial stress state near the boundary. This can lead to directional striped patterns or herringbone buckling patterns, as observed by Breid *et al.* [27]. Figure 2(a) shows the change in the free energy densities with respect to ε , using $\lambda = 1.1 \times 10^{-6}$ and $\bar{\psi} = -58$ based on the material parameters of the PDMS film-substrate system [19]. Note that ε is related to the applied stress level and serves here as a phenomenological temperature-like parameter, as the corresponding ε_ρ in the 2D PFC model represents the thermodynamic temperature [28]. At high phenomenological temperature (ε is low and σ_0 is small), i.e., the blue region in Fig. 2(a), the system is in the homogeneous liquid phase (buckle-free state), because all the inhomogeneous solid phases have a minimized free energy

at zero amplitude. Decreasing the temperature (increasing ε and σ_0) results in the emergence of a hexagonal lattice phase. Further decreasing the temperature leads to the striped phase, as also implied by the 2D PFC model [6]. Note that as ε changes, the free energy density of the square lattice phase f_s is always higher than those of the above two phases, i.e., f_h and f_t , and thus it is metastable and thermodynamically unfavored. This is also the case throughout the $(\lambda, \varepsilon, \bar{\psi})$ space. A classical two-parameter phase diagram, analogous to those from PFC theories, is constructed in Fig. 2(b). It suggests that increasing the average out-of-plane displacement, as achieved by increasing the out-of-plane surface pressure, can suppress the system buckling. The theoretical ε - $\bar{\psi}$ phase diagram can be converted into a single-variable (σ_0) controlled curve while fixing the material parameters [Fig. 2(c)]. As the compressive stress σ_0 increases to the critical value $\sigma_{0,c}$, a hexagonal buckling pattern emerges. Further increasing σ_0 to $\sigma_{0,c}^{\text{Striped}} \approx 1.17\sigma_{0,c}$ gives the striped buckling pattern formation. Previous theoretical analysis of elastic buckling predicted the formation of a checkerboard square lattice phase in a film-substrate system at a small overstress under equi-biaxial stress, which contradicts the experimental observations [13,27]. In comparison, the present theory with a generalized SH functional successfully predicts the hexagonal phase over the metastable square lattice phase, agreeing with the experiment [13,27]. It is worth emphasizing that the

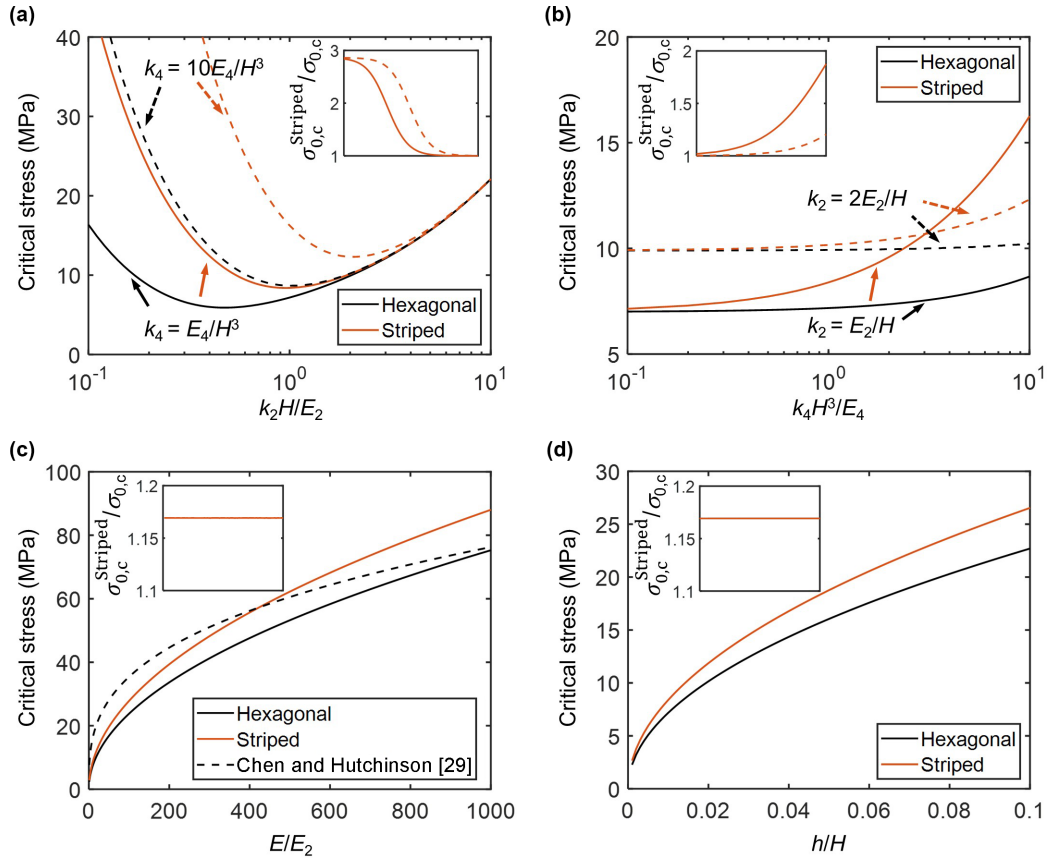


FIG. 3. (a), (b) Critical compressive stresses for the formation of hexagonal ($\sigma_{0,c}$) and striped ($\sigma_{0,c}^{\text{Striped}}$) phases vs the normalized substrate spring constants k_2H/E_2 and k_4H^3/E_4 , with the film parameters $E = 10$ GPa, $\nu = 0.5$ and $h = 0.01H$. (c), (d) Critical compressive stresses vs E/E_2 and h/H . Dashed line is the prediction from the elastic buckling theory (Ref. [29]). Insets show the ratio of critical stresses.

nonlinear elasticity of the compliant substrate is crucial for the selection of the hexagonal phase within the proposed framework. We provide analysis of the effect of the substrate stiffness on the critical stresses in Fig. 3. As k_2 increases, $\sigma_{0,c}^{\text{Striped}}/\sigma_{0,c}$ tends to 1, and the regime of the hexagonal lattice vanishes [Fig. 3(a)]. The hexagonal lattice phase also vanishes when k_4 decreases and tends to zero [Fig. 3(b)]. In the case of linear elasticity ($k_4 = 0$), local energy minimum with respect to the amplitude does not exist for all the solid phases, such that the phase selection is undetermined by energy minimization. The absence of nonlinear elasticity in previous continuum mechanics models may also account for the discrepancy between the model predictions and the experiment. We further plot the critical stresses of elastic buckling as a function of the film modulus and thickness in Figs. 3(c) and 3(d), respectively. The critical stress for the onset of the hexagonal phase is shown to increase with increasing the film-substrate modulus ratio E/E_2 (with fixed $h = 0.01H$) and the thickness ratio h/H (with fixed $E = 10$ GPa). E_2 is the second-order elastic modulus of the substrate. This is also rationalized by the analytical solution of elastic buckling in 1D [19]. Linear perturbation analysis by Chen and Hutchinson [29] gave the same trend, although the derived scaling law is different. Nevertheless, the present theory suggests that the ratio of the critical stress of the

striped phase to that of the hexagonal phase, $\sigma_{0,c}^{\text{Striped}}/\sigma_{0,c}$, remains a constant for a variety of thin film materials with different thickness and elastic moduli. This criterion can guide the controllable formation of hexagonal wrinkles in different film-substrate systems. Discussions on the potential applications of controllable pattern formation will be given later in the main text.

Besides the phase diagram and the critical stress, it is necessary to delve into the structural characteristics of the buckling patterns. First, the theoretical model is validated regarding the postbuckling amplitude and wavelength. Numerical simulations of elastic buckling are carried out based on the free energy functional [Eq. (4)] and the PFC model. For comparison, simulations are also performed using the Fourier spectral method (FSM) [19]. As shown in Fig. 4, the postbuckling amplitude and the wavelength predicted by the present model agree well with the simulation results using the PFC model and FSM. As the applied compressive stress increases, the amplitude increases while the wavelength decreases, with jumps occurred at the liquid-hexagonal lattice transition as well as the hexagonal lattice-to-striped phase transition. Next, we focus on the regime of hexagonal lattice analogous to a 2D crystalline system. Temporal evolutions of the ψ field are monitored at different overstresses (different phenomenological temperatures). At a stress slightly higher than the critical

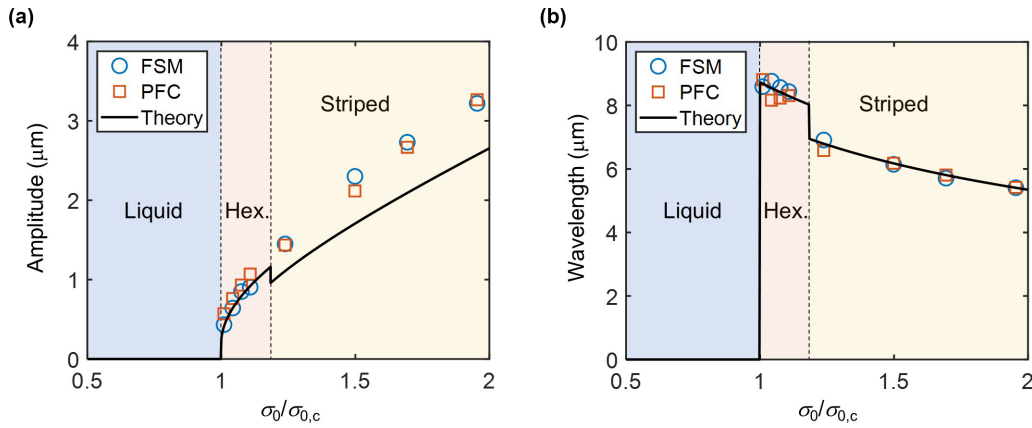


FIG. 4. Buckling pattern characteristics from the theory and the simulations using Fourier spectral method and PFC model. (a) Postbuckling amplitude vs $\sigma_0/\sigma_{0,c}$. (b) Postbuckling wavelength vs $\sigma_0/\sigma_{0,c}$.

value, i.e., $\sigma_0/\sigma_{0,c} = 1.01$ (temperature slightly below the solidification temperature), a defect-free hexagonal lattice forms with uniform orientation [Fig. 4(a)]. The corresponding fast Fourier transform (FFT) mapping of the entire domain also supports the hexagonal ordering [Fig. 5(c)]. In comparison, increasing the overstress ($\sigma_0/\sigma_{0,c} = 1.05$) leads to a polycrystalline morphology with grain boundary defects [Fig. 5(b)]. This aligns with the experimental observation of a polycrystalline buckling pattern at a deep buckling stage ($\sigma_0/\sigma_{0,c} \approx 1.3$), as shown in Fig. 5(d). The stress-dependent buckling pattern formation manifests an equivalent temperature-dependent solidification process. When a system is cooled down deeply to a low temperature, a large number of precursor crystal nuclei emerge and grow, as the crystallization condition is satisfied

throughout the system. Since the nuclei orientations generally exhibit a random distribution, it facilitates a polycrystalline structure with random orientation that forms after grain coarsening. Moreover, we evaluate the lattice constant, which is defined as $\sqrt{3}$ times the side length of the hexagon. Without any adjustable parameter, the simulated lattice constant is in excellent agreement with the experiment, as can be seen from the matching peak position of the FFT intensity [Fig. 5(e)]. In fact, the proposed theory also gives a consistent prediction: Energy minimization of a hexagonal lattice based on Eq. (4) gives $q_h = \sqrt{3}/2$ [19]. Thus, the lattice constant equals $2\pi/(q_h q) \approx 51 \mu\text{m}$, given that the characteristic wavenumber $q \approx 0.14 \mu\text{m}^{-1}$ for $\sigma_{0,c} < \sigma_0 < \sigma_{0,c}^{\text{Striped}}$.

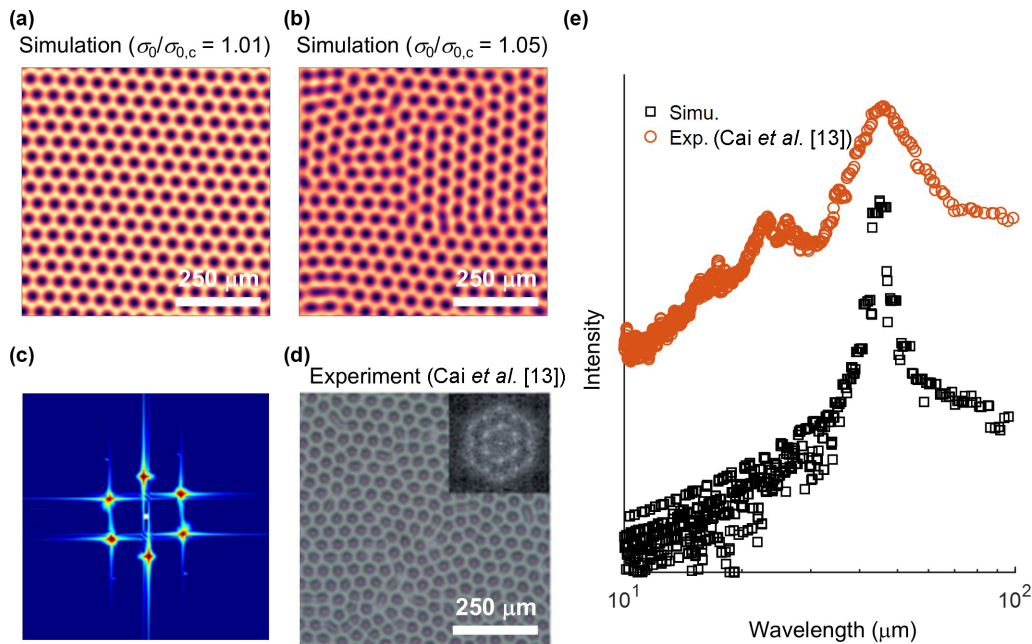


FIG. 5. Simulations of hexagonal pattern formation under constant compressive stress. (a), (b) ψ field at the overstress $\sigma_0/\sigma_{0,c} = 1.01$ and 1.05, respectively. (c) The corresponding FFT of the ψ field at $\sigma_0/\sigma_{0,c} = 1.01$. (d) Experimental buckling pattern adopted from previous study (Ref. [13]). Inset shows the corresponding FFT. Reproduced with permission from Ref. [13]. (e) Intensity of the FFT vs the wavelength. The characteristic wavelength (lattice constant) $\approx 45 \mu\text{m}$ of the hexagonal buckling pattern is identified as the peak position.

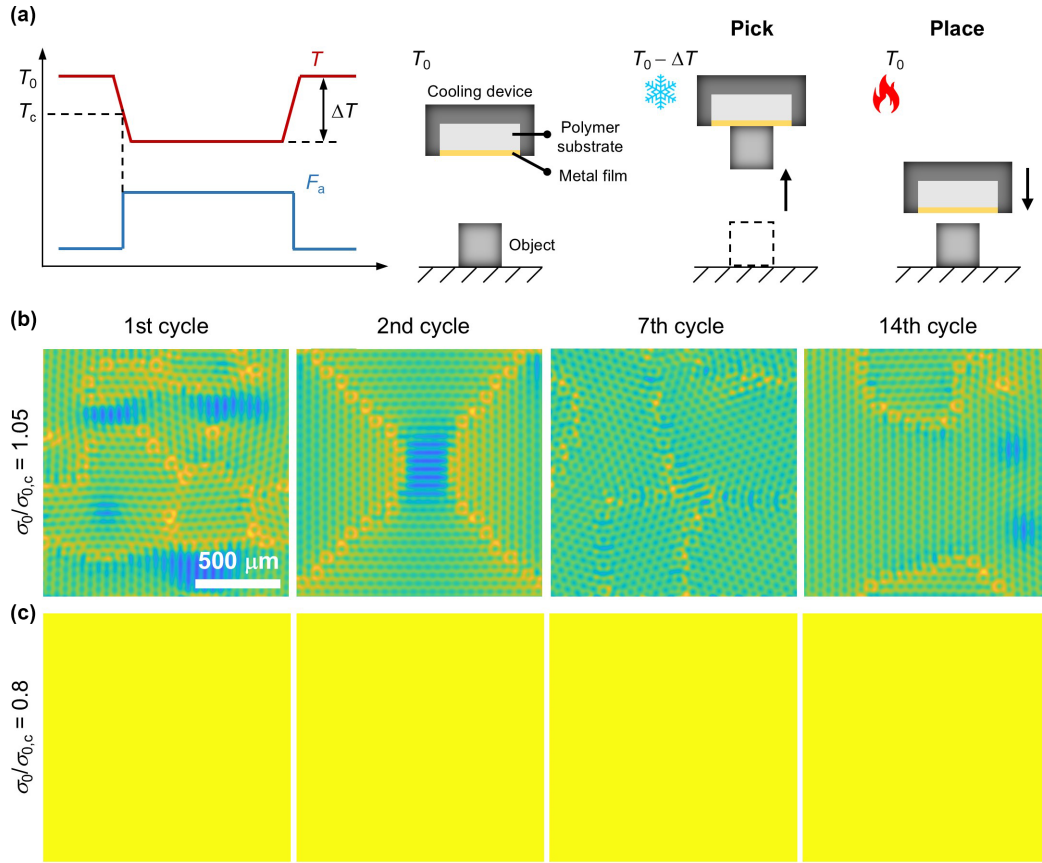


FIG. 6. Cyclic buckling (crystallization) for smart adhesive applications. (a) Schematic of pick-and-place manipulation with the adhesive force modulated by temperature. (b), (c) Smoothed free energy density maps at $\sigma_0/\sigma_{0,c} = 1.05$ and 0.8 for different loading cycles showing grain boundaries of the 2D crystal.

The buckling behavior resembling the crystallization of a hexagonal lattice is crucial for modulating the surface properties of the film-substrate system. Here, we propose a design of a metasurface with tunable adhesive ability via controlling the magnitude of equi-biaxial compressive stress in the elastic film. This can be achieved by introducing cooling-induced thermal strain [Fig. 6(a)]. Given the temperature drop ΔT , the compressive stress $\sigma_0 = (\alpha_{\text{sub}} - \alpha_f)\Delta T/E$; α_f and α_{sub} are the thermal expansion coefficients of the film and substrate, respectively. Inputting $\alpha_f = 10^{-5} - 10^{-4} \text{ K}^{-1}$ for metal films and $\alpha_{\text{sub}} = 2.5 \times 10^{-4} \text{ K}^{-1}$ for PDMS [30], the compressive stress is around 20 MPa using a small temperature drop of 1 K, suitable to induce surface buckling. In a pick-and-place task of object movement, a hexagonal surface buckling pattern can significantly enhance the surface adhesive force F_a , as inspired by tree frog's adhesive pads [31], enabling the lifting of an object. When returned to room temperature conditions, the buckling pattern vanishes and F_a decreases, releasing the object. We conduct finite element simulations of adhesion using a bilinear cohesive law. It shows that increasing the postbuckling amplitude gives an enhanced adhesive force, which can be up to 10 times higher than that of an unbuckled surface (Fig. S2 within the Supplemental Material [19]). Smart adhesion has also been achieved by heating to modify the polymeric material properties [32,33], but it requires a larger temperature change to exceed the glass transition temperature, which is not energy cost-effective. The design proposed herein requires

a small temperature variation and does not introduce large hysteresis among pick-and-place cycling. We perform simulations of repeated buckling with cyclic compressive stress. During each cycle, when the stress magnitude exceeds the critical value, a polycrystalline pattern emerges [Fig. 6(b)]. When the stress magnitude is below the critical stress, an unbuckled liquid state is retrieved [Fig. 6(c)]. Note that the emerging polycrystalline structure at each cycle varies, having entirely different lattice orientation and grain boundary defects. It suggests the absence of a memory effect for the unbuckled-buckled cycling, which is also the case for melting and recrystallization of a solid. Finally, it is worth stating that the physical temperature drop in the film-substrate system indeed leads to crystallization, in analogy to crystallization by cooling from high temperature in a condensed matter system, despite that they hold different temperature scaling and critical onset temperatures.

Besides the nonlinear elasticity discussed in the present study, tension-compression asymmetry of the substrate can also trigger the formation of a hexagonal lattice phase, as revealed by Huang *et al.* [22]. Curvature of the substrate surface also influences the buckling pattern selection. It has been shown that a hexagonal lattice phase emerges at large curvature values of a spherical surface [20]. Moreover, Zhang *et al.* found that hexagonal buckling patterns can form in a torus by introducing a stiff core [34]. In the future, it will be interesting to explore the correlation between curvature-induced elastic

buckling and crystal growth on a curved surface, where topological defects exist because of geometric frustration [35,36].

In summary, we prove that an equi-biaxially compressed elastic film-substrate system is equivalent to a model 2D crystal and exhibits a standard phase diagram predicted by dynamical density functional theory. The proposed theoretical model clarifies the selection of hexagonal buckled state at a small overstress, which could not be predicted by linear perturbation analysis. Importantly, the elastic film-substrate system can serve as a model crystal for understanding history-dependent structural evolution. Using time-resolved and nondestructive optical approaches [37], it is expected to uncover detailed crystallization processes and defect dy-

namics. This is somehow more challenging to be revealed by *in situ* observation of crystalline materials under electron microscopy, given the ultrashort timescale of atomic motion and influence of electron irradiation. These findings provide deeper insight into elastic instability and offer a pathway to establishing a unified physical interpretation between crystallization and diverse phenomena of pattern formation such as Turing morphogenesis and hydrodynamic convection [12].

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Data availability. The data that support the findings of this study are available from the authors upon reasonable request.

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