Stress driven fractionalization of vacancies in regular packings of elastic particles

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Elucidating the interplay of defect and stress at the microscopic level is a fundamental physical problem that has strong connection with materials science. Here, based on the two-dimensional crystal model, we show that the instability mode of vacancies with varying size and morphology conforms to a common scenario. A vacancy under compression is fissioned into a pair of dislocations that glide and vanish at the boundary. This neat process is triggered by the local shear stress around the vacancy. The remarkable fractionalization of vacancies creates rich modes of interaction between vacancies and other topological defects, and provides a new dimension for mechanical engineering of defects in extensive crystalline structures.

I. INTRODUCTION

Topological defects are emergent structures commonly seen in various ordered condensed media [1–5]. As a fundamentally important crystallographic defect, vacancies are highly involved in several important physical processes in both two- and three-dimensional systems, such as in facilitating migration of atoms, and crystallization of DNA-programmable nanoparticles, and in resolving geometric frustrations in curved crystals. Vacancies, together with other defects, are crucial for the characteristic of heterogeneous stress distributions in the packing of particles, which has connections with the formation of force chain structures and the resulting jamming transition in granular media. Understanding the interplay of vacancy and stress yields insights into a variety of crystallographic phenomena. While these problems have been extensively studied from the statistical perspective, the microscopic behavior of vacancies in stressed particle arrays is still not clear. Furthermore, vacancies in particulate systems tend to suppress the relaxation towards the densest state. As such, elucidating the microscopic evolution of vacancies under stress provides a dynamical perspective on the classical close packing problem, which is a fruitful fundamental problem with extensive applications in multiple fields.

The goal of this work is to explore the instability mode in the mechanical behaviors of vacancies under compression, and to reveal the underlying microscopic mechanism. To this end, we employ a two-dimensional model system consisting of elastic particles of uniform size, and resort to the combination of theoretical analysis and numerical experiment. This model system provides the opportunity to address a host of conceptual questions regarding the interplay of vacancy and stress, such as: How and under which condition does a vacancy become unstable? Do vacancies of varying size and morphology conform to a common instability mode? What is the microscopic mechanism of vacancy instability? Illustrating these questions lays a theoretical foundation for the strategy of vacancy based stress engineering in extensive crystalline structures. Note that while vacancies in regular crystals at non-zero temperature equilibrium could exhibit rich physics, this work focuses on the purely mechanical behavior of vacancies.

In this work, we first show the concentration of stress around the vacancy by analytical elasticity analysis. By Delaunay triangulation, we further reveal the underlying topological defect motif of the vacancy. A vacancy consists of a pair of dislocations. The combination of the concentrated local stress and the unique topological structure of the vacancy constitutes the key element for the instability of the vacancy structure. By numerical experiments, we observe the remarkable fission of the vacancy into a pair of dislocations under compression. The dislocations glide along opposite directions and vanish at the boundary. This neat process, which is triggered by the local shear stress around the vacancy, corresponds to a highly coordinated movement of particle arrays. Similar fission process is also observed for two- and three-point vacancies, and in systems consisting of stiffer particles. All these observations boil down to a common scenario of stress driven fractionalization of vacancies. This discovery advances our understanding on the interplay of defect and stress at the microscopic level in crystalline structures.

II. MODEL AND METHOD

Our model consists of elastic particles with regular arrangement confined in the two-dimensional box as shown in Fig. 1. The regular particle array in the simulation box consists of $n$ layers. The number of particles in each layer alternates between $m$ and $m−1$. In the initial state, the particle-particle and particle-wall are in tangential contact, and the system is thus initially stress-free. The system is subject to compression through a piston, which creates a uniform stress field in the triangular lattice of particles free of vacancies. An $n$-point vacancy is cre-

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pressed by removing \( n \) particles in the triangular lattice. In the background of such a uniform, initially stress-free setting, the effect of the vacancy on the stress pattern becomes prominent. We note that in real systems the particles are generally not of uniform size. Size polydispersity brings randomness, which could lead to non-linear elastic and dynamical behaviors, unique mechanical properties, and proliferation of topological defects. While rich physics is underlying the mixture of vacancies and size polydispersity, here we focus on the case of uniform particle size to reveal the microscopic behaviors of vacancies under compression. To highlight the effect of compression, the system is also free of friction and gravity.

The particle-particle and particle-wall interactions are modelled by the Hertzian potential. Specifically, the particle-particle interaction potential is

\[
V_{pp}(r) = \begin{cases} 
\frac{2\sqrt{2}}{3E}(2R - r)^{3/2} & r \leq 2R \\
0 & r > 2R,
\end{cases}
\]

where \( r \) the distance between the centers of the particles, and \( R \) is the radius of the particle. \( D = 3(1 - \nu^2)/(2E) \), where \( E \) is the Young’s modulus, and \( \nu \) is the Poisson ratio. The interaction potential between the particle and the rigid wall is

\[
V_{pw}(r) = \begin{cases} 
\frac{2\sqrt{2}}{3E}(R - r)^{3/2} & r \leq R \\
0 & r > R,
\end{cases}
\]

where \( r \) the distance from the center of the particle to the wall, and \( D' = D/2 \). The Hertzian model has been used to investigate phase transitions in soft particle arrays on interfaces and to reveal the screening effect of topological defect in polydispersity phenomena.

We employ continuum elasticity theory to analyze the linear mechanical response of the system to small compression, and resort to numerical approach based on the steepest descend method to investigate the nonlinear microscopic behaviors of the particle array under large compression. In our numerical experiment, the piston is pushed downward by a small displacement at each time, and the system is then relaxed by updating the particle configuration towards the lowest energy state. As such, the system evolves along a quasi-static process. The variation of the height of the system is controlled within 1% in each push of the piston. We employ the steepest descend method to determine the lowest energy particle configuration under the given compression; the particle positions are updated collectively and individually in sequence to reduce the energy of the system. The typical step size is \( s = 10^{-3}R \).

As the termination condition, the magnitude of the total force on each particle in the final state is required to be sufficiently small. Specifically, upon each small push of the piston, we track the variation of the maximum magnitude of the total force \( f_{\text{max}} \) on the particles in the relaxation process. Typically, the evolution of particle configuration is terminated when \( f_{\text{max}} \) reduces to about 0.1% of its original value. Simulation tests show that this level of precision is sufficient for our problem.

### III. RESULTS AND DISCUSSION

#### A. Stress analysis of vacancy structure

We first analyze the mechanical response of the particle array to small compression by continuum elasticity theory. The particulate system is approximated by a two-dimensional isotropic continuum elastic medium of Young’s modulus \( E \), Poisson’s ratio \( \nu \), and shear modulus \( \mu = E/(2(1+\nu)) \). The in-plane stress distribution is governed by the stress balance equation:

\[
\partial_i \sigma_{ij} = 0,
\]

where \( i, j = x, y \). The system is subject to the compression force \( P \) per unit length on the piston and the shear-free boundary conditions on the three walls. We thus have \( \sigma_{xx} = -\nu P \), \( \sigma_{yy} = -P \), and \( \sigma_{xy} = \sigma_{yx} = 0 \). The corresponding strain field is: \( u_{xx} = u_{xy} = u_{yx} = 0 \), \( u_{yy} = -P(1 - \nu^2)/E \). Here, the Poisson’s ratio \( \nu \) enters the solution through the boundary condition. In general, the stress distribution in an elastic plate subject to given forces at its edges is independent of the elastic constants of the material.

How will a vacancy influence the uniform stress field in the particle array under compression? To address this question, we consider the general case that the two-dimensional continuum elastic medium with a circular hole of radius \( a \) is horizontally compressed by the force \( P_1 \) per unit length, and vertically compressed by the force \( P_2 \) per unit length. By linear superposition of these two forces, the equilibrium stress field can be derived from the stress balance equation. The expressions for
FIG. 2: Fractionalization process of a one-point vacancy into a pair of dislocations under external strain $\epsilon_y$. A dislocation is a dipole of five- and seven-fold disclinations. The Delaunay triangulation and elastic energy distribution are presented in the upper and lower figures, respectively. The red and blue dots represent five- and seven-fold disclinations. The green dots at the boundary represent four-fold disclinations. Brighter particles have larger elastic energy. The solid (yellow) and dashed (blue) arrows in (g) and (h) indicate the collective movement of the particle arrays underlying the fractionalization process. The red arrow in (d) and (i) indicate the location of the square lattice belt that is geometrically incompatible with the triangular lattice.

To characterize the discrete structure of the vacancy that is absent in the preceding elasticity analysis, we perform a Delaunay triangulation of the particles, and reveal a pair of dislocations as the underlying topological structure of a compressed vacancy as shown in Fig. 2(a). A dislocation consists of a pair of five- and seven-fold disclinations, which are indicated by the red and blue dots in Fig. 2. As the elementary topological defect in two-dimensional triangular lattice, an $n$-fold disclination refers to a vertex whose coordination number $n$ deviates from six, and it carries a topological charge $q = (6 - n)\pi/3$. Elasticity theory shows attraction between oppositely charged disclinations and repulsion between like-charge disclinations, which is in analogy with electric charges [43]. A dislocation is thus analogous to an electric dipole. The pair of dislocations in Fig. 2(a) are associated with two Burgers vectors of opposite signs; the Burgers vectors are perpendicular to the line connecting the five- and seven-fold disclinations [43]. Note that in a perfect triangular lattice, a vacancy is represented by a ring of three dislocations [12]. A slight compression leads to the annihilation of a dislocation; the $C_6$ symmetry is broken, but the total topological charge is invariant in this process [1, 12]. The revealed underlying topological defect structure of the vacancy provides the conceptual foundation for the stability analysis of the vacancy under compression. It is natural to ask if the dislocation pair associated with the vacancy will be separated under the external compression.

To address this question, we shall analyze the Peach-Koehler force acting on a dislocation by the external stress field are presented below:

$$\sigma_{rr}(r, \theta) = -\frac{1}{2}(P_1 + P_2)(1 - \frac{a^2}{r^2}) - \frac{1}{2}(P_1 - P_2) \times \left(1 + \frac{3a^4}{r^4} - \frac{4a^2}{r^2}\right) \cos 2\theta, \quad (2)$$

$$\sigma_{\theta\theta}(r, \theta) = -\frac{1}{2}(P_1 + P_2)(1 + \frac{a^2}{r^2}) + \frac{1}{2}(P_1 - P_2) \times \left(1 + \frac{3a^4}{r^4}\right) \cos 2\theta, \quad (3)$$

$$\sigma_{r\theta}(r, \theta) = \frac{1}{2}(P_1 - P_2) \left(1 - \frac{3a^4}{r^4} + \frac{2a^2}{r^2}\right) \sin 2\theta. \quad (4)$$

where $(r, \theta)$ are the polar coordinates with the origin at the center of the hole structure. Now, we apply Eqs. (2) - (4) to our system by setting $P_1 = \nu P$ and $P_2 = P$. The nonvanishing $\sigma_{r\theta}$-component indicates that the hole structure is under a shear stress field. The shear stress could lead to the glide of dislocation, and it is related to the fractionalization of the vacancy, which will be discussed later. Furthermore, analysis of Eq. (3) shows that the azimuthal stress $\sigma_{\theta\theta}$ at $\theta = 0$ and $\pi$ around the vacancy is amplified by a factor of $3 - \nu$ in comparison with the vacancy free case. The concentration of stress may lead to morphological instability of the vacancy.
stress field \( \sigma_{ij}^{(e)} \) [14]:
\[
f_i = -\epsilon_{ij} \sigma_{lm}^{(e)} b_m,
\]
where \( \epsilon_{ij} \) is the anti-symmetric tensor, and \( \vec{b} \) is the Burgers vector. Note that the assumption underlying Eq. (5) is that the external stress field is unaffected by the presence of the dislocation, which resembles the case of a point charge in an external electric field. The parallel component of the Peach-Koehler force along the line of the Burgers vector drives the glide motion of the dislocation [11, 40, 45]. Dislocation glide occurs at a much lower stress than that for the perpendicular motion of the dislocation. The latter kind of motion is known as dislocation climb, and it involves the addition or reduction of material. As such, glide is much easier than climb for a dislocation under stress. For convenience, we set the direction of the Burgers vector \( \vec{b} \) as the \( x' \)-axis, and introduce the \( y' \)-axis by counterclockwise rotation of the \( x' \)-axis by \( \pi/2 \). For example, in the case of Fig. 2(a), the Burgers vector \( \vec{b} \) of the upper dislocation, which is perpendicular to the line connecting the five- and seven-fold disclinations, makes an angle of \( \pi/3 \) with respect to the horizontal line (the \( x \)-axis).

We analyze Eq. (5) in the \( (x', y') \) coordinates. Since \( m = x' \) and \( \epsilon_{ij} \) is the anti-symmetric tensor, it is required that \( \sigma_{ij}^{(e)} \neq 0 \) to ensure that \( f_{x'} \), the component of the Peach-Koehler force in the \( x' \)-axis, is nonzero. According to the previously solved stress field \( \sigma_{ij} \) \( (i, j = x, y) \) under the compression \( \vec{P} \), we derive for the stress field in the \( (x', y') \) coordinates: \( \sigma_{x'y'} = -(3 + \nu)P/4, \sigma_{y'y'} = -(3\nu + 1)P/4, \sigma_{x'y'} = \sigma_{y'x'} = \sqrt{3}(\nu - 1)P/4 \). We see that the external compression produces the nonzero component \( \sigma_{y'y'} \). In combination with the analysis of Eq. (5), we conclude that the compression \( \vec{P} \) provides the driving force for the glide of the pair of dislocations in Fig. 2(a) along opposite directions; the Peach-Koehler forces on the pair of dislocations have opposite signs. As such, a vacancy under compression that is composed of a dislocation pair carries the element of self-destruction upon its creation. We resort to numerical approach to determine the critical strain at which the fission of the vacancy occurs. Note that the presence of friction and particle bonding may suppress the occurrence of fractionalization.

B. Fractionalization of vacancies

In Fig. 2 we show the morphological transformation of the vacancy with the increase of the external strain \( \epsilon_y \). A vacancy under compression is represented by a pair of dislocations; each dislocation consists of a five- and seven-fold disclinations, as indicated by the red and blue dots in Fig. 2(a). Figure 2(b) shows that fractionalization of the vacancy occurs when \( \epsilon_y \) reaches about 6%. Under stronger compression, the dislocations continuously glide and ultimately vanish at the boundary in response to a small change of the external strain \( \epsilon_y \).

Notably, this instability mode persists as the system size and the vacancy location are changed.

In Fig. 2(d), we notice the appearance of a square lattice belt indicated by the red arrow. This geometrically incompatible structure embedded in the triangular lattice is eliminated by a further increase of the strain, leaving out a perfect triangular lattice as shown in Fig. 2(c). We also notice that the number of layers decreases from 19 to 18 from Fig. 2(c) to Fig. 2(d). This layering transition, which is triggered mechanically, occurs with the vanishing of the dislocations at the boundary; the number of layers is invariant as vacancy fractionalization occurs from Fig. 2(a) to Fig. 2(c). Layering transition driven by thermal fluctuations is commonly seen in confined solids of hard spheres and colloidal particles where there are no vacancies [46–51]. As such, the presence of vacancies is not a necessary condition for the occurrence of layering transition. Both vacancy fractionalization and layering transition require sufficiently high strain. In connection with real systems, high strain may be realized in a series of deformable mesoparticle systems like ultrasmall colloids and hydrogel beads: both soft interaction and particle size can be engineered in these soft particle systems [52–54].

To rationalize the numerically observed fractionalization of the vacancy, we present analytical analysis based on the concepts of resolved shear stress and passing stress [45, 55]. Resolved shear stress refers to the component of the stress in the glide direction of a dislocation. The resolved shear stress to split a dislocation dipole of separation \( h \) is referred to the passing stress. By linear elasticity theory, the passing stress is \( \tau_P = \mu b/(8\pi(1 - \nu)h) \), where \( \mu \) and \( \nu \) are the shear modulus and Poisson ratio, and \( b = |\vec{b}| \) [55]. From the previously solved stress field \( \sigma_{ij} \) \( (i, j = x, y) \), we compute for the magnitude of the resolved shear stress as \( |\tau| = \sqrt{3}(1 - \nu)P/4 \). Inserting the critical value for \( P \) to the previously derived expression for the strain \( \epsilon_y \), and with \( h = b \) and \( \nu = 1/3 \) (for 2D triangular lattice com-
posed of linear springs), we estimate the passing strain $u_{yy,P}$ as

$$u_{yy,P} = \frac{1}{4\sqrt{3}\pi} \frac{1}{1-\nu} = \frac{\sqrt{3}}{8\pi} \approx 6.89\%,$$  \hspace{1cm} (6)$$

which is close to the numerically observed critical strain (6%).

Here, we shall emphasize that the observed instability mode of the vacancy is specific to two-dimensional systems. It is of interest to investigate the transformation of vacancies under compression in three-dimensional systems, which is beyond the scope of this work. The stress driven fractionalization of the vacancy is fundamentally different from the thermally driven diffusive motion of the vacancy as a whole in a two-dimensional Lennard-Jones crystal; the integrity of the vacancy is well preserved and no fission occurs in the latter case [12]. Curvature driven fractionalization of interstitial in two-dimensional crystal lattice has also been reported [56, 57]. Here, fractionalization occurs on vacancies and is driven by external stress, which enlarges the space of manipulation in stress and defect engineering [32].

From the distribution of the strain energy in Fig. 2(f)-(h), where the brighter particles have larger elastic energy, we see that the fractionalization process of the vacancy is realized by the highly coordinated movement of particle arrays as indicated by the solid (yellow) and dashed (blue) arrows. The vacancy driven heterogeneous stress distribution also demonstrates the intrinsic connection between defects and the formation of force chains in granular matter [19]. In the rearrangement of the particles around the vacancy, the total energy of the system experiences an abrupt reduction, as shown in Fig. 3(a). Figure 3(a) also shows a significant stiffening of the system with the disappearance of a single one-point vacancy. The coefficient of the quadratic term of the red fitting curve, which corresponds to the mechanical rigidity of the system, increases by as large as 70% in comparison with that of the blue fitting curve. In Fig. 3(b), we show that the magnitude of the energy gap increases with the size of the system.

We proceed to discuss the microscopic evolution of two- and three-point vacancies under compression. The main results are presented in Fig. 4. Like the case of one-point vacancy in Fig. 2, both two- and three-point vacancies are uniformly fissioned to a single pair of dislocations under compression regardless of their morphology. Figure 4 shows that the disclinations (the colors dots) around the vacancies experience a series of reconfiguration and annihilation events, and converge to a single pair of dislocations prior to fission. Simulations show that two- and three-point vacancies with varying orientations and morphologies conform to the same fractionalization scenario as in Fig. 4. Notably, the critical strain for the fractionalization of the two- and three-point vacancies is reduced to about 4.8%, which is much lower than the value of 6% for the one-point vacancy case. We further investigate the microscopic behavior of multiple one-point vacancies under compression. Figure 5 shows the consecutive fractionalizations of the vacancies into pairs of dislocations that ultimately vanish at the boundary. The critical strain reduces to about 4%. We also place the vacancy at different layers and still observe vacancy fractionalization. To conclude, the instability mode of vacancies of varying size and morphology conforms to a common fractionalization scenario.

C. Effects of particle stiffness, system size and simulation parameters

Finally, we briefly discuss the effect of particle stiffness on the instability of vacancies, and the dependence of the critical strain on system size and relevant simulation parameters.

The phenomenon of vacancy fractionalization is also found in systems consisting of stiffer particles. By increasing the power of 5/2 in the expressions for $V_{pp}(r)$ and $V_{pp}(r)$ to 7/2, the critical strain for the fractionalization of the vacancy increases accordingly from about 6% to 10%. The increase of the critical strain can be attributed to the enhanced binding energy of the dislocation pair that is proportional to the Young’s modulus [1]. The stiffening of the particles leads to the enhanced Young’s modulus and thus the increase of the binding energy of the dislocation pair. Stronger external stress is therefore required to pull a dislocation pair apart.

Regarding the dependence of the critical strain on the
number of particles and relevant simulation parameters, the results are summarized in Fig. 5 and Table I, respectively. From Fig. 5 we see that the value for the critical strain weakly depends on the number of particles. We also investigate the influence of the step size and the termination condition on the value of the critical strain, and present the results in Table I. It shows that the variation of these simulation parameters by an order of magnitude only leads to a 1% – 2% variation of the value for the critical strain.

### IV. CONCLUSIONS

In summary, we revealed a common instability mode of vacancies with varying size and morphology under compression. Triggered by the local shear stress, the vacancy is fissioned into a pair of dislocations that ultimately glide to the boundary. The remarkable fractionalization of vacancies creates rich modes of interaction between vacancies and other defects, and provides a new dimension for mechanical engineering of defects in extensive crystalline materials. This discovery may also be exploited to explore the classical close packing problem from the dynamical perspective by using the softness of particles to eliminate vacancies and achieve higher particle density.

### V. ACKNOWLEDGEMENTS

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