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Symmetry-Dependent Kinetics of Dislocation Reaction

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Reactions between dislocations are investigated in two-dimensional colloidal crystals. It is found that, because of the conservation of total Burgers vectors, the kinetics of the reaction is dependent on the the symmetry of the crystal lattice. Merging is possible only when the total Burgers vector of the reacting dislocations is in line with existing crystal lines. In non-merging reactions, the number of dislocations cannot be reduced but the interacting dislocations can exchange their Burgers vectors and migrate to different gliding lines. The changing of gliding lines promises additional annihilation in multi-dislocation reactions. The bonding of non-merging dislocations determines the configuration and the orientation of the grain boundaries. The findings in this study may shed new light on understanding of dislocations and have potential applications in fabrication of crystalline materials.

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Formation of defects, such as interstitials, vacancies and dislocations, is a universal phenomenon in growth of crystals.[1−4] Dislocation is the most popular and active defect in crystals.[5−7] Presence of dislocations can significantly modify mechanical and optoelectronic properties of solid materials.[8−11] Dislocation results from the termination of a crystal plane in the middle of a crystal. At the end of a terminating plane, the crystal lattice becomes locally mismatched and deformed.[8,12,13] Each dislocation can be characterized by a Burgers vector that describes the magnitude and the direction of the structure deformation. Generally, dislocations are confined on a plane that is perpendicular to and crosses the terminating plane. The cross line defines the location of the dislocation. A single dislocation can never disappear. However, as two dislocations with opposite Burgers vectors glide on the same plane, the two half crystal planes can merge into one full plane, giving rise to annihilation of dislocations.[14,15] Dislocations which are not antiparallel or glide on different planes cannot merge directly. The reactions between non-merging dislocations play a critical role in the response to plastic deformation and have a significant impact on the mechanical and electrical properties of crystals.[8,9,16,17]

Despite their critical role, knowledge about the reactions of non-merging dislocations is incomplete. The challenge is that in typical atomic systems, visualizing and following the dynamic processes of reactions at the single-particle level is inhibited because of the small length and the short time scales. As an alternative approach, colloidal systems have been widely employed in the last decades to explore the mechanisms underlying liquid-solid transitions, including gelation,[18] glass transition[19] and crystallization.[20−24] Colloidal particles in a solution behave like 'big atoms' and exhibit similar phase behaviors as observed in atomic systems.[23,25,26] The advantages of colloidal systems are such that colloidal particles are big enough to be observed via normal light microscopes and the dynamic processes are slow enough to follow in real time.

Facilitated by colloidal systems, many insights have been achieved on nucleation and crystal growth. Theories about the classical nucleation[27] and non-classical nucleation[28−30] have been verified. The diffusion of vacancies was successfully visualized at the single-particle level,[9,31] and the observations revealed that stress relaxation in crystalline solids is governed by the formation and diffusion of defects.[13,22,31,32] In recent studies, diffusion, merging and recombination of dislocations were visualized in colloidal crystals as well.[14,15] However, in these studies, the general principles and the consequence of reactions of non-merging dislocations were not addressed. In this study, the dynamic processes of reactions of non-merging dislocations are visualized and followed at the single-particle level in two-dimensional colloidal crystals. It is found that the kinetics and the results of dislocation reactions are dependent on the symmetry of the crystal lattice and the crossing angle between Burgers vectors. These observations greatly extended our understanding on the dynamic properties of dislocation and shed new light on the control of crystal growth and the annealing of defects.

Methods. Polystyrene spheres with a radius of $r_c = 1.0\,\mu m$ (Duke standards, std dev. $< 1.5\%$) were dispersed in a mixture of water and lutidine (vol-

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ume ratio 4 : 1). The colloidal suspension was sealed between two conducting glass plates coated with indium tin oxide (ITO).[28] The ITO-coated surface was processed with a mixture of 5 mg/mL poly(sodium 4-styrenesulfonate) (PSS) solution and 0.5 M potassium chloride (KCl) solution so that the surface becomes more negatively charged.[33] The gap between the two ITO-coated glass plates was $110 \pm 10\,\mu$m. When an alternating-current field was applied, the colloidal particles were transported by the fluid flow onto the surface of the glass plates, where they form a two-dimensional (2D) dense phase with an area fraction $\varphi_c = 0.1$. The phase behavior of the 2D system is dominated by two competing interactions: dipole–dipole repulsion and electrohydrodynamic (EHD) attraction. The repulsion is mainly determined by the field strength E, whereas the attraction depends on the frequency as well as the field strength.[34,35] Decreasing frequency and increasing E can enhance the attraction and thus 'cool' the system for crystallization.[23] The system was first equilibrated in a 2D liquid phase at the peak voltage $V_p = 19.0\,$V ($E_p = 0.17\,$V/μm) and $f = 4000\,$Hz,[24] and then cooled for crystallization by decreasing the frequency to $f = 1000\,$Hz. The dynamic processes of crystallization were observed using a Nikon (Niu) microscope with a 60× objective, and recorded with an SMOS camera (Andor Zyla) at 17 frames per second (FPS). The positions of the single particles were extracted with IDL routines at an accuracy of ∼10 nm.[36]

To identify and localize the position of the dislocations, Voronoi cell construction is performed on the snapshots. In an ideal 2D triangular lattice, all particles are sixfold coordinated so that the corresponding Voronoi polygons have six sides. The center of dislocations is characterized by a pair of a five-coordinated particle and a seven-coordinated particle, namely a 5–7 pair.[11,37] In this study, all Voronoi polygons of five-coordinated particles are colored green while that of seven-coordinated particles are colored red. Other polygons are colored white. A dislocation is then visualized as a red and green pair. The size and the orientation of the dislocation is characterized by the Burgers vector $\boldsymbol{B}$ that is centered on the line joining the five-coordinated particle and the paired seven-coordinated particle.[37]

Results. Driven by the stress, two dislocations which have antiparallel Burgers vectors and glide on the same crystal line attract each other. As they meet, the two dislocations annihilate and the two half crystal lines merge into one full line (Fig. 1). Annihilation is the simplest reaction to recover a perfect lattice and reduce the number of dislocations.[11,14,15] In the early stage of crystallization, annihilation is common. However, in the later stage of crystallization, as most dislocations on the same lattice lines vanish via annihilation, the remaining dislocations are generally characterized by different gliding lines or different orientations.

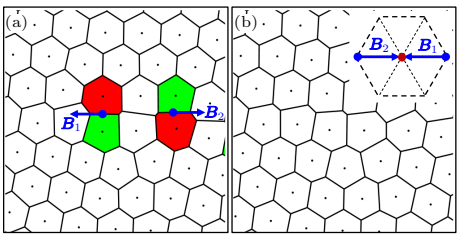

Fig. 1. The annihilation of antiparallel dislocations gliding on the same crystal lines. (a) Dislocations before annihilation. (b) Inset: the configuration of Burgers vectors on the triangular lattice. Blue arrows represent the orientations of Burgers vectors before reaction.

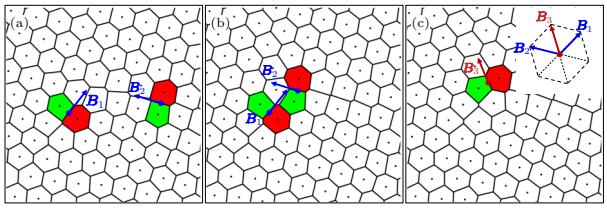

Fig. 2. The merging of dislocations with a crossing angle of 120°. Blue arrows represent the orientations of Burgers vectors before reaction. Red arrow represents the Burgers vectors after the merging.

In a triangular lattice, there are six orientations available for Burgers vectors. The angles between Burgers vectors can be 0°, 60°, 120° and 180°. For two parallel (0°) dislocations, the interaction between them is repulsive.[11] They do not like to meet each other. Two antiparallel dislocations (180°) attract each other but annihilation cannot occur if they do not stay on the same gliding line. For dislocations with crossed gliding lines, there are two possible configurations for their Burgers vectors: 60° and 120°. Figure 2 exhibits a reaction process of two dislocations with a crossing angle of 120° between the Burgers vectors $\boldsymbol{B}_1$ and $\boldsymbol{B}_2$. After the reaction, the two dislocations merge into one. The resulting dislocation is characterized by a distinct Burgers vector $\boldsymbol{B}_3$, which is exactly equal to $\boldsymbol{B}_1 + \boldsymbol{B}_2$. The total Burgers vector is conserved in the reaction. In principle, a dislocation can also reversibly dissociate into two distinct dislocations with a crossing angle of 120° if the thermal energy is comparable with the bonding energy between two particles.[14]

The conservation of total Burgers vector is a general principle followed by the reaction of dislocations.[38,39] For two dislocations with a crossing angle of 60°, the total Burgers vector is not in line with existing crystalline lines such that the merging cannot be accomplished. The behavior and the interaction between non-merging dislocations plays a central role in the response to plastic deformation. However, the related information is little and incomplete so far. Figure 3 exhibits a full reaction process of two dislocations with a crossing angle 60° between their Burgers

vectors $\boldsymbol{B}_1$ and $\boldsymbol{B}_2$ [Fig. 3(a)]. As is expected, the two dislocations meet but cannot merge [Fig. 3(b)]. However, during the reaction, they exchange their Burgers vectors [Fig. 3(c)]. Most importantly, the Burgers vectors that have the same orientation before and after the exchange stay on two parallel crystal lines that are separated by the lattice constant. It follows that in the reaction, the exchanging of Burgers vectors is accompanied by a hopping of gliding lines. In this process, no defect is removed. However, the hopping of gliding lines offers further opportunity for annihilation.

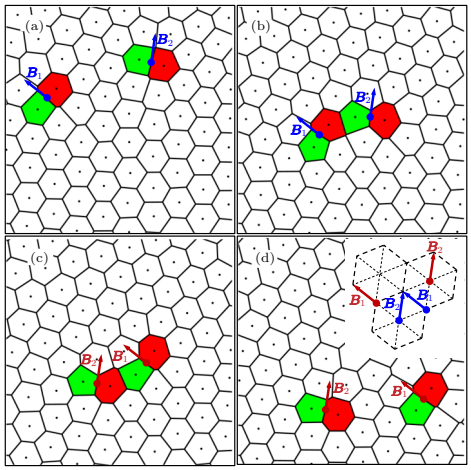

Fig. 3. The exchanging and hopping of Burgers vectors in the reaction of dislocations with a crossing angle of 60°.

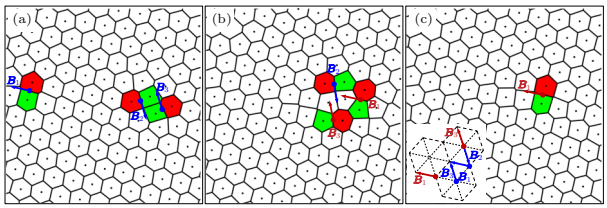

Fig. 4. Exchanging facilitated annihilation in a multi-dislocation reaction. Blue arrows represent the orientations of Burgers vectors before reaction. Red arrows represent the Burgers vectors after the merging.

Figure 4 exhibits an annihilation process facilitated by the exchanging and hopping of Burgers vectors in a multi-dislocation reaction. $\boldsymbol{B}_2$ and $\boldsymbol{B}_3$ are antiparallel but they stay on two parallel gliding lines that are separated by one lattice constant. $\boldsymbol{B}_2$ and $\boldsymbol{B}_3$ are bonded by the attraction but cannot annihilate.[40] $\boldsymbol{B}_1$ is approaching at the angle 60° with respect to $\boldsymbol{B}_3$. The reaction between $\boldsymbol{B}_1$ and $\boldsymbol{B}_3$ shifts $\boldsymbol{B}_3$ over to the gliding line of $\boldsymbol{B}_2$ such that the annihilation between $\boldsymbol{B}_2$ and $\boldsymbol{B}_3$ becomes possible. As a result, $\boldsymbol{B}_2$ and $\boldsymbol{B}_3$ vanish by annihilation, and $\boldsymbol{B}_1$ hops to a neighbored gliding line.

At the end of crystal growth, non-merging dislocations aggregate as grain boundaries. In grain boundaries, dislocations are connected and bonded.[24,37,41] As demonstrated in Fig. 2, a pair of dislocations with a crossing angle of 120° are not stable and tend to merge. Therefore, the configuration of 120° cannot persist in a grain boundary. Figure 5(a) exhibits a steady grain boundary observed at $E = 0.17\,\mathrm{V/\mu m}$ and $f = 1000\,\mathrm{Hz}$. As is expected, the configuration of 120° is not present. Figure 5(b) presents the probability distributions of the crossing angles in grain boundaries achieved at three different E. At the field of $E = 0.17\,\mathrm{V/\mu m}$ and $E = 0.11\,\mathrm{V/\mu m}$, the distributions are dominated by 0° and 60°, which happen with comparable probabilities, and no configuration of 120° is identified. However, as the field E decreases to $0.07\,\mathrm{V/\mu m}$, the probability of 120° becomes significant. The understanding is that decreasing the field E weakens the attraction between particles such that the crystal lattice becomes heated. In this case, the merging as illustrated in Fig. 2 becomes reversible such that the dissociation becomes active in practice. The dissociation in grain boundaries gives rise to the presence of the 120° configuration. The reversible merging and dissociation give rise to the fluctuation of the length of grain boundaries. To quantify the stability of the grain boundary, the length of the grain boundary in the observation window is measured and averaged over thousands of snapshots. The results are then normalized by the mean length. Figure 5(c) presents the probability distributions of the length of grain boundaries. The width of the distribution offers a quantitative estimation on the magnitude of fluctuation. At $E = 0.17\,\mathrm{V/\mu m}$ and $0.11\,\mathrm{V/\mu m}$, the grain boundaries are pretty stable and the fluctuation of length is within 15%. At the low field of $0.07\,\mathrm{V/\mu m}$, the fluctuation goes up to 25%, suggesting that the grain boundary becomes active due to the 'high temperature', being consistent with the presence of the 120° configuration.

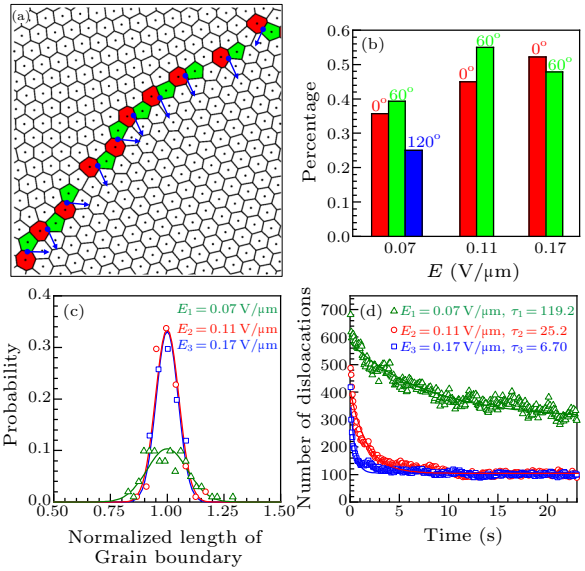

Fig. 5. Grain boundaries and statistics of dislocations. (a) The grain boundary at $E = 0.17\,\mathrm{V/\mu m}$. (b) Probabilities of the crossing angle between dislocations in grain boundaries. (c) Probabilities of the length of grain boundaries. (d) Time evolution of the number of dislocations.

A stronger attraction between the particles means a stronger stress induced by defects and thus a higher driving force for annihilation, merging and the nucleation of grain boundaries. To quantify the effect of attraction on the dynamics of dislocation, the number of dislocations as a function of time is investigated. It is shown that the number of dislocations decreases exponentially with time by $\exp(-t/\tau)$ in the early stage [Fig. 5(d)]. The characteristic time τ drops sharply with E, suggesting that the dynamic processes of dislocation reaction are accelerated by increasing the attraction strength. In the later stage, the number of dislocations reaches an equilibrium value. The equilibrium number at $E = 0.17\,\mathrm{V}/\mu\mathrm{m}$ and $E = 0.11\,\mathrm{V}/\mu\mathrm{m}$ are essentially the same. The understanding is that at these two fields, the excitation of dislocations from grain boundary (the ground state of dislocation) is suppressed. Nevertheless, at the low field $E = 0.07\,\mathrm{V}/\mu\mathrm{m}$, the equilibrium number of dislocations are much higher than the ground state due to the excitation, being consistent with the larger fluctuation of the length of grain boundary.

In summary, our investigations reveal that the dynamics of the dislocation reaction is determined and limited by the lattice symmetry as well as the interaction between units. Direct merging of dislocations is possible only as the resulting Burgers vector can be accommodated by the existing crystal lines. However, non-merging dynamic processes can shift the gliding lines of the included dislocations by exchanging their Burgers vectors. The shift of gliding lines can facilitate additional annihilation or merging in multiple-dislocation reactions. These findings shed new light on the understanding of the dynamic behavior of dislocations and have potential applications in controlling the quality of crystalline materials. The dynamic behaviors of dislocations revealed in this study arise from two basic principles. First, Burgers vectors are in line with existing crystal planes (3D) or crystal lines (2D). Second, the total of Burgers vectors is conserved in reactions. These two principles are general and valid both in 2D and 3D crystals. Therefore, the conclusion in this study that the results of the dislocation reaction are determined and confined by the symmetry of the lattice is applicable to both 2D and 3D. Nevertheless, the detailed dynamics in 3D will be more complicated due to the increased freedom and crystal orientations. In 3D systems, however, following the reaction processes at single-particle level is inhibited technically.

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