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Editorial

Computer Simulations: Essential Tools for Crystal Growth Studies

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Abstract: This special issue discusses recent advances in computer simulation studies of crystal growth. Crystal growth is a key to innovation in science and technology. Owing to recent progress in computer performance, computer simulation studies of crystal growth have become increasingly important. This special issue covers a variety of simulation methods, including the Monte Carlo, molecular dynamics, first-principles, multiscale, and continuum simulation methods, which are used for studies on the fundamentals and applications of crystal growth and related phenomena for different materials, such as hard-sphere systems, ice, organic crystals, semiconductors, and graphene.

Keywords: molecular dynamics (MD); Monte Carlo (MC); first-principles (FP) simulation; continuum simulation; multiscale simulation

1. Introduction

Crystals are ubiquitous in daily life and technology. Many kinds of crystalline products, such as salt, sugar, and fat, are used in cooking, and electronic devices are made from semiconductor crystals. Crystals also play an important role in life and the global environment. Living organisms produce mineral crystals to maintain biogenic activity, and snow and ice crystals play a crucial role in climate change. For most topics related to crystals, crystal growth is an important research area.

Owing to recent progress in computer performance, computer simulation studies of crystal growth have become increasingly important. Computer simulations can be used to analyze and predict various aspects of the crystal growth process, such as growth and nucleation mechanisms, as well as the structures and dynamics of surfaces and interfaces, and pattern formation.

This special issue discusses recent advances in computer simulation studies of crystal growth. We present 10 papers, covering fundamental studies and applications of crystal growth or related phenomena. A variety of simulation methodologies are used in the studies.

2. Methodologies of Crystal Growth Computer Simulations

2.1. Molecular Simulation

Molecular simulations, such as molecular dynamics (MD) and Monte Carlo (MC) simulations, are powerful tools for investigating the growth mechanisms and interface structures of crystals at the molecular scale [1–3]. MD simulations analyze the structure, dynamics, mechanical properties, electrical properties, and optical properties of a condensed phase by solving the Newtonian equations of motion for each atom or molecule. MC simulations generate new states of the atomic or molecular arrangement stochastically according to a Boltzmann probability distribution. In addition to single condensed phases, crystal–liquid interfaces or crystal–vapor interfaces can be examined by MD or MC simulations. Therefore, we can analyze the atomic- or molecular-scale mechanisms of crystal growth by these simulation methods.

In this special issue, Mori [4], Qiu and Molinero [5], Barcaro et al. [6], Elts et al. [7], Hagiwara et al. [8], and Y.-P. Liu et al. [9] used MD simulations. Ito and Akiyama [10] and Akutsu [11] used MC simulations. Elts et al. also used a kinetic MC method, which is a simulation method for the mesoscale growth or dissolution of a crystal [7].

2.2. First-Principles Simulation

First-principles (FP) methods, such as density functional theory (DFT) [12], can reproduce the atomic-scale structure and energetic state of a real material precisely. In the FP method, the electronic structures of atoms and molecules in a material are computed by solving the Schrödinger wave equation. The FP method can be combined with the MD simulation method (FP-MD method) [13]. In principle, the FP-MD method can provide precise information on the crystal growth mechanism and interface structure of a real material. However, FP-MD simulations of crystal growth for a large system are too time-consuming. Thus, for many cases, the FP method can be used effectively in parts of computer simulation studies of crystal growth or related phenomena.

In this special issue, Ito and Akiyama used the DFT method for calculating chemical potential precisely in their simulation studies [10]. Barcaro et al. used the DFT method to obtain energetic and structural information about small Si clusters, which was then used to optimize the reactive force-field parameters [6].

2.3. Continuum Simulations

Continuum simulations can be used to study mesoscale or macroscale phenomena related to crystal growth, such as crystal morphology and fluid dynamics in crystal growth. There are a variety of continuum simulation methods. The phase-field method is a popular continuum simulation method for studies of crystal growth kinetics and crystal growth morphology [3,14], although this issue does not include studies using the phase-field method.

In this special issue, Elts et al. used continuum simulations in their multiscale simulation studies of crystal growth [7]. Related to continuum simulations, Ruan used a morphology evolution model and the MC method for simulations of polymer crystallization in a shear flow [15]. Z. Liu et al. used a particle level set method [16] for simulations of polymer crystallization under isothermal and temperature gradient conditions [17].

2.4. Multiscale Simulations

Recently, multiscale simulations have attracted a great deal of attention in the field of crystal growth. In this special issue, Elts et al. review the recent progress made by their group in multiscale simulations, including MD simulations, kinetic MC simulations, and continuum simulations for crystal growth [7]. Using multiscale simulations, they predicted the macroscopic morphology and growth or dissolution rate of a crystal from the molecular structure.

3. Materials for Crystal Growth Computer Simulations

3.1. Hard-Sphere System

The hard-sphere system is the simplest off-lattice model used for computer simulation studies of crystal growth. Crystallization of a hard-sphere system is often used as a model for crystallization of a colloidal system. In this special issue, Mori reviews computer simulation studies of crystal growth using a hard-sphere model for crystal-fluid coexistence in the equilibrium state and hard-sphere systems in gravity [4].

3.2. Organic Molecules

Crystal growth of organic molecules, such as carbohydrates, amino acids, peptides, proteins, and polymers, is related to many phenomena in nature and in industries such as the biology, pharmaceutical,

nutraceutical, food, and cosmetic industries. Computer simulations have been used for studies of the growth mechanism, growth morphology, and equilibrium morphology of organic molecule crystals.

In this special issue, Qiu and Molinero report MD simulations of the crystal growth of alkanes [5]. They found that the strength of the alkane–fluid attractive interaction controls the interfacial orientation of liquid alkanes and their crystallization. Elts et al. performed multiscale simulations of the growth and dissolution of aspirin crystals [7]. They predicted the aspirin dissolution rates, which agreed well with the experimental rates. Ruan performed MC simulations of polymers crystallizing in a shear flow [15], and provided simulation results for the growth kinetics, morphology, and rheology of the polymer crystals that agreed well with earlier experimental and theoretical studies. Z. Liu et al. performed simulations of polymer crystallization using a particle level set method [17]. They clarified the development of crystallinity during crystallization under quiescent isothermal conditions, and their results were consistent with theory.

3.3. Ice

Ice is a familiar material in daily life and studies of its crystal growth are important, both scientifically and practically, in connection with topics such as the freezing of water in biological systems, pattern formation of snow crystals, artificial snow, cryopreservation of tissues, and food processing. In this special issue, Hagiwara et al. studied the structural and dynamic properties of an aqueous solution including a winter flounder antifreeze protein and salt ions near the secondary prismatic and pyramidal planes of ice [8]. Their MD simulation indicated that hydrogen bonding between water molecules in the solution is inhibited, which may be related to the fact that the antifreeze activity of the protein is enhanced if salt ions are present.

3.4. Functional Materials

Controlling the growth, size, and morphology of crystals is essential for developing functional materials, which can be used for applications including devices, solar cells, and optical materials. Computer simulations have been used for studies of the growth of various crystals of functional materials, such as semiconductors and graphene.

In this special issue, Ito and Akiyama review recent progress in computational materials science in the area of semiconductor epitaxial growth [10]. They present their computer simulation studies of the heteroepitaxial growth of InAs on GaAs and the formation of InP nanowires with their ab initio approach. Barcaro et al. performed a computer simulation study of the nucleation and growth of Si nanoclusters [6]. They proposed a theoretical approach that can be used to model the nucleation and growth of small particles for which experimental studies are difficult to perform. Akutsu studied the surface tension, growth rate, and size of macrosteps on the surface of 4H-SiC crystals using the restricted solid-on-solid model [11]. The effects of the driving force on the size of a faceted macrostep and on the growth rate of the vicinal surface were discussed. Y.-P. Liu et al. studied the growth of graphene sheets embedded with single-wall carbon nanocones (SWCNCs) and suggested conditions suitable for SWCNCs growing on a Cu substrate [9].

4. Conclusions

This special issue presents advances in computer simulation studies of crystal growth. Crystal growth is important in many fields of science and technology. Because the performance of computers is still improving, computer simulations will continue to be essential tools. By covering various types of computer simulation studies of crystal growth and related phenomena from fundamental research to practical applications, this special issue provides helpful information for future simulation studies.

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Conflicts of Interest: The authors declare no conflict of interest.

References

1. Frenkel, D.; Smit, B. *Understanding Molecular Simulation: From Algorithms to Applications*; Frenkel, D., Klein, M., Parrinello, M., Smit, B., Eds.; Academic Press, A division of Harcourt, Inc.: San Diego, CA, USA, 1996.
2. Allen, M.P.; Tildesley, D.J. *Computer Simulation of Liquids*; Oxford University Press: Oxford, UK, 1987.
3. Nada, H.; Miura, H.; Kawano, J.; Irisawa, T. Observing Crystal Growth Processes in Computer Simulations. *Prog. Cryst. Growth Charact.* **2016**, *62*, 404–407. [CrossRef]
4. Mori, A. Computer Simulations of Crystal Growth using a Hard-Sphere Model. *Crystals* **2017**, *7*, 102. [CrossRef]
5. Qiu, Y.; Molinero, V. Strength of Alkane-Fluid Attraction Determines the Interfacial Orientation of Liquid Alkanes and Their Crystallization through Heterogeneous or Homogeneous Mechanisms. *Crystals* **2017**, *7*, 86. [CrossRef]
6. Barcaro, G.; Monti, S.; Sementa, L.; Carravetta, V. Atomistic Modelling of Si Nanoparticles Synthesis. *Crystals* **2017**, *7*, 54. [CrossRef]
7. Elts, E.; Greiner, M.; Briesen, H. In Silico Prediction of Growth and Dissolution Rates for Organic Molecular Crystals: A Multiscale Approach. *Crystals* **2017**, *7*, 288. [CrossRef]
8. Yasui, T.; Kaijuma, T.; Nishio, K.; Hagiwara, Y. Molecular Dynamics Analysis of Synergistic Effects of Ions and Winter Flounder Antifreeze Protein Adjacent to Ice-Solution Surfaces. *Crystals* **2018**, *8*, 302. [CrossRef]
9. Liu, Y.P.; Li, J.T.; Song, Q.; Zhuang, J.; Ning, X.J. A Scheme for the Growth of Graphene Sheets Embedded with Nanocones. *Crystals* **2017**, *7*, 35. [CrossRef]
10. Ito, T.; Akiyama, T. Recent Progress in Computational Materials Science for Semiconductor Epitaxial Growth. *Crystals* **2017**, *7*, 46. [CrossRef]
11. Akutsu, N. Disassembly of Faceted Macrosteps in the Step Droplet Zone in Non-Equilibrium Steady State. *Crystals* **2017**, *7*, 42. [CrossRef]
12. Kohn, W.; Becke, A.D.; Parr, R.G. Density Functional Theory of Electronic Structure. *J. Phys. Chem.* **1996**, *100*, 12974–12980. [CrossRef]
13. Car, R.; Parrinello, M. Unified Approach for Molecular Dynamics and Density Functional Theory. *Phys. Rev. Lett.* **1985**, *55*, 2471–2474. [CrossRef] [PubMed]
14. Sekerka, R.F. Fundamentals of Phase Field Theory. In *Advances in Crystal Growth Research*; Saito, K., Furukawa, Y., Nakajima, K., Eds.; Elsevier Science B.V: Amsterdam, The Netherlands, 2001; Chapter 2; p. 21.
15. Ruan, C. Kinetics and Morphology of Flow Induced Polymer Crystallization in 3D Shear Flow Investigated by Monte Carlo Simulation. *Crystals* **2017**, *7*, 51. [CrossRef]
16. Enright, D.; Fedkiw, R.; Ferziger, J.; Mitchell, I. A Hybrid Particle Level Set Method for Improved Interface Capturing. *J. Comput. Phys.* **2002**, *183*, 83–116. [CrossRef]
17. Liu, Z.; Ouyang, J.; Ruan, C.; Liu, Q. Simulation of Polymer Crystallization under Isothermal and Temperature Gradient Conditions Using Particle Level Set Method. *Crystals* **2016**, *6*, 90. [CrossRef]