Crystal growth rates in supercooled atomic liquid mixtures

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ABSTRACT

Crystalization is a fundamental process in materials science, providing the primary route for the realization of a wide range of new materials. Crystalization rates are also considered to be useful probes of glass-forming ability. At the microscopic level, it is phenomenologically described by the classical crystal nucleation and growth theories. Yet, solid formation is a far more complex process and markedly different crystal growth regimes in many binary liquid mixtures greatly challenge our understanding of crystallization. Here, we study, by experiments, theory and computer simulations, the crystallization of supercooled mixtures of argon and krypton, showing that crystal growth rates in these systems can be reconciled with existing classical models only by explicitly accounting for non-ideal mixing effects. Our results highlight the importance of thermodynamic aspects in describing the crystal growth kinetics, providing a substantial step towards a more sophisticated theory of crystal growth.

CRYSTAL GROWTH

Crystalization rates in many systems to maximum rate and huge variances

Technological goal: identification of glass-formers (extreme crystal growth rate slowdown)

Locally2d Frenkel Structures, diffusion, quantum effects, liquid pre-ordering...

ballistic at short times, no activation barrier, weak T dependence

diffusive at longer times, activated process, strong T dependence

Collision-limited (CL)

WF model

Collision-limited (CL)

WF model

Wilson-Frenkel (WF)

WF model

ε(T) = f(x(T),ε(T)) exp [ΔG/ kT] (1-exp [-ΔG/kT])

u(T) = f(x(T),ε(T)) exp [ΔG/ kT] (1-exp [-ΔG/kT])

Crystal growth rate depends on pressure, temperature, concentration, vibrational frequencies, etc.

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Ground state energy of the crystal, $E_{cr}$

Lammps simulation, $E_{cr}$

Structural order parameter

Individual atoms are fixed, others move independently

Average Local Bond Order Parameter

Crystal structures detection

Unified picture of crystal growth mechanism?

THE EXPERIMENT

Experiments on Ar-Kr @ DESY and @ EU-XFEL

- Crystal growth rates — metallic alloys
- Miscibility in the whole phase diagram (study several Kr fractions)
- Many data for $\Delta G$ and $\Delta S$ available
- Straightforward comparison with the model
- Least-squares interactions: easy to simulate
- r of the jet constant: distance from nozzle -> time evolution at x=y=z

WHICH MODEL?

- Experimental data also include nucleation events (composition-dependent)
- Many crystallizes in the microcr as seeded (based) growth
- Nontrivial temperature-dependent behavior

MD simulations capture relevant details

RESULTS & PERSPECTIVES

Only simulations: fixed x, y, z

Modified CL also fits relative growth rates as a function of Kr fraction

Next steps:
- Study nucleation (in collaboration with EU-XFEL)
- Study model systems (role of interaction parameters)
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- Study of nucleation

REFERENCES

1. Scheidt and Risse, Nat. Mat. 2012
2. Tang et al., Nat. Mat. 2013
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4. Jackson, Kinetic Processes 2004

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