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Nanoscale Patterning of Chemical Order in Irradiated Metallic Alloys

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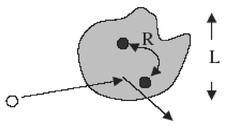
Introduction

Irradiation with energetic ions creates displacement cascades, resulting in disordered zones in chemically ordered alloys. At finite temperatures, this disorder competes with thermally activated reordering.



*Schematic drawings illustrating the characteristic lengths introduced by irradiation: the relocation range R and the cascade size L (In this study, the number of forced pair exchanges in one cascade **b** is used to represent the cascade size).*

Materials under irradiation are dissipative systems, and as such, they are susceptible to self-organize (SO). There are twofold interests:
- Fundamental: excellent test bed of the theory driven systems since microscopic mechanisms are well identified and can be varied in a controlled manner experimentally.
- Practical: self-organization can be used to synthesize functional nanocomposites with tunable scales

Objective: study the effect of cascade size **b** on self-organize.

Main results: we identify a new patterning reaction in the case of ordered alloys: when the cascade size b is large enough, the degree of chemical order develops nanoscale patterns, under appropriate irradiation flux and temperature.

Kinetic Monte Carlo Simulations

Kinetic Monte Carlo simulations are employed to study the effect of the size of displacement cascades in an ordered alloy. A particular binary $A_{1-x}B_x$ model alloy undergoing $L1_0$ ordering is selected.

The forced mixing and disordering produced in displacement cascades is introduced by forcing exchanges of atoms in the cascade. In the present model, four parameters are used to specify this forced mixing.

- Γ_c the rate of introduction of cascade per atom.
- **b** the cascade size.
- ¶ cascade density: set to 80% in this work in order to create fully or near fully disordered zones in L10 phases.
- **R** the relocation distance: forced exchanges are here performed between nearest neighbor atoms, in order to isolate cascade size effects from relocation range effects on patterning reactions.

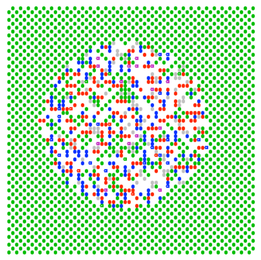
Finally, for comparison purposes, it is useful to introduce the ballistic jump frequency, $\Gamma_b = b^* \Gamma_c$, which is a measure of the disordering rate. $2\Gamma_b$ is the number of replacements par atom per second.

The simulation temperature is set at 60% T_c.

Simulation results are analyzed using complementary methods, encompassing direct visualization of the configurations in 2D and 3D, determination of the size and number of ordered clusters belonging to any variant, and calculation of the spherically averaged structure factor, centered on an $L1_0$ superlattice position

Cascade model used in the simulations

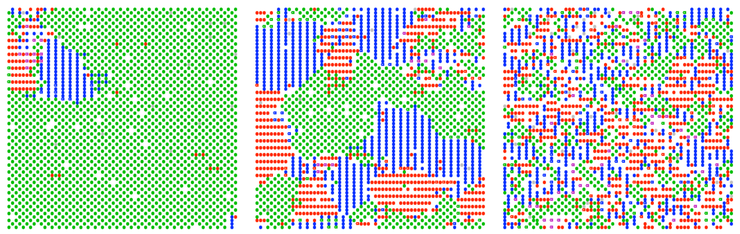
For simplicity, the volume of the cascade is assumed to be spherical, and only one cascade size is used during each KMC run.



*(100) cut of instantaneous configuration after the introduction of one large dense cascade (**b**=8000, density=80%) in an initially long-range ordered phase;*

Only B atoms are displayed. Three colors correspond to three rotational $L1_0$ variants.

Three steady states under sustained irradiation



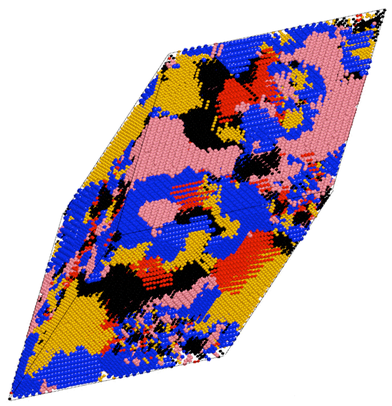
Low $\__b$ — **Long-Range Ordered**

Intermediate $\__b$ — **Patterning of order**

High $\__b$ — **Disordered**

*Steady states of AB alloy under irradiation with large dense cascade (**b**=4002, density=80%) at increasing disordering frequency: (a) $\__b = 1000s^{-1}$; (b) $\__b = 3000s^{-1}$; (c) $\__b = 30000s^{-1}$.*

Three distinct steady-states are identified. Besides the expected long-range ordered (LRO) and disordered states, for large enough cascade, there is a new state in the sense that the alloy is well ordered, but all six variants are present in domains of finite size; this is a state of **patterning of the chemical order**.



*3D view of atomic configuration of **patterning of chemical order** steady state. All six variants of $L1_0$ structure are present in domains of finite size. Domains from same variant form connected structure in 3D.*

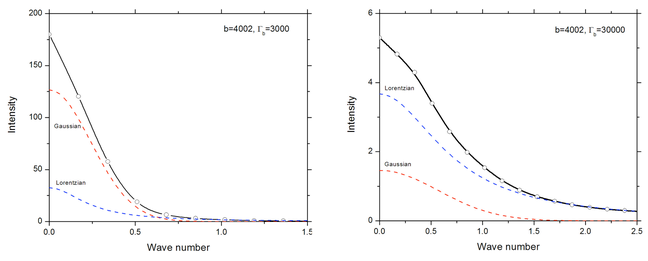
Simulation parameters:

***b**=4002, $\__b = 3000s^{-1}$.*

Identification of Patterning-disordered boundary

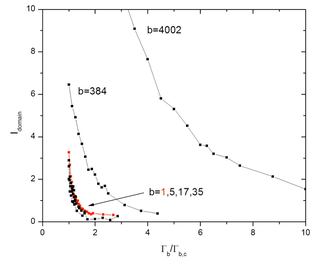
$$I(k) = \frac{I^{SRO}\xi}{1+\xi^2 k^2} + \sqrt{\pi \ln 2}\, I^D l \exp(-\ln 2 l^2 k^2)$$

Two components of structure factor:
1. Gaussian: Ordered domain, approximation of dynamical scaling function.
2. Lorentzian: SRO, exponential decay of fluctuation of order.

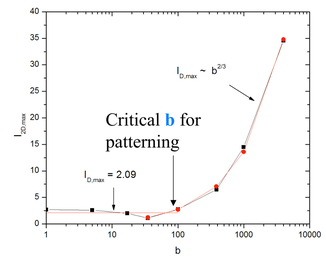


*Decomposed structure factors of two steady state: **patterning of chemical order** and disordered. It is clearly shown that the domain intensity is much higher than the SRO intensity in patterning state.*

Simulation parameters are as marked.

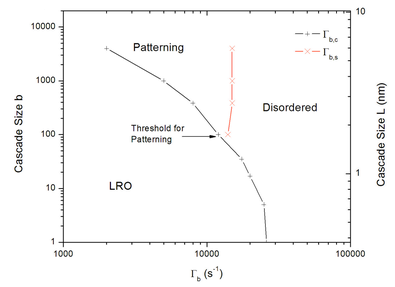


*With small cascade size **b** (b=1,5,17 and 35), all I_{domain} vs. normalized $\__b/\__{b,c}$ curves can be rescaled to one curve. For large **b** (b=394 and 4002), the behaviors of the system are different. Rescaling requires the introduce of new flux, $\__{b,s}$.*



- *Two different regions: Constant $I_{D,max}$ at small **b** region and abnormal $I_{D,max}$ value at large **b** region*
- *Consistent with direct observation of the atom configurations.*
- *Threshold for **patterning** at crossover.*

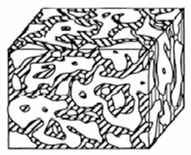
Dynamical steady-state phase diagram

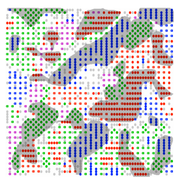


1) *there is a minimum cascade size for patterning of degree of order to be possible. This threshold value is around **b=100**.*
2) *The patterning-disorder boundary is almost independent of the cascade size.*
3) *At small b, I_{domain} obeys scaling with $\__b/\__{b,c}$. At large b, it obeys scaling with $\__b/\__{b,s}$.*

Summary

- **A new steady state, patterning of chemical order, is predicted under irradiation with large and dense cascades.**
- **A dynamical phase diagram is built that yields the stable steady states as a function of the cascade size and the irradiation flux.**
- **KMC results are supported by analytical model (see PRB 70,094105(2004)).**
- **Patterning of chemical order is a general phenomenon. It can be used to rationalize previous experimental results by Nelson et al and Schmitz et al for $Ni_{88}Al_{12}$ alloy.**
- **Direct synthesis of nano-composites by large and dense cascades (heavy ion irradiation) is possible.**
- **Potential applications for exchange-spring magnets (Kneller and Hawig, 1991).**

(a) 

(b) 

(a) Sketch of exchange-spring magnet. (b) KMC result of patterning state from c_B=37.5%, b=4002. White region: soft magnet ($L1_2$ phase). Dark region: hard magnet ($L1_0$ phase).

Acknowledgement

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