Atomistic Simulation Study of the Crystal-Melt Interface Excess Stresses

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Introduction

For the solid-liquid interface system, the EIS plays a role in many aspects. i) The magnitude of τ_{mn} and its crystalline anisotropy provide practical knowledge of γ in predicting solidification microstructure evolution; ii) The divergence of the EIS contributes to the mechanical force balances at crystal-melt interfaces (CMIs); The EIS is an essential factor in the non-equilibrium nucleation process.; The non-uniformly distributed local stress shifts the equilibrium composition or the mutual miscibilities at the interfacial region away from their unstressed bulk values.

For quantifying the work of elastic stretching/compressing a solid surface, e.g., Shuttleworth derived the equation that associating the surface (or solid-vapor interface) free energy γ with the surface (or solid-vapor interface) stress tensor τ_{mn},

$$\tau_{mn} = \delta_{mn}\gamma + \frac{\partial \gamma}{\partial e_{mn}}$$

in which, δmn is the Kronecker delta, and e_{mn} is the surface strain tensor. $m, n = x, y$ are the two Cartesian axial directions parallel to the surface.[Phys. Rev. B 79, 054109(2009); Phys. Rev. B 79,045430(2009); J. Chem. Phys. 131,114110(2009)]

Microscopic Pressure Definitions

The first definition method is so-called the virial method.

$$s_i^{\alpha\beta} = -\left[m v_{i\alpha} v_{i\beta} + \frac{1}{2}\sum_{j=1}^{N_n}\left(r_{i_\alpha} f_{i_\beta} + r_{j_\alpha} f_{j_\beta} \right) \right] \qquad P_{\alpha\beta}^{\mathrm{VM}}(z) = -\frac{\left\langle \sum_i^{N_z} s_i^{\alpha\beta}(z) \right\rangle}{A\Delta z}$$

where α, β take on values x, y, z, m is particle mass, v_i is the velocity of particle i. r_{ij} and f_{ij} are the distance and force between particle i and particle j connected with a pairwise potential. N_n is the number of particle i's neighboring particles. The VM pressure components profiles along the CMI normal, $P_{\alpha\beta}^{VM}(z)$, are determined in fine-graining z axis (with bin size Δz, see details below), and calculated as the sum of the negative per-particle stress tensors $S_i^{\alpha\beta}$ divided by bin volume and the summation run over Nz particles located between z and $z+\Delta z$, here A is the cross-section area.

The second definition method is the well-known Irving-Kirkwood contour method (hereafter, referred as IK for simplicity).

$$P_{\alpha\beta}^{\mathrm{IK}}(z) = \rho(z) k_B T \delta_{\alpha\beta} + \frac{1}{A\Delta z}\left\langle \sum_{i=1}^{N-1}\sum_{j>i}^{N} r_{ij_\alpha} f_{ij_\beta} \theta(z) \right\rangle \qquad \theta(z) = \begin{cases} \Delta z / |z_{ij}|, & Z_i + \Delta z/2 < z < Z_j - \Delta z/2 \\ \Delta_i / |z_{ij}|, & Z_i - \Delta z/2 < z \le Z_i + \Delta z/2 \\ \Delta_j / |z_{ij}|, & Z_j - \Delta z/2 < z \le Z_j + \Delta z/2 \\ 1, & Z_i = Z_j \end{cases}$$

k_B is the Boltzmann constant, ρ is number density, $\delta_{\alpha\beta}$ is the Dirac delta function, N is the number of particles in the whole system.

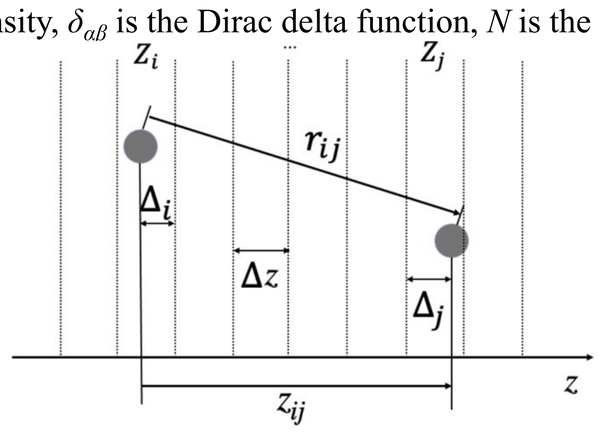
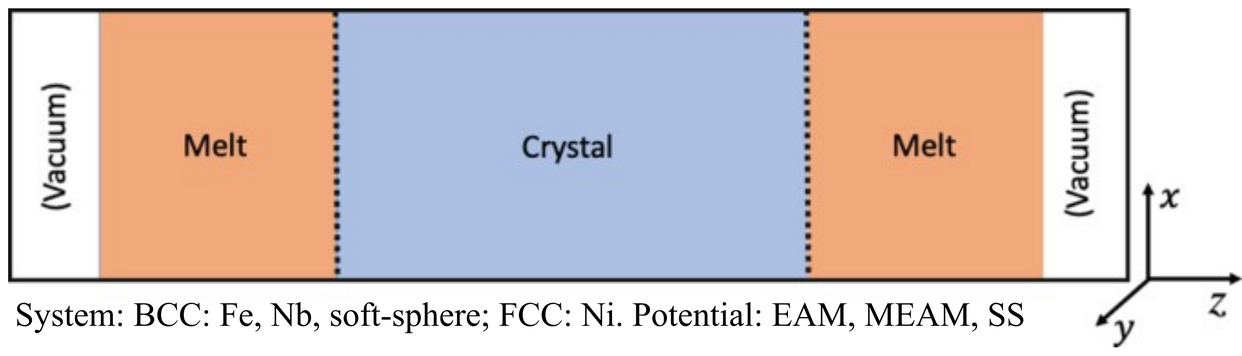


FIG.1 Illustration of a particle pair (i and j) that contributes to the fine-scale IK pressure, in a flat plane in Cartesian coordinates. The double-heads arrows denote different integration cases. [Mol. Simul. 29, 101–109(2003); Mol. Phys. 48,1357–1368(1983)]

Simulation Details



System: BCC: Fe, Nb, soft-sphere; FCC: Ni. Potential: EAM, MEAM, SS

FIG.2 Schematic illustration of the CMIs built in the current MD simulations. The two CMIs parallel to the xy plane (separated over 20 times of the lattice parameters in z) giving a total of forty independent samples (20 replica runs) to determine statistical uncertainty. The simulation box contains crystal, melt, and vapor phases for the metal systems, while for the SS system, the vapor regions are removed due to the crystal and melt phases coexist under the finite value of the hydrostatic pressures.
[Philos. Mag. 83 (35) 3977–3994(2002); Phys. Rev. B 81, 144119(2010); Phys. Rev. B 33 (12) (1986) ; Comput. Mater. Sci. 161 (2019); Phys. Rev. Lett. 94 (8) (2005), 086102]

Interface Characterization

The fine-grained density profile across the interface, $\rho(z)$, is computed as the average number of atoms in each discrete bin of spacing Δz divided by the volume of the bin, $A\Delta z$,

$$\rho(z) = \frac{\langle N_z \rangle}{A\Delta z}$$

The fine-grained stress profile is defined as the between the fine-grained normal and transverse components of the pressure tensor. τ is calculated by integrating the stress profile over the entire length of the simulation cell.

$$S^{\mathrm{VM,IK}}(z) = P_{zz}^{\mathrm{VM,IK}}(z) - \frac{1}{2}\left[P_{xx}^{\mathrm{VM,IK}}(z) + P_{yy}^{\mathrm{VM,IK}}(z) \right] \qquad \tau^{\mathrm{VM,IK}} = \int_0^{L_z} S^{\mathrm{VM,IK}}(z)\mathrm{d}z$$

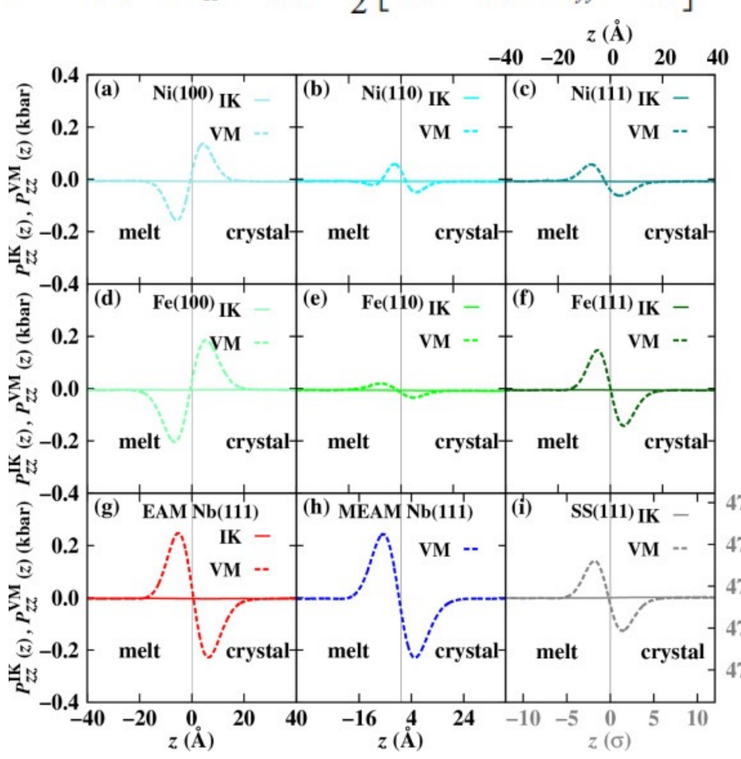


FIG.3 Significant difference found in the pressure component profiles, i.e., "VM"(dashed line) and "IK" (solid line) in the superscript correspond to the virial method and Irving-Kirkwood method, respectively. Vertical lines denote $z = 0$, which corresponds to the GDS with $z < 0$ for melt and $z > 0$ for crystal.

Result and Discussion

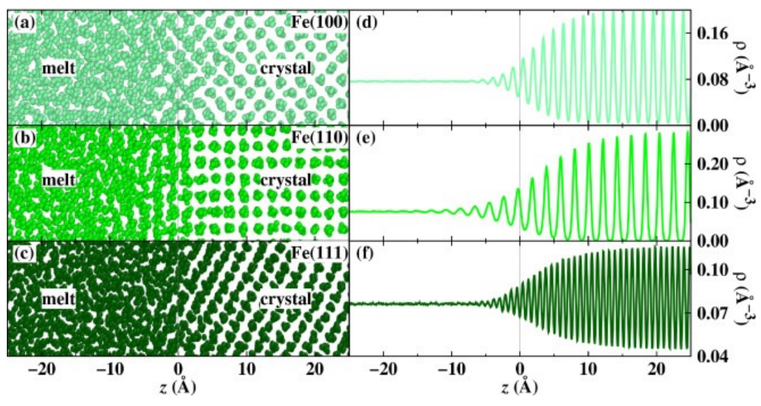


FIG.4 (a)-(c) snapshots of the equilibrated BCC Fe (100), (110), and (111) CMIs ; (d)-(f) the fine-grained density profiles for the three BCC CMIs

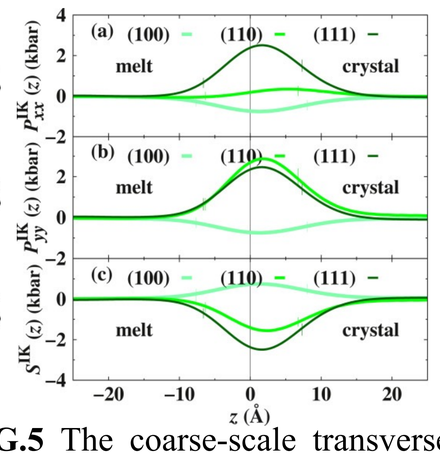


FIG.5 The coarse-scale transverse pressure tensor components(a)(b), and stress profiles(c)

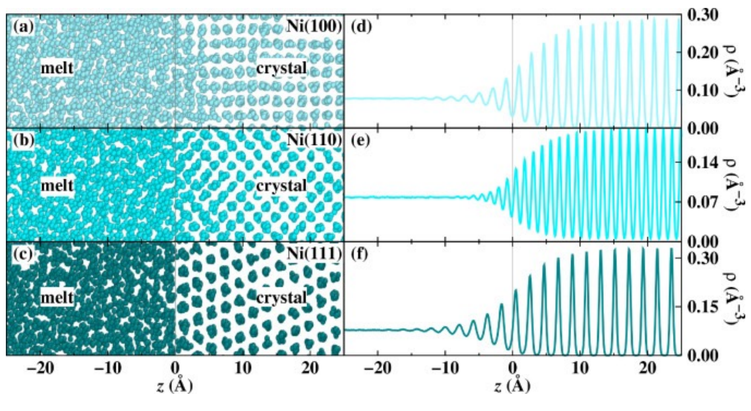


FIG.6 (a)-(c) snapshots of the equilibrated BCC Fe (100), (110), and (111) CMIs ; (d)-(f) the fine-grained density profiles for the three BCC CMIs

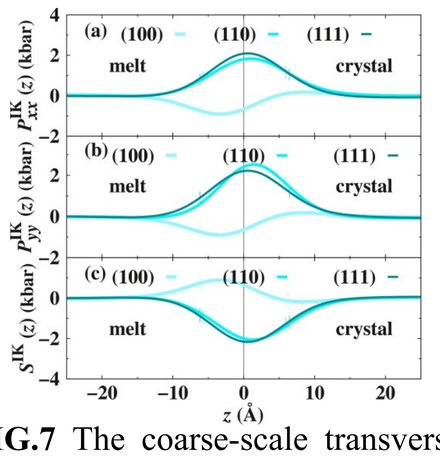


FIG.7 The coarse-scale transverse pressure tensor components(a)(b), and stress profiles(c)

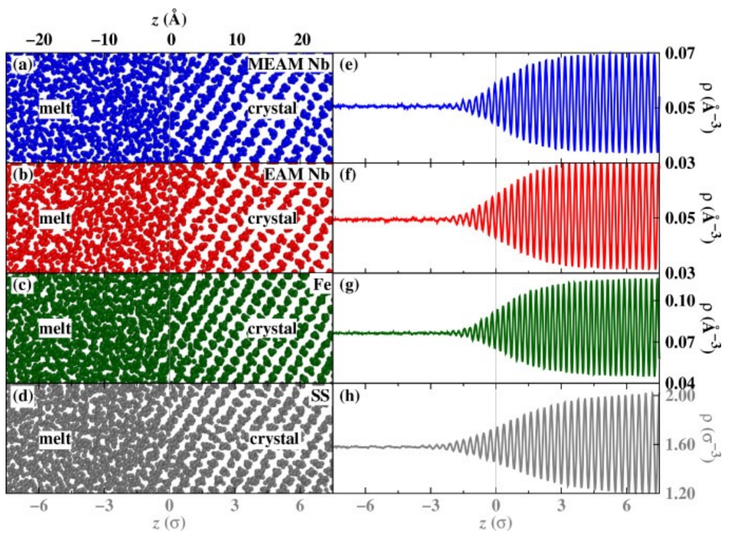


Fig. 8. (a)-(d) show representative snapshots from equilibrated BCC (111) CMIs modeled with four different potentials, under each melting point temperatures. Viewpoints are same as Fig. 4(a). Panel (e)-(h) show the corresponding fine-grained density profiles. The zero points of z corresponding to the GDS are chosen that the excess number of particles equals zero.

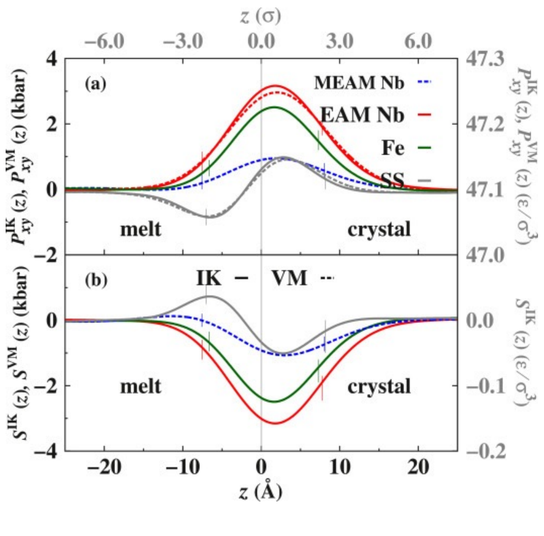


Fig. 9. The coarse-scale transverse pressure tensor components profiles (a) and stress profiles $S(z)$ (c) across the four BCC (111) CMIs. The two (x and y) components for BCC (111) CMI are isotropic. The result of MEAM Nb (blue dashed line) are only calculated with VM definition.

Acknowledgements

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