

Ritz-type surface homogenization: from atomistic to continuum surface models of copper despite imperfect bulk models

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Surfaces possess mechanical features on smaller scales that stand out against bulk phases, e.g., scaling of stiffness, curvature-dependence, surfactant control and anchoring-induced anisotropy. Continuum properties for the respective scales are often derived from ab initio simulations. This scale-bridging however bears conceptual challenges and we highlight three aspects for the example of pure copper. First, free surface atoms relax and alter the boundary region in terms of interatomic distances and resulting initial stresses. Second, eliminating the influence of finite thickness on the two-dimensional continuum surface can be achieved by different averages or limit definitions, not all being physically consistent. Third, the continuum model of the surface is usually coupled to a continuum model of the bulk, which causes an approximation error itself. However, the bulk phase can not be eliminated directly from the examination and simple averaging may even mask the aforementioned influences on the surface mechanics. A thermodynamically sound parameter identification across the scales is hence required. We present a Ritz-type modeling approach for surfaces that ensures energy equivalence between atomistic and continuum simulations. The influences of relaxation, finite thickness and bulk approximation are identified by a mismatch in the energy contributions and accounted for by using appropriate homogenization limits.

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1 Introduction and homogenization by energy equivalence

Continuum modeling on smaller scales must account for interface contributions to, e.g., effective moduli, resonance or shape changes [1, 2, 5, 6, 9]. This applies particularly to novel engineering materials such as solid nano structures or liquid crystals [3, 4]. A valuable benchmark for continuum models is provided by molecular dynamics simulations [7, 8]. Challenges of experimental work at the atomistic scale further increase the relevance of molecular dynamics simulations. The atomistic description, however, uses fundamentally different physical properties when compared to continuum mechanics. On the one hand, atomistic potentials are embedded in an electron density. On the other hand, continuous stress and strain fields describe the behaviour of a homogenized material point. It is thus important to apply a physically consistent homogenization.

We employ an energy-based Ritz-type approach, i.e., we minimize the energy difference between the atomistic description (index a) and the continuum description (index c), $|\Psi^a - \Psi^c|$. More specifically, the material continuum properties are determined by the minimal residuum

$$\mathcal{R} := \sum_{i=1}^n (\Psi_i^a - \Psi_i^c)^2 \rightarrow \min. \quad (1)$$

Index i represents one of n load cases that combine tension and/or shear with various amplitudes and orientations. See [8] for further information and for the numerical implementation in the molecular dynamics code LAMMPS and the finite element framework FEAP.

2 Deriving continuum surface properties

Having a continuum model at hand, a reliable estimation of the material parameters is required. This task is usually straightforward for bulk parameters but bears conceptual difficulties for surface parameters. We present peculiarities for the example of pure copper with free surfaces.

2.1 Relaxation depth and influence of surface thickness

Being a two-dimensional continuum idealisation, the surface's individual contribution is required to determine its material parameters. In practical applications, though, a surface does not exist without an underlying bulk. A common approach is thus to let the bulk contribution vanish by reducing the thickness of the sample. There is, however, a lower bound dictated by the molecular physics. Several molecular boundary layers adapt to the outer environment (that is vacuum in the present example) by relaxation, Fig. 1 (left). We observed a depth of approximately six unit cells to be affected within the range of numerical accuracy. A simple zero-thickness limit should thus not be used for the determination of the surface parameters.

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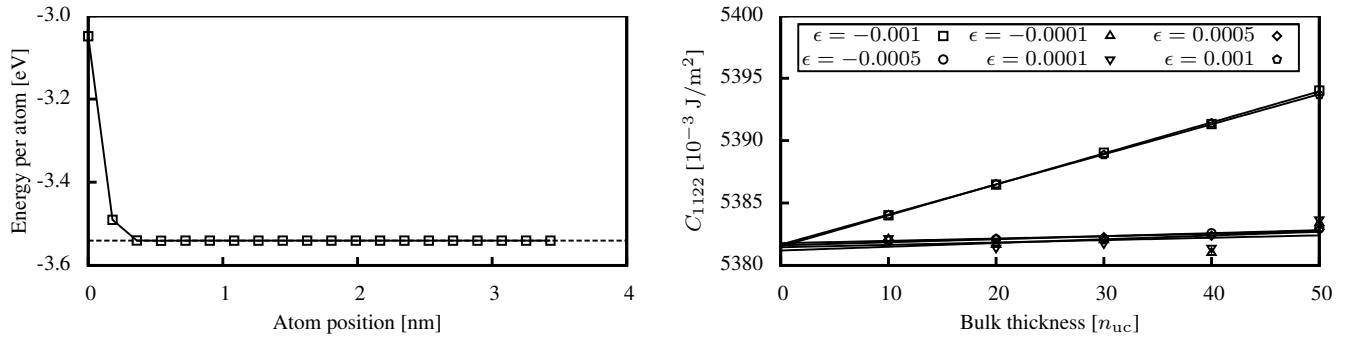


Fig. 1: Energy per atom in thickness direction (left). Surface stiffness C_{1122} as a function of bulk thickness (measured by unit cells n_{uc}) for various strain amplitudes ϵ as applied in the load settings with inter- and extrapolation (right).

An extrapolation from fully developed surface boundary regions is recommended instead. The present results are based on an extrapolation with a lower bound of twenty unit cells of copper.

2.2 Influence of the bulk approximation

A typical problem in continuum mechanics comprises the modeling of both bulk and surfaces, e.g., for crystal plasticity of fine grain steel. Often, a calibrated continuum bulk model is given or already implemented and should be extended by a surface description. We implemented quadratic energy potentials in the present example. Certainly, such bulk models remain an approximation. Higher-order strain influences, for instance, originate from the atomistic potential and cannot be captured completely. Further note, that higher-order terms may be masked when material parameters are determined from averages over strain variations instead of explicitly approximating the strain dependency.

Unfortunately, the error of the bulk approximation will likely propagate to the surface model as the latter is typically set up to account for the missing energy contribution. If the bulk contribution cannot be eliminated initially (e.g., by comparing molecular simulations), an indicator for a propagating bulk error can be found in the form of unphysical strain-dependency. In the present example of copper, we found a strain-dependent equilibrium energy of the surface. Even more so, part of the strain-dependence of the material parameters (equilibrium energy, initial stresses, elastic stiffness tensor) linearly correlate with the bulk mass. Reducing the bulk mass reduced the strain sensitivity with a correlation factor of ≥ 0.999 for most parameters, except constant ones.

Such unphysical strain-dependence of the surface parameters can be eliminated by a suitable limit definition. For the present example of pure copper, we found two options to limit the propagating bulk error: reducing bulk thickness and reducing strain amplitude, both lowering the absolute error of the approximated bulk energy. Both approaches converge to the same parameters within numerical accuracy, see Fig. 1 (right).

3 Conclusion

Deriving surface continuum parameters from molecular simulations is a valuable approach. An energy-based approach guarantees a physically consistent homogenization between the atomistic and the continuum description. The determination of the model parameter must consider at least two peculiarities. First, the surface contribution cannot be isolated by a simple zero-thickness limit. It must be derived from an extrapolation that is based on fully developed boundary regions in the molecular system. Second, an approximative bulk model can cause errors for a coupled surface model. This can be identified and eliminated by a variation and reduction of the bulk mass. Still, fully developed boundary regions must be allowed in molecular simulations.

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