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Calculation of excess interfacial entropy, stress and energy for solid-liquid interfaces BRIAN B. LAIRD, Dept. of Chemistry, Univ. of Kansas, RUSLAN L. DAVIDCHACK, Dept. of Mathematics, Univ. of Leicester, UK, MARK ASTA, YANG YANG, Dept. of Chem. Eng. and Materials Sci., UC Davis — The solid-liquid interfacial free energy, $\gamma_{\rm sl}$, governs a number of important phenomena, e.g., crystal nucleation and growth, and wetting. For an equilibrium crystal-melt interface, $\gamma_{\rm sl}$ can be calculated via simulation using thermodynamic integration or capillary fluctuations [Phys. Chem. B **109**, 17802 (2005)]. The calculation of $\gamma_{\rm sl}$ away from coexistence requires the temperature and strain dependence of $\gamma_{\rm sl}$, which can be determined from the excess interfacial entropy, $\eta_{\rm sl}$, and stress tensor, $\boldsymbol{\tau}_{\rm sl}$. We determine $\eta_{\rm sl}$ and $\boldsymbol{\tau}_{\rm sl}$ for a system of Lennard-Jones particles and for particles with an inverse-power interaction $[\phi(r) = \epsilon(\sigma/r)^n]$ for $n = 6$, 8 (fcc and bcc) and 12, 20 (fcc). We determine $\eta_{\rm sl}$ and $\boldsymbol{\tau}_{\rm sl}$ for the (100), (110) and (111) orientations. We calculate $\eta_{\rm sl}$ using two methods, both using the Gibbs dividing surface defined so that the excess interfacial particle number is zero. In the first, we calculate $\eta_{\rm sl}$ from the temperature dependence of $\gamma_{\rm sl}$, $\boldsymbol{\tau}_{\rm sl}$ and the number density, ρ, along the coexistence curve. In the second, we calculate the excess interfacial energy, $e_{\rm sl}$, and use the equation $\gamma_{\rm sl} = e_{\rm sl} - T\eta_{\rm sl}$. The results agree within estimated errors. One surprising observation is that $\eta_{\rm sl}$, $e_{\rm sl}$ and $\boldsymbol{\tau}_{\rm sl}$ are significantly more anisotropic than $\gamma_{\rm sl}$.

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