

## Fluid-Fluid Interfacial Tension Using a Predictive Square Gradient Theory

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Rigorous approaches to the fluid-fluid interfacial tension (IFT), such as the family of density functional theories [1], provide predictive insight into the thermodynamics of the interface. However, such approaches are computationally intensive and scale badly with the number of components when applied to mixtures. One alternative is the square gradient theory (SGT) [1]. This reduces the computational effort but adds a free parameter, the influence parameter, CIP, making the theory non-predictive – a fact only partially mitigated by model-specific correlations of CIP. In this work we develop a predictive SGT by means of the molecular model underlying SAFT-VR Mie [2]. The free energy is described within an SGT formalism, in which CIP has a straightforward link to the direct correlation function (DCF) of the fluid. We propose a simple approximation for the DCF of a Mie segment. Together with detail of the molecular model, and by means of an effective mapping in state variables, this provides a predictive, closed-form expression for CIP, which can be used in the normal SGT formalism, together with SAFT-VR Mie, to calculate the IFT of single-segment molecules. The mapping is calibrated for a range of Mie potentials using IFT data from molecular simulations of Mie spheres. Extension of the approach to multi-segment chains, and to mixtures, is straightforward. We test our approach by predicting the IFT of Mie fluids not included in the calibration, and demonstrate its predictive power through the calculation of IFTs of multi-segment Mie-chain molecules, as well as real fluids.

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### References

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