Comment on "Multicomponent Order Parameter for Surface Melting"

Recently, Lipowsky et al.[1] described melting of a planar crystal surface using a phenomenological square-gradient Landau theory with a multicomponent order parameter (OP). Each OP component is the amplitude of a density oscillation with a reciprocal-lattice vector (RLV), $\mathbf{Q}^{\parallel}$, of the 2D lattice planes parallel to the surface, and decays in the quasiliquid film with a decay length $a(\mathbf{Q}^{\parallel})$. Within the Landau theory, the decay lengths decrease with increasing $|\mathbf{Q}^{\parallel}|$, satisfying the simple algebraic relation

$$a(\mathbf{Q}^{\parallel}) = a(0)/\{1 + [a(0)\mathbf{Q}^{\parallel}]^2\}^{1/2}. \qquad (1)$$

It is the aim of this Comment to calculate the decay lengths $a(\mathbf{Q}^{\parallel})$ within a microscopic theory. As a result, $a(\mathbf{Q}^{\parallel})$ indeed decreases with increasing $|\mathbf{Q}^{\parallel}|$. However, the relation (1) is no longer valid.

Consider a solid-liquid interface. I use a more detailed multicomponent OP: Each OP component $\rho_{\mathbf{Q}}$ is the amplitude of a density oscillation with a RLV $\mathbf{Q} = (\mathbf{Q}^{\parallel}, \mathbf{Q}^{\perp})$ of the *full* bulk crystal lattice. Then, the local density is

$$\rho(\mathbf{r}) = \rho_l + \sum_{\mathbf{Q}} \rho_{\mathbf{Q}}(z) \exp(i\mathbf{Q} \cdot \mathbf{r}), \qquad (2)$$

where z is the coordinate perpendicular to the surface plane and ρ_l is the liquid density. $\rho_{\mathbf{Q}}(z)$ is complex, in general, with $\rho_{\mathbf{Q}}^*(z) = \rho_{-\mathbf{Q}}(z)$, and fulfills the boundary conditions $\rho_{\mathbf{Q}}(z \to -\infty) = \rho_{\mathbf{Q}s}$ and $\rho_{\mathbf{Q}}(z \to \infty) = 0$ corresponding to the solid and liquid bulk values.

An asymptotic analysis which includes gradients to *arbitrary* order bilinear in the OP's shows[2] that for $z \to \infty$,

$$\rho_{\mathbf{Q}}(z) \sim \exp(ik_{\mathbf{Q}}z) \exp(-z/l_{\mathbf{Q}}), \qquad (3)$$

where $k_{\mathbf{Q}}$ denotes an oscillation mismatch between the solid and liquid density, whereas $l_{\mathbf{Q}}$ is the decay length of $\rho_{\mathbf{Q}}$ in the liquid. Let $q = u + iv$ be a complex zero of $1/S(q)$, $S(q)$ being the liquid structure factor. Then, with $w \equiv (u^2 - v^2 - Q^{\parallel 2})/2$, one has

$$l_{\mathbf{Q}}^{-2} = \min[w + (w^2 + u^2 v^2)^{1/2}]. \qquad (4)$$

Here the minimum extends over all complex zeros q whose wave vector $uv[w + (w^2 + u^2v^2)^{1/2}] + Q^{\perp}$ lies in the projection of the first Brillouin zone of the solid lattice on the z axis. $a(\mathbf{Q}^{\parallel})$ is related to $l_{\mathbf{Q}}$ via

$$a(\mathbf{Q}^{\parallel}) = \max_{Q^{\perp}}(l_{(\mathbf{Q}^{\parallel}, \mathbf{Q}^{\perp})}). \qquad (5)$$
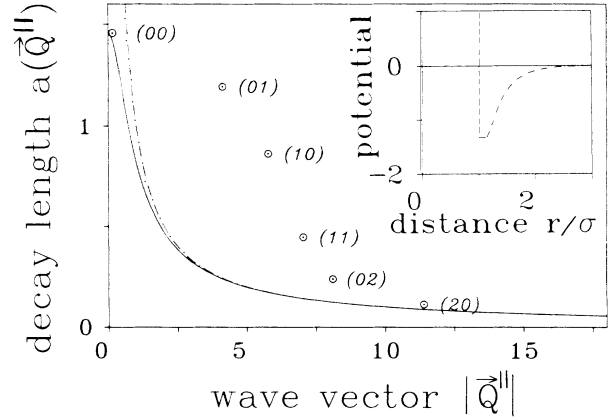

FIG. 1. Microscopically calculated decay lengths $a(\mathbf{Q}^{\parallel})$ vs $|\mathbf{Q}^{\parallel}|$, (○) for a potential with a hard core of diameter σ (see inset) near the triple point. Units are in σ^{-1} and σ. I choose an fcc lattice with first RLV $|\mathbf{Q}_1| = 6.89/\sigma$ and a (110) plane. Each point is labeled by the pair of integers (nm) resulting from $\sqrt{3}\mathbf{Q}^{\parallel}/|\mathbf{Q}_1| = n\mathbf{Q}_1^{\parallel} + m\mathbf{Q}_2^{\parallel}$, with $\mathbf{Q}_1^{\parallel} = (1, -1, 0)$, $\mathbf{Q}_2^{\parallel} = (0, 0, 1)$. For comparison, the relation (1) is shown (solid line), which, for any choice of $a(0)$, is bounded by $1/|\mathbf{Q}^{\parallel}|$ (dot-dashed line).

The zeros q can be found either by using experimental data of $S(q)$ or by assuming a microscopic interparticle potential and calculating $S(q)$ with methods from the theory of liquids (see, e.g., Ref. 3).

As an example, I choose a potential with a hard core and a short-ranged attractive part and use the optimized random-phase approximation to obtain $S(q)$. The calculated $a(\mathbf{Q}^{\parallel})$'s decrease with increasing $|\mathbf{Q}^{\parallel}|$, but, as is shown in Fig. 1, they do not fulfill the algebraic relation (1).

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