PNAS LETTER

# Reply to Fernández-Quevedo García et al.: Surface tension in phase-separated active Brownian particles

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The determination of surface tension $\gamma$ of phase-separated active Brownian particles (ABPs) has sparked considerable debate during last decade (1–4). Recently, using intrinsic pressure of ABPs, Omar et al. (3) and we (4) derived distinct expressions of active $\gamma$ by calculating the mechanical work required to create a unit area of surface through deforming the system. The difference between ref. 3 and ref. 4 essentially arises from distinct deformation strategies used. In ref. 5, García et al. observe that the surface tensions in refs. 3 and 4 obtained via continuous finite-thickness surface treatment are equivalent to choosing different dividing surfaces within Gibbs' discontinuous zero-thickness surface framework, and argue that $\gamma$ in ref. 3 comes from a firmer mechanical interpretation. Below, we respond to the observation by García et al.

1. In Gibbs' scheme (6), the exact location of dividing surface cannot be a priori determined. For phase coexistence with spatially uniform normal pressure, $\gamma$ can be expressed as the integral of difference between normal and tangential pressures and is independent of the dividing surface location. However, when normal pressure is nonuniform across the interface region (as the case of ABPs), distinct choices of dividing surface lead to different $\gamma$, as noticed in ref. 5. Moreover, the simple pressure excess-based interpretation of $\gamma$, established from uniform normal pressure, could be inaccurate. Therefore, to unambiguously determine $\gamma$ of ABPs, the interface should be considered as a continuous transition region of finite thickness [as pioneered by Van der Waals for equilibrium-state interfaces (7, 8)]. Such routes have been used to derive active $\gamma$ by refs. 3 and 4.
2. In ref. 3, Omar et al. deformed the ABP system (Fig. 1*A*), only with its volume conserved, $l_i l_y = (l_i - \delta x)(l_y + \delta y)$. The resulted $\gamma$ reads

$$\gamma = \int_0^{l_i} \left[ P(x) - P_i^T(x) \right] \mathrm{d}x, \quad [1]$$

   where $P(x)$ and $P_i^T(x)$ are the normal and tangential pressures, respectively.

   However, in their deformation protocol, the interfacial profiles of intensive quantities (e.g., density) deform accordingly and thus deviate from intrinsic interfacial profiles uniquely determined by spontaneous phase coexistence. Consequently, the interface state being probed is no longer the original one, rendering Eq. **1** improper for characterizing the true $\gamma$ of the original system. Moreover, the changed interfacial profiles are inconsistent with a quasistatic deformation process (with temperature, activity, and bulk pressures fixed), which is required for a proper derivation of $\gamma$. Notably, the $\gamma$ from Eq. **1** significantly deviates from that independently obtained from the Young–Laplace equation (4).
3. To address these inconsistencies, the deformation protocol employed in our derivation (4) maintains both volume and interfacial profiles invariant (Fig. 1*B*). This constitutes a genuine quasistatic deformation, yielding,

$$\gamma = \int_0^{l_i} \left[ P_g \frac{\rho_l - \rho_i(x)}{\rho_l - \rho_g} + P_l \frac{\rho_i(x) - \rho_g}{\rho_l - \rho_g} - P_i^T(x) \right] \mathrm{d}x, \quad [2]$$

   with $P_g$ ($P_l$) the bulk-gas (liquid) pressure, and $\rho_i(x)$ the local density. Though our expression is formally equivalent to that from Gibbs' framework with an equimolar dividing surface, our derivation is completely based on the finite-thickness surface treatment. Furthermore, the $\gamma$ from Eq. **2** agrees well with that independently measured from the Young–Laplace equation (4), strongly confirming the validity of Eq. **2**. Thus, Eq. **2** offers a proper mechanical surface tension expression for ABPs.

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The authors declare no competing interest.



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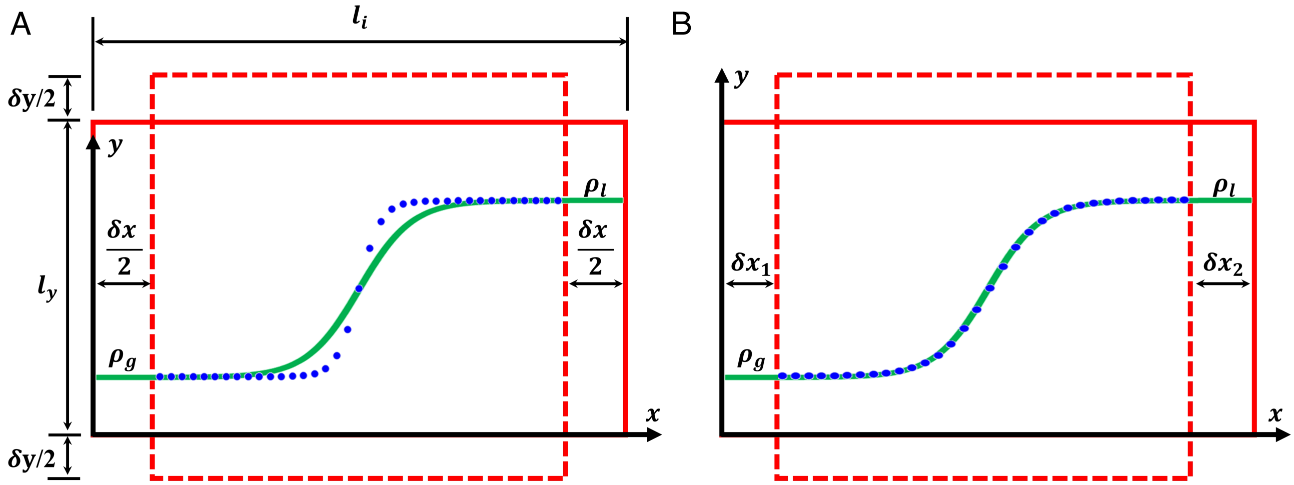


**Fig. 1.** System deformation for the derivation of $\gamma$. The deformation protocols employed in ref. 3 (*A*) and in ref. 4 (*B*). The original and deformed surface regions are outlined by solid and dashed red squares, respectively. The *x*-axis is perpendicular to the interface. The green solid and blue dotted curves, respectively, correspond to the density profiles before and after deformation. $\rho_g$ ($\rho_l$) is the bulk-gas (liquid) density.


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