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Uniform bound on the number of partitions for optimal configurations of the Ohta–Kawasaki energy in 3D

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Abstract. We study a 3D ternary system which combines an interface energy with a long-range interaction term. Several such systems were derived as a sharp-interface limit of the Nakazawa–Ohta density functional theory of triblock copolymers. Both the binary case in 2D and 3D, and the ternary case in 2D, are quite well understood, whereas very little is known about the ternary case in 3D. In particular, it is even unclear whether minimizers are made of finitely many components. In this paper, we provide a positive answer to this, by proving that the number of components in a minimizer is bounded from above by a computable quantity depending only on the total masses and the interaction coefficients. There are two key difficulties, namely, the impossibility to decouple the long-range interaction from the perimeter term, and the absence of a quantitative isoperimetric inequality with two mass constraints in 3D. Therefore, the actual shape of minimizers is unknown, even for small masses, making the construction of suitable competing configurations significantly more delicate.

1 Introduction

Energy functionals entailing a direct competition between an attractive short-range force and a repulsive Coulombic long-range force have been studied intensively in recent years, to understand physical problems such as Gamow's liquid-drop problem, and self-assembly of block copolymers. In Gamow's liquid-drop model [10], the volume of the nucleus $\Omega \subset \mathbb{R}^3$ is fixed, i.e., $|\Omega| = m$ with the parameter m being referred to as "mass." The binding energy is given by

$$\mathcal{E}_{\text{liquid}}(\Omega) := \text{Per}(\Omega) + \frac{1}{8\pi} \int_{\Omega\times\Omega} \frac{dxdy}{|x-y|},$$

where the first term is the perimeter (or surface area) of Ω, which arises due to the lower nucleon density near the nucleus boundary; the second term is a Coulomb-type one, introduced to account for the presence of positively charged protons [3].

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In Ohta and Kawasaki's diblock copolymer model [22], the free energy is given by

$$\mathcal{E}_{\text{diblock}}(\Omega) := \text{Per}(\Omega) + \gamma \int_{\Omega\times\Omega} G(x,y)\,\mathrm{d}x\mathrm{d}y,$$

where the first term, i.e., the perimeter, favors a large ball; the second term prefers splitting, and models long-range interactions between monomers due to the connectivity of different subchains in copolymer molecules. Here,

$$G(x,y) = \frac{1}{4\pi|x-y|} + R(x,y)$$

is the zero-average Green's function of the Laplace operator in $\mathbb{R}^3$, $R(x,y)$ is the regular part of $G(x,y)$, and γ is the long-range interaction coefficient, determined by the percentage of each type monomer, the total number of monomers in a chain molecule, the repulsion between different monomers, and the average distance between two adjacent monomers [6]. During each experiment, the total mass of each type monomer is fixed. So the energy is minimized under the mass constraint $|\Omega| = m$.

In this paper, we study a model in ternary systems, introduced by Nakazawa and Ohta to study triblock copolymers [21], with vanishing mass fraction. A triblock copolymer is a chain molecule consisting of three types of subchains: a subchain of type A monomers is connected to a subchain of type B monomers, and then connected to a subchain of type C monomers. Block copolymers can be used as a material in artificial organ technology and controlled drug delivery.

The free energy of triblock copolymers, for the sharp interface model in $\mathbb{R}^2$, was derived by Ren and Wei in [24, 25] as the Γ-limit of Nakazawa and Ohta's diffuse interface model:

$$\mathcal{E}_{\text{triblock}}(\Omega_1,\Omega_2) := \frac{1}{2}\sum_{i=0}^{2}\text{Per}(\Omega_i) + \sum_{i,j=1}^{2}\gamma_{ij}\int_{\Omega_i\times\Omega_j} G(x,y)\,\mathrm{d}x\mathrm{d}y.$$

Here, $\Omega_0 = (\Omega_1\cup\Omega_2)^c$, the perimeter term is defined by

$$\frac{1}{2}\sum_{i=0}^{2}\text{Per}(\Omega_i) = \sum_{0\le i<j\le 2}\mathcal{H}^2(\partial\Omega_i\cap\partial\Omega_j),$$

with ∂ denoting the (reduced) boundary, and the long-range interaction coefficients γ_{ij} form a 2×2 symmetric matrix. The choice of γ_{ij} is nontrivial, as it must ensure that the perimeter and long-range interaction terms must be of comparable order. Therefore, finding the correct order of γ_{ij} can be somewhat delicate, and can depend on the total masses. Using a "droplet" scaling argument, as done by Choksi and Peletier in [4, Section 3], [5, Section 3], and by Alama, Bronsard, the first author, and Wang in [1], it can be shown that the leading order of the free energy takes the form

$$E_0(\Omega_1,\Omega_2) = \sum_k e_0\big(|\Omega_{1,k}|,|\Omega_{2,k}|\big), \qquad \Omega_i = \bigcup_k \Omega_{i,k}, \quad i=1,2, \tag{1.1}$$

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with

$$e_0 : [0,+\infty) \times [0,+\infty) \longrightarrow \mathbb{R},$$
$$e_0(m_1,m_2) := \inf\left\{ \sum_{0\le i<j\le 2} \mathcal{H}^2(\partial\Omega_i \cap \partial\Omega_j) + \sum_{i,j=1}^{2} \frac{\Gamma_{ij}}{4\pi} \int_{\Omega_i\times\Omega_j} \frac{\mathrm{d}x\mathrm{d}y}{|x-y|} : |\Omega_i| = m_i,\ i=1,2 \right\},$$

where Γ_{ij} is a suitable scaling of γ_{ij}. That is, E_0 seeks the optimal partition $\Omega_i = \bigcup_k \Omega_{i,k}$, with each couple $(\Omega_{1,k}, \Omega_{2,k})$ minimizing e_0. Note that, generally, Ω_0 represents the "background," while Ω_i, $i = 1, 2$ represent the two types of "materials." The fact that e_0 does not penalize interactions between Ω_0 and Ω_i, $i = 1, 2$, thus, corresponds to the (rather natural) assumption that neither type of material interacts with the background.

The fact that $\Omega_{i,k}$ and $\Omega_{j,l}$ do not interact when $k \neq l$, is due to the fact that these represent masses located in different clusters. In the small volume-fraction limit, as argued in [1, 4], the interaction terms between different mass clusters become negligible compared to the interaction within the same cluster.

Choksi and Peletier showed in [4, Theorem 4.2] that, when the domain is the unit torus $\mathbb{T}^3$, in the small mass volume fraction regime, the first-order Γ-limit of the free energies (see [4, Equation (1.8)])

$$E_\eta^{3d}(v) := \begin{cases} \eta \int_{\mathbb{T}^3} |\nabla v|\mathrm{d}x + \eta \left\| v - \frac{1}{|\mathbb{T}^3|}\int_{\mathbb{T}^3} v\mathrm{d}x \right\|^2_{\mathcal{H}^{-1}(\mathbb{T}^3),} & \text{if } v \in BV(\mathbb{T}^3;\{0,\eta^{-3}\}), \\ +\infty, & \text{otherwise,} \end{cases}$$

is of the form

$$\text{perimeter} + \text{long-range interaction},$$

i.e., (see [4, Equation (4.1)], and more in general [4, Section 4]),

$$E_0^{3d}(v) := \begin{cases} \sum_{k=0}^{\infty} e_0(m_k), & \text{if } v = \sum_{k=0}^{\infty} m_k \delta_{x_k},\ \sum_{k=0}^{\infty} m_k = M = \text{total mass}, \\ +\infty, & \text{otherwise,} \end{cases}$$

with

$$e_0^{3d}(m) = \inf\left\{ \int_{\mathbb{R}^3} |\nabla z|\mathrm{d}x + \|z\|^2_{H^{-1}(\mathbb{R}^3)} : z \in BV(\mathbb{R}^3;\{0,1\}),\ \|z\|_{L^1(\mathbb{R}^3)} = M \right\}.$$

The H^{-1} norm can be made explicit:

$$\|z\|^2_{H^{-1}(\mathbb{R}^3)} = \int_{\mathbb{R}^3\times\mathbb{R}^3} G(|x-y|)z(x)z(y)\mathrm{d}x\mathrm{d}y,$$

where G denotes the Green's function of the Laplacian in $\mathbb{R}^3$. That is, the minima seeks the optimal partition, in which each component minimizes the energy e_0^{3d}. An analogous result, but for ternary systems in the two-dimensional torus, was obtained by Alama, Bronsard, the first author, and Wang in [1, Theorem 3.2].

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With the same arguments from [1, 4], it is possible to show that, again, with the domain being the unit torus $\mathbb{T}^3$, in the small mass volume fraction regime, the first-order Γ-limit of the free energies (which are the analog of [1, Equation (1.8)] for ternary systems in 3D)

$$E^{3d}_{\text{ternary},\eta}(v_{1,\eta}, v_{2,\eta}) := \begin{cases} f_\eta(v_{1,\eta}, v_{2,\eta}), & \text{if } v_{1,\eta}, v_{2,\eta} \in BV(\mathbb{T}^3; \{0, \frac{1}{\eta^3}\}), \\ +\infty, & \text{otherwise,} \end{cases}$$

$$f_\eta(v_{1,\eta}, v_{2,\eta}) := \frac{\eta}{2} \sum_{i=0}^{2} \int_{\mathbb{T}^3} |\nabla v_{i,\eta}| \mathrm{d}x + \eta^4 \sum_{i,j=1}^{2} \gamma_{ij} \int_{\mathbb{T}^3 \times \mathbb{T}^3} G_{\mathbb{T}^3}(|x-y|) v_{i,\eta}(x) v_{i,\eta}(y) \mathrm{d}x \mathrm{d}y,$$

$$G_{\mathbb{T}^3} := \text{Green's function of the Laplacian in } \mathbb{T}^3 \text{ with zero average,}$$

can be again written in the form

$$E^{3d}_{\text{ternary},0}(v_1, v_2) := \begin{cases} \sum_{k=0}^{\infty} e_0(m_{1,k}, m_{2,k}), & \text{if } v_i = \sum_{k=0}^{\infty} m_{i,k} \delta_{x_{i,k}}, \ \sum_{k=0}^{\infty} m_{i,k} = M_i, \\ +\infty, & \text{otherwise,} \end{cases} \tag{1.2}$$

$$M_i = \text{total mass of type } i \text{ constituent}, \qquad i = 1, 2,$$

and $\Gamma_{ij}\eta^{-3} = \gamma_{ij} \geq 0$ are coefficients penalizing the Coulomb interaction. Observe that the problem of minimizing $E^{3d}_{\text{ternary},0}$ is again fully determined once we fix the total masses M_i and the interaction coefficients Γ_{ij}. Each couple of sets (Ω_1, Ω_2), with the appropriate masses, and minimizing e_0, is referred to as a "cluster."

Next, we introduce the main energy of this paper: given connected sets Ω_i, with $\mathbf{1}_{\Omega_i} \in BV(\mathbb{R}^3; \{0,1\})$, $i = 1, 2$, and $|\Omega_1 \cap \Omega_2| = 0$, define the energy

$$E(\Omega_1, \Omega_2) := \sum_{0 \leq i < j \leq 2} \mathcal{H}^2(\partial\Omega_i \cap \partial\Omega_j) + \sum_{i,j=1}^{2} \gamma_{ij} \int_{\Omega_i \times \Omega_j} |x-y|^{-1} \mathrm{d}x \mathrm{d}y, \tag{1.3}$$

where $\Omega_0 = (\Omega_1 \cup \Omega_2)^c$. Here, γ_{ij} denote the interaction strengths, and are positive, of order $O(1)$. Note that E is the analog of e_0 from (1.1), (1.2), and [1], and of e_0^{3d} from [4], for ternary systems with domain $\mathbb{R}^3$. Then, given disjoint unions

$$\Big(\bigsqcup_k \Omega_{1,k}, \bigsqcup_k \Omega_{2,k} \Big),$$

with $\Omega_{i,k}$ being the connected components, the total energy of this configuration is defined by

$$\mathcal{E}\Big(\bigsqcup_k \Omega_{1,k}, \bigsqcup_k \Omega_{2,k} \Big) := \sum_k E(\Omega_{1,k}, \Omega_{2,k}).$$

Observe that $\mathcal{E}$ is the analog of [1, Equation (3.5)] and [4, Equation (4.1)], for 3D ternary systems. It is also worthy noting that $\mathcal{E}$ is similar to $\mathcal{E}_{\text{liquid}}$, $\mathcal{E}_{\text{diblock}}$, and $\mathcal{E}_{\text{triblock}}$, as they are all of the form

$$\text{perimeter} + \text{long-range interaction},$$

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with the main difference being that $\mathcal{E}$ suppresses the interaction between different connected components.

In the following, when we say "optimal configuration," unless otherwise specified, we mean a configuration $(\bigsqcup_k \Omega_{1,k}, \bigsqcup_k \Omega_{2,k})$ minimizing $\mathcal{E}$.

Existence of minimizers has been proven in [4], whose Sections 4 and 5 were dedicated to the 3D case, whereas Section 6 deal with the 2D case. In 2D, due to the fact that the Green's function is a logarithmic term, the interaction was simply the product of the masses, hence it was equivalent to minimize the perimeter, subject to two mass constraints. It is well known that the double bubble is the unique such minimizer (see, e.g., [8, 19] for the 2D case, and [12] for the 3D case, and also [7, 17, 18, 23]). In the ternary 3D case, however, such simplification is not available, and the shape of the minimizers is unclear, even for small masses. This is a significant hurdle, and studying the shape of minimizers is hindered by the lack of a quantitative isoperimetric inequality with two mass constraints in 3D

Therefore, a priori, it is even unclear whether optimal configurations have finitely many clusters, as we cannot exclude the presence of infinitely many components with very small masses. Our main result is to show that this is not the case:

Theorem 1.1 *Given total masses M_1, M_2, and interaction coefficients $\gamma_{11}, \gamma_{12}, \gamma_{22}$, there exists a computable constant $K = K(M_1, M_2, \gamma_{11}, \gamma_{22})$ such that any optimal configuration has at most K clusters.*

Existence of optimal configurations can be shown using the same arguments as in Sections 4 and 5 of [4]. The proof of Theorem 1.1 is split into several steps: first, in Lemma 2.1, we show that the number of clusters made of one constituent type is bounded from above. Then, in Lemma 2.2, we show that there is at least *one* cluster that is relatively massive, i.e., we bound from below the mass of the largest cluster. Finally, in the crucial Lemma 2.3, and the subsequent Lemma 2.4, we bound from below the mass of *any* cluster. Since there the total amount of masses is given a priori, this allows us to infer Theorem 1.1.

1.1 Notation

Since the position of the clusters is rarely relevant, in this paper, we denote by B_m, a ball of mass m.

2 Uniform upper bound on the number of clusters

The proof of Theorem 1.1 will be split over several lemmas. Throughout the entire section, M_i, $i = 1, 2$, will denote the total masses of type i constituent, and γ_{ij}, $i, j = 1, 2$ will denote the interaction coefficients. These parameters completely determine the minimization problem for $\mathcal{E}$ in 3D. All the M_i and γ_{ij} will assumed to be given, and do not change throughout the section. Our proof will proceed as follows:

(1) First, in Lemma 2.1, we bound from above the number of clusters made purely of one constituent type. Such upper bound will depend only on M_i, γ_{ii}, $i = 1, 2$.

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(2) Then, in Lemma 2.2, we show that the total mass of the largest clusters cannot be too small. Such lower bound on the mass will depend only on M_i, γ_{ii}, $i = 1, 2$.
(3) Finally, in Lemmas 2.3 and 2.4, we show that the total mass of each cluster is bounded from below by a constant depending only on M_i, γ_{ii}, $i = 1, 2$. Since there is only so much total mass (i.e., $M_1 + M_2$), this allows us to infer Theorem 1.1.

As we have no information on the shape of optimal configurations, we will often compare their energy against that of a suitable standard double bubble. Further information about the geometry of standard double bubbles are available in the Appendix.

Lemma 2.1 *Consider an optimal configuration, made of clusters* $(\Omega_{1,k}, \Omega_{2,k})$, $k \geq 1$. *Then*

$$\#\{k : |\Omega_{1,k}||\Omega_{2,k}| = 0\}$$

is bounded from above by a constant depending only on M_i, γ_{ii}, $i = 1, 2$.

Proof The proof follows arguments similar to those used to show the sub-addictivity formula (2.1) in [16]. It is well known (see, e.g., [2, 9, 13–15, 20], and the references therein) that there exist $m_{i,B} = m_{i,B}(\gamma_{ii}) > 0$, $i = 1, 2$, such that, for all $m \leq m_{i,B}(\gamma_{ii})$, the minimizer of

$$\inf_{|X|=m} \left\{ \mathcal{H}^2(\partial X) + \gamma_{ii} \int_{X\times X} |x - y|^{-1} \mathrm{d}x\mathrm{d}y \right\}$$

is given by B_m. Since $\mathcal{H}^2(\partial B_m)$ (resp. $\int_{X\times X} |x - y|^{-1}\mathrm{d}x\mathrm{d}y$) scales like $m^{2/3}$ (resp. $m^{5/3}$), the perimeter term is dominating for all sufficiently small masses. Thus, there exist geometric constants $m_{i,S} = m_{i,S}(\gamma_{ii}) \leq m_{i,B}(\gamma_{ii})$ such that

$$\begin{aligned} \mathcal{H}^2(\partial B_{m_1}) + \gamma_{ii} \int_{B_{m_1}\times B_{m_1}} |x - y|^{-1}\mathrm{d}x\mathrm{d}y \\ + \mathcal{H}^2(\partial B_{m_2}) + \gamma_{ii} \int_{B_{m_2}\times B_{m_2}} |x - y|^{-1}\mathrm{d}x\mathrm{d}y \\ > \mathcal{H}^2(\partial B_{m_1+m_2}) + \gamma_{ii} \int_{B_{m_1+m_2}\times B_{m_1+m_2}} |x - y|^{-1}\mathrm{d}x\mathrm{d}y, \end{aligned}$$

for all $m_1, m_2 \leq m_{i,S}(\gamma_{ii})$, i.e., combining the two balls is energetically favorable whenever $m_1, m_2 \leq m_{i,S}(\gamma_{ii})$. Thus, we cannot have two balls of the type i constituent, both with masses less than $m_{i,S}(\gamma_{ii})$. Since the total mass is $M_1 + M_2 < +\infty$, the proof is complete. ∎

Lemma 2.2 *Consider an optimal configuration, made of clusters* $(\Omega_{1,k}, \Omega_{2,k})$, $k \geq 1$. *Then*

$$m_i^+ := \sup_k m_{i,k}, \quad m_{i,k} := |\Omega_{i,k}|, \qquad i = 1, 2,$$

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is bounded from below by

$$\min\left\{\frac{M_i}{2}, \left(\frac{\sqrt[3]{36\pi}M_i}{4\sum_{i=1}^{2}[\sqrt[3]{36\pi}M_i^{2/3} + \gamma_{ii}\int_{B_{M_i}\times B_{M_i}}|x-y|^{-1}dxdy]}\right)^3\right\}, \qquad i = 1, 2.$$

Note that, curiously, this lower bound is independent of γ_{12}. As it will be clear from the proof, this is due to the fact that an upper bound for the energy of an optimal configuration is given by the energy of two balls of masses M_1 and M_2, respectively. Such bound is clearly independent of γ_{12}.

Proof The idea is that, for very small masses, the perimeter term is sub-addictive and dominating. Assume $m_i^+ \le M_i/2$, as otherwise $M_i/2$ is already a lower bound. Note that

$$E(\Omega_{1,k}, \Omega_{2,k}) \ge \mathcal{S}(m_{1,k}, m_{2,k}) \qquad \forall k \ge 1,$$

where

(2.1)

$$\mathcal{S}(m_1, m_2) = \text{perimeter of the standard double bubble with masses } m_1 \text{ and } m_2,$$

and, by [11, Theorem 4.2] (applied with $v_1 = m_1$, $x = v_2 = m_2$, $n = 3$), we get the following isoperimetric inequality for clusters

$$\mathcal{S}(m_1, m_2) \ge \frac{\sqrt[3]{36\pi}}{2}\sum_{i=1}^{2} m_i^{2/3}.$$

Thus, the total energy of our optimal configuration satisfies

$$\sum_{k\ge1} E(\Omega_{1,k}, \Omega_{2,k}) \ge \frac{\sqrt[3]{36\pi}}{2}\sum_{i=1}^{2}\sum_{k\ge1} m_{i,k}^{2/3}.$$

By the concavity of the function $t \mapsto t^{2/3}$, the sum $\sum_{k\ge1} m_{i,k}^{2/3}$ is minimum when $m_{i,k} \in \{0, m_i^+\}$ for all k. Since $\sum_{k\ge1} m_{i,k} = M_i$, there are at least $\lfloor \frac{M_i}{m_i^+} \rfloor$ many clusters containing type i constituents, thus

$$\begin{aligned}\sum_{k\ge1} E(\Omega_{1,k}, \Omega_{2,k}) &\ge \frac{\sqrt[3]{36\pi}}{2}\sum_{i=1}^{2}\sum_{k\ge1} m_{i,k}^{2/3} \ge \frac{\sqrt[3]{36\pi}}{2}\sum_{i=1}^{2}\left\lfloor\frac{M_i}{m_i^+}\right\rfloor (m_i^+)^{2/3}\\ &\ge \frac{\sqrt[3]{36\pi}}{2}\sum_{i=1}^{2}\frac{M_i - m_i^+}{(m_i^+)^{1/3}} \ge \frac{\sqrt[3]{36\pi}}{4}\sum_{i=1}^{2}\frac{M_i}{(m_i^+)^{1/3}}.\end{aligned}$$

Since our configuration was an optimal one, its energy does not exceed that of two balls, which we denote by B_{M_1} and B_{M_2}, of masses M_1 and M_2, respectively. Thus, the above line continues as

$$\begin{aligned}\frac{\sqrt[3]{36\pi}}{4}\sum_{i=1}^{2}\frac{M_i}{(m_i^+)^{1/3}} &\le \sum_{k\ge1} E(\Omega_{1,k}, \Omega_{2,k})\\ &\le \sum_{i=1}^{2}\left[\sqrt[3]{36\pi}M_i^{2/3} + \gamma_{ii}\int_{B_{M_i}\times B_{M_i}}|x-y|^{-1}\mathrm{d}x\mathrm{d}y\right],\end{aligned}$$

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hence

$$(m_i^+)^{1/3} \geq \frac{\sqrt[3]{36\pi}}{4} \frac{M_i}{\sum_{i=1}^2 [\sqrt[3]{36\pi} M_i^{2/3} + \gamma_{ii} \int_{B_{M_i} \times B_{M_i}} |x-y|^{-1} \mathrm{d}x\mathrm{d}y]},$$

and the proof is complete. ∎

Lemma 2.3 *Consider an optimal configuration, made of clusters* $(\Omega_{1,k}, \Omega_{2,k})$, $k \geq 1$. *Assume* $\sup_k |\Omega_{1,k}|$ *and* $\sup_k |\Omega_{2,k}|$ *are achieved on different clusters, i.e., without loss of generality,*

$$|\Omega_{1,1}| = m_1^+ = \sup_k |\Omega_{1,k}|, \qquad |\Omega_{2,2}| = n_2^+ = \sup_k |\Omega_{2,k}|.$$

Then

$$\inf_k \sum_{i=1}^2 |\Omega_{i,k}|$$

is bounded from below by a constant depending only on M_i, γ_{ii}, $i = 1, 2$.

Proof Consider a cluster $(\Omega_{1,k}, \Omega_{2,k})$, with $k \geq 3$, and let

$$m_2 := |\Omega_{2,1}|, \qquad n_1 := |\Omega_{1,2}|, \qquad \varepsilon_i := |\Omega_{i,k}| > 0, \ i = 1, 2.$$

Note that $m_1^+ \geq n_1$, $n_2^+ \geq m_2$. The construction will be slightly different depending on the values of $\frac{m_1^+}{m_2}$, $\frac{m_2}{n_2^+}$, and $\frac{\varepsilon_1}{\varepsilon_2}$.

Case 1: $\frac{m_1^+}{m_2} \geq \frac{\varepsilon_1}{\varepsilon_2}$. Consider the competitor constructed in the following way (see Figure 1).

- Move mass ε_1 (resp. rm_2, with $r := \frac{\varepsilon_1}{m_1^+} \leq 1$) of type I (resp. type II) constituent from the cluster $(\Omega_{1,k}, \Omega_{2,k})$ to $(\Omega_{1,1}, \Omega_{2,1})$. This is possible, since we are discussing the case $\frac{m_1^+}{m_2} \geq \frac{\varepsilon_1}{\varepsilon_2}$, i.e., $rm_2 = \varepsilon_1 \frac{m_2}{m_1^+} \leq \varepsilon_2$.
- Replace $(\Omega_{1,k}, \Omega_{2,k})$ and $(\Omega_{1,1}, \Omega_{2,1})$ with $B_{\varepsilon_2 - rm_2}$ (of type II constituent) and $(\tilde{\Omega}_{1,1}, \tilde{\Omega}_{2,1}) := (1+r)^{1/3}(\Omega_{1,1}, \Omega_{2,1})$, while every other cluster remains unaltered.

Now, we estimate the change in energy. Since our initial configuration was optimal,

$$\begin{aligned} 0 \leq {} & E((1+r)^{1/3}(\Omega_{1,1}, \Omega_{2,1})) + E(\emptyset, B_{\varepsilon_2 - rm_2}) \\ & - E(\Omega_{1,1}, \Omega_{2,1}) - E(\Omega_{1,k}, \Omega_{2,k}). \end{aligned} \tag{2.2}$$

By a straightforward scaling argument,

$$\begin{aligned} & E((1+r)^{1/3}(\Omega_{1,1}, \Omega_{2,1})) \\ & \quad = (1+r)^{2/3} \sum_{0 \leq i < j \leq 2} \mathcal{H}^2(\partial\Omega_{i,1} \cap \partial\Omega_{j,1}), \qquad \Omega_{0,1} := (\Omega_{1,1} \cup \Omega_{2,1})^c, \\ & \qquad + (1+r)^{5/3} \sum_{i,j=1}^2 \gamma_{ij} \int_{\Omega_{i,1} \times \Omega_{j,1}} |x-y|^{-1} \mathrm{d}x\mathrm{d}y \end{aligned}$$

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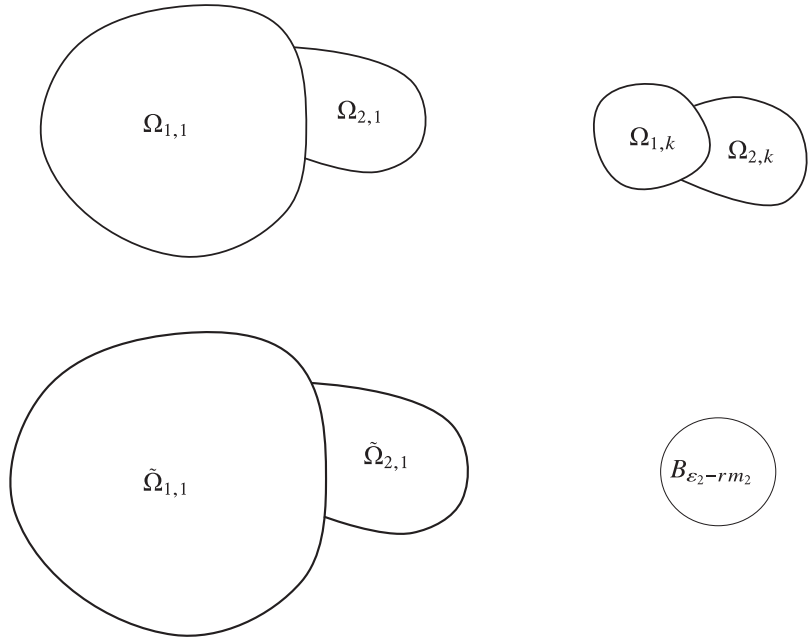

Figure 1: Schematic representation of the construction of the competitor: original clusters (top), and modified clusters (bottom). Though the objects in question are three-dimensional, for better clarity, we represented the construction in two dimensions. Only the affected clusters are represented here. The clusters are drawn deliberately deformed, to emphasize the fact that we do not know the clusters' precise shapes.

$$\leq (1+r)\sum_{0\leq i<j\leq 2}\mathcal{H}^2(\partial\Omega_{i,1}\cap\partial\Omega_{j,1}) + (1+3r)\sum_{i,j=1}^{2}\gamma_{ij}\int_{\Omega_{i,1}\times\Omega_{j,1}}|x-y|^{-1}\mathrm{d}x\mathrm{d}y$$
$$\leq (1+3r)\underbrace{\left[\sum_{0\leq i<j\leq 2}\mathcal{H}^2(\partial\Omega_{i,1}\cap\partial\Omega_{j,1}) + \sum_{i,j=1}^{2}\gamma_{ij}\int_{\Omega_{i,1}\times\Omega_{j,1}}|x-y|^{-1}\mathrm{d}x\mathrm{d}y\right]}_{=E(\Omega_{1,1},\Omega_{2,1})},$$

where we used the estimates

$$(1+r)^{2/3}\leq 1+r\leq 1+3r, \qquad (1+r)^{5/3}\leq (1+r)^2 \overset{(r\leq 1)}{\leq} 1+3r.$$

Thus, in view of Lemma 2.2,

$$\begin{aligned} &E((1+r)^{1/3}(\Omega_{1,1},\Omega_{2,1})) - E(\Omega_{1,1},\Omega_{2,1}) \\ &\qquad\leq 3rE(\Omega_{1,1},\Omega_{2,1}) \leq \varepsilon_1 H_1(M_1,M_2,\gamma_{11},\gamma_{22}), \end{aligned} \tag{2.3}$$

$$H_1(M_1,M_2,\gamma_{11},\gamma_{22}) := \sum_{i=1}^{2}\frac{3}{m_1^+}\left[\sqrt[3]{36\pi}M_i^{2/3} + \gamma_{ii}\int_{B_{M_i}\times B_{M_i}}|x-y|^{-1}\mathrm{d}x\mathrm{d}y\right].$$

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Now, we estimate $E(\emptyset, B_{\varepsilon_2 - rm_2}) - E(\Omega_{1,k}, \Omega_{2,k})$:

$$\begin{aligned} E(\emptyset, B_{\varepsilon_2 - rm_2}) - E(\Omega_{1,k}, \Omega_{2,k}) &\le \mathcal{S}(0, \varepsilon_2 - rm_2) - \mathcal{S}(\varepsilon_1, \varepsilon_2) \\ &= \mathcal{S}(0, \varepsilon_2 - rm_2) - \mathcal{S}(\varepsilon_1, \varepsilon_2 - rm_2) + \mathcal{S}(\varepsilon_1, \varepsilon_2 - rm_2) - \mathcal{S}(\varepsilon_1, \varepsilon_2) \\ &\le -c_1 \varepsilon_1^{2/3}, \qquad c_1 := \frac{\sqrt[3]{36\pi}}{2}, \end{aligned}$$

where the last line is due to [11, Theorem 3.2], which gives

$$\mathcal{S}(\varepsilon_1, \varepsilon_2 - rm_2) - \mathcal{S}(\varepsilon_1, \varepsilon_2) \le 0,$$

and [11, Theorem 4.2] (applied with $\nu_1 = \varepsilon_1$, $x = \nu_2 = \varepsilon_2 - rm_2$, $n = 3$), which gives

$$\begin{aligned} \mathcal{S}(\varepsilon_1, \varepsilon_2 - rm_2) &\ge \frac{\sqrt[3]{36\pi}}{2} \left[\varepsilon_1^{2/3} + (\varepsilon_2 - rm_2)^{2/3} + (\varepsilon_1 + \varepsilon_2 - rm_2)^{2/3} \right] \\ &\ge \frac{\sqrt[3]{36\pi}}{2} \left[\varepsilon_1^{2/3} + 2(\varepsilon_2 - rm_2)^{2/3} \right] = \frac{\sqrt[3]{36\pi}}{2} \varepsilon_1^{2/3} + \underbrace{\sqrt[3]{36\pi} (\varepsilon_2 - rm_2)^{2/3}}_{= \mathcal{S}(0, \varepsilon_2 - rm_2)}. \end{aligned}$$

Combining with (2.2) and (2.3) gives the necessary condition

$$\begin{aligned} 0 &\le E((1+r)^{1/3}(\Omega_{1,1}, \Omega_{2,1})) + E(\emptyset, B_{\varepsilon_2 - rm_2}) - E(\Omega_{1,1}, \Omega_{2,1}) - E(\Omega_{1,k}, \Omega_{2,k}) \\ &\le \varepsilon_1 H_1(M_1, M_2, \gamma_{11}, \gamma_{22}) - c_1 \varepsilon_1^{2/3}, \end{aligned} \tag{2.4}$$

hence

$$\varepsilon_1^{1/3} \ge H_1(M_1, M_2, \gamma_{11}, \gamma_{22}) c_1^{-1},$$

thus, completing the proof for this case.

Case 2: $\frac{n_2^+}{n_1} \ge \frac{\varepsilon_2}{\varepsilon_1}$. The competitor constructed in a way similar to the previous case.

- Move mass ε_2 (resp. rn_1, with $r := \frac{\varepsilon_2}{n_2^+} \le 1$) of type II (resp. type I) constituent from the cluster $(\Omega_{1,k}, \Omega_{2,k})$ to $(\Omega_{1,2}, \Omega_{2,2})$. This is possible since we are discussing the case $\frac{n_2^+}{n_1} \ge \frac{\varepsilon_2}{\varepsilon_1}$, i.e., $rn_1 = \varepsilon_2 \frac{n_1}{n_2^+} \le \varepsilon_1$.
- Replace $(\Omega_{1,k}, \Omega_{2,k})$ and $(\Omega_{1,2}, \Omega_{2,2})$ with $B_{\varepsilon_1 - rn_1}$ (of type I constituent) and $(1+r)^{1/3}(\Omega_{1,2}, \Omega_{2,2})$, while every other cluster remains unaltered.

Then the proof proceeds like in the previous case. With the same arguments from Case 1, we obtain

$$\begin{aligned} &E((1+r)^{1/3}(\Omega_{1,2}, \Omega_{2,2})) - E(\Omega_{1,2}, \Omega_{2,2}) \le 3rE(\Omega_{1,2}, \Omega_{2,2}) \le \varepsilon_2 H_2(M_1, M_2, \gamma_{11}, \gamma_{22}), \\ &H_2(M_1, M_2, \gamma_{11}, \gamma_{22}) := \sum_{i=1}^{2} \frac{3}{n_2^+} \left[\sqrt[3]{36\pi} M_i^{2/3} + \gamma_{ii} \int_{B_{M_i} \times B_{M_i}} |x - y|^{-1} \mathrm{d}x \mathrm{d}y \right], \end{aligned}$$

which is the analog of (2.3), and

$$\begin{aligned} 0 &\le E((1+r)^{1/3}(\Omega_{1,2}, \Omega_{2,2})) + E(\emptyset, B_{\varepsilon_1 - rn_1}) - E(\Omega_{1,2}, \Omega_{2,2}) - E(\Omega_{1,k}, \Omega_{2,k}) \\ &\le \varepsilon_2 H_2(M_1, M_2, \gamma_{11}, \gamma_{22}) - c_2 \varepsilon_2^{2/3}, \end{aligned}$$

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for some computable, purely geometric constant $c_2 > 0$, which is the analog of (2.4). Thus

$$\varepsilon_2^{1/3} \geq H_2(M_1, M_2, \gamma_{11}, \gamma_{22}) c_2^{-1},$$

concluding the proof for this case.

Finally, note that the above two cases are exhaustive: if Case 1 does not hold, i.e., $\frac{\varepsilon_2}{\varepsilon_1} < \frac{m_2}{m_1^+}$, using $m_1^+ \geq n_1$, $n_2^+ \geq m_2$, we get

$$\frac{\varepsilon_2}{\varepsilon_1} < \frac{m_2}{m_1^+} \leq \frac{n_2^+}{n_1},$$

i.e., Case 2 holds. The proof is thus complete. ∎

Lemma 2.4 *Consider an optimal configuration, made of clusters* $(\Omega_{1,k}, \Omega_{2,k})$, $k \geq 1$. *Assume* $\sup_k |\Omega_{1,k}|$ *and* $\sup_k |\Omega_{2,k}|$ *are achieved on the same clusters, i.e., without loss of generality,*

$$|\Omega_{i,1}| = m_i^+ = \sup_k |\Omega_{i,k}|, \quad i = 1, 2.$$

Then

$$\inf_k \sum_{i=1}^{2} |\Omega_{i,k}|$$

is again bounded from below by a constant depending only on M_i, γ_{ii}, $i = 1, 2$.

Proof We rely on Lemma 2.3: Consider another cluster $(\Omega_{1,k}, \Omega_{2,k})$, $k \geq 2$. Let $|\Omega_{1,k}| = \varepsilon_1 > 0$, $|\Omega_{2,k}| = \varepsilon_2 > 0$, and note that one of the following cases must hold.

(1) If $\frac{m_1^+}{m_2^+} \geq \frac{\varepsilon_1}{\varepsilon_2}$, then we can use the construction from Case 1 of Lemma 2.3.

(2) If $\frac{m_1^+}{m_2^+} \leq \frac{\varepsilon_1}{\varepsilon_2}$, i.e., $\frac{m_2^+}{m_1^+} \geq \frac{\varepsilon_2}{\varepsilon_1}$, then we can use the construction from Case 2 of Lemma 2.3.

The proof is thus complete. ∎

A Appendix: geometry of the standard double bubble

In [12], it was shown that the three-dimensional standard double bubbles has the least surface area among all sets enclosing two regions of given volumes.

Geometrically, the standard double bubble is a surface of revolution, with all the three surfaces being part of spheres, meeting at 120 degrees (see Figures A.1 and A.2).

Below, we collect several results, used in the proof of Theorem 1.1, on the function $\mathfrak{S}$ introduced in (2.1).

Lemma A.1 *[11, Theorem 3.2] The function* $\mathfrak{S}$ *is strictly concave: given* $m_i, n_i \geq 0$, $i = 1, 2$, *it holds*

$$\mathfrak{S}((1-t)m_1 + tn_1, (1-t)m_2 + tn_2) > (1-t)\mathfrak{S}(m_1, m_2) + t\mathfrak{S}(n_1, n_2)$$

for all $t > 0$.

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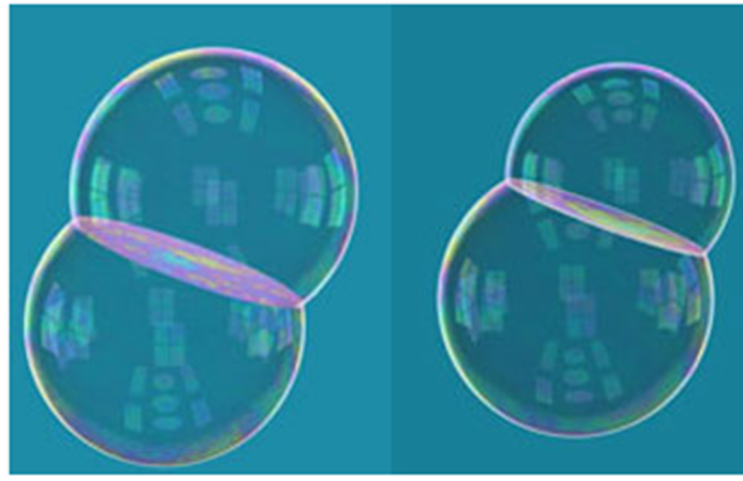

Figure A.1: The standard double bubble in $\mathbb{R}^3$: if the two bubbles that meet have equal volumes, the shared surface between them is a flat disk. But in the case of unequal volumes, the smaller bubble, given its larger internal pressure, will bow slightly into the larger bubble. In either scenario, the two bubbles always meet at angles of 120 degrees. Credit: John M. Sullivan, Technical University of Berlin and University of Illinois at Urbana–Champaign.

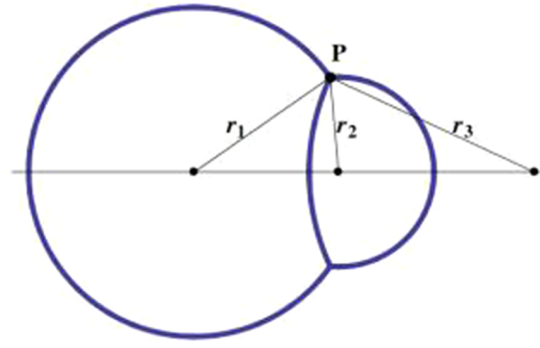

Figure A.2: Cross section of a standard double bubble.

Lemma A.2 *[11, Corollary 3.3] The function $\mathcal{S}(m_1, m_2)$ is increasing in both variables.*

Lemma A.3 *[11, Theorem 4.2] Suppose that in a minimal enclosure of volumes m_1 and m_2 in $\mathbb{R}^3$, with the latter having a connected component with volume $x > 0$. Then*

$$\frac{2\mathcal{S}(m_1, m_2)}{c_1} \geq m_2 x^{-1/3} + m_1^{2/3} + (m_1 + m_2)^{2/3},$$

$$c_1 := \sqrt[3]{36\pi} = \text{surface area of the unit ball in } \mathbb{R}^3.$$

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