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Droplet Minimizers for the Cahn-Hilliard Free Energy Functional

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ABSTRACT. We prove theorems characterizing the minimizers for the Cahn-Hilliard free energy functional, which is used to describe the liquid vapor phase transition (or the 2 state magnetization transition). In particular, we exactly determine the critical density for droplet formation, and the geometry of the droplets.

1. Introduction

1.1. The variational problem

Let Ω be the d-dimensional square torus with volume L^d. Consider the free energy functional $\mathcal{F}(m)$ defined by

$$\mathcal{F}(m) = \frac{\theta^2}{2}\int_\Omega |\nabla m|^2\,\mathrm{d}x + \int_\Omega F(m)\,\mathrm{d}x\ , \tag{1.1}$$

where θ is a parameter with the units of length, and

$$F(m) = \frac{1}{4}\left(m^2 - 1\right)^2\ .$$

This is a "double well" potential with minima at $m = \pm 1$. The two minima are the two "phases" of the system. The function $m(x)$ is an "order parameter field," representing a summary of the microscopic information about the underlying system locally at the point x that is necessary to compute a density for the Helmholz free energy. The particular free energy density considered here is phenomenological; it does not arise from any particular microscopic model, but it is

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a simple caricature of the Helmholz free energy densities that do arise from scaling limits of actual microscopic systems with phase transitions. See [9] for discussion on the relation with the microscopic models. The free energy functional (1.1) is frequently called the Cahn-Hilliard free energy functional, but it has a fairly ancient history. It was already discussed by van der Walls [19] in the nineteenth century.

For any number n with $-1 < n < 1$, define the *minimal free energy function* $f_L(n)$ by

$$f_L(n) = \inf \left\{ \mathcal{F}(m) \ : \ \frac{1}{L^d} \int_\Omega m(x)\,\mathrm{d}x = n \right\} . \tag{1.2}$$

In what follows we shall work in units with

$$\theta = 1 .$$

This makes L and x dimensionless. That is, we are implicitly introducing dimensionless coordinates $\tilde{x} = x/\theta$ and a dimensionless scale parameter $\tilde{L} = L/\theta$. However, setting $\theta = 1$, we can drop the tildes, and work directly in dimensionless coordinates. The central problem under discussion here is what the minimizers look like.

If we fix a value of n with $-1 < n < 1$, the minimization problem (1.2) simplifies as L becomes very large: In the bulk of Ω it must be the case that $m(x) \approx \pm 1$ to high accuracy. Consider any order parameter field $m(x)$ that takes on only the two pure phase values ± 1. Let V_+ be the volume of the region in which $m(x) = 1$. While such an order parameter field would be discontinuous, and would therefore yield an infinite free energy, the quantity V_+ is quite relevant to (1.2) for large L. From the constraint $\int_\Omega m(x)\,\mathrm{d}x = nL^d$, $V_+ - (L^d - V_+) = nL^d$ so that V_+ and n are related by

$$V_+ = \frac{n+1}{2} L^d \qquad \text{and} \qquad n = -1 + 2\frac{V_+}{L^d} . \tag{1.3}$$

Any order parameter field $m(x)$ that is a minimizer for (1.2) will be continuous, and in fact, C^∞ as a consequence of the Euler-Lagrange equation that it must satisfy. Define the interface Γ between the two phases by

$$\Gamma = \{ x \ : \ m(x) = 0 \} .$$

Let $|\Gamma|$ denote $\mathcal{H}^{d-1}(\Gamma)$, the $d-1$ dimensional Hausdorff measure of Γ. Because of the gradient terms in the free energy functional, a minimizer m cannot make a sudden transition for $+1$ to -1 in crossing Γ, and so one would expect to pay a price for making such a transition that is proportional to $|\Gamma|$. That is, we would expect that

$$f_L(n) \approx S|\Gamma| \tag{1.4}$$

for some proportionality constant S, which is called the *surface tension.* For the physics behind this terminology and this approximation, see [18].

The approximation (1.4) becomes more and more accurate in the *sharp interface limit* $\theta/L \to 0$. In this limit, the minimization problem reduces to an isoperimetric problem [13], [14], as has been rigorously proved in the Γ-convergence framework by Mortola, Modica, and Luckhaus.

Though we do not study the sharp interface limit, the isoperimetric inequality will play an important role in our analysis. The isoperimetry problem in the torus is complicated by the fact that it can be advantageous to arrange a volume V so that it "wraps around" the torus, and reduces the perimeter below what it would be in the Euclidean space of the same dimension. However, for smaller volumes, the optimal geometry for the isoperimetric inequality is still a ball. For $d > 2$, this is a surprinsigly recent result, due to Morgan and Johnson [15]. The Appendix to this article contains a discussion of isoperimetry on the torus which recalls these results, and explains several others that we use. Roughly, the result of Morgan and Johnson provides a constant $\epsilon > 0$ so that with V denoting an open set in Ω, and $P(V)$ its perimeter,

$$|V| < \epsilon L^d \quad \Rightarrow \quad P(V) \geq \sigma_d \left(|V| / (\sigma_d/d)\right)^{1-1/d} \tag{1.5}$$

In what follows, we shall be concerned with values of n sufficiently close to -1, so that, with $V = V_+$, as given by (1.3), the minimal bounding surface is a sphere.

The *equimolar radius,* r_0, is defined to be the radius of the sphere whose enclosed volume is V_+, where V_+ is given in terms of n by (1.3). Since $(\sigma_d/d)r_0^d = V_+$, we have from (1.3) that

$$r_0 = \left(\frac{d}{2\sigma_d}(n+1)\right)^{1/d} L \quad = \left(\frac{V_+}{\sigma_d/d}\right)^{1/d} . \tag{1.6}$$

Here we shall only consider values of n for which

$$r_0 < \epsilon L \tag{1.7}$$

where ϵ is sufficiently small to ensure that a sphere of radius ϵL has minimal perimeter for the volume it encloses in the torus. The Appendix provides a lower bound on ϵ.

1.2. Two simple trial functions

Under the condition (1.7) on n and L, there are two natural trial functions to consider for the variational problem (1.2).

The first of these is the *equimolar droplet trial function*

$$m_{\text{emd}}(x) = m_0\left(|x| - r_0(n)\right) , \tag{1.8}$$

where we are representing Ω as the centered cube in $\mathbb{R}^d$ with side length L and periodic boundary conditions, and $m_0(z)$ is some function such that $\lim_{z\to\pm\infty} m_0(z) = \mp 1$, with the transition from $+1$ to -1 being made in such a way as to minimize the cost in free energy. In fact, we require that the limits $\lim_{z\to\pm\infty} m_0(z) = \mp 1$ are achieved at finite values of z such that (1.8) does indeed define a smooth function on Ω.

The second of these is the *uniform trial function*

$$m_{\text{uni}}(x) = n , \tag{1.9}$$

corresponding to a "supersaturated" state with the order parameter strictly between the minimizing values.

Accepting the validity of the approximation (1.4), we have $\mathcal{F}(m_{\text{emd}}) \approx S\sigma_d r_0^{d-1}$, and therefore, from (1.6)

$$\mathcal{F}(m_{\text{emd}}) \;\approx\; S\sigma_d \left(\frac{d}{2\sigma_d}(n+1)\right)^{1-1/d} L^{d-1} \;=\; S\sigma_d \left(\frac{V_+}{\sigma_d/d}\right)^{1-1/d} , \tag{1.10}$$

one easily computes that

$$\mathcal{F}(m_{\text{uni}}) \;=\; \frac{1}{4}\big(n^2-1\big)^2 L^d \;=\; 4\frac{V_+^2}{L^d}\left(1-\frac{V_+}{L^d}\right)^2 . \tag{1.11}$$

Which of these trial functions provides a better description of the minimizers in (1.2)? That depends on n, or what is the same, on the ratio V_+/L^d. In fact, there are now two obvious scaling regimes to consider: We can take L to infinity while keeping either n or V_+ constant. For this reason, we have expressed $\mathcal{F}(m_{\text{uni}})$ and $\mathcal{F}(m_{\text{emd}})$ in terms of both n and V_+.

If one holds n constant, and takes $L \to \infty$, then m_{emd} does much better than m_{uni}. On the other hand, if one holds V_+ constant as L tends to infinity, we see from (1.11) and (1.10) that m_{uni} does much better than m_{emd} for large L: This suggests that a droplet of the $+1$ phase will always "evaporate" into the surrounding -1 phase if the ambient volume $|\Omega|$ is sufficiently large compared to V_+.

1.3. The critical scaling regime

The situation is much more interesting if one considers $f_L(n)$ with n tending towards -1 at the same time that L tends to infinity: We seek the smallest value of $n(L)$ for which droplets are stable in a box of volume L^d, and seek also to determine the structure of such critical minimizing droplets.

In this sort of scaling regime, V_+/L^d will be very small, and we can express $\mathcal{F}(m_{\text{uni}})$ in more physically meaningful terms as follows: Define the *compressibility* χ by

$$\chi = \frac{1}{F''(-1)} . \tag{1.12}$$

Then since $F(-1) = F'(-1) = 0$,

$$F(n) = F\big(-1 + 2V_+/L^d\big) \approx \frac{1}{2\chi}\left(\frac{2V_+}{L^d}\right)^2 ,$$

this gives us the approximation

$$\mathcal{F}(m_{\text{uni}}) \approx \frac{1}{2\chi}\,\frac{4V_+^2}{L^d} . \tag{1.13}$$

Of course, in our problem, $\chi = 1/2$. But introducing the compressibility highlights a competition between surface and bulk terms in minimizing the free energy.

When $V_+^{1+1/d} \asymp L^d$,

$$\mathcal{F}(m_{\text{emd}}) \approx S\sigma_d\left(\frac{V_+}{\sigma_d/d}\right)^{1-1/d} \qquad \text{and} \qquad \mathcal{F}(m_{\text{uni}}) \approx \frac{2}{\chi}\,\frac{V_+^2}{L^d}$$

are comparable. For this reason, we refer to $V_+^{1+1/d} \asymp L^d$ as the *critical scaling regime.* In terms of n and the equimolar radius r_0, the critical scaling regime is characterized by

$$n+1 \asymp L^{-d/(d+1)} \qquad \text{or, equivalently} \qquad r_0 \asymp L^{d/(d+1)} . \tag{1.14}$$

What should one expect for the minimizing free energy in the critical scaling regime, and will the minimizers be given by some sort of droplet, or not?

In a recent and incisive investigation of droplet formation in two-dimensional Ising model [4], Biskup, Chayes, and Kotecky proposed that to answer this question, one should introduce a *volume fraction* η, and put ηV_+ into the drop, and $(1-\eta)V_+$ into the uniform background. They then constructed a phenomenological thermodynamic free energy function $\Phi(\eta)$ which is the sum of the surface tension term and the uniform background term:

$$\Phi(\eta) = S\sigma_d \left(\frac{\eta V_+}{\sigma_d/d}\right)^{1-1/d} + \frac{1}{2\chi}\frac{(1-\eta)^2 V_+^2}{L^d} \ . \tag{1.15}$$

Here, $0 \le \eta \le 1$, and the suggestion in [4] is that in great generality, one can resolve a competition between surface and bulk energy effects by choosing η to minimize Φ. Defining $C(n)$ by

$$C(n) = \frac{\sigma_d}{2\chi S}\left(\frac{2}{d}\right)^2 \left(\frac{r_0^{d+1}}{L^d}\right) = \frac{2}{d\chi S}\left(\frac{\sigma_d}{d}\right)^{-1/d}\left(\frac{n+1}{2}\right)^{(d+1)/d} L \tag{1.16}$$

and $|\Gamma_0|$ by $|\Gamma_0| = \sigma_d r_0^{d-1}$, the quantity in (1.15) can be written as

$$\Phi(\eta) = S|\Gamma_0|\left(\eta^{1-1/d} + C(n)(1-\eta)^2\right) \ .$$

Notice that

$$\frac{\Phi(\eta)-\Phi(0)}{S|\Gamma_0|} = \eta\left(\eta^{-1/d} + C(n)\eta - 2C(n)\right) \ .$$

By the arithmetic-geometric mean,

$$\begin{aligned} \eta^{-1/d} + C\eta &= \frac{d}{d+1}\left(\frac{d+1}{d}\eta^{-1/d}\right) + \frac{1}{d+1}\left((d+1)C\eta\right) \\ &\ge \left(\frac{d+1}{d}\eta^{-1/d}\right)^{d/(d+1)} \left((d+1)C\eta\right)^{1/(d+1)} \\ &= C^{1/(d+1)}\frac{d+1}{d^{d/(d+1)}} \ . \end{aligned} \tag{1.17}$$

Therefore, a minimum occurs at $\eta > 0$ if and only if

$$C^{1/(d+1)}\frac{d+1}{d^{d/(d+1)}} \le 2C \ .$$

Let $C_\star$ be the value of C that gives equality in this last inequality. One finds, as in [4],

$$C_\star = \frac{1}{d}\left(\frac{d+1}{2}\right)^{(d+1)/2} \ . \tag{1.18}$$

Moreover, with $C = C_\star$, there is equality in the application made above of the arithmetic geometric mean inequality if and only if $\eta^{-1/d}/d = C_\star$. Therefore, define $\eta_\star$ by $\eta_\star = (dC_\star)^{-d}$. One finds

$$\eta_\star = \left(\frac{d+1}{2}\right)^{(d+1)/2d} \ . \tag{1.19}$$

The heuristic argument of [4] suggests that when Φ is minimized at $\eta = 0$, one puts all of the mass into the uniform supersaturated state, and there is no droplet. This is the case if $C(n) < C_\star$. On the other hand, if Φ is minimized at a strictly positive value of η, then a strictly positive fraction of the mass should go into a droplet. This is the case if $C(n) > C_\star$. Moreover, it is easy to see that for all $C(n) > C_\star$, the minimizing value η_c of η satisfies $\eta_c \geq \eta_\star$. As emphasized in [4], this suggests that there are never drops containing a volume fraction less than $\eta_\star$. That is, at least according to this heuristic analysis, droplet sizes always fall in the universal range

$$\eta_\star(\sigma_d/d)r_0^d < \text{droplet size} \leq (\sigma_d/d)r_0^d \ .$$

Note that for $d = 1$, $\eta_\star = 1$, and the droplets always have the equimolar radius, when they exist. This is what one expects; shrinking the droplet does not shrink its boundary in one dimension.

The validity of this was rigorously established for the two-dimensional Ising model in [5]. We show here that the same heuristic analysis is correct for the minimization problem (1.2) concerning the Cahn-Hilliard free energy function $\mathcal{F}$. The first result concerns the value of the ratio $\frac{f_L}{|\Gamma_0|}(n)$ for $n = -1 + KL^{-d/(d+1)}$, for any $K > 0$, as L tends to infinity.

Theorem 1.1. *For all $K > 0$,*

$$\lim_{L\to\infty} \frac{f_L}{|\Gamma_0|}\left(-1 + KL^{-d/(d+1)}\right) = \inf_{0\leq\eta\leq 1} S\left(\eta^{1-1/d} + D(K)(1-\eta)^2\right) \tag{1.20}$$

where

$$D(K) = C\left(-1 + KL^{-d/(d+1)}\right) = \frac{2}{d\chi S}\left(\frac{\sigma_d}{d}\right)^{-1/d}\left(\frac{K}{2}\right)^{(d+1)/d}$$

and $S = 2^{3/2}/3$.

Furthermore, let $K_\star$ be defined by

$$K_\star = 2\left(\frac{d+1}{2}\right)^{d/2}\left(\frac{\sigma_d}{d}\right)^{1/(d+1)}\left(\frac{\chi S}{2}\right)^{d/(d+1)} . \tag{1.21}$$

Then for all $K < K_\star$, and all L sufficiently large, the infimum in (1.20) is a minimum attained uniquely at $\eta = 0$, while for all $K > K_\star$, and all L sufficiently large, the infimum in (1.20) is a minimum attained uniquely at $\eta = \eta_c$ where $\eta_c \geq \eta_\star$.

To prove Theorem 1.1, we prove precise upper and lower bounds on $f_L(n)$ for values of n in the critical scaling regime, and from these bounds deduce (1.20). The remaining statements in the theorem then follow from the discussion just above concerning the minimization of $\Phi(\eta)$. For example, note that $K_\star$ is obtained by solving $D(K) = C_\star$ for K. The upper and lower bounds on $f_L(n)$ will be presented and proved in Sections 2 and 3, respectively. We conclude Section 3 with the proof of Theorem 1.1.

The theorem suggests that the curve $n(L) = -1 + K_\star L^{-d/(d+1)}$ is critical for droplet formation, so that for large L and densities n significantly below this level, the minimizers will be uniform, while for large L and densities n significantly above this level, the minimizers will correspond to droplets of a reduced radius $\eta_c^{1/d} r_0$. The following theorems bear this out.

Theorem 1.2. *For all $K < K_\star$ and L sufficiently large, when*

$$-1 \leq n \leq -1 + KL^{-d/(d+1)} ,$$

the unique minimizer for (1.2) is the uniform order parameter field $m(x) = n$.

Before stating the result concerning droplet minimizers, we must make this notion precise. To facilitate this, regard Ω as the centered cube in $\mathbb{R}^d$ with side length L and periodic boundary conditions.

For given η and n, and hence for given η and r_0, define a *sharp interface reduced radius droplet* order parameter field $m^{\sharp}_{\eta,n}(x)$ by

$$m^{\sharp}_{\eta,n}(x) = \begin{cases} 1 & \text{if} \quad |x| < \eta^{1/d} r_0 \\ -1 & \text{if} \quad |x| \geq \eta^{1/d} r_0 \end{cases} .$$

Theorem 1.3. *For all $K > K_\star$, $\epsilon > 0$, and L sufficiently large, when*

$$-1 + KL^{-d/(d+1)} \leq n \leq -1 + L^{-1/2} ,$$

any minimizer m for (1.2) *is such that, after a possible translation on the torus Ω,*

$$\frac{1}{|r_0^d|} \int_\Omega \left| m(x) - m^{\sharp}_{\eta_c,n}(x) \right|^4 \, \mathrm{d}x \leq \epsilon$$

where η_c is the minimizing value of η in (1.20).

This theorem says that for large L, the set on which m and $m^{\sharp}_{\eta_c,n}$ differ by an appreciable amount is small compared to $(\sigma_d/d)r_0^d$. In particular, on the ball where $m^{\sharp}_{\eta_c,n} = 1$, m must be very close to 1 on all but a negligibly small percentage of the volume of that ball. Likewise, on the set where $m^{\sharp}_{\eta_c,n} = -1$, m must be very close to -1 on all but a set whose measure is a negligibly small percentage of the volume of the ball. The role of the fourth power is to make this small difference even smaller so that it is not overwhelmed by the large volume of the region external to the ball. Any power larger than three would work just as well in our argument.

In this sense, m "looks like" $m^{\sharp}_{\eta_c,n}$ for large L, and thus describes a droplet of the radius predicted by the heuristic argument of [4].

Theorems 1.2 and 1.3 are proved in Section 4. Finally, in Section 5, we give an explanation for the remarkable efficacy of the simple trial function used in Section 2. In particular, we see which features of the free energy functional (1.1) are responsible for this. The point is that for the free energy functionals coming from models with a nonlocal interaction, such as the ones considered in [8], as well as in [2] and [3], these features are not present. However, the analysis in Section 5 leads to a method for constructing trial functions of high accuracy that does apply to such cases, as well as to (1.1).

2. The upper bound

2.1. The interpolating family of trial functions

For $0 \leq \eta \leq 1$, let $r_\eta = \eta^{1/d} r_0$ be the radius of a ball whose volume is η times the volume of a ball with the equimolar radius, r_0. The arguments of Biskup, Chayes, and Kotecky suggest that one should use as a trial function a function of the form

$$m_{\eta\,\mathrm{dr}}(x) = m_0(|x| - r_\eta) + \alpha(\eta) , \tag{2.1}$$

where m_0 is a transition profile that very nearly minimizes the cost in free energy of making the transition from $m = +1$ to $m = -1$, and $\alpha(\eta)$ is a constant determined by the constraint $\int_\Omega m_{\eta\,\mathrm{dr}}(x)\,\mathrm{d}x = n|\Omega|$. As in Section 1, we are taking Ω to be the centered cube in $\mathbb{R}^d$ with side length L.

As η varies in the interval $0 < \eta < 1$, the family of "fractional droplet" trial functions defined in (2.1) interpolates between m_{uni}, for $\eta = 0$ and m_{emd}, for $\eta = 1$. Of course, it remains to choose m_0.

2.2. Planar surface tension and the choice of m_0

The natural choice for m_0 is given by considering the problem of minimizing the cost per unit area in free energy of an infinite planar interface between the $+1$ and -1 phase. Denote this quantity by S; it will turn out to be the same constant S that appears in (1.4). That is,

$$S = \inf \left\{ \int_{\mathbb{R}} \left(\frac{1}{2} \left| m'(z) \right|^2 + F(m(z)) \right) \mathrm{d}z \ : \ \lim_{z \to \pm\infty} m(z) = \mp 1 \right\} .$$

Let $\bar{m}$ denote minimizer for this variational problem with $\bar{m}(0) = 0$. The Euler-Lagrange equation satisfied by $\bar{m}$ is $\bar{m}''(z) = F'(\bar{m}(z))$. Multiplying both sides by $\bar{m}'(z)$, and integrating from $-\infty$ to z, we obtain

$$\left(\bar{m}'(z)\right)^2 = 2F(\bar{m}(z)) , \tag{2.2}$$

since $\lim_{z\to-\infty} m'(z) = \lim_{z\to-\infty} F(m(z)) = 0$.

One now easily deduces that $\bar{m}(z) = -\tanh(z/\sqrt{2})$, from which one could compute S. However, there is anther route that is more informative and useful in what follows: From (2.2), we see that

$$S = 2\int_{-\infty}^{\infty} F(\bar{m}(z))\, \mathrm{d}z ,$$

and furthermore,

$$\begin{aligned} \int_{-\infty}^{\infty} F(\bar{m}(z))\, \mathrm{d}x &= \int_{-\infty}^{\infty} \frac{F(\bar{m}(z))}{\bar{m}'(z)} \bar{m}'(z)\, \mathrm{d}x \\ &= \int_{-\infty}^{\infty} -\sqrt{\frac{F(\bar{m}(z))}{2}} \bar{m}'(z)\, \mathrm{d}z \\ &= \int_{-1}^{1} \sqrt{\frac{F(h)}{2}}\, \mathrm{d}h . \end{aligned} \tag{2.3}$$

Thus,

$$S = \int_{-1}^{1} \sqrt{2F(h)}\, \mathrm{d}h , \tag{2.4}$$

and hence $S = 2^{3/2}/3$, which provides the numerical value for S that is quoted in Theorem 1.1. However, in what follows, it is the integral formula, and not so much the numerical value, that turns out to matter.

We now choose m_0. We cannot simply choose $m_0 = \bar{m}$ since then $m_0(|x| - r_\eta)$ would not define a smooth, or even continuous, function on Ω. However, only mild modifications are required. Since we are interested in values of r with $r = \mathcal{O}\left(L^{d/(d+1)}\right)$, define

$$m_0(z) = \begin{cases} \bar{m}(z) & \text{if} \quad |z| < L^{(d-1)/(d+1)} \\ -\mathrm{sgn}(z) & \text{if} \quad |z| > 2L^{(d-1)/(d+1)} \end{cases} ,$$

and smoothly interpolate in such a way that m_0, like $\bar{m}$, is odd. With any such interpolation, $m_0(|x| - r_\eta)$ defines a smooth function on Ω, and the difference between m_0 and $\bar{m}$ goes to zero exponentially fast as L tends to infinity.

2.3. The determination of $\alpha(\eta)$

The constraint equation is

$$\int_\Omega m_{\eta\,\mathrm{dr}}(x)\,\mathrm{d}x = nL^d = \left[2(\sigma_d/d)r_0^d - L^d\right]$$

and hence

$$\alpha(\eta)L^d = \left[2(\sigma_d/d)r_0^d - L^d\right] - \int_\Omega m_0(|x| - r_\eta)\,\mathrm{d}x\ . \tag{2.5}$$

We require sharp estimates on the integral on the right.

Lemma 2.1. *Define the constant M by*

$$M = \int_{\mathbb{R}} \left(\mathrm{sgn}(z) - \tanh\left(z/\sqrt{2}\right)\right)z\,\mathrm{d}z\ .$$

For all $L^{(d-1)/(d+1)} < r_\eta < r_0$,

$$\int_\Omega m_0(|x| - r_\eta)\,\mathrm{d}x = \left[2(\sigma_d/d)r_0^d\eta - L^d\right] - (d-1)M\sigma_d r_0^{d-2}\eta^{(d-2)/d} + \mathcal{O}\left(e^{-L^{1/4}}\right)$$

when $d = 1, 2$ and 3. For higher dimension, the only difference is that the error term is $\mathcal{O}(r_0^{d-4})$.

Proof. Note that

$$\int_\Omega m_0(|x| - r_\eta)\,\mathrm{d}x = \left[2(\sigma_d/d)r_\eta^d - L^d\right] - \int_\Omega (\mathrm{sgn}(|x| - r_\eta) + m_0(|x| - r_\eta))\,\mathrm{d}x\ .$$

Define

$$I_1 = \int_{|x|\le 2r_\eta} (\mathrm{sgn}(|x| - r_\eta) + m_0(|x| - r_\eta))\,\mathrm{d}x \quad \text{and} \quad I_2 = \int_{|x|>2r_\eta} (\mathrm{sgn}(|x| - r_\eta) + m_0(|x| - r_\eta))\,\mathrm{d}x.$$

We easily see that for all dimensions d, $I_2 = \mathcal{O}(e^{-L^{1/4}})$. Moreover, using polar coordinates,

$$I_1 = \sigma_d \int_0^{2r_\eta} (\mathrm{sgn}(s - r_\eta) + m_0(s - r_\eta))s^{d-1}\,\mathrm{d}s\ .$$

Introducing the new variable $z = s - r_\eta$, we see that if we extend the integration in z over the whole real line, we only make an error of size $\mathcal{O}(e^{-L^{1/4}})$ at most, and so

$$I_1 = \sigma_d r^{d-1} \int_{\mathbb{R}} (\mathrm{sgn}(z) + m_0(z))\left(1 - \frac{z}{r_\eta}\right)^{d-1}\,\mathrm{d}z + \mathcal{O}\left(e^{-L^{1/4}}\right).$$

Taking into account the fact that $(\mathrm{sgn}(z) + m_0(z))$ is odd and rapidly decaying, we see that for $d = 1$, 2, and 3,

$$\int_{\mathbb{R}} (\mathrm{sgn}(z) + m_0(z))\left(1 - \frac{z}{r_\eta}\right)^{d-1}\,\mathrm{d}z = \frac{d-1}{r_\eta}\int_{\mathbb{R}} (\mathrm{sgn}(z) + m_0(z))z\,\mathrm{d}z\ .$$

In any dimension, this gives the leading order correction. This, together with the definition of $m_0(z)$ in terms of $\bar{m}(z) = -\tanh(z/\sqrt{2})$, yields the result. □

Therefore, (2.5) together with Lemma 2.1 yield for $d = 2$ or $d = 3$ that

$$\alpha(\eta) = 2(\sigma_d/d)\frac{r_0^d}{L^d}(1-\eta) + (d-1)M\sigma_d\frac{r_0^{d-2}}{L^d}\eta^{(d-2)/d} + \mathcal{O}\big(e^{-L^{1/4}}\big)\,. \tag{2.6}$$

The only difference for higher dimensions d is that $\mathcal{O}(e^{-L^{1/4}})$ must be replaced by $\mathcal{O}(r_0^{d-4}/L^d)$. Notice that unless $\eta = 1$, the first explicit correction is already very small compared to the leading term; it is smaller by a factor of r_0^{-2}. In the critical regime, by (1.14), $r_0^{-2} \asymp L^{-2d/d+1)}$. Moreover, we see that in the critical scaling regime, except when $\eta = 1$,

$$\alpha(\eta) \asymp L^{-d/(d+1)}\,. \tag{2.7}$$

2.4. Computation of $\mathcal{F}(m_{\eta\,\mathrm{dr}})$

With the trial function specified, we now compute $\mathcal{F}(m_{\eta\,\mathrm{dr}})$.

Lemma 2.2. *In the critical scaling regime $r_0 \asymp L^{d/(d+1)}$,*

$$\mathcal{F}(m_{\eta\,\mathrm{dr}}) \le \Phi(\eta) - 8(\sigma_d/d)^3\frac{r_0^{3d}}{L^{2d}}(1-\eta)^3 + \mathcal{O}\left(L^{(d^2-3d)/(d+1)}\right)\,, \tag{2.8}$$

where $\Phi(\eta)$ is $\mathcal{O}\big(L^{(d^2-d)/(d+1)}\big)$ and the second term on the right is $\mathcal{O}\big(L^{(d^2-2d)/(d+1)}\big)$.

Notice that the leading term in the upper bound is exactly $\Phi(\eta)$, and that the next term is negative.

Proof. To simplify the notation, we write m_0 to denote $m_0(|x| - r_\eta)$ and α to denote $\alpha(\eta)$ so that $m_{\eta\,\mathrm{dr}} = m_0 + \alpha$. Then

$$F(m_{\eta\,\mathrm{dr}}) = F(m_0) + F'(m_0)\alpha + \frac{1}{2}F''(m_0)\alpha^2 + \frac{1}{6}F'''(m_0)\alpha^3 + \frac{1}{4}\alpha^4\,.$$

We are required to produce a close upper bound on the integral of each of these terms over Ω. We start with $\alpha\int_\Omega F'(m_0)\,\mathrm{d}x$.

Note that $F'(m) = m^3 - m$. Since $m_0^3(z) - m_0(z)$ is an odd, rapidly decaying function of z, estimates just like the ones employed in the proof of Lemma 2.1 show that

$$\int_\Omega F'(m_0)\,\mathrm{d}x = \sigma_d r_\eta^{d-1}\int_{\mathbb{R}}\big(\bar m^3(z) - \bar m(z)\big)\left(1 - \frac{z}{r_\eta}\right)^{d-1}\,\mathrm{d}z + \mathcal{O}\big(e^{-L^{1/4}}\big)\,.$$

Then, with the constant B defined by

$$B = \int_{\mathbb{R}}\big(\bar m^3(z) - \bar m(z)\big)z\,\mathrm{d}z\,,$$

we have for $d = 2$ or $d = 3$ that

$$\int_\Omega F'(m_0)\,\mathrm{d}x = \sigma_d r_0^{d-2}B\eta^{(d-2)/d} + \mathcal{O}\big(e^{-L^{1/4}}\big)\,, \tag{2.9}$$

and the same is true for $d \ge 4$ except that the error term must be replaced by $\mathcal{O}(r_0^{d-4})$.

Next, $F''(m_0) = 3m_0^2 - 1 \le 2 = 1/\chi$. Therefore

$$\int_\Omega \frac{1}{2} F''(m_0)\,\mathrm{d}x \le \frac{1}{2\chi} L^d \ . \tag{2.10}$$

Finally, $F'''(m) = 6m$, and so

$$\int_\Omega \frac{1}{6} F'''(m_0)\,\mathrm{d}x = \int_\Omega m_0\,\mathrm{d}x \ , \tag{2.11}$$

and this integral has been computed in Lemma 2.1.

Now let I denote the integral

$$I = \int_\Omega \left[F'(m_0)\alpha + \frac{1}{6} F'''(m_0)\alpha^3 + \frac{1}{4}\alpha^4 \right] \mathrm{d}x \ .$$

In the critical scaling regime, the dominant contribution to I comes from the F''' term, and is $-L^d\alpha^3 \asymp L^{(d^2-2d)/(d+1)}$. Each of the terms in the integrand contributes at the order $L^{(d^2-3d)/(d+1)}$ in the critical scaling regime, and we have

$$I = -L^d\alpha^3 + \left[\sigma_d r_0^{d-2} B \eta^{(d-2)/d}\alpha + 2(\sigma_d/d) r_0^d \eta\alpha^3 + \alpha^4 L^d/4\right] + \mathcal{O}\left(L^{(d^2-4d)/(d+1)}\right) \ .$$

Using (2.5), we can express this as

$$\begin{aligned} I = &-8(\sigma_d/d)^3 \frac{r_0^{3d}}{L^{2d}}(1-\eta)^3 + \left[2d(\sigma_d/d)^2 B \frac{r_0^{2d-2}}{L^d}\eta^{2-2/d} + 4(\sigma_d/d)^4 \frac{r_0^{4d}}{L^{3d}}(1-\eta)^3(1+3\eta) \right] \\ &+ \mathcal{O}\left(L^{(d^2-4d)/(d+1)}\right) \ , \end{aligned} \tag{2.12}$$

where the first term on the right is proportional to $L^{(d^2-2d)/(d+1)}$, and the second is proportional to $L^{(d^2-3d)/(d+1)}$.

Finally, we have to estimate $\int_\Omega \left[|\nabla m_0|^2 + F(m_0)\right] \mathrm{d}x$. Once more, estimates just like the ones employed in the proof of Lemma 2.1 show that

$$\int_\Omega \left[|\nabla m_0|^2 + F(m_0)\right] \mathrm{d}x \approx \sigma_d r_\eta^{d-1} \int_{\mathbb{R}} \left[\left|\bar{m}'(z)\right|^2 + F(\bar{m}(z))\right] (1 + z/r_\eta)^{d-1}\,\mathrm{d}z$$

where the errors are exponentially small in $L^{1/4}$. But because m_0 is so close to $\bar{m}$, this only differs from $S|\Gamma_0|\eta^{1-1/d}$ by errors that are $\mathcal{O}(r_0^{d-3})$. In the asymptotic scaling regime, $r_0^{d-3} \asymp L^{(d^2-3d)/(d+1)}$.

Combining estimates, we have

$$\begin{aligned} \mathcal{F}(m_{\eta\,\mathrm{dr}}) &\le S\sigma_d r^{d-1} + \frac{1}{2\chi}\left(\frac{2\sigma_d}{d}\left(\frac{r_0}{L}\right)^d - \frac{2\sigma_d}{d}\left(\frac{r}{L}\right)^d\right)^2 L^d + \mathcal{O}(1) \\ &= S\sigma_d r_0^{d-1}\left(\frac{r}{r_0}\right)^{d-1} + \frac{L^d}{2\chi}\left(\frac{2\sigma_d r_0^d}{dL^d}\right)^2 \left(1 - \left(\frac{r}{r_0}\right)^d\right)^2 + \mathcal{O}(1) \\ &= S|\Gamma_0|\big(\eta^{1/d} + C(n)(1-\eta)^2\big) + \mathcal{O}\left(L^{(d^2-2d)/(d+1)}\right) \ . \quad \square \end{aligned} \tag{2.13}$$

3. The lower bound

3.1. An *a priori* pointwise upper bound

Standard compactness arguments show that the infimum in (1.2) is attained at a minimizer $m(x)$ which satisfies the Euler-Lagrange equation

$$-\Delta m(x) + m^3(x) - m(x) + \mu = 0\ , \tag{3.1}$$

where μ is a Lagrange multiplier corresponding to the constraint in (1.2).

Our immediate goal is to prove an *a priori* pointwise upper bound on a minimizer m that is very close to 1 in the critical scaling regime. Such a bound can be obtained from the Euler-Lagrange equation and the maximum principle.

Let $x_{\min}$ and $x_{\max}$ be such that for all x,

$$m(x_{\min}) \le m(x) \le m(x_{\max})\ .$$

These exist since any solution of the Euler-Lagrange equation is continuous.

We will now show that $m(x_{\max})$ cannot be too large. Define numbers λ and ν by

$$1 + \lambda = m(x_{\max}) \qquad \text{and} \qquad -1 + \nu = m(x_{\min})\ . \tag{3.2}$$

It will be convenient in the arguments leading to the proof to write n in the form

$$n = -1 + \delta\ . \tag{3.3}$$

Notice that in the critical scaling regime, $\delta \asymp L^{-d/(d+1)}$. Also, from (1.3) and (1.6),

$$\delta = 2\frac{V_+}{L^d} = 2(\sigma_d/d)\frac{r_0^d}{L^d}\ . \tag{3.4}$$

Lemma 3.1. *For any solution of the Euler-Lagrange Equation* (3.1)*, let* λ *and* ν *be given by* (3.2)*. Then* $\nu \ge \lambda$*. Consequently, if* m *is any minimizer for* (3.1)*,*

$$\delta \ge \lambda\ . \tag{3.5}$$

Proof. Evidently, $\Delta m(x_{\max}) \le 0$, and so from (3.1) and (3.2), $(1+\lambda)^3 - (1+\lambda) + \mu \le 0$, or

$$2\lambda + 3\lambda^2 + \lambda^3 \le -\mu\ .$$

In the same way, from (3.1) and (3.2) we have

$$2\nu - 3\nu^2 + \nu^3 + \mu \ge -\mu\ .$$

Therefore,

$$2\nu - 3\nu^2 + \nu^3 \ge 2\lambda + 3\lambda^2 + \lambda^3\ .$$

It evidently follows that

$$2\nu + \nu^3 \ge 2\lambda + \lambda^3\ ,$$

and since $f(x) = 2x + x^3$ is monotone increasing, it follows that $\nu \ge \lambda$.

Next, since the average value of any function is no less than its minimum, it follows that $n \ge -1 + \nu$, and by (3.4), this means $\delta \ge \nu$. Combining estimates, we have (3.5). □

3.2. An *a priori* lower bound on $m(x_{\max})$

We next show that any for nonconstant minimizer m, it cannot be that $m(x_{\max})$ is much smaller than 1. For this purpose, define w by $w(x) = m(x) - n$. For m satisfying the constraint in (1.2),

$$\int_\Omega w(x)\,\mathrm{d}x = 0\,. \tag{3.6}$$

Clearly, $\int_\Omega |\nabla m|^2\,\mathrm{d}x = \int_\Omega |\nabla w|^2\,\mathrm{d}x$, and

$$\int_\Omega \frac{1}{4}\big(m^2-1\big)^2\,\mathrm{d}x = L^2\big(n^2-1\big)^2 + \int_\Omega \frac{1}{4}\left(\frac{1}{2}\big(3n^2-1\big)w^2 + nw^3 + \frac{1}{4}w^4\right)\mathrm{d}x\,,$$

since the terms linear in w drop out due to (5b), and

$$nw^3 + \frac{1}{4}w^4 = \frac{1}{4}w^2(w+2n)^2 - n^2w^2\,.$$

Hence, if we define the functional $\mathcal{G}$ by

$$\mathcal{G}(w) = \frac{1}{2}\int_\Omega |\nabla w|^2\,\mathrm{d}x + \int_\Omega G(w)\,\mathrm{d}x\,, \tag{3.7}$$

where

$$G(w) = \frac{n^2-1}{2}w^2 + \frac{1}{4}w^2(w+2n)^2\,, \tag{3.8}$$

we have

$$\mathcal{F}(m) = \mathcal{F}(n) + \mathcal{G}(w) \tag{3.9}$$

whenever m satisfies the constraint in (1.2).

Lemma 3.2. *Let m be any minimizer for* (1.2)*, and suppose that m is not constant. Then*

$$m(x_{\max}) \geq 1 - \delta - 2\sqrt{\delta}\sqrt{1-\delta/2}\,. \tag{3.10}$$

Proof. Notice that

$$\frac{n^2-1}{2} + \frac{1}{4}(w+2n)^2 < 0$$

if and only if

$$z_- < w < z_+$$

where

$$z_\pm = -2n \pm \sqrt{2-2n^2} = 2 - 2\delta \pm 2\sqrt{\delta}\sqrt{1-\delta/2}\,.$$

Since m is not constant $\int_\Omega |\nabla w|^2\,\mathrm{d}x > 0$. Thus, $\mathcal{G}(w) > 0$ unless $w(x_{\max}) \geq 2 - 2\delta - 2\sqrt{\delta}\sqrt{1-\delta/2}$. If m is a minimizer, $\mathcal{G}(w) > 0$ is impossible, on account of (3.9). Since $m(x_{\max}) = n + w(x_{\max})$, we have the estimate. □

3.3. A partition of Ω

We now partition Ω in to three pieces, one of which will contribute a surface tension term to the free energy, another of which will contribute a compressibility term, and another that will be negligible.

Returning to the original dependent variable m, we fix a number $\kappa > 0$ to be determined below. However, to fix our ideas for the time being, suppose that $\kappa = \mathcal{O}(\delta^{1/3})$. Define numbers h_+ and h_- by

$$h_+ = 1 - \kappa \qquad \text{and} \qquad h_- = -1 + \kappa \ . \tag{3.11}$$

Define the sets A, B and C by

$$A = \{ \, x \ : \ h_- \le m(x) \le h_+ \, \} \qquad B = \{ \, x \ : \ m(x) \le h_- \, \} \ ,$$

and

$$C = \{ \, x \ : \ m(x) \ge h_+ \, \} \ .$$

If m is a nonconstant minimizer, and $\kappa = \mathcal{O}(\delta^{1/3})$, then for L large enough, C will be nonempty by Lemma 3.2. Define a radius R by

$$(\sigma_d/d)R^d = |C| \ , \tag{3.12}$$

where the right-hand side denotes the measure of C. Evidently R is the radius of the ball with the same volume as C.

We shall obtain a lower bound on $f_L(n)$ by separately estimating the integrals

$$I_A = \int_A \left[\frac{1}{2}|\nabla m|^2 + F(m) \right] \mathrm{d}x \qquad \text{and} \qquad I_B = \int_B \left[\frac{1}{2}|\nabla m|^2 + F(m) \right] \mathrm{d}x \ . \tag{3.13}$$

3.4. The surface tension contribution

We now prove a lower bound on I_A, which corresponds to the surface tension contribution to the free energy. The lower bound is obtained through use of the co-area formula [1, 11], which expresses the volume element in Ω as

$$\mathrm{d}x = \frac{1}{|\nabla m(x)|} \, \mathrm{d}\sigma_h \, \mathrm{d}h \ ,$$

where $\mathrm{d}\sigma_h$ is the surface area along Γ_h, the level set $\{m(x) = h\}$, or, more properly put, the $d-1$ dimensional Hausdorff measure on this set. L. Modica has earlier made [13] a similar use of the co-area formula in studying a different scaling limit of essentially the same functional. His work draws on an earlier article of Modica and Mortola [14]. We thank E. Pressuti for bringing these references to our attention.

In applying the co-area formula, we shall gloss over certain standard technical issues. These are all explained, for example, in the discussion of the Faber-Krahn inequality in [7] or [10], where the co-area formula is applied to another variational problem, namely the one for the fundamental eigenvalue for the Laplacian in a domain in $\mathbb{R}^d$. Those readers who are not familiar with the use of the co-area formula in proving inequalities such as the Faber-Krahn inequality may wish to consult the references cited above.

It is worth noting, however, that the rearrangement inequalities of the sort discussed in [8] apply in this case, and allow us to conclude that for any minimizer m, the level sets are *symmetric* *monotone.* If we translate so that the maximum of m is at 0, then this means that for any h, and any of the standard basis vectors $\vec{e}_j$, $j = 1, \ldots, d$, the set of t for which $m(t\vec{e}_j) > h$ is a symmetric interval. In particular, Γ_h is a rectifiable, simply connected curve.

Lemma 3.3. *Let m be any nonconstant minimizer for* (1.2)*, and suppose that $\kappa = \mathcal{O}(\delta^{1/3})$ and let R be given by* (3.12)*. Then for L large enough,*

$$I_A \geq (\sigma_d) R^{d-1} (S - 2\kappa) \ . \tag{3.14}$$

Proof. By the co-area formula,

$$I_A = \int_{h_-}^{h_+} \int_{\Gamma_h} \left(\frac{1}{2} |\nabla m(x)| + \frac{F(h)}{|\nabla m(x)|} \right) \mathrm{d}\sigma_h \, \mathrm{d}h \ .$$

By the arithmetic-geometric mean inequality,

$$\frac{1}{2} \left(|\nabla m(x)| + 2 \frac{F(h)}{|\nabla m(x)|} \right) \geq \sqrt{2F(h)} \ ,$$

and therefore,

$$I_A \geq \int_{h_-}^{h_+} |\Gamma_h| \sqrt{2F(h)} \, \mathrm{d}h$$

where $|\Gamma_h|$ denote the one-dimensional Hausdorff measure of Γ_h.

Note that Γ_h encloses a region whose volume is at least $|C|$. Let $P(V)$ be the isoperimetric function on the torus Ω, as explained in the Appendix.

By the isoperimetric inequality on the torus, the length of the boundary of such a region is at least $\sigma_d^{1/d} (dV)^{1-1/d}$ provided $|C|/L^d \leq \epsilon$ for some ϵ depending only on d. Since $P(V)$ is an increasing function of V for $V \leq L^d/2$, we have

$$|\Gamma_h| \geq P\,(|C|)$$

for all $h_- \leq h \leq h_+$. Hence,

$$I_A \geq P\,(|C|) \int_{h_-}^{h_+} \sqrt{2F(h)} \, \mathrm{d}h \ .$$

By (2.4), this yields

$$I_A \geq P\,(|C|) \left(S - \int_{-1}^{h_-} \sqrt{2F(h)} \, \mathrm{d}h - \int_{h_+}^{1} \sqrt{2F(h)} \, \mathrm{d}h \right) \ .$$

Furthermore,

$$\sqrt{2F(h)} = \left(1 - h^2\right)/\sqrt{2} \leq 1 \ ,$$

and hence $\int_{-1}^{h_-} \sqrt{2F(h)} \, \mathrm{d}h \leq \kappa$ and $\int_{h_+}^{1} \sqrt{2F(h)} \, \mathrm{d}h \leq \kappa$. This gives us $I_A \geq P(|C|)(S - 2\kappa)$. Now if $|C| \geq \epsilon L^d$, where ϵ is the constant in the Morgan-Johnson isoperimetric inequality, this would imply $I_A \geq \text{const} \times L^{d-1}$, which is much larger than $\mathcal{F}(n)$ for large L. This is impossible when m is a minimizer for (1.2), and so $P(|C|) \geq \sigma_d(|C|)/(\sigma_d/d))^{1-1/d}$. Then, with R defined by (3.12), we have the bound (3.14). □

3.5. The bulk contribution

In this subsection, we prove a lower bound on the contribution to the free energy from B. For this purpose, we first require an upper bound on $|A|$ which shows that, for large L, $|A|$ is negligible compared to $|C|$. Ideally, one might hope that A is an annular region about C, and to obtain a "surface term" type bound for A, showing that is bounded by a multiple of $r_0^{d-1} \asymp L^{d/(d+1)}$. However, as one can see from the proof of Lemma 3.3, even if C were spherical and A was an annulus about it, one would have to take the annulus to be fairly "thick" in order to capture most of S in the estimate (3.14). Thus, the following simple estimate is rather sharp.

Lemma 3.4. *Let m be any minimizer for* (1.2)*. Then*

$$|A| \le 2F(n)\frac{L^d}{\kappa^2} \le 2\frac{\delta^2}{\kappa^2}L^d \ .$$

Proof. Since

$$F(h_+) = \kappa^2(1-\kappa/2)^2 = F(h_-) \ ,$$

it is easy to see that uniformly on A,

$$F(m(x)) \ge \kappa^2(1-\kappa/2)^2 \ . \tag{12}$$

Therefore

$$I_A \ge |A|\kappa^2(1-\kappa/2)^2 \ .$$

On the other hand, since m is a minimizer,

$$I_A < \mathcal{F}(n) = F(n)L^2 \ .$$

In the range of δ being considered, $(1-\kappa/2)^2 \ge 1/2$. □

Our next goal is a lower bound on I_B. Notice that on $(-\infty, h_-)$, F is strictly convex. In fact, $F''(h) \ge 3h_-^2 - 1$. Define the quantity χ_- by

$$\frac{1}{\chi_-} = F''(h_-) = 3h_-^2 - 1 \ .$$

Then, by Taylor's Theorem, and using the fact that $F(-1) = F'(-1) = 0$, we have

$$F(m(x)) \ge \frac{1}{2\chi_-}(m(x)+1)^2$$

everywhere on $\{m \le h_-\}$.

Therefore,

$$\begin{aligned}
\int_B F(m(x))\,\mathrm{d}x &= |B|\left(\frac{1}{|B|}\int_B F(m(x))\,\mathrm{d}x\right)\\
&\ge |B|\frac{1}{2\chi_-}\left(\frac{1}{|B|}\int_B (m(x)+1)^2\,\mathrm{d}x\right)\\
&\ge |B|\frac{1}{2\chi_-}\left(\frac{1}{|B|}\int_B (m(x)+1)\,\mathrm{d}x\right)^2\\
&= \frac{1}{2\chi_-|B|}\left(\int_B m(x)\,\mathrm{d}x + |B|\right)^2 \ .
\end{aligned} \tag{3.15}$$

We now need an upper bound and lower bounds on $|B|$ and $\int_B m(x)\,\mathrm{d}x$. Note that

$$\int_B m(x)\,\mathrm{d}x = nL^d - \int_C m(x)\,\mathrm{d}x - \int_A m(x)\,\mathrm{d}x\ .$$

By Lemma 3.1 and the definition of R,

$$(1-\kappa)(\sigma_d/d)R^d \le \int_C m(x)\,\mathrm{d}x \le (1+\delta)(\sigma_d/d)R^d\ .$$

By Lemma 3.4,

$$-|A| \le h_-|A| \le \int_A m(x)\,\mathrm{d}x \le h_+|A| \le |A|\ .$$

Thus, since $\kappa > \delta$,

$$\left|\int_B m(x)\,\mathrm{d}x - \big(nL^d - (\sigma_d/d)R^d\big)\right| \le |A| + \kappa(\sigma_d/d)R^d\ .$$

Next, it is evident that $|B| = L^d - (\sigma_d/d)R^d - |A|$. Therefore

$$\left|\left(\int_B m(x)\,\mathrm{d}x + |B|\right) - \big(\delta L^d - 2(\sigma_d/d)R^d\big)\right| \le 2|A| + \kappa(\sigma_d/d)R^d\ .$$

Hence, if we define ϵ by

$$\epsilon = 2\big(2|A|/L^d + \kappa(\sigma_d/d)(R/L)^d\big)\left|\delta - 2(\sigma_d/d)(R/L)^d\right|,$$

we have

$$\left(\int_B m(x)\,\mathrm{d}x + |B|\right)^2 \ge \big(\delta L^d - 2(\sigma_d/d)R^d\big)^2 - \epsilon L^{2d}\ .$$

By Lemma 3.4, $|A|/L^2 = \mathcal{O}(\delta^2/\kappa^2)$, and for any minimizer, we must have $R = \mathcal{O}(L^{d/(d+1)}$, since otherwise, if R were any larger, the contribution from the interface as estimated in Lemma 3.3 would already exceed the free energy for the uniform trial function. Thus, $(R/L)^d = \mathcal{O}(L^{-d/(d+1)}) = \mathcal{O}(\delta)$. Therefore,

$$\epsilon = \mathcal{O}\left(\frac{\delta^3}{\kappa^2} + \kappa\delta^2\right)\ .$$

With the choice $\kappa = \delta^{1/3}$, this gives us

$$\epsilon = \mathcal{O}\big(\delta^{7/3}\big) = \mathcal{O}\left(L^{-\frac{7}{3}\frac{d}{d+1}}\right)\ , \tag{3.16}$$

with the essential point being that this is negligible compared to δ^2 as L tends to infinity in the critical scaling regime.

Finally, since $|B| < L^d$, this proves the following bound.

Lemma 3.5. *Let m be any nonconstant minimizer for* (1.2)*. Then, with ϵ given as above,*

$$I_B \ge \frac{L^d}{2\chi_-}\left(\big(\delta - 2(\sigma_d/d)(R/L)^d\big)^2 - \epsilon\right)\ .$$

It now follows from Lemmas 3.3 and 3.5 that for any nonconstant minimizer m,

$$\begin{aligned} \mathcal{F}(m) &\geq I_A + I_B \\ &\geq \sigma_d R^{d-1}(S - 2\kappa) + \frac{L^d}{2\chi_-}\big(\delta - 2(\sigma_d/d)(R/L)^d\big)^2 - \frac{L^d}{2\chi_-}\epsilon\ . \end{aligned} \tag{3.17}$$

It now remains to optimize this over R.

Now introduce $S_- = (S - 2\kappa)$ and $\eta = R^d/r_0^d$. Then we can rewrite this lower bound as

$$\mathcal{F}(m) \geq S_-|\Gamma_0|\left(\eta^{1-1/d} + \frac{S\chi}{S_-\chi_-}C(n)(1-\eta)^2\right) - \frac{L^d}{2\chi_-}\epsilon\ . \tag{3.18}$$

Proof of Theorem 1.1. As $L \to \infty$ in the critical scaling regime, $S_- \to S$ and $\chi_- \to \chi$. Moreover, from (3.16), (3.4), and the definition of ϵ, $L^d\epsilon/|\Gamma_0| \to 0$ as $L \to \infty$. Thus, (3.18) provides the lower bound needed to prove (1.20). The upper bound is provided by Lemma 2.2. The remaining statements follow from the analysis of the minimization of the phenomenological free energy function (1.15) that was explained in the introduction. □

4. The structure of the minimizers

4.1. The proof of Theorem 1.2

Suppose that $n = -1 + KL^{d/(d+1)}$ where $K < K_\star$. We would like to conclude from (3.18) that any nonconstant trial function m has a higher free energy than the uniform trial function $m(x) = n$, at least for all sufficiently large L.

Recalling that $S|\Gamma_0|C(n) = \mathcal{F}(n)$, define $\bar\eta$ by

$$\bar\eta = \sup\left\{\eta\ :\ S_-|\Gamma_0|\left(\eta^{1-1/d} + \frac{S\chi}{S_-\chi_-}C(n)(1-\eta)^2\right) - \frac{L^d}{2\chi_-}\epsilon < S|\Gamma_0|C(n)\right\}\ .$$

As in the proof of Theorem 1.1, for all L sufficiently large, S_- is sufficiently close to S, and χ_- is sufficiently close to χ that

$$\frac{S\chi}{S_-\chi_-}C(n) < C$$

for some $C < C_\star$. For $C < C_\star$, the unique minimizer of

$$\eta \mapsto \eta^{1-1/d} + C(1-\eta)^2$$

is $\eta = 0$. Since by (3.16) $\epsilon L^d/|\Gamma_0| \to 0$ as $L \to \infty$, it follows that $\bar\eta \to 0$ as $L \to \infty$.

Now, as in the previous section, for any nonuniform minimizer m, there is a relation between η and the size of the level set $|\{m > 1-\kappa\}|$ given by $\eta = (R/r_0)^d$ and $|\{m > 1-\kappa\}| = (\sigma_d/d)R^d$. Here, as in the last section, $\kappa = \delta^{1/3}$ with δ given by (3.3). It follows from (3.18) and the definition of $\bar\eta$ that for any nonconstant minimizer m, $\eta < \bar\eta$, and so $|\{m > 1-\kappa\}|$ is negligibly small compared with the volume of the equimolar ball; that is, $(\sigma_d/d)r_0^d$, when L is large.

In other words, if $n = -1 + KL^{d/(d+1)}$ where $K < K_\star$, and L is large, then any droplet in any minimizer must be extremely small. To prove Theorem 1.2, it therefore suffices to show that such extremely small drops are impossible in a minimizing order parameter field. We do this in the next lemma.

Lemma 4.1. *For all $K > 0$, there is a constant $C_K > 0$ depending only on K so that if $n \le -1 + KL^{-d/(d+1)}$ and m is any nonuniform minimizer for* (1.2), *then*

$$|\{m > 1 - \kappa\}| \ge C_K r_0^d \ .$$

Moreover, C_K is uniformly strictly positive for all K in an interval around $K_\star$.

Proof. We again work with the functional $\mathcal{G}(w)$ which is defined in (3.7) and related to $\mathcal{F}(m) - \mathcal{F}(n)$ by (3.9). Clearly, if $\mathcal{F}(m) < \mathcal{F}(n)$, then the potential $G(w)$, defined in (3.8), must become negative. However, as seen in the proof of Lemma 3.2, $G(w) < 0$ if and only if $z_- < w < z_+$ where $z_\pm = 2 - 2\delta \pm 2\sqrt{\delta}\sqrt{1-\delta/2}$.

Moreover, $G(w) \ge (n^2-1)w^2/2$ for all w. Since $w = m - n$ and by Lemma 3.1, $m \le 1+\delta$, while by definition $n = -1+\delta$, $w \le 2$. Therefore,

$$G(w(x)) \ge -4\delta(1-\delta/2)$$

for all x.

Define the set $\tilde{C}$ by $\tilde{C} = \{x \ : \ w(x) \ge z_- \}$, and define the number $\tilde{R}$ by

$$(\sigma_d/d)\tilde{R}^d = \left|\tilde{C}\right| \ .$$

We have

$$\int_{\tilde{C}} \left[\frac{1}{2}|\nabla w|^2 + G(w)\right] \mathrm{d}x \ge \int_{\tilde{C}} G(w)\,\mathrm{d}x \ge -4\delta(\sigma_d/d)\tilde{R}^d \ . \tag{4.1}$$

Define the set $\tilde{A}$ by $\tilde{A} = \{x \ : 0 \le \ w(x) \le z_- \}$ where the lower bound 0 is arbitrary but convenient. The same argument used to prove Lemma 3.3 shows that

$$\int_{\tilde{A}} \left[\frac{1}{2}|\nabla w|^2 + G(w)\right] \mathrm{d}x \ge \tilde{S}\sigma_d \tilde{R}^{d-1} \ , \tag{4.2}$$

where $\tilde{S} = \int_0^{z_-} \sqrt{2G(h)}\,\mathrm{d}h$.

It now follows from (3.9) that $\mathcal{F}(m) > \mathcal{F}(n)$ unless

$$4\delta(\sigma_d/d)\tilde{R}^d \ge \tilde{S}\sigma_d\tilde{R}^{d-1} \ .$$

However, $\mathcal{F}(m) > \mathcal{F}(n)$ is impossible if m is a minimizer. Hence, $\tilde{R} \ge d\tilde{S}/(4\delta)$. From (3.3), $\delta = 2(\sigma_d/d)(r_0^{d+1}/L^d)\dfrac{1}{r_0}$. Hence,

$$\tilde{R} \ge \frac{d\tilde{S}}{8(\sigma_d/d)}\frac{L^d}{r_0^{d+1}} r_0 \ .$$

The condition $n \le -1 + KL^{-d/(d+1)}$ yields the bound

$$r_0 \le \left(\frac{K}{2(\sigma_d/d)}\right)^{1/d} L^{d/(d+1)} \ .$$

Thus, there is a constant C_K depending only on K so that $\tilde{R} \ge C_K r_0$.

The final observation to make is $w > z_-$ if and only if $m > 1 - \delta = 2\sqrt{\delta}\sqrt{1-\delta/2}$. Therefore, since $\kappa = \delta^{1/3}$, $\tilde{C} \subset C$ for all L large enough. □

Proof of Theorem 1.2. By Lemma 4.1, whenever m is a minimizer for (1.2) with $n < -1 + KL^{-d/(d+1)}$ and $K < K_\star$, the corresponding value of η is bounded away from zero by a strictly positive quantity depending only on K. But by the remarks preceding Lemma 4.1, the η value of any minimizer cannot exceed $\bar{\eta}$, which tends to zero as L increases. Hence, for L sufficiently large, there are no nonconstant minimizers with $n < -1 + KL^{-d/(d+1)}$ and $K < K_\star$. □

4.2. The proof of Theorem 1.3

The essential tool here is quantitative version of the isoperimetric inequality. The classical version, valid in $d = 2$, is due to Bonnesen [6], who worked in the setting of convex geometry. For the form used here, see [16]. To formulate the inequality, let U be a domain in $\mathbb{R}^2$ bounded by a simply connected rectifiable curve $|\Gamma|$. Suppose that $\rho_{\rm out}(U)$ is the infimum of the radii of circles containing U, and $\rho_{\rm in}(U)$ is the supremum of the radii of circles contained in U. $\rho_{\rm out}(U)$ is called the *outradius* of U, and $\rho_{\rm in}(U)$ is called the *inradius* of U. The inequality of Bonessen states that

$$|\Gamma|^2 - 4\pi|U| \geq \pi(\rho_{\rm out}(U) - \rho_{\rm in}(U))^2 \tag{4.3}$$

where $|\Gamma|$ denotes $\mathcal{H}^1(\Gamma)$, the one-dimensional Hausdorff measure of Γ. That is,

$$\frac{|\Gamma|}{\sqrt{|U|}} \geq \sqrt{\pi}\left(2 + \frac{|\rho_{\rm out}(U) - \rho_{\rm in}(U)|}{\sqrt{|U|}}\right) . \tag{4.4}$$

We shall show how this inequality may be used to prove Theorem 1.3 for $d = 2$, and then shall explain how a recent extension by Hall [12] of Bonnesen's inequality to higher dimensions yields the result for $d > 2$. The essential points are clearest for $d = 2$, and so we begin with that case.

We first have to justify the application of this inequality on the torus. For this, see the Appendix. The basic point is that for a set to take advantage of being in the torus by "wrapping around" to reduce its perimeter, it must already have a very large perimeter anyway — large enough to wrap around.

To apply (4.4), we return to the proof of Lemma 3.3, and take $U = U_h = \{m > h\}$ for $h_- \leq h \leq h_+$. Then, using the notation of Section 3,

$$|U_{h_+}| = |C| \qquad \text{and} \qquad |U_{h_-}| = |C| + |A| .$$

The essential point is that in the critical scaling regime, $|A|$ is negligibly small compared to $|C|$ for large L. This is the content of Lemma 3.4. Therefore, for all $\varepsilon > 0$, if L is sufficiently large,

$$|U_{h_-}| \leq (1+\varepsilon)^2|U_{h_+}| = (1+\varepsilon)^2|C| . \tag{4.5}$$

Recall that in Section 3 we have defined R by $\pi R^2 = |C|$. Now suppose that

$$\rho_{\rm out}(U_{h_+}) \geq (1+2\varepsilon)R . \tag{4.6}$$

Then since for all $h < h_+$, $U_{h_+} \subset U_h$,

$$\rho_{\rm out}(U_h) \geq (1+2\varepsilon)R \qquad \text{for all} \qquad h_- \leq h \leq h_+ . \tag{4.7}$$

On the other hand, since for all $h < h_+$, $U_h \subset U_{h_-}$, so that

$$|U_h| \leq |U_{h_-}| \leq \pi(1+\varepsilon)^2 R^2 \ ,$$

it follows that

$$\rho_{\rm in}(U_h) \leq (1+\varepsilon)R \qquad \text{for all} \qquad h_- \leq h \leq h_+ \ . \tag{4.8}$$

Combining (4.4), (4.7), and (4.8), and letting Γ_h denote the boundary of U_h, we have

$$\frac{|\Gamma_h|}{\sqrt{|U_h|}} \geq \sqrt{\pi}\left(2+\frac{\varepsilon}{1+\varepsilon}\right) \qquad \text{for all} \qquad h_- \leq h \leq h_+ \ . \tag{4.9}$$

Thus, under the hypothesis (4.6),

$$|\Gamma_h| \geq 2\pi\left(1+\frac{\varepsilon}{2+2\varepsilon}\right)R \qquad \text{for all} \qquad h_- \leq h \leq h_+ \ . \tag{4.10}$$

Going back to the proof of Lemma 3.3, one sees that effectively, the hypothesis (4.6) would introduce an extra factor of $(1+\varepsilon/(2+2\varepsilon))$ into the surface tension S. This would increase the leading order contribution to the free energy over what obtained in Section 2 with the trial function $m_{\eta\,\rm dr}$. Therefore, for all sufficiently large L, the hypothesis (4.6) is incompatible with m being a minimizer.

The same conclusion can be obtained from Hall's theorem [12] in higher dimension, in essentially the same way. The version of Hall's theorem found in Theorem 4.1 of [17] is particularly useful for this purpose. We summarize the discussion in a lemma, using the notation introduced above.

Lemma 4.2. *Let $K > K_\star$ and $n \geq -1 + KL^{-d/(d+1)}$. For any $\varepsilon > 0$ and all L sufficiently large, if m is any minimizer for (1.2), then with $B(x_0, R)$ denoting the ball of radius R centered on x_0, there is a point x_0 with*

$$\frac{|C\Delta B(x_0,R)|}{|B(x_0,R)|} \leq \varepsilon \ ,$$

where Δ denotes the symmetric difference $C \cup B(x_0,R)\backslash(C \cap B(x_0,R))$.

Proof of Theorem 1.3. Let m be any minimizer. After translating, we may assume that

$$\frac{|C\Delta B(0,R)|}{|B(0,R)|} \leq \varepsilon \ .$$

Also, since $K > K_\star$, we know that the value of R must be very close to $\eta_c^{1/d} r_0$. Therefore, for all L large enough, we have

$$\frac{\left|C\Delta B\big(0,\eta_c^{1/d} r_0\big)\right|}{\left|B\big(0,\eta_c^{1/d} r_0\big)\right|} \leq 2\varepsilon \ .$$

Now, if $|m(x) - m^\sharp_{n,\eta}(x)\}| > 2\kappa$, it must be that either $x \in C\Delta B(0,\eta_c^{1/d} r_0)$, or else $x \in A$. Hence, the set

$$\left\{\ x\ :\ \left|m(x) - m^\sharp_{n,\eta}(x)\}\right| > 2\kappa\ \right\}$$

has a volume no greater than

$$2\varepsilon|B(0,r_0)| + |A| \ ,$$

and we recall that by Lemma 3.4, $|A|$ is negligibly small compared to r_0^d for all sufficiently large L.

Thus, we obtain

$$\left|\left\{\, x \;:\; \left|m(x) - m^\sharp_{n,\eta}(x)\right\}\right| > 2\kappa \,\right\}\right| \le 3\varepsilon|B(0,r_0)|$$

for all sufficiently large L. Of course we have the globally valid bound $|m(x) - m^\sharp_{n,\eta}(x)\}| \le 2$. Therefore,

$$\int_\Omega \left|m(x) - m^\sharp_{n,\eta}(x)\right\}\right|^4 \mathrm{d}x \le 48\varepsilon|B(0,r_0)| + 16\kappa^4 L^d \;.$$

Recall that $\kappa = \delta^{1/3}$, and that in the critical scaling regime,

$$\delta L^d \asymp L^{d/(d+1)} \asymp |B(0,r_0)| \;.$$

Therefore, for any $\epsilon > 0$

$$\frac{1}{|B(0,r_0)|}\int_\Omega \left|m(x) - m^\sharp_{n,\eta}(x)\right\}\right|^4 \mathrm{d}x < \epsilon$$

for all L sufficiently large. □

5. The construction of good trial functions

5.1. A Chapman-Enskog-Hilbert expansion approach

As we have seen, the simple trial function $m_{\eta\,\mathrm{dr}}$ was sufficient to provide the upper bounds required here. In fact, it is quite likely that the upper bounds computed in Section 2 are accurate to at least the first two orders in powers of L. Our goal here is to present a systematic construction of high order trial functions. This will explain the remarkable efficacy of the simple prescription $m_{\eta\,\mathrm{dr}}$ for (1.1), and will also suggest how one should construct trial functions of similar efficacy for other free energy functionals. To keep this section concise, we only treat the case $d = 2$. This is fully representative, except that the formulas are much simpler.

For $d = 2$, in the critical scaling regime, $r_0 \asymp L^{2/3}$. We introduce the scaling parameter $\lambda = r_0^{-1}$. After the rescaling $x' = \lambda x$, so that $\Omega \to \Omega^\lambda = \lambda\Omega$, so that $|\Omega^\lambda| = L^2\lambda^2 = L^2 r_0^{-2}$, the Euler-Lagrange Equation (3.1) becomes

$$-\lambda^2\Delta m + F'(m) + \mu^\lambda = 0 \;. \tag{5.1}$$

We will drop the prime on the new coordinate x for sake of simplicity.

An approximate solution of order N to the Euler-Lagrange equation is a function $m^{(N)}$ such that

$$-\lambda^2\Delta m^{(N)} + f\big(m^{(N)}\big) + \mu^{(N)} = O\big(\lambda^{N+1}\big) \;. \tag{5.2}$$

Since we are interested in solutions m to the Euler-Lagrange equation such that

$$\frac{1}{\left|\Omega^\lambda\right|}\int_{\Omega^\lambda} m(x)\,\mathrm{d}x = n \;,$$

we require that the constraint on the mass be approximately satisfied in the sense that the approximate solution $m^{(N)}$ satisfies

$$\frac{1}{\left|\Omega^\lambda\right|}\int_{\Omega^\lambda} m^{(N)}(x)\,\mathrm{d}x = n + O\big(\lambda^{N+1}\big) \;. \tag{5.3}$$

Our aim here is to develop an expansion method, based on the Chapman-Hilbert-Enskog expansion of kinetic theory to construct such approximate solutions, and to use them as trial function for (1.2), after adding a small constant so that the constraint is exactly satisfied.

To do this, we first introduce local coordinates in a neighborhood of the curve $\Gamma^{(N)}$, which will be determined in the course of the expansion. Let s denote an arc length parameter along $\Gamma^{(N)}$. The starting point of the parameterization is immaterial. We denote by $d(x, \Gamma^{(N)})$ the signed distance of x from $\Gamma^{(N)}$, with $d(x, \Gamma^{(N)}) > 0$ when x is in the interior of $\Gamma^{(N)}$ (i.e., the smaller of the two regions into which $\Gamma^{(N)}$ divides the tours). Define a "fast" variable z by $z = d(x, \Gamma^{(N)})/\lambda$. Then (s, z) give us a system of coordinates on a tubular neighborhood around $\Gamma^{(N)}$.

To construct the approximate solutions, we make the following prescription, which has several parts. For any positive integer N:
(1) The interfacial curve $\Gamma^{(N)}$ will be a circle of radius $r^{(N)}$ to be determined at each order, essentially by the condition (5.3). Note that, because of the rescaling the radius $r^{(N)}$ is actually measured in units r_0 and the original units of the radius of the circle is $r^{(N)}r_0$.
(2) The chemical potential $\mu^{(N)}$ has an expansion of the form

$$\mu^{(N)} = \lambda\mu_1 + \lambda^2\mu_2 + \cdots + \lambda^N\mu_N \ .$$

(3) We then construct

$$m^{(N)} = \bar{m}\left(\frac{d\big(x, \Gamma^{(N)}\big)}{\lambda}\right) + \sum_{n=1}^{N} \lambda^n \left[h_n + \phi_n\right] \tag{5.4}$$

where: (i) m_0 is the approximation to $\bar{m}$ introduced in Section 2, (ii) ϕ_j will be a bounded continuous function that is nearly constant away from $\Gamma^{(N)}$. (iii) h_j is a function that has the form

$$h_j\left(\frac{d\big(x, \Gamma^{(N)}\big)}{\lambda}\right) ,$$

where on the right, $h_j(z)$ denotes a rapidly decaying function of the variable z. (The notation is such that the symbol h_n plays two roles, but this should cause no confusion.)

By using the local coordinates (s, z) around the curve $\Gamma^{(N)}$, we write the Laplacian as

$$\lambda^2\Delta f = \frac{\partial^2}{\partial z^2} f + \frac{\lambda K^{(N)}}{\big(1 - z\lambda K^{(N)}\big)} \frac{\partial}{\partial z} f + \frac{\lambda^2}{\big(1 - z\lambda K^{(N)}\big)^2} \frac{\partial^2}{\partial s^2} \tag{5.5}$$

and

$$\frac{1}{1 - z\lambda K^{(N)}} = \sum_{n=0} (-1)^n \frac{\big(\lambda K^{(N)} z\big)^n}{n} := \sum_{n=0} \lambda^n k_n \ . \tag{5.6}$$

Note that our simplifying assumption that $\Gamma^{(N)}$ is a circle amount to assume that $K^{(N)}$ does not depend on s, thus dropping a few terms of higher order in λ in (5.5). Note also, for future reference that the area element in these coordinates is

$$\big(1 + K^{(N)}\lambda z\big)\lambda \, \mathrm{d}z \, \mathrm{d}s \tag{5.7}$$

and recall that the surface tension, S, is given by

$$S = \int_{\mathbb{R}} \left|\bar{m}'(z)\right|^2 \mathrm{d}z = 2\int_{\mathbb{R}} F\big(\bar{m}'(z)\big) \, \mathrm{d}z \ . \tag{5.8}$$

We expand $f = F'$ as

$$f\big(m^{(N)}\big) = f(\bar m) + f'(\bar m)\sum_{n=1}^{N}\lambda^n(h_n+\phi_n) + \frac{1}{2}f''(\bar m)\left(\sum_{n=1}^{N}\lambda^n(h_n+\phi_n)\right)^2 + \frac{1}{3!}f'''(\bar m)\left(\sum_{n=1}^{N}\lambda^n(h_n+\phi_n)\right)^3 . \tag{5.9}$$

Our goal is to show that we can choose the $r^{(j)}$, μ_j, h_j and ϕ_j so that $m^{(N)}(r)$ is a high order approximate solution of (5.1). At the j-th stage, to determine ϕ_j, we will solve an equation in $L^\infty(\mathbb{R})$, and to determine h_j, we will solve an equation in $L^2(\mathbb{R})$. The Fredholm criterion for solvability of this equation will relate μ_j to $r^{(j)}$, and the constraint Equation (5.3) will then determine $r^{(j)}$.

5.2. The first-order approximate solution

To see how this goes, we insert

$$m^{(1)}(r) = \bar m\left(\frac{d\big(x,\Gamma^{(1)}\big)}{\lambda}\right) + \lambda(h_1+\phi_1)$$

into (5.1), collect terms by order in λ, and solve the resulting equations to find r_1, μ_1, h_1 and ϕ_1.

The result is:

$$-\bar m'' + F'(\bar m) + \lambda\Big(-K^{(1)}\bar m' - h_1'' - \phi_1'' + F''(\bar m)h_1 + F''(\bar m)(\phi_1+\mu_1)\Big) + \mathcal{O}\big(\lambda^2\big) .$$

Since, by the definition of $\bar m$ the term multiplied by λ^0 vanishes, we need to equate to 0 the coefficient of the term of order λ. This corresponds to put

$$-K^{(1)}\bar m' + \big(-h_1'' + F''(\bar m)h_1\big) + \big(-\phi_1'' + F''(\bar m)\phi_1\big) + \mu_1 = 0 . \tag{5.10}$$

Define the operator $\mathcal{L}$ by

$$\mathcal{L} = -\frac{\mathrm{d}^2}{\mathrm{d}z^2} + F''(\bar m) .$$

Also define $\mathcal{L}_0$ by

$$\mathcal{L}_0 = -\frac{\mathrm{d}^2}{\mathrm{d}z^2} + F''(1) = -\frac{\mathrm{d}^2}{\mathrm{d}z^2} + \frac{1}{\chi} .$$

In the present example, $1/\chi = 2$, and so $\mathcal{L}^{-1}$ is given by convolution with the Helmholtz Green's function

$$R(z) = \frac{1}{2\sqrt{2}}e^{-\sqrt{2}|z|} .$$

$\mathcal{L}_0^{-1}$ maps constants into constants and preserve the parity properties of functions.

Then we can rewrite (5.10) as

$$\mathcal{L}h_1 = K^{(1)}\bar m' + \big(F''(1) - F''(\bar m)\big)\phi_1 - \mathcal{L}_0\phi_1 - \mu_1 . \tag{5.11}$$

The first two terms on the right-hand side are rapidly decaying in z. In order for the entire right-hand side to rapidly decay, and thus belong to $L^2(\mathbb{R})$, we require that the last to terms cancel. That is, we require

$$\mathcal{L}_0\phi_1 + \mu_1 = 0 \ .$$

This is solved by

$$\phi_1 = -\chi\mu_1 \ , \tag{5.12}$$

and so (5.11) reduces to

$$\mathcal{L}h_1 = K^{(1)}\bar{m}' + \big(1 - \chi F''(\bar{m})\big)\mu_1 \ . \tag{5.13}$$

We have

$$\phi_1 = \mathcal{L}_0^{-1}\mu_1 = -\frac{1}{\chi}\mu_1 \ . \tag{5.14}$$

The equation for h_1 becomes

$$\mathcal{L}h_1 = K^{(1)}\bar{m}' + \big(1 - \chi f'(\bar{m})\mu_1 = 0 \ . \tag{5.15}$$

The null space of $\mathcal{L}$ is spanned by $\bar{m}'$, and so the Fredholm criterion says that (5.15) is solvable if and only if $K^{(1)}\bar{m}' + (1 - \chi f'(\bar{m}))\mu_1$ is orthogonal to $\bar{m}'$. Multiply by $\bar{m}'$ and integrate. Using the fact that $\int_{\mathbb{R}} f'(\bar{m})\bar{m}' \, \mathrm{d}z = 0$, and $\int_{\mathbb{R}} \bar{m}' \, \mathrm{d}z = -2$, we obtain,

$$K^{(1)}\left(\int_{\mathbb{R}} \left(\bar{m}'\right)^2 \mathrm{d}z\right) = -2\mu_1 \ .$$

We see that to leading order in λ, the curvature must be constant, and so $\Gamma^{(N)}$ must be a circle, since we are considering values of n that are close to -1. Let $r^{(1)}$ be the radius of the circle, which is, as yet, undetermined.

Using (5.8) to express the integral in terms of S, we obtain

$$\mu_1 = -\frac{K^{(1)}S}{2} \ . \tag{5.16}$$

Moreover, since $\bar{m}(z) = -\tanh(z/\sqrt{2})$, it is easy to see that both $\bar{m}'(z)$ and $(1 - \chi f'(\bar{m}))$ are proportional to $\mathrm{sech}^2(z/\sqrt{2})$, and so with μ_1 given by (5.16), the right-hand side of (5.15) vanishes identically, and we see that $h_1 = 0$. From (5.14) and (5.16), we have

$$\phi_1 = \chi\frac{K^{(1)}S}{2} \ . \tag{5.17}$$

Finally, we determine $r^{(1)}$ using the approximate constraint (5.3). Since $K^{(1)} = 1/r^{(1)}$, we have that

$$m^{(1)} = \bar{m}\left(\frac{d\big(x, \Gamma^{(1)}\big)}{\lambda}\right) + \frac{\chi S}{2r^{(1)}} \ .$$

Toward this end, note that by (5.7),

$$\frac{1}{\left|\Omega^\lambda\right|}\int_{\Omega^\lambda} \bar{m}\left(\frac{d\big(x, \Gamma^{(1)}\big)}{\lambda}\right) \mathrm{d}x = -1 + 2\pi\frac{\big(r^{(1)}\big)^2}{\left|\Omega^\lambda\right|} + O\big(\lambda^2\big) \ .$$

Thus,

$$\begin{aligned}\frac{1}{|\Omega^\lambda|}\int_{\Omega^\lambda} m^{(1)}(x)\,\mathrm{d}x &= -1+2\pi\frac{\big(r^{(1)}\big)^2}{\big|\Omega^\lambda\big|}+\lambda\phi_1+O\big(\lambda^2\big)\\ &= -1+2\pi\frac{\big(r^{(1)}\big)^2}{\big|\Omega^\lambda\big|}+\lambda\frac{\chi S}{2r^{(1)}}+O\big(\lambda^2\big)\\ &= n+O\big(\lambda^2\big)=-1+\frac{2\pi}{\big|\Omega^\lambda\big|}+O\big(\lambda^2\big)\,. \end{aligned} \tag{5.18}$$

This yields

$$2\pi\big[\big(r^{(1)}\big)^2-1\big]+\lambda\big|\Omega^\lambda\big|\frac{\chi S}{2r^{(1)}}=0\,, \tag{5.19}$$

which is a cubic equation determining $r^{(1)}$. One could solve it explicitly — it is, after all, a depressed cubic. However, the important point is that the of the three roots, only two are positive, and the largest one is exactly the radius determined by the Biskup-Kotecky-Chayes prescription.

To see this, we compute the free energy $\mathcal{F}(m^{(1)})$. First note that, with the scaling we used, we have

$$\mathcal{F}(m^{(1)})=\lambda^{-2}\int_{\Omega^\lambda}\left[\frac{\lambda^2}{2}\big|\nabla m^{(1)}\big|^2+F\big(m^{(1)}\big)\right]\mathrm{d}x\,.$$

By using the expression of $m^{(1)}$ and (5.7) to pass to the variables (s,z), we get

$$\mathcal{F}\big(m^{(1)}\big)=\lambda^{-1}2\pi Sr^{(1)}+\frac{\lambda^{-2}\big|\Omega^\lambda\big|}{2\chi}\lambda^2\phi_1^2\,.$$

From (5.18) and (1.6) we compute

$$\phi_1=\frac{2\pi\big(1-\big(r^{(1)}\big)^2\big)}{\lambda\big|\Omega^\lambda\big|}+O(\lambda)\,. \tag{5.20}$$

Therefore

$$\mathcal{F}\big(m^{(1)}\big)=\lambda^{-1}\left(2\pi r^{(1)}S+\frac{\lambda^{-3}L^{-2}}{\chi}2\pi^2\big(1-\big(r^{(1)}\big)^2\big)^2\right)+O(1)\,.$$

This is exactly the phenomenological free energy of [4] for the free energy functional (1.1). Also, the Euler-Lagrange equation for it reduces to (5.19). Hence, $m^{(1)}$, with $r^{(1)}$ chosen to be the minimizing solution of (5.19) is essentially exactly the trial function we used to obtain the upper bound in Section 2. The only difference is a slight adjustment of the additive constant so that the constraint in (1.2) is exactly satisfied.

The crucial feature in the free energy function (1.1) that is responsible for this is that in this case, we found $h_1=0$.

5.3. The second order approximate solution

Going on to second order is not difficult since $h_1=0$, and ϕ_1 is an explicit constant. The next order displays some new features, but once the calculations are carried out to second order, it will be clear how to go on to arbitrary order.

The equations for h_2 and ϕ_2 are

$$-\frac{d^2}{dz^2}(h_2+\phi_2) - K^{(2)}\bar{m}' + f'(\bar{m})(h_2+\phi_2)\frac{1}{2}f''(\bar{m})(h_1+\phi_1)^2 + \mu_2 = 0\ .$$

As before, write

$$\mathcal{L}\phi_2 = \mathcal{L}_0\phi_2 + \big(f'(\bar{m}) - f'(1)\big)\phi_2\ ,$$

and we then have

$$\mathcal{L}h_2 = -\Big[K^{(2)}\bar{m}' + \big(F''(\bar{m}) - f'(1)\big)\phi_2\Big] - \mathcal{L}_0\phi_2 - \frac{1}{2}f''(\bar{m})\phi_1^2 - \mu_2\ .$$

The terms in square brackets are rapidly decaying as long as ϕ_2 is bounded. To eliminate the other terms, we choose

$$\mathcal{L}_0\phi_2 = -\frac{1}{2}F'''(\bar{m})\phi_1^2 - \mu_2\ .$$

This is easily solved in $L^\infty(\mathbb{R})$ using the Helmholtz Green's function. With this choice of ϕ_2, we determine h_2 through

$$\mathcal{L}h_2 = -K^{(2)}\bar{m}' - \big(F''(\bar{m}) - F''(1)\big)\phi_2\ .$$

The right-hand side must be orthogonal to $\bar{m}'$. Recall that $(f'(\bar{m}) - f'(1))$ is even, and in fact is a multiple of $\bar{m}'$. Since $f''(\bar{m}) = 6\bar{m}$, which is odd, only the even part of ϕ_2 is relevant in the solvability condition. But clearly, the even part of ϕ_2 is given by

$$(\phi_2)_{\text{even}} = -\chi\mu_2\ .$$

Thus, the solvability condition becomes

$$K^{(2)}\left(\int_{\mathbb{R}}\big(\bar{m}'(z)\big)^2\,\mathrm{d}z\right)^2 + \frac{1}{\chi}\chi 2\mu_2 = 0\ .$$

As before, with this choice,

$$K^{(2)}\bar{m}' + \big(f'(\bar{m}) - f'(1)\big)(\phi_2)_{\text{even}} = 0\ ,$$

since it is a multiple of $\bar{m}'$, and the equation for h_2 reduces to

$$\mathcal{L}h_2 = -\big(f'(\bar{m}) - f'(1)\big)(\phi_2)_{\text{odd}}\ .$$

The mass condition is

$$\begin{aligned}
\frac{1}{|\Omega_\lambda|}\int_{\Omega_\lambda} m^{(2)}(x)\,\mathrm{d}x &= \frac{1}{|\Omega_\lambda|}\left[\int_{\Omega_\lambda}\bar{m}(x) + \lambda|\Omega_\lambda|\phi_1 + \lambda^2|\Omega_\lambda|(\phi_2)_{\text{even}} + \lambda^2\int_{\Omega_\lambda}(\phi_2)_{\text{odd}}(x)\right]\\
&= -1 + \frac{2\pi\big(r^{(2)}\big)^2}{|\Omega_\lambda|} + \lambda\phi_1 + \lambda^2[(\phi_2)_{\text{even}} + (\phi_2)_{\text{odd}}(+\infty)] + \mathcal{O}\big(\lambda^3\big)\\
&= n + O\big(\lambda^3\big) = -1 + \frac{2\pi}{|\Omega^l|} + O\big(\lambda^3\big)\ .
\end{aligned}$$

From 5.19, by choosing $r^{(2)} = r^{(1)} + \lambda r_2$, we get

$$r_2 = -\frac{(\phi_2)_{\text{even}} + (\phi_2)_{\text{odd}}(+\infty)}{r^{(1)}4\pi}\ . \tag{5.21}$$

Introducing the notation $\hat{\phi}^{(2)} := (\phi_2)_{\text{even}} + (\phi_2)_{\text{odd}}(+\infty)$ and recalling that $\phi_1 = \dfrac{\chi S}{2r^{(1)}}$ we get

$$r_2 = \frac{\phi_1 \hat{\phi}_2}{\chi S 2\pi} \,. \tag{5.22}$$

The correction $\hat{\phi}^{(2)}$ is the most significant term among the new correction. However, it is a constant, so that even keeping the most significant term at second order, we still have a trial function of the simple type considered in Section 2. For this reason, one can expect that the upper bounds obtained in Section 2 are in fact sharp not only in the leading order, but in the first two orders in powers of L. To see that this is indeed the case, we compute the free energy $\mathcal{F}(m^{(2)})$

$$\mathcal{F}\big(m^{(2)}\big) = \lambda^{-2} \int_{\Omega_\lambda} \frac{1}{2} \left[\lambda^2 \big|\nabla m^{(2)}\big|^2 + F\big(m^{(2)}\big) \right] dx \,.$$

We have

$$\int_{\Omega_\lambda} \frac{1}{2} \lambda^2 \big|\nabla m^{(2)}\big|^2 = \lambda \big|\Gamma^{(2)}\big| \frac{1}{2} \int_{\mathbb{R}} \big(\bar{m}'\big)^2 \, dz + \lambda^3 \big|\Gamma^{(2)}\big| \int_{\mathbb{R}} \big(\bar{m}'\big)(h_2 + (\phi_2)_{\text{odd}})' \, dz + \mathcal{O}\big(\lambda^4\big)$$

$$\begin{aligned} F\big(m^{(2)}\big) &= F(\bar{m}) + F'(\bar{m})\big[\lambda\phi_1 + \lambda^2(h_2 + \phi_2) + \lambda^3(h_3 + \phi_3)\big] \\ &\quad + \frac{1}{2} F''(\bar{m})\big[\lambda^2(\phi_1)^2 + \lambda^3 2\phi_1(\phi_2 + h_2)\big] + \frac{1}{3!} F'''(\bar{m})\lambda^3(\phi_1)^3 + O\big(\lambda^4\big) \,. \end{aligned} \tag{5.23}$$

$$\int_{\Omega_\lambda} F''(\bar{m}) \, \mathrm{d}x = F''(-1)|\Omega_\lambda| + F''(1)\pi \big(r^{(2)}\big)^2 + \mathcal{O}\big(e^{-L^{1/4}}\big) \,, \tag{5.24}$$

$$\int_{\Omega_\lambda} F'(\bar{m}) \, \mathrm{d}x = \mathcal{O}\big(\lambda^2\big) \,, \quad \int_{\Omega_\lambda} \frac{1}{3!} F'''(m_0) \, \mathrm{d}x = \left[\pi \big(r^{(2)}\big)^2 - |\Omega_\lambda|\right] + \mathcal{O}\left(\lambda^2\right) \,.$$

Putting everything together and using that $F(\pm 1) = F'(\pm 1) = 0$, $F''(\pm 1) = \chi^{-1}$, $F'''(m) = 6m$ and $\lambda|\Omega_\lambda| = 1$ we get

$$\begin{aligned} \mathcal{F}\big(m^{(2)}\big) &= \lambda^{-1} \left[2\pi r^{(2)} S + \frac{1}{2}(\phi_1)^2 \chi^{-1} \lambda |\Omega_\lambda| \right] + \phi_1 \hat{\phi}_2 \chi^{-1} \lambda |\Omega_\lambda| - \lambda |\Omega_\lambda| (\phi_1)^3 + \mathcal{O}(\lambda) \\ &= \lambda^{-1} \left[2\pi r^{(2)} S + \frac{1}{2}(\phi_1)^2 \chi^{-1} \right] + \left[\phi_1 \hat{\phi}_2 \chi^{-1} - (\phi_1)^3\right] + \mathcal{O}(\lambda) \,. \end{aligned}$$

Finally, by (5.22) we have

$$\begin{aligned} \mathcal{F}\big(m^{(2)}\big) &= \lambda^{-1} \left[2\pi r^{(1)} S - \lambda\big(\phi_1 \hat{\phi}_2 \chi^{-1}\big) + \frac{1}{2}(\phi_1)^2 \chi^{-1} \right] + \left[\phi_1 \hat{\phi}_2 \chi^{-1} - (\phi_1)^3\right] + \mathcal{O}(\lambda) \\ &= \lambda^{-1} \big[2\pi r^{(1)} S - \lambda(\phi_1)^3 \big] + O(\lambda) \,. \end{aligned}$$

The expression of ϕ_1 in this context is determined by the condition (5.20) which is the analogous of (2.5). The comparison allows to identify α_η with ϕ_1 for a droplet of radius $\eta = r^{(1)}$. The correction is exactly of the same form as in (2.8). On the critical value of $r^{(1)}$ the first term reduces to $\lambda^{-1}\mathcal{F}(n)$.

The procedure can be continued along the same lines to higher order thus producing approximate solutions to the Euler-Lagrange equations. However, what is probably more significant is that it can be applied to other free energy functional with nonlocal interactions for which it will not be the case that $h_1 = 0$, and hence the construction of a suitable trial function is not so simple.

6. Appendix: Isoperimetry on the flat torus

The purpose of this Appendix is to gather some facts concerning isoperimetry in the that flat torus in $\mathbb{R}^d$, $d \geq 2$. By the flat torus, we mean simply the unit cube in $\mathbb{R}^d$ with periodic boundary conditions; i.e., with the usual identification of points on opposite faces. The article of Morgan and Johnson, [15], is a basic reference for this problem.

The situation for $d = 2$ is relatively trivial as explained in [15], since extremal sets will be bounded by curves of constant curvature, and there are not many of these. In higher dimension, there are many surfaces of constant mean curvature, so that what one learns about the problem from the Euler-Lagrange equation is less and less helpful as the dimension increases. Even in $d = 3$, the situation is not well understood except for small volumes. However, as small volumes are our main concern here, Theorem 4.4 of [15] provides a sufficient basis for our analysis here.

To formulate this result, let $P(V)$ denote the minimal perimeter of a set of volume V in the unit flat torus. By the regularity of the minimizing sets, see [15], $P(V)$ will be the $d - 1$ dimensional Hausdorff measure of the boundary of an extremal set. Clearly, on the unit flat torus, $P(V) = P(1 - V)$. Also, as shown in [15], P is a concave function of V, and so it has its maximum at $V = 1/2$, and we need only consider V in the range $0 \leq V \leq 1/2$, on which P is an increasing function of V. The following is a special case of Theorem 4.4 in [15], which provides the deep lower bound on P, and consideration of a ball, which trivially provides the upper bound on P.

Theorem 6.1 (Morgan and Johnson). *For all $d \geq 2$, there is an $\epsilon > 0$ so that for $V \leq \epsilon$, the isoperimetric function $P(V)$ on the unit flat d-dimensional torus satisfies*

$$P(V) = \sigma_d^{1/d}(dV)^{1-1/d} \ . \tag{6.1}$$

Note that the right-hand side is exactly the isoperimetric function in $\mathbb{R}^d$. Thus, the theorem asserts the intuitively clear result that for small volumes it is impossible to take advantage of "wrapping around" in the torus to reduce the perimeter.

The proof in [15] does not give an explicit value for ϵ. It is, however, proved in a much wider setting. If one specialized the argument proving Theorem 4.4 in [15] to the torus, it would simplify considerably, and yield an explicit value of ϵ.

There is an alternative to the use of the above result, which is useful for obtaining lower bounds on the perimeter of sets that are not balls. It also fairly simple, so we explain it here.

Let A be a set in the unit flat d-dimensional tous, regarded as the unit cube in $\mathbb{R}^d$ with periodic boundary. Let $a_1(t)$ denote the $d - 1$ dimensional Hausdorff measure of the "slice" cut through A by the plane $x_1 = t$. Let a_1 be the essential infimum of this function on [0, 1]. Pick any $\epsilon > 0$. By translating A in the torus, we may arrange the $a_1(0) \leq a_1 + \epsilon$. Since the volume of A is $V = \int_0^1 a_1(t)\,\mathrm{d}t \geq a_1$, it follows that

$$a_1(1) = a_1(0) \leq V + \epsilon \ .$$

In the same way, we obtain a similar bound on the $d - 1$ dimensional Hausdorff measure of the "slice" cut through A by the other bounding planes — note that the translation do not interfere with one another, and may be taken in any order.

Therefore, after making suitable translations, if we imbed the torus in $\mathbb{R}^d$ by simply dropping the periodic boundary identification we increase the boundary of A by at most

$$2d(V + \epsilon) \ .$$

Now let P_E denote the Euclidean isoperimetric function in $\mathbb{R}^d$. The boundary of A considered as a subset of $\mathbb{R}^d$ is at least $P_E(V)$. Some part of this may have come from additional boundary points created by removing the periodic identification. By we have just bounded the contribution of these. Since $\epsilon > 0$ is arbitrary, we obtain the following.

Theorem 6.2. *For all $d \geq 2$, let $P(V)$ denote the isoperimetric function in the unit flat d-dimensional torus, and let $P_E(V)$ denote the Euclidean isoperimetric function in $\mathbb{R}^d$. The*

$$P(V) \geq P_E(V) - 2dV .$$

The point of this result is that for small V and $d \geq 2$, V is a small correction to $P(V)$, which is proportional to $V^{(d-1)/d}$. Thus, for small sets V, the leading term in the isoperimetric function for the flat torus is simply the euclidean isoperimetric function. We could use Theorem 6.2 just as well as Theorem 6.1 to justify the proofs of the Theorems 1.1 and 1.2. The same sort of reasoning also enables us to apply Bonessen type estimates in the torus, such as are used in Theorem 1.3.

To explain this, consider the case $d = 2$ where we have (4.3), which we recall is

$$|\Gamma|^2 - 4\pi|U| \geq \pi(\rho_{\text{out}}(U) - \rho_{\text{in}}(U))^2 .$$

Consider this in the unit torus. If

$$\rho_{\text{out}}(U) \leq 1/2$$

then, after a translation, U does not intersect the boundaries of the cube. Hence, when we drop the periodic boundary identification, we do not create any new boundary of U. We may therefore regard U as a subset of $\mathbb{R}^2$, and apply the Bonessen inequality as usual.

On the other hand, suppose that $|U|$ is small, but that

$$\rho_{\text{out}}(U) \geq 1/2 .$$

Since clearly

$$|U| \geq \pi\rho_{\text{in}}^2(U) ,$$
$$\rho_{\text{in}}(U) \leq \sqrt{|U|/\pi} .$$

Now applying the Bonessen inequality to U considered as a subset of $\mathbb{R}^2$ — with the periodic boundary identification dropped — we find that the length L_E of its perimeter satisfies

$$L_E^2 \geq 4\pi|U| + \pi\big(1/2 - \sqrt{|U|/\pi}\big)^2 .$$

However, at most a length of $4|U|$ was added to $|\Gamma|$ when we removed the periodic boundary identification (after an appropriate translation). Thus,

$$|\Gamma|^2 \geq 4\pi|U| + \pi\big(1/2 - \sqrt{|U|/\pi}\big)^2 - 4|U| .$$

The right-hand side tends to $\pi/4$ as $|U|$ tends to zero. This provides a large lower bound on $|\Gamma|$ in the case that $\rho_{\text{out}}(U) \geq 1/2$, and otherwise we use Bonessen's inequality exactly as we would in $\mathbb{R}^2$.

Similar arguments apply for Hall's higher-dimensional generalization [12] of Bonessen's inequality.

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