Free Energy Minimizers for a Two–Species Model with Segregation and Liquid-Vapor Transition.

by

E. A. Carlen[(1)], M. C. Carvalho [(2)], R. Esposito[(3)], J. L. Lebowitz[(4)] and R. Marra[(5)]

Abstract

We study the coexistence of phases in a two–species model whose free energy is given by the scaling limit of a system with long range interactions (Kac potentials) which are attractive between particles of the same species and repulsive between different species.

Key words: phase transition, segregation, convexity, rearrangement inequalities

1. Introduction.

The rigorous *ab initio* derivation of the full phase diagram of even the simplest realistic model of a physical system, is still far beyond our mathematical grasp. This is so even when we restrict ourselves to simple classical one component system, e.g., point particles with Lennard-Jones type pair interactions. Only the low density–high temperature properties of such a system can be fully derived from statistical mechanics: there one can prove analytic behavior of the free energy as a function of the fugacity (or density) and temperature. Beyond that we have almost no means to prove the commonly observed facts of the phase diagram such as the Gibbs phase rule, the smoothness of the boundaries between the phases, etc. [17]

To overcome this deficiency one can consider even more simplified models such as lattice systems. There, thanks to the Pirogov-Sinai [15], [22] theory, some exactly solvable models, inequalities, etc., one has much better control over the low temperature region of the phase diagram [17], [20], [7], [19]. Alternatively, one can consider, following the pioneering work of van der Waals [21], [18], [16], [5], mean-field-theories (MFT) yielding approximate equations for state or free energies which indeed exhibit most of the qualitative and many quantitative features of real world phase diagrams. These results were initially obtained in a heuristic manner and had to be supplemented by additional rules, e.g. the Gibbs double tangent or Maxwell equal area rule, to make them thermodynamically consistent. Subsequently, following the work of van Kampen [11], Kac, Uhlenbeck and Hemmer [9], Lebowitz and Penrose [12] were able to derive generalized mean field models in a more consistent mathematical way by considering systems in which part of the interactions were explicitly modeled by Kac potentials. Kac potentials are simply potentials of the form $\gamma^d U(\gamma x)$, which contain a range parameter γ^{-1} [9], [12]. For fixed $\gamma > 0$ these interactions are essentially finite ranged but in the limit $\gamma \to 0$ they become mean field like [12]. Depending on whether γ^{-1} is small or comparable to the macroscopic scale (but always very large compared to the interparticle distances) one gets either the usual MFT (including the supplemental rules) or a macroscopic continuum theory (MCT) from which one can also obtain the surface tension associated with phase transitions caused by these Kac potentials [8], [1].

(1) School of Mathematics, Georgia Institute of Technology, Atlanta, GA 30332–0160

(2) Department of Mathematics and GFM, University of Lisbon, Av. Prof. Gama Pinto no 2 1649-003 Lisbon Portugal

(3) Centro Linceo "Beniamino Segre", and Dip. di Matematica, Università di L'Aquila, Coppito, 67100 AQ, Italy

(4) Departments of Mathematics and Physics, Rutgers University, New Brunswick, NJ 08903, USA

(5) Dipartimento di Fisica and Unità INFM, Università di Roma Tor Vergata, 00133 Roma, Italy.

More recently it has been possible to prove phase transitions close to the mean field ones for large finite γ^{-1} [13].

The structure of the coexisting phases in the MCT for a one component system has been extensively investigated in recent years by Presutti et al [6] and references therein. In multicomponent systems the MCT is rather unexplored. In the present work we study binary mixtures where the phase diagram has new qualitative features, resulting from the possibility of segregation of the system into regions rich in one of the species, e.g. oil and water. Such systems were already considered by van der Waals [21] and by Korteweg [10] using MFT expressions for the free energy. MFT however neglects the spatial structures of the phase domains so the starting point of our analysis is a MCT expression for the free energy $\mathcal{F}$ of a binary mixture in a macroscopic domain Λ, which we shall take for simplicity to be a d-dimensional torus of side length L and volume $|\Lambda| = L^d$.

The free energy functional that we consider satisfies certain rearrangement inequalities, and we study the consequences of these for the phase diagram. We will later focus on special cases of physical interest, such as a mixture of van der Waals gases with a repulsive interaction between the species. However, to make clear the role of the rearrangements, we begin in a general setting in which these can be applied.

Let ρ_1 and ρ_2 denote the densities for the two species, and consider a "free energy" functional $\mathcal{F}$ that depends on ρ_1 and ρ_2 through

$$\begin{aligned}\mathcal{F}(\rho_1,\rho_2) = \int_\Lambda F(\rho_1(x),\rho_2(x))\mathrm{d}x + \int_\Lambda\int_\Lambda V(x-y)\rho_1(x)\rho_2(y)\mathrm{d}x\mathrm{d}y \\ - \frac{1}{2}\int_\Lambda\int_\Lambda U(x-y)[\rho_1(x)\rho_1(y)+\rho_2(x)\rho_2(y)]\mathrm{d}x\mathrm{d}y\ .\end{aligned} \tag{1.1}$$

The function F represents the free energy density of a system with only short range interactions. We require that F be separately convex in ρ_1 and ρ_2, and that the set of all (ρ_1,ρ_2) for which F is finite, which we call the domain of F, is an open convex set. Finally we require that

$$\frac{\partial^2}{\partial\rho_1\partial\rho_2}F(\rho_1,\rho_2) \geq 0 \tag{1.2}$$

for all (ρ_1,ρ_2) in the domain of F. An example of particular interest is given by

$$F(\rho_1,\rho_2) = \frac{1}{\beta}\left[G(\rho_1)+G(\rho_2)+D(\rho_1+\rho_2)-\mu_1\rho_1-\mu_2\rho_2\right] \tag{1.3}$$

where β is the inverse temperature, μ_1 and μ_2 are chemical potentials, G is a convex function, and D takes account of short range interactions between the particles which we have taken for simplicity to be species independent. Examples to keep in mind are:

$$G(t) = t\log(t), \tag{1.4}$$

$$D(t) = \begin{cases} -t\ \log(1-bt), & \text{if } x < b^{-1}, \\ +\infty \text{ otherwise} \end{cases} \tag{1.5}$$

for some $b > 0$, where b^{-1} can be thought of as the close packing density. In this case D is defined on a bounded interval, and the domain of F is the set $0 < \rho_1, \rho_2 < b^{-1}$. In particular the case $b = 1$ corresponds to the lattice gas with exclusion rule.

As for the long range interaction terms, we assume that $V(x)$ and $U(x)$ are non-negative radial decreasing functions of $x \in I\!R^d$ with compact support and

$$\int_{I\!R^d} V(x)\mathrm{d}x = \alpha, \qquad \int_{I\!R^d} U(x)\mathrm{d}x = \sigma \tag{1.6}$$

We further define

$$\ell = \int_{\mathbb{R}} |x|[\frac{1}{\alpha}V(x) + \frac{1}{\sigma}U(x)]\,\mathrm{d}x \ , \tag{1.7}$$

which characterizes the length scale of the interactions. We remark that it is not essential, but convenient, to have the same attractive interaction U for the two species.

Notice that F is not required to be jointly convex or affine in ρ_1 and ρ_2, although this is expected on physical grounds, and, in any case, in general $\mathcal{F}$ would not be because of the interaction terms. Convexity of the interaction terms would require positive definiteness of the 2×2 matrix valued function

$$\begin{pmatrix} -\widehat{U} & \widehat{V} \\ \widehat{V} & -\widehat{U} \end{pmatrix},$$

where the hat denotes the Fourier transform.. We do not make such an assumption in this paper. Instead we make use in our analysis of the fact, proven later, that the free energy functional that we consider satisfies certain rearrangement inequalities. These give a certain monotonicity to the spatial density profile of the two species. We study the consequences of these for the phase diagram which originates from the competition between the repulsive interaction terms, which would prefer to have the two species segregated as completely as possible, and the entropy term, which would prefer to have the densities of the two species be uniform over Λ.

Our aim here is to study the minimizers of this functional either under constraints on the integrals of ρ_1 and ρ_2, or without such constraints. Specifically, let n_1 and n_2 be two given positive numbers. We define the sets $\mathcal{D}(n_1, n_2)$ and $\mathcal{D}$ of constrained and unconstrained pairs of densities: $\mathcal{D}(n_1, n_2)$ and $\mathcal{D}$ by

$$\mathcal{D}(n_1,n_2) = \left\{ (\rho_1,\rho_2) \in I\!R_+ \times I\!R_+ \ : \ \frac{1}{|\Lambda|}\int_\Lambda \rho_1 \mathrm{d}x = n_1 \qquad \text{and} \qquad \frac{1}{|\Lambda|}\int_\Lambda \rho_2 \mathrm{d}x = n_2 \ \right\} \ , \tag{1.8}$$

$$\mathcal{D} = \bigcup_{n_1,n_2 \geq 0} \mathcal{D}(n_1,n_2). \tag{1.9}$$

The canonical (Helmholtz) equilibrium free energy per unit volume or the grand canonical (Gibbs) equilibrium free energy per unit volume will be obtained by minimizing $\mathcal{F}(\rho_1, \rho_2)/|\Lambda|$ under the constraint (1.8), or the unconstrained as in (1.9). In the former case the μ_i are irrelevant and we shall set them equal to zero.

The two cases will give different equilibrium density profiles $\bar{\rho}_i(x)$, defined as the minimizer of $\mathcal{F}$ in (1.1), when there is a coexistence of phases, i.e. when the minimizer for specified chemical potentials is not unique. This may happen when $\mathcal{F}(\rho_1, \rho_2)$ is not strictly convex and the system undergoes a phase transition.

In the case where the diameter of the torus is large compared to ℓ, the interactions become approximately local. In this case, for densities ρ_1 and ρ_2 that are effectively constant on the length scale ℓ over most of Λ, $\mathcal{F}(\rho_1, \rho_2)$ is well approximated by $\mathcal{F}_0(\rho_1, \rho_2)$ the MFT free energy functional where

$$\mathcal{F}_0(\rho_1,\rho_2) = \int_\Lambda F(\rho_1(x),\rho_2(x))\mathrm{d}x + \alpha\int_\Lambda \rho_1(x)\rho_2(x)\mathrm{d}x - \frac{\sigma}{2}\int_\Lambda [\rho_1^2(x)+\rho_2^2(x)]\mathrm{d}x \ . \tag{1.10}$$

When ρ_1, ρ_2 are restricted to be constants, n_1, n_2 respectively, then $\mathcal{F}_0(n_1, n_2)$ is essentially the function studied by van der Waals [21] and by Korteweg [10]. (Korteweg however considered the case $\alpha < 0$ so the rearrangement inequalities used here would not apply in his case.) There are interesting questions regarding the minimization of $\mathcal{F}_0$ in case ρ_1, ρ_2 are not restricted to be constant. For example, knowledge of the minimizing profiles enables one to compute the equilibrium internal (interaction) energy at given temperature under the constraints in (1.8). An application of this is given in Section 6.

We will show in section 2 that

$$\inf_{(\rho_1,\rho_2)\in\mathcal{D}} \mathcal{F}(\rho_1, \rho_2) \qquad \text{and} \qquad \inf_{(\rho_1,\rho_2)\in\mathcal{D}} \mathcal{F}_0(\rho_1, \rho_2) \tag{1.11}$$

are both attained, and the same statement holds for the constrained problem. Clearly

$$f_0 := \frac{1}{|\Lambda|} \inf_{(\rho_1,\rho_2)\in\mathcal{D}} \mathcal{F}_0(\rho_1, \rho_2) \tag{1.12}$$

is independent of Λ. We shall see in section 3 that, using of the rearrangement inequalities, we also have

$$\lim_{|\Lambda|\to\infty} \frac{1}{|\Lambda|} \inf_{(\rho_1,\rho_2)\in\mathcal{D}} \mathcal{F}(\rho_1, \rho_2) = f_0 \ , \tag{1.13}$$

and moreover there is a close relation between the minimizers in both infimums in (1.11).

The equality in (1.13) may look natural, but an example of Gates and Penrose [8] shows that the analogous statement need not hold for simple one component systems. Specifically, their one component system free energy functional is given by

$$\mathcal{G}(\rho) = \frac{1}{\beta} \int_\Lambda \rho(x) \log \rho(x) \mathrm{d}x + \int_\Lambda \int_\Lambda \rho(x) V(x-y) \rho(y) \mathrm{d}x \mathrm{d}y \tag{1.14}$$

and

$$\mathcal{G}_0(\rho) = \frac{1}{\beta} \int_\Lambda \rho(x) \log \rho(x) \mathrm{d}x + \alpha \int_\Lambda \int_\Lambda \rho(x)^2 \mathrm{d}x \tag{1.15}$$

where α and V are related as before. Since $\alpha > 0$, $\mathcal{G}_0$ is strictly convex, and hence is minimized at the constant density $\rho(x) = n$. However, if $\widehat{V}(k_0) < 0$ for some k_0, then for sufficiently small $\epsilon > 0$, there is a k close k_0 and a $\delta > 0$ so that with $\rho_\epsilon(x) = n + \epsilon \sin(kx)$,

$$\frac{1}{|\Lambda|} \mathcal{G}(\rho_\epsilon) \le \frac{1}{|\Lambda|} \mathcal{G}_0(n) - \delta \tag{1.16}$$

for all Λ sufficiently large. In our model, the rearrangement inequalities prevent oscillations from lowering the free energy.

In more detail, observe that

$$\mathcal{F}(\rho_1, \rho_2) = \mathcal{F}_0(\rho_1, \rho_2) - \frac{1}{2} \sum_{i,j} \int \int_\Lambda \xi_i(x, y) V_{ij}(x-y) \xi_j(x, y) \mathrm{d}x \mathrm{d}y \tag{1.17}$$

where

$$V_{11} = V_{22} = -U \qquad \text{and} \qquad V_{12} = V_{21} = V \ , \tag{1.18}$$

and

$$\xi_i(x, y) = \rho_i(x) - \rho_i(y) \ . \tag{1.19}$$

The rearrangement inequalities will ensure that final integral in the right of (1.17) is a surface term. This will be proved in Section 3 below.

The rearrangement inequalities have other interesting consequences for the phase diagram of our system. To be concrete, we will investigate these in the case of the van der Waals free energy functional given by (1.3) and (1.5). Because of (1.13), the rearrangement inequalities allow us to draw conclusions about the phase diagram of $\mathcal{F}_0$ even though rearrangement of ρ_1 and ρ_2 may not affect the value of $\mathcal{F}_0(\rho_1, \rho_2)$.

We will see from the structure of the minimizers that the system, for appropriate choices of F, V and U, exhibits two phase transitions: A *liquid-vapor* transition and a *segregation* transition. In the first of these, below a precisely determined critical temperature, the system separates into two regions, one a *liquid region* in which the total bulk density is uniformly very close to some number ρ_ℓ, and the other a *vapor region* in which the total density is very close to some number ρ_v. This is similar to a van der Waals type transition in a one component system. The other transition is a segregation transition. The liquid or vapor homogeneous phases can split into two sub regions in which the densities of the two species are very close to certain preferred concentrations ρ_ℓ^+ and ρ_ℓ^- for the liquid, and ρ_v^+ and ρ_v^- for the vapor. The values of these densities depend on the temperature and the parameters of the system, and a fairly detailed analysis is required to determine them in any given case. Nonetheless, there are general conclusions we can draw as a consequence of the rearrangement inequalities; for example, we will see in broad generality that for our system one always has

$$\rho_\ell^- \le \rho_v^- \le \rho_v^+ \le \rho_\ell^+ \tag{1.20}$$

and never

$$\rho_v^- \le \rho_v^+ \le \rho_\ell^- \le \rho_\ell^+ \ . \tag{1.21}$$

The paper is organized as follows: In section 2, we state precisely the rearrangement inequalities referred to above, and apply them to prove the existence of minimizers. We also prove that minimizers necessarily have certain regularity properties. In section 3 we prove a general theorem showing that as a consequence of the rearrangement inequalities, minimizing $\mathcal{F}$ and minimizing $\mathcal{F}_0$ are essential by the same problem for L much larger than ℓ. The general theorem requires certain assumptions on F. These assumptions are verified for the physical model that we study in the next two sections. Section 4 is devoted to the application of convex analysis methods to deduce general features of the minimizers for our system. We will see that there are generically one, two, three or four coexisting equilibrium states in this model. The general convex analysis does however permit more bizarre coexistence of phases. In section 5 we impose further restrictions (physically natural) and obtain a detailed phase portrait, ruling out these bizarre non–generic behaviors. Section 6 gives an application of the results obtained here to the study of the equilibrium states of an energy conserving kinetic model. In particular, the results obtained here allow us to determine the equilibrium partition of the total energy into kinetic and interaction terms. Thus, given the energy, we are able to determine the equilibrium temperature in such a system.

2. Existence and Regularity of Minimizers.

In this section we show that minimizing densities exist, and begin the determination of their nature. Since particles of the same species attract and of different species repel, one can expect that the minimizing densities should be "well organized" in some sense, and we use a rearrangement inequality to prove this.

Introduce coordinates x_i, $i = 1, \ldots, d$ with $-L \le x_i < L$ for each $i = 1, \ldots, d$ where $L > 0$, and we make the usual identifications to parametrize our torus Λ.

We say that a function g on Λ is *symmetric monotone decreasing* about the origin $0, \ldots, 0$ if it is a symmetric decreasing function of each coordinate x_i for $-L \le x_i < L$. That is, for each i

$$g(x_1, x_2, \ldots, x_i, \ldots x_d) = g(x_1, x_2, \ldots, -x_i, \ldots x_d) \tag{2.1}$$

and whenever $L \geq y_i > x_i \geq 0$, then

$$g(x_1, x_2, \ldots, y_i, \ldots x_d) \leq g(x_1, x_2, \ldots, x_i, \ldots x_d) \ . \tag{2.2}$$

We define *symmetric monotone increasing* in the analogous way. Finally we say that a function is symmetric monotone if it is either symmetric monotone decreasing or increasing.

Given a non–negative integrable function g on Λ, we say that a non–negative integrable function $g^\star$ is a symmetric monotone decreasing rearrangement of g if it is a symmetric monotone decreasing function such that for all $t > 0$ $\{x|g(x) > t\}$ has the same measure as $\{x|g^\star(x) > t\}$. Analogously we define a symmetric monotone increasing rearrangement $h_\star$ of a non–negative integrable function h.

We will see below that for an appropriate choice of $\rho_1^\star$ and $\rho_{2\star}$, $\mathcal{F}(\rho_1^\star, \rho_{2\star}) \leq \mathcal{F}(\rho_1, \rho_2)$, with equality only in case ρ_1 and ρ_2 are already symmetric monotone up to a common translation. The relevance of these definitions lies in the following lemma, which says that the "total variation", $\|\nabla g\|_1$, of a symmetric monotone function g is a "surface term". (Here and in what follows, $\|\cdot\|_p$ denotes the $L^p(\Lambda)$ norm.)

Lemma 2.1 *Suppose that g is a bounded and non-negative integrable function on Λ that is symmetric monotone. Then g has an integrable distributional gradient, and the total variation of g, that is $\|\nabla g\|_1$, satisfies*

$$\int_\Lambda |\nabla g(x)| \, \mathrm{d}x \leq |\Lambda|^{(d-1)/d} 2d \|g\|_\infty \ . \tag{2.3}$$

Note that the right hand side is proportional to $|\Lambda|^{(d-1)/d}$, which is what makes it a surface term.

Proof: We suppose first that g is smooth and symmetric monotone decreasing, and derive the bound (2.3) under this hypothesis. Any such function g has the property that as one goes around any of the circles where only one coordinate varies, say x_i, one proceeds monotonically from the minimum of g on this circle, to the maximum, and then back down to the minimum. Since $g \geq 0$, the difference between the maximum and the minimum is no more than $\|g\|_\infty$. Hence, since g is smooth,

$$\int_\Lambda \left| \frac{\partial g}{\partial x_i} \right| \mathrm{d}x \leq \frac{|\Lambda|}{L} \left(2\|g\|_\infty \right) \ . \tag{2.4}$$

Now integrating over the remaining coordinates one obtains (2.3).

For the general case, one may approximate g by applying the heat kernel on the torus to it. This approximation preserves both the positivity and the property of being symmetric monotone. Also, the heat evolution is continuous in both the total variation norm and the L^∞ norm, so (2.3) is preserved as the approximation is relaxed. ■

This lemma will play a crucial role in determining the structure of our minimizers, since, as we will see below, our minimizers are symmetric monotone, up to translation on the torus, and thus, they satisfy the bound (2.3) as well as *a–priori* L^∞ bounds.

Lemma 2.2 *Let F be a function on $\mathbb{R}_+ \times \mathbb{R}_+$ that satisfies (1.2) everywhere on its domain. Let U be a non-negative strictly monotone decreasing radial function on $\mathbb{R}^d$. Suppose also that the diameter of its support is less than the smallest period of Λ. Then for all non–negative functions g and h on Λ, there is a symmetric monotone decreasing function $g^\star$ and a symmetric monotone increasing $h_\star$ so that*

$$\int\int_\Lambda g(x) U(x-y) h(y) \mathrm{d}x\mathrm{d}y \geq \int\int_\Lambda g^\star(x) U(x-y) h_\star(y) \mathrm{d}x\mathrm{d}y \tag{2.5}$$

and

$$\int_\Lambda F(g(x),h(x))\mathrm{d}x \geq \int_\Lambda F(g^\star(x),h_\star(x))\mathrm{d}x \ . \tag{2.6}$$

Moreover, there is equality in (2.5) only in case $g = g^\star$ and $h = h_\star$ up to a common translation.

These estimates (2.5) could be deduced from related rearrangement inequalities of Baernstein and Taylor [3], and Luttinger [14]. However, a direct proof is provided in Appendix A. The second inequality is reminiscent of an inequality of Almgren and Lieb [2] with the opposite sense for symmetric decreasing rearrangement on $I\!R^n$ in which both functions are "piled up" around the *same* point. The proofs though are different.

We now turn to the existence of minimizers for the variational problem of determining

$$\inf \{\mathcal{F}(\rho_1,\rho_2) \ : \ (\rho_1,\rho_2) \in \mathcal{D}(n_1,n_2) \ \} \tag{2.7}$$

where $\mathcal{D}(n_1,n_2)$ is given by (1.8).

The lemmas collected above enable this to be proved in a wide variety of circumstances. Theorem 2.3 below applies to one of these that is of particular physical interest. Mathematically, it is a very direct consequence of the lemmas since when D is given by (1.5), there is a trivial *a-priori* bound on $\|\rho_1\|_\infty$ and $\|\rho_2\|_\infty$ – namely b^{-1}.

Without such a term to enforce *a–priori* L^∞ bounds, and without an entropy that prevents a vacuum, the proof is more involved, but still works. Mathematically, this context is much more interesting, if less physical. To keep the focus on the main physical case, we prove here the theorem only in the simple case required to treat the van der Waals gas and put in Appendix B the statement and the proof for the other case.

The role of the rearrangement inequalities is this: Since the interaction is not positive definite, lowering the interaction energy may well favor oscillations in the densities ρ_1 and ρ_2, and there is no mechanism to prevent a minimizing sequence from oscillating more and more rapidly so that some part of the mass vanishes in a weak limit, with the consequence that the limit no longer belongs to $\mathcal{D}(n_1,n_2)$. This cannot happen here because of Lemma 2.2.

Theorem 2.3 *Let F be given by (1.3), where in particular D has the property that $D(t) = \infty$ for $t > b^{-1}$ for some $b > 0$. Then, for all n_1 and n_2 and all β,*

$$\mathcal{F}(n_1,n_2,\beta) = \inf_{(\rho_1,\rho_2)\in\mathcal{D}(n_1,n_2)} \mathcal{F}(\rho_1,\rho_2) \tag{2.8}$$

is achieved at least at one minimizing pair (ρ_1,ρ_2). Any such pair satisfies the Euler–Lagrange equations

$$G'(\rho_1)+D'(\rho_1+\rho_2)+\beta V*\rho_2-\beta U*\rho_1 = C_1 \quad \text{and} \quad G'(\rho_2)+D'(\rho_1+\rho_2)+\beta V*\rho_1-\beta U*\rho_2 = C_1 \ . \tag{2.9}$$

Moreover,

$$\|\nabla\rho_1\|_1 \ , \|\nabla\rho_2\|_1 \ \leq |\Lambda|^{(d-1)/d} 2db^{-1} \ , \tag{2.10}$$

and if (ρ_1,ρ_2) is any minimizing pair, then $\rho_1 = \rho_1^\star$ and $\rho_2 = \rho_{2\star}$ up to a common translation.

Likewise, for any μ_1, μ_2, let

$$\mathcal{F}(\mu_1,\mu_2,\beta) = \inf_{(\rho_1,\rho_2)\in\mathcal{D}} \left\{ \mathcal{F}(\rho_1,\rho_2) - \mu_1\int_\Lambda \rho_1(x)\mathrm{d}x - \mu_2\int_\Lambda \rho_2(x)\mathrm{d}x \right\} \ . \tag{2.11}$$

Then the minimizers do exist and satisfy the above conditions with C_1 and C_2 in (2.9) replaced by μ_1, μ_2.

Proof: We first observe that we may assume $\rho_1, \rho_2 \leq b^{-1}$. These *a–priori* bound makes this case easier.

The functional

$$\rho_1, \rho_2 \mapsto \mathcal{F}(\rho_1, \rho_2) \tag{2.12}$$

is jointly continuous in the L^1 topology by the dominated convergence theorem.

Now let $(\rho_1^{(k)}, \rho_2^{k)})$ be a minimizing sequence satisfying the constraint $(1.8)_1$, and such that $(\rho_1^{(k)}, \rho_2^{k)})$ is a symmetric monotone pair. Such a sequence exists by Lemma 2.2. By the Helly selection principle and the *a–priori* pointwise bound, this sequence is strongly compact in $L^1 \times L^1$. Passing to a convergent subsequence, we have our minimizing pair.

The Euler–Lagrange equations now easily follow, and then (2.10) follows from (2.3) and the *a–priori* sup norm bound provided by D. ■

3. Large Volume.

In this section we focus on the case in which L, the period of the torus Λ, is large compared to ℓ, the length scale of the interaction defined in (1.7).

Lemma 3.1 *Let (ρ_1, ρ_2) be any minimizer for the free energy functional. Then, using $*$ for convolution,*

$$\sum_{j=1}^{2} \int_\Lambda |V * \rho_j - \alpha\rho_j| \,\mathrm{d}x + \sum_{j=1}^{2} \int_\Lambda |U * \rho_j - \sigma\rho_j| \,\mathrm{d}x \leq C|\Lambda|^{(d-1)/d} \tag{3.1}$$

for some constant C depending only on β and n_1, n_2. As a consequence,

$$\sum_{j=1}^{2} \left| \int_\Lambda \rho_i V * \rho_j \mathrm{d}x - \alpha \int_\Lambda \rho_i\rho_j \mathrm{d}x \right| + \sum_{j=1}^{2} \left| \int_\Lambda \rho_i U * \rho_j \mathrm{d}x - \sigma \int_\Lambda \rho_i\rho_j \mathrm{d}x \right| \leq C|\Lambda|^{(d-1)/d} \tag{3.2}$$

where again, C depends only on β and n_1, n_2.

Proof:

$$\rho_j * V(x) - \alpha\rho_j(x) = \int_\Lambda V(y)\left[\rho_j(x-y) - \rho_j(x)\right] \mathrm{d}y = -\int_0^1 \int_\Lambda V(y) \nabla\rho_j(x - ty) \cdot y \mathrm{d}y\mathrm{d}t \ . \tag{3.3}$$

Hence

$$\int_\Lambda |\rho_j * V(x) - \alpha\rho_j(x)| \,\mathrm{d}x \leq \left(\int_\Lambda V(y)|y|\mathrm{d}y \right) \|\nabla\rho_1\|_1 \ , \tag{3.4}$$

and from Lemma 2.1,

$$\int_\Lambda |\rho_j * V(x) - \alpha\rho_j(x)| \,\mathrm{d}x \leq C \left(\int_\Lambda V(y)|y|\mathrm{d}y \right) |\Lambda|^{(d-1)/d} \ . \tag{3.5}$$

The same argument applies to the terms involving U. ■

Now fix any $\epsilon > 0$. A simple Chebyshev argument based on the integral bounds of Lemma 3.1 shows that, by choosing Λ sufficiently large, off of a set of "surface term size", $V * \rho_i = \alpha\rho_i \pm \epsilon$ and $U * \rho_i = \sigma\rho_i \pm \epsilon$. More precisely, with $|A|$ denoting the Lebesgue measure of a set A,

$$|\{x \mid |V * \rho_i(x) - \alpha\rho_i(x)| \geq \epsilon \ \}| \leq C\epsilon^{-1}\ell|\Lambda|^{(d-1)/d}, \quad i = 1, 2 \ , \tag{3.6}$$

and likewise for U.

Let G_ϵ be the "good" set

$$\left(\bigcap_{j=1}^{2}\{x \mid |V*\rho_j(x)-\alpha\rho_j(x)|<\epsilon\ \}\right)\bigcap\left(\bigcap_{j=1}^{2}\{x \mid |U*\rho_j(x)-\sigma\rho_j(x)|<\epsilon\ \}\right)\ . \tag{3.7}$$

Any minimizing pair of densities (ρ_1,ρ_2) for $\mathcal{F}$ with n_1 and n_2 fixed must satisfy the Euler Lagrange equations

$$\begin{aligned}\frac{\partial F}{\partial \rho_1}(\rho_1,\rho_2)+V*\rho_2-U*\rho_1 &= C_1\\ \frac{\partial F}{\partial \rho_2}(\rho_1,\rho_2)+V*\rho_1-U*\rho_2 &= C_2\end{aligned} \tag{3.8}$$

for some C_1 and C_2.

Likewise, any minimizing pair of densities $(\widetilde{\rho}_1,\widetilde{\rho}_2)$ for $\mathcal{F}_0$ with n_1 and n_2 fixed must satisfy the Euler Lagrange equations

$$\begin{aligned}\widetilde{C}_1 &= \frac{\partial F}{\partial \widetilde{\rho}_1}(\widetilde{\rho}_1,\widetilde{\rho}_2)+\alpha\widetilde{\rho}_2-\sigma\widetilde{\rho}_1,\\ \widetilde{C}_2 &= \frac{\partial F}{\partial \widetilde{\rho}_2}(\widetilde{\rho}_1,\widetilde{\rho}_2)+\alpha\widetilde{\rho}_1-\sigma\widetilde{\rho}_2\ .\end{aligned} \tag{3.9}$$

Given a minimizing pair (ρ_1,ρ_2) for $\mathcal{F}$, define functions $C_1(x)$ and $C_2(x)$ through

$$\begin{aligned}C_1(x) &:= \frac{\partial F}{\partial \rho_1}(\rho_1,\rho_2)+\alpha\rho_2-\sigma\rho_1,\\ C_2(x) &:= \frac{\partial F}{\partial \rho_2}(\rho_1,\rho_2)+\alpha\rho_1-\sigma\rho_2\ .\end{aligned} \tag{3.10}$$

It follows from the definitions that on G_ϵ

$$|C_1(x)-C_1|\le\epsilon \qquad \text{and} \qquad |C_2(x)-C_2|\le\epsilon\ . \tag{3.11}$$

Therefore, on the set G_ϵ, $(\rho_1(x),\rho_2(x))$ is a solution of (3.10) with $C_1(x)$ and $C_2(x)$ very close to C_1 and C_2. We wish to conclude that on G_ϵ, the values of $(\rho_1(x),\rho_2(x))$ are essentially those of a minimizer of F_0.

Toward this end, we make a "stability" assumption on the Euler Lagrange equations describing minimizers of $\mathcal{F}$ under the constraint for fixed n_1 and n_2. This condition is easy to verify for many particular choices of F, as we shall see.

Definition: (Amenable Free Energy Function) We say that the free energy function $F(\rho_1,\rho_2)+\alpha\rho_1\rho_2-(\rho_1^2+\rho_2^2)$ is *amenable* when for any given constants C_1 and C_2, there are at most a finite number N of pairs of numbers

$$(\rho_1^{(i)},\rho_2^{(i)}) \qquad i=1,\ldots,N \tag{3.12}$$

so that for any given $\epsilon>0$, there is a $\delta>0$ depending only on ϵ, C_1 and C_2 such that the following is true:

Whenever any pair of numbers $(\widetilde{\rho}_1,\widetilde{\rho}_2)$ satisfies

$$\frac{\partial F}{\partial \rho_1}(\widetilde{\rho}_1,\widetilde{\rho}_2)+\alpha\widetilde{\rho}_2-\sigma\widetilde{\rho}_1=\widetilde{C}_1 \qquad \text{and} \qquad \frac{\partial F}{\partial \rho_2}(\widetilde{\rho}_1,\widetilde{\rho}_2)+\alpha\widetilde{\rho}_1-\sigma\widetilde{\rho}_2=\widetilde{C}_2\ . \tag{3.13}$$

for some $\widetilde{C}_1$ and $\widetilde{C}_2$ with $|\widetilde{C}_1 - C_1| + |\widetilde{C}_2 - C_2| < \delta$, it follows that

$$|\widetilde{\rho}_1 - \rho_1^{(i)}| + |\widetilde{\rho}_2 - \rho_2^{(i)}| < \epsilon, \quad \text{for some} \quad 1 \le i \le N \; . \tag{3.14}$$

Checking this in practice amounts to checking that there is continuous dependence of solutions to (3.9) from (C_1, C_2). Just to concrete, consider the simple case in which $F(\rho_1, \rho_2) = (\rho_1 \log \rho_1 + \rho_2 \log \rho_2)/\beta$. Then (3.9) are

$$\log \rho_1 + \beta\alpha\rho_2 = C_1 \qquad \text{and} \qquad \log \rho_2 + \beta\alpha\rho_1 = C_2 \tag{3.15}$$

Then with

$$h_{C_1,C_2}(\rho) = e^{C_1} \exp(-\beta\alpha e^{C_2}(\exp(-\beta\alpha\rho))) \; ,$$

ρ_1 and ρ_2 satisfy the fixed point equations

$$\rho = h_{C_1,C_2}(\rho) \qquad \text{and} \qquad \rho = h_{C_2,C_1}(\rho)$$

respectively. For each C_1 and C_2 there is a number R depending only on C_1 and C_2 so that $h_{C_1,C_2}(\rho)$ is convex for $\rho < R$, and concave for $\rho > R$. One sees that there are always one, two or three solutions to (3.15) in this case. As C_1 and C_2 vary in small intervals, there are at most three small pairs intervals required to hold all of the solutions. This is the situation described in general by the definition.

We will give further physical examples in the next section, and here we focus on the general consequences of amenability.

The main consequence is that on the "good set" G_ϵ, any minimizers ρ_1 and ρ_2 of $\mathcal{F}$ are essentially discrete, taking their values in the union of a finite number of short intervals. We now identify these intervals, and relate the minimization of $\mathcal{F}$ and $\mathcal{F}_0$.

Theorem 3.2 *Suppose that the free energy function is amenable. For any fixed (n_1, n_2), let (ρ_1, ρ_2) be a minimizer for $\mathcal{F}$ in $\mathcal{D}(n_1, n_2)$. Then there is a finite set of pairs of numbers*

$$(\rho_1^{(i)}, \rho_2^{(i)}) \qquad i = 1, \ldots, N \tag{3.16}$$

such that for all $\epsilon > 0$, and all L sufficiently large, there is a set $G \subset \Lambda$ such that

$$\frac{|G|}{|\Lambda|} < C|\Lambda|^{-1/d}$$

such that for all x in G_ϵ,

$$|\rho_1(x) - \rho_1^{(i)}| + |\rho_2(x) - \rho_2^{(i)}| < \epsilon \tag{3.17}$$

for some $1 \le i \le N$. Moreover, there is a pair $(\widetilde{\rho}_1, \widetilde{\rho}_2)$ satisfying $|\widetilde{\rho}_1 - \rho_1^{(i)}| + |\widetilde{\rho}_2 - \rho_2^{(i)}| < \epsilon$ that satisfies the Euler–Lagrange equation (3.9) for a minimizer of $\mathcal{F}_0$ in $\mathcal{D}(\widetilde{n}_1, \widetilde{n}_2)$ for some $\widetilde{n}_1$ and $\widetilde{n}_2$ with $|\widetilde{n}_1 - n_1| + |\widetilde{n}_2 - n_2| < \epsilon$.

Proof: For some κ to be determined later, let $G = G_\kappa$. Then, as a consequence of Lemma 3.1, $\rho_1(x)$ and $\rho_2(x)$ satisfy (3.10) for $C_1(x)$ and $C_2(x)$ nearly constant on G. Let C_1 and C_2 be the respective average values of $C_1(x)$ and $C_2(x)$ on Λ. It follows that on G, ρ_1 and ρ_2 have values lying finite number of pairs of intervals. As κ is decreased, so is the width of these intervals. Each pair of values in these intervals is a solution of the Euler–Lagrange equations (3.9) for some values of $\widetilde{C}_1$ and $\widetilde{C}_2$ close to C_1 and C_2 respectively.

By decreasing κ, we can ensure that $|\widetilde{C}_1 - C_1| + |\widetilde{C}_2 - C_2| < \delta$, where δ is related to ϵ as in the definition of amenability. ■

We remark that in specific cases, it is possible to carry the analysis further, and to show that the pairs of values $(\widetilde{\rho}_1, \widetilde{\rho}_2)$ correspond to values of minimizers for $\mathcal{F}_0$, and not simply solutions of the Euler–Lagrange equation. At this level of generality, that is not possible.

In this section we have only considered the constrained problem because in the unconstrained case the situation is much simpler. In the unconstrained case surface tension would discourage partitioning Λ among different minimizing phases.

4. Local Interactions.

In this section we consider the case in which $V(x) = \alpha\delta(x)$ and $U(x) = \sigma\delta(x)$. This will enable us to obtain a very complete picture of the minimizers when σ is sufficiently small and a rather clear understanding for arbitrary σ, as we show in the next section. Note that this corresponds exactly to replacing $\mathcal{F}$ by $\mathcal{F}_0$.

We begin our analysis by looking for the unconstrained spatially homogeneous minimizers of $\mathcal{F}_0$ with additional chemical potentials μ_1 and μ_2 (*grand-canonical ensemble*) which will play the role of Lagrange multipliers later on. Therefore, fixing μ_1 and μ_2, we look at the minimizers of grand canonical free energy density on $I\!R_+ \times I\!R_+$ given by

$$f_{\mu_1,\mu_2}(\rho_1, \rho_2) = F(\rho_1, \rho_2) + \alpha\rho_1\rho_2 - \frac{\sigma}{2}(\rho_1^2 + \rho_2^2) - \mu_1\rho_1 - \mu_2\rho_2 \tag{4.1}$$

Since this model exhibits both condensation-evaporation transition and segregation transition the natural variables are not ρ_1 and ρ_2 but ρ and ϕ where

$$\rho(x) = \rho_1(x) + \rho_2(x) \qquad \text{and} \qquad \phi(x) = \frac{\rho_1(x) - \rho_2(x)}{\rho(x)} \ . \tag{4.2}$$

Then the free energy $\mathcal{F}_0(\rho_1, \rho_2)$ can be written as

$$\mathcal{F}_0(\rho_1, \rho_2) = \int_\Lambda g_{\mu,h}(\rho(x), \phi(x))\mathrm{d}x \ , \tag{4.3}$$

where

$$g_{\mu,h}(\rho, \phi) = F\left(\frac{\rho}{2}(1+\phi), \frac{\rho}{2}(1-\phi)\right) + \frac{\alpha}{4}\rho^2(1-\phi^2) - \frac{\sigma}{4}\rho^2(1+\phi^2) - \mu\rho - h\rho\phi \tag{4.4}$$

where

$$\mu = \frac{\mu_1 + \mu_2}{2}, \quad h = \frac{\mu_1 - \mu_2}{2}. \tag{4.5}$$

It turns out that, under our assumptions, minimizers of this functional have a very special structure: Each of ρ_1 and ρ_2 can take on at most four values. The region Λ is decomposed into at most four subregions in which both ρ_1 and ρ_2 are constant. The four possible values of the densities result from a possible condensation-evaporation transition, together with a possible segregation transition.

Let $\widetilde{g}_{\mu,h}$ be the *convex minorant* of $g_{\mu,h}$. That is,

$$\widetilde{g}_{\mu,h}(\rho, \phi) = \sup_{\ell \in \mathcal{L}} \{\ell(\rho, \phi) \mid \ell(\rho, \phi) \le g_{\mu,h}(\rho, \phi)\} \tag{4.6}$$

where $\mathcal{L}$ is the set of all the linear functions $\ell(\rho, \phi) = a\rho + b\phi + c,\ a, b, c \in I\!R$. Let B be the set of ρ and ϕ for which

$$g_{\mu,h}(\rho, \phi) > \widetilde{g}_{\mu,h}(\rho, \phi) \ . \tag{4.7}$$

B is the set where there are "flat spots" in the graph of $\widetilde{g}_{\mu,h}$, and as is clear and well known, these are irrelevant to the minimization problem so that

$$\inf_{\rho_1,\rho_2} \frac{\mathcal{F}_0(\rho_1,\rho_2)}{|\Lambda|} = \inf_{\rho,\phi} \frac{1}{|\Lambda|}\int_\Lambda \widetilde{g}_{\mu,h}(\rho(x),\phi(x))\mathrm{d}x \ . \tag{4.8}$$

Next, define

$$\psi_{\mu,h}(\rho) = \inf_{\phi\in[-1,1]} g_{\mu,h}(\rho,\phi) \ , \tag{4.9}$$

and

$$\widetilde{\psi}_{\mu,h}(\rho) = \inf_{\phi\in[-1,1]} \widetilde{g}_{\mu,h}(\rho,\phi). \tag{4.10}$$

Then $\widetilde{\psi}_{\mu,h}$ is the convex minorant of $\psi_{\mu,h}$. Indeed, the epigraph of $\widetilde{g}_{\mu,h}$; i.e., the set

$$\{(\rho,\phi,z) \ : \ z \geq \widetilde{g}_{\mu,h}(\rho,\phi) \ \} \tag{4.11}$$

is convex, and the epigraph of $\widetilde{\psi}_{\mu,h}$ is simply the projection of the epigraph of $\widetilde{g}_{\mu,h}$ onto the ρ, z plane.

It is clear that

$$\frac{1}{|\Lambda|}\int_\Lambda \widetilde{g}_{\mu,h}(\rho(x),\phi(x))\mathrm{d}x \geq \frac{1}{|\Lambda|}\int_\Lambda \widetilde{\psi}_{\mu,h}(\rho(x))\mathrm{d}x \tag{4.12}$$

with equality if and only if for almost every x, $\phi(x)$ minimizes $\widetilde{g}_{\mu,h}(\rho(x),\phi)$ considered as a function of ϕ.

Now let $\rho_{\rm ave}$ be given by

$$\rho_{\rm ave} = \frac{1}{|\Lambda|}\int_\Lambda \rho(x)\mathrm{d}x \ . \tag{4.13}$$

Then by Jensen's inequality,

$$\frac{1}{|\Lambda|}\int_\Lambda \widetilde{\psi}_{\mu,h}(\rho(x))\mathrm{d}x \geq \widetilde{\psi}_{\mu,h}(\rho_{\rm ave}) \ . \tag{4.14}$$

It is well known that if $\widetilde{\psi}_{\beta,\mu,h}$ is strictly convex, then there is equality if and only if $\rho(x) = \rho_{\rm ave}$ almost everywhere. However, even if $\widetilde{\psi}_{\mu,h}$ is not strictly convex, the proof of Jensen's inequality has important consequences for our problem.

To explain these, we first recall that if ψ is any convex function on the positive axis, then an affine function $\ell(\rho) = a\rho + b$ is a *supporting line* for ψ in case $\psi(\rho) \geq \ell(\rho)$ for all ρ, and if $\psi(\rho_0) = \ell(\rho_0)$ for some ρ_0. If ℓ is any supporting line for ψ, then the set

$$\{\rho \ : \ \ell(\rho) = \psi(\rho) \ \} \tag{4.15}$$

is a closed interval. Such an interval is called a *support interval* of ψ. The following is simply Jensen's inequality, with the only novel feature being that the statement about the cases of equality that applies outside the strictly convex case.

Lemma 4.1 *Let $(\Omega,\mathcal{S},\nu)$ be a probability measure space, ψ a convex function on the positive axis, and ρ a non–negative measurable function. Then*

$$\int \psi(\rho)\mathrm{d}\nu \geq \psi\left(\int \rho \mathrm{d}\nu\right) \ , \tag{4.16}$$

and there is equality if and only if, up to a set of measure zero, ρ takes it values in a single support interval of ψ.

Proof: This follows from a close examination of the standard proof of Jensen's inequality, which turns on the fact that if f and g are two measurable functions,

$$\int \max\{f,g\}\mathrm{d}\nu \geq \max\left\{\int f\mathrm{d}\nu\ ,\ \int g\mathrm{d}\nu\right\}\ , \tag{4.17}$$

with equality if and only if either $f = \max\{f,g\}$ or $g = \max\{f,g\}$ almost everywhere. This applied to

$$\psi(\rho) = \sup_{\ell} \ell(\rho) \tag{4.18}$$

where the supremum is taken over all supporting lines of ψ yields the inequality. By the above, there is equality if and only if for any two supporting lines ℓ_1 and ℓ_2, either $\ell_1(\rho(x)) = \max\{\ell_1(\rho(x)), \ell_2(\rho(x))\}$ or $\ell_2(\rho(x)) = \max\{\ell_1(\rho(x)), \ell_2(\rho(x))\}$ for almost every x. This can only happen if, almost everywhere, ρ takes on all of its values in a single supporting interval. ■

This has the immediate consequence that for any minimizer of the local interaction problem, the values of $\rho(x)$ all lie in a single support interval of $\widetilde{\psi}_{\mu,h}$. Now in most cases that we will consider,

$$\psi_{\mu,h}(\rho) > \widetilde{\psi}_{\mu,h}(\rho) \tag{4.19}$$

for all ρ in any support interval, except at the endpoints. In particular, this is the case if $\psi_{\mu,h}$ is almost everywhere either strictly convex or strictly concave, and there are no lines that are tangent at three or more points. Then a minimizer for our problem cannot have $\rho(x)$ in the interior of the support interval because of (4.19). In exceptional cases, there may be physical values in the interior of a support interval. This occurs when there is a point of triple tangency, or higher.

Putting aside this exceptional case for the moment, there are only two possibilities for any minimizer in our problem: Either

- *The support interval of $\widetilde{\psi}_{\mu,h}$ that contains ρ_{ave} consists of ρ_{ave} alone. In this case, $\rho(x) = \rho_{\mathrm{ave}}$ almost everywhere.*

- *The support interval of $\widetilde{\psi}_{\mu,h}$ that contains ρ_{ave} consists of a closed interval $[\rho_v, \rho_\ell]$ with $\rho_v < \rho_\ell$. In this case, $\Lambda = \Lambda_v \cup \Lambda_\ell$ with $\rho(x) = \rho_v$ almost everywhere in Λ_v and $\rho(x) = \rho_\ell$ almost everywhere in Λ_ℓ.*

We say that there is a *condensation-evaporation* transition in the second case; the region Λ_v contains the vapor state and Λ_ℓ contains the liquid state. The volumes of these two regions are given by

$$|\Lambda_\ell|\rho_\ell + |\Lambda_v|\rho_v = |\Lambda|\rho_{\mathrm{ave}} = n_1 + n_2\ . \tag{4.20}$$

Note that the volume fractions of the liquid and vapor states are determined by this relation alone, before we begin considering any possible segregation in either the vapor or the liquid.

We note that there is a condensation-evaporation transition exactly where $\psi_{\mu,h}$ has an interval of concavity. Any such interval is contained in an interval $[\rho_v, \rho_\ell]$ with

$$\psi_{\mu,h}(\rho_v) = \widetilde{\psi}_{\mu,h}(\rho_v) \qquad \text{and} \qquad \psi_{\mu,h}(\rho_\ell) = \widetilde{\psi}_{\mu,h}(\rho_\ell) \tag{4.21}$$

and

$$\psi'_{\mu,h}(\rho_v) = \psi'_{\mu,h}(\rho_\ell)\ .$$

This means that

$$\int_{\rho_v}^{\rho_\ell} \psi''_{\mu,h}\mathrm{d}\rho = 0 \tag{4.22}$$

which is Maxwell's equal area rule for determining the values of ρ_v and ρ_ℓ.

Suppose for all $t \in (0,1)$

$$\psi_{\mu,h}((1-t)\rho_v + t\rho_\ell) > (1-t)\psi_{\mu,h}(\rho_v) + t\psi_{\mu,h}(\rho_\ell). \tag{4.23}$$

Suppose also that the infimum in (4.9) is attained exactly at one or two values of ϕ when $\rho = \rho_v$ and $\rho = \rho_\ell$. Then, depending whether ρ_v equals or not ρ_ℓ and whether there are one or two minimizers for ϕ in (4.9) there will be one, two, three or four states. To determine if these hypotheses hold we must examine a particular free energy.

5. van der Waals Gas.

In this section, we focus on the van der Waals gas for which

$$f_{\beta,\mu_1,\mu_2}(\rho_1,\rho_2) = \frac{1}{\beta}\big[G(\rho_1) + G(\rho_2) + D(\rho)\big] + \alpha\rho_1\rho_2 - \frac{\sigma}{2}(\rho_1^2 + \rho_2^2) - \mu_1\rho_1 - \mu_2\rho_2 \tag{5.1}$$

and hence

$$g_{\beta,\mu,h}(\rho,\phi) = \frac{1}{\beta}\Big[G\left(\frac{\rho}{2}(1+\phi)\right) + G\left(\frac{\rho}{2}(1-\phi)\right) + D(\rho)\Big] + \frac{\alpha}{4}\rho^2(1-\phi^2) - \frac{\sigma}{4}\rho^2(1+\phi^2) - \mu\rho - h\rho\phi \tag{5.2}$$

The function G, defined in $I\!R_+$, is assumed to be smooth and moreover

(1) G and G'' strictly convex;

(2) $G'(x) \to -\infty, \quad G''(x) \to +\infty \quad$ for $x \to 0$.

The function D is defined in some interval $(0, b^{-1})$ contained in (and possibly coinciding with) $I\!R_+$, and is smooth and strictly convex. For example, with $G(t) = t \log\, t$ and D given by (1.5), these conditions are satisfied. This is the usual two components van der Waals gas.

The existence of minimizers follows from the considerations of Section 2, which apply also in the case of local interactions. Note however that we have no symmetric monotonicity of the minimizers and, in general, no reason to have regularity properties. Moreover, the same argument that we used in the proof of Theorem 2.3 shows that any minimizer satisfies the Euler–Lagrange equations

$$\begin{aligned} G'(\rho_1) + D'(\rho_1+\rho_2) + \beta\alpha\rho_2 - \beta\sigma\rho_1 &= C_1 \\ G'(\rho_2) + D'(\rho_1+\rho_2) + \beta\alpha\rho_1 - \beta\sigma\rho_2 &= C_2. \end{aligned} \tag{5.3}$$

We now turn to the study of the structure of these minimizers. The following theorem gives a criterion for segregation:

Theorem 5.1. *With g given by (5.2), the infimum in (4.9) is attained in exactly one point $\widehat{\phi}(\rho;\beta,\mu,h)$ if $h \neq 0$, independently of ρ, or if $h = 0$ and $\beta(\sigma+\alpha) \le G''(\rho/2)$. If $h = 0$ and $\beta(\sigma+\alpha) > G''(\rho/2)$, then the infimum is attained in two points $\phi_\pm = \pm\widehat{\phi}(\rho;\beta,\mu,0)$*

Proof: When there is no ambiguity, we omit subscript β,μ,h from $g_{\beta,\mu,h}$. For fixed values of β, μ and h we look for the minimizers of g. To do this, we first fix ρ and look at the solutions of the equation

$$\frac{\partial g}{\partial \phi}(\rho,\phi) = 0 \tag{5.4}$$

for fixed ρ. Let $\widehat{\phi} = \widehat{\phi}(\rho; \beta, \mu, h)$ be one of the solutions to (5.4). Explicitly, for fixed $\rho > 0$, $\widehat{\phi}$ solves the equation

$$H_\rho(\widehat{\phi}) = \beta(\alpha + \sigma)\rho\widehat{\phi} + 2h, \tag{5.5}$$

with

$$H_\rho(\phi) := G'(\frac{\rho}{2}(1+\phi)) - G'(\frac{\rho}{2}(1-\phi)) \tag{5.6}$$

The following properties of the function H_ρ will be relevant below. Assume $\rho > 0$:

$$H_\rho(-\phi) = -H_\rho(\phi), \quad H_\rho(0) = 0; \tag{5.7}$$

$$H'_\rho(\phi) > 0, \forall \phi \in (-1, 1); \tag{5.8}$$

$$H_\rho(\phi) \to \pm\infty, \quad H'_\rho(\phi) \to +\infty \quad \text{for } \phi \to \pm 1; \tag{5.9}$$

$$\phi H''_\rho(\phi) > 0 \quad \text{for } \phi \neq 0, \quad H''_\rho(0) = 0; \tag{5.10}$$

(Notice that if $G(t) = t \log t$ then $H_\rho(\phi) = 2\tanh^{-1}(\phi)$). In particular, (5.10) implies that

$$\inf_{\phi\in[-1,1]} H'_\rho(\phi) = H'_\rho(0) = \rho G''(\frac{\rho}{2}) \quad \text{and } H'_\rho(\phi) > \rho G''(\frac{\rho}{2}) \text{ if } \phi \neq 0. \tag{5.11}$$

The above relations are immediate consequences of the assumptions on G: in fact (5.7) just follows from (5.6); (5.8) follows from

$$H'_\rho(\phi) = \frac{\rho}{2} G''(\frac{\rho}{2}(1+\phi)) + G''(\frac{\rho}{2}(1-\phi)) \tag{5.12}$$

and the fact that G is strictly convex. (5.9) follow from condition (2) on G and finally, (5.10) follows from

$$H''_\rho(\phi) = \frac{\rho^2}{4} G'''(\frac{\rho}{2}(1+\phi)) - G'''(\frac{\rho}{2}(1-\phi)) \tag{5.13}$$

and the strict monotonicity of G'''.

We have

$$\frac{\partial^2 g}{\partial \phi^2}(\rho, \phi) = \frac{\rho}{2}\left(H'_\rho(\phi) - (\alpha+\sigma)\beta\rho\right) \geq \frac{\rho^2}{2}\left(G''(\frac{\rho}{2}) - (\alpha+\sigma)\beta\right). \tag{5.14}$$

Therefore, if

$$\beta(\alpha + \sigma) < G''(\frac{\rho}{2}), \tag{5.15}$$

the r.h.s. of (5.14) is positive, the function $g(\rho, \phi)$, for ρ fixed is a strictly convex function of ϕ and hence it has a unique minimizer solving $\widehat{\phi}(\rho; \beta, \mu, h)$ solving (5.5), because of condition (2) on G permits to exclude that it is a corner solution. On the other hand, if

$$\beta(\alpha + \sigma) > G''(\frac{\rho}{2}), \tag{5.16}$$

by (5.10), there is an interval $(-\phi_s, \phi_s)$ where the function $g(\rho, \phi)$, for ρ fixed is concave while in the complement it is convex. The value ϕ_s is determined by the condition

$$H'_\rho(\phi_s) = (\alpha + \sigma)\beta\rho. \tag{5.17}$$

Therefore, it is possible to get more than one stationary point, satisfying (5.5).

We assume that (5.16) is verified and look at the stationary points. We distinguish two cases: $h = 0$ and $h \neq 0$.

We assume first $h = 0$. Then, clearly, $\phi = 0$ solves (5.5). The condition (5.16) ensures that $g(\rho, \phi)$ is concave in $\phi = 0$ and hence $\phi = 0$ is not a minimizer. Moreover, by the symmetry, if $\widehat{\phi}(\rho; \beta, \mu, 0)$ solves (5.5) the same is true for $-\widehat{\phi}(\rho; \beta, \mu, 0)$. (5.8) and (5.10) show that there is exactly one positive value $\widehat{\phi}(\rho; \beta, \mu, 0) \in (\phi_s, 1)$ solving (5.5). Consequently, we have the two stationary points

$$\phi_{\pm} = \pm\widehat{\phi}(\rho; \beta, \mu, 0). \tag{5.18}$$

Now we take $h \neq 0$. One immediately realizes that there is $\bar{h}(\rho)$ such that, if $h \notin [-\bar{h}(\rho), \bar{h}(\rho)]$ then there is only one solution to (5.5), for $h = \pm\bar{h}(\rho)$ there are two solutions, one of them in $(-\phi_s, \phi_s)$ and the other one is the minimizer. Finally, if $h \in (-\bar{h}(\rho), \bar{h}(\rho))$ there are three solutions, one in $(-\phi_s, \phi_s)$ and the others, in the complement, corresponding to local minimizers; moreover they have different signs. Let $\widehat{\phi}_1(\rho; \beta, \mu, h) > 0$ and $\widehat{\phi}_2(\rho; \beta, \mu, h) < 0$ denote the two local minimizers. We show that $g(\rho, \widehat{\phi}_1(\rho; \beta, \mu, h)) \neq g(\rho, \widehat{\phi}_2(\rho; \beta, \mu, h))$ and hence there is only one absolute minimizer. To show this, we compare with the case $h = 0$, where $\widehat{\phi}_2(\rho; \beta, \mu, 0)) = -\widehat{\phi}_1(\rho; \beta, \mu, 0))$ and the corresponding values of g are equal. We define

$$J_i(h) = g_{\beta,\mu,\eta}(\rho, \widehat{\phi}_i(\rho; \beta, \mu, h)), \quad i = 1, 2. \tag{5.19}$$

Since $\widehat{\phi}_i$ are stationary points for g, we have

$$\begin{aligned} \frac{d}{dh} J_i &= \frac{\partial g_{\beta,\mu,\eta}}{\partial \phi}(\rho, \widehat{\phi}_i(\rho; \beta, \mu, h)) \frac{\partial \widehat{\phi}_i}{\partial h} + \frac{\partial g_{\beta,\mu,\eta}}{\partial h}(\rho, \widehat{\phi}_i(\rho; \beta, \mu, h)) \\ &= \frac{\partial g_{\beta,\mu,\eta}}{\partial h}(\rho, \widehat{\phi}_i(\rho; \beta, \mu, h)) = -\rho\widehat{\phi}_i(\rho; \beta, \mu, h). \end{aligned} \tag{5.20}$$

Therefore, for $h > 0$

$$J_1(h) < J_1(0) = J_2(0) < J_2(h) \tag{5.21}$$

and *vice–versa* for $h < 0$. ■

Next we give a criterion for condensation-evaporation transition. Such a transition occurs exactly for those values of $\alpha, \sigma, \beta, \mu, h$ for which the function $\psi_{\beta,\mu,h}(\rho)$ fails to be strictly convex. Our first result pertaining to this is the following:

Lemma 5.2 *Let $\widehat{\phi}(\rho)$ be any minimizer of $g(\rho, \phi)$ in (5.2) with respect to ϕ, so that $\psi(\rho) = g(\rho, \phi(\rho))$. Then $\psi''(\rho)$ is strictly positive if the point $(\rho, \widehat{\phi}(\rho))$ is in the regions where the Hessian of g, $D^2 g$, is positive definite.*

Proof.

We have:

$$\psi' = \frac{\partial g}{\partial \rho} + \frac{\partial g}{\partial \phi}\widehat{\phi}' , \tag{5.22}$$

$$\begin{aligned} \psi'' &= \frac{\partial^2 g}{\partial \rho^2} + 2\frac{\partial^2 g}{\partial \phi \partial \rho}\widehat{\phi}' + \frac{\partial^2 g}{\partial \phi^2}(\widehat{\phi}')^2 + \frac{\partial g}{\partial \phi}\widehat{\phi}'' \\ &= \langle (1, \widehat{\phi}'), D^2 g(1, \widehat{\phi}') \rangle + \frac{\partial g}{\partial \phi}\widehat{\phi}'', \end{aligned} \tag{5.23}$$

$D^2 g$ being the Hessian matrix of g and $\langle\,\cdot\,,\,\cdot\,\rangle$ the usual scalar product in $I\!R^2$.

When $\widehat{\phi}(\rho)$ is a minimizer, above relation reduces to

$$\psi'' = \langle (1, \widehat{\phi}'), D^2 g (1, \widehat{\phi}') \rangle \tag{5.24}$$

Therefore, $\psi''(\rho)$ is strictly positive if the point $(\rho, \widehat{\phi}(\rho))$ is in the regions where $D^2 g$ is positive definite. ■

The above result can be restated in terms of the function f. Since $\widehat{\phi}(\rho)$ solves (5.5), it is plain that $\psi''(\rho) = \langle j^T(1, \widehat{\phi}'), D^2 f j^T(1, \widehat{\phi}') \rangle$ where

$$j = \begin{pmatrix} \dfrac{\partial \rho}{\partial \rho_1} & \dfrac{\partial \rho}{\partial \rho_2} \\ \dfrac{\partial \phi}{\partial \rho_1} & \dfrac{\partial \phi}{\partial \rho_2} \end{pmatrix} . \tag{5.25}$$

Therefore we study the positivity properties of $D^2 f$. Its expression is:

$$D^2 f = \begin{pmatrix} G''(\rho_1) - \beta\sigma + D''(\rho) & D''(\rho) + \beta\alpha \\ D''(\rho) + \beta\alpha & G''(\rho_2) - \beta\sigma + D''(\rho) \end{pmatrix} . \tag{5.26}$$

As a consequence of Lemma 5.1, when the Hessian of f is positive definite, there is no condensation-evaporation transition. However, if σ is sufficiently large, the diagonal terms may become negative, while for α sufficiently large the off diagonal terms may make $\det(D^2 f) < 0$.

We now consider explicitly some examples for the case $G(t) = t\ \log\ t$.

1) $\sigma = 0$ and $D = 0$ (ideal gas). Then $D^2 f$ is positive definite in the region $\mathcal{P}$ defined by

$$\mathcal{P} = \{(\rho_1, \rho_2)\,|\,\rho_1\rho_2 < \alpha^{-2}\beta^{-2}\}. \tag{5.27}$$

To check this, first suppose $\alpha\beta\rho > 2$. With our choice of G, (5.5) reduces to

$$\phi = \tanh\left(\frac{\alpha\beta\phi\rho}{2}\right) . \tag{5.28}$$

With $\xi = \alpha\beta\rho/2$ it becomes

$$\phi = \tanh(\xi\phi)\ . \tag{5.29}$$

For all strictly positive values of $\xi\phi$,

$$\sinh(\xi\phi) > \xi\phi\ . \tag{5.30}$$

By (5.29) we can divide the left hand side by $\tanh(\xi\phi)$ and the right hand side by ϕ without affecting the inequality. It follows that $\cosh(\xi\phi) > \xi$, and hence that $(1 - \tanh^2(\xi\phi))^{-1} > \xi^2$. By (5.29), this means that

$$(1 - \phi^2)^{-1} \geq \left(\frac{\alpha\beta\rho}{2}\right)^2 \tag{5.31}$$

whenever $\alpha\beta\rho > 2$. Of course if $\alpha\beta\rho \leq 2$, then, by Theorem 5.1 $\phi = 0$, and so (5.31) holds in all cases, and there is equality if and only if $\alpha\beta\rho = 2$. Since

$$\rho_1\rho_2 = \frac{\rho^2}{4}(1 - \phi^2) \leq \frac{1}{\alpha^2\beta^2}, \tag{5.32}$$

by (5.29), the condition in (5.27) is fulfilled for all the values of the parameters but for $\alpha\beta\rho = 2$. In this case however, $\phi = 0$ and by (5.23) $\psi'' = \frac{2}{\rho} > 0$. Hence $\psi(\rho)$ is strictly convex and there is no condensation-evaporation transition for any value of the parameters.

2) $D(\rho)$ is given by (1.5) and $\sigma = 0$. Then the strict convexity of G and D ensures that the diagonal terms are positive and we only need to check $\det(D^2 f) > 0$. Hence, in this case $D^2 f$ is positive definite in

$$\mathcal{Q} = \{(\rho_1, \rho_2) \mid \det(D^2 f) > 0\} \tag{5.33}$$

and its boundary,

$$\partial\mathcal{Q} = \{(\rho_1, \rho_2) \mid \det(D^2 f) = 0\}$$

separates it from the region where $D^2 f$ is not positive definite. Explicitly, the condition $\det(D^2 f) > 0$ can be written

$$\frac{1}{\rho_1\rho_2} - \alpha^2\beta^2 + D''(\rho)(\frac{1}{\rho_1} + \frac{1}{\rho_2} - 2\alpha\beta) > 0. \tag{5.34}$$

Note that the set $\mathcal{Q}$ contains the set

$$\mathcal{P} = \{(\rho_1, \rho_2) \mid \frac{1}{\rho_1\rho_2} - \alpha^2\beta^2 > 0\} \tag{5.35}$$

because the arithmetic mean of two positive numbers is not less than the geometric mean. Since the equation for ϕ is unaffected by the presence of D, $\psi(\rho)$ is strictly convex and we get the same conclusion as before.

3) If $\sigma > 0$, the set $\mathcal{P}$ is replaced by

$$\mathcal{P}_\sigma = \{(\rho_1, \rho_2) \mid (\frac{1}{\rho_1} - \beta\sigma)(\frac{1}{\rho_2} - \beta\sigma) - \alpha^2\beta^2 > 0\}. \tag{5.36}$$

Therefore, as σ increases the set $\mathcal{P}_\sigma$ shrinks thus encouraging the condensation-evaporation transition.

Korteweg [10] has also discussed a situation with segregation and four phases in equilibrium in a mixture of two van der Waals gases. Korteweg's paper concerns the $\alpha < 0$ case where there is attraction between different species too, and to which rearrangement arguments do not apply, though segregation is still possible because it is controlled by $\sigma + \alpha$.

Up to now we just gave general conditions for the absence of condensation-evaporation transition. We now derive a formula for the pressure which allows us to draw conclusions about the not strict convexity of the function ψ. Introduce the specific volume $v = \rho^{-1}$ and define the function

$$q(v) = v\psi(v^{-1}), \tag{5.37}$$

which has the same convexity properties of ψ in the corresponding points. A simple calculation shows that,

$$q(v) = -\log(v - b) - \frac{\beta\widetilde{\sigma}(\widehat{\phi}(v^{-1}))}{4v} + \gamma(\widehat{\phi}(v^{-1})) \tag{5.38}$$

with

$$\gamma(\widehat{\phi}) = \frac{1}{2}\log\left(\frac{1-\widehat{\phi}^2}{4}\right) + \frac{\widehat{\phi}}{2}\log\left(\frac{1+\widehat{\phi}}{1-\widehat{\phi}}\right) - h\widehat{\phi} - \mu \tag{5.39}$$

and

$$\widetilde{\sigma}(\widehat{\phi}) = \sigma - \alpha + \widehat{\phi}^2(\sigma + \alpha). \tag{5.40}$$

Now, by (5.5), we have

$$\frac{\beta\widetilde{\sigma}'}{4v} + \gamma' = 0. \tag{5.41}$$

Hence the pressure p as function of the specific volume is given by

$$\beta p(v) := -q'(v) = \frac{1}{v-b} - \frac{\beta\widetilde{\sigma}(\widehat{\phi}(v^{-1}))}{4v^2}. \tag{5.42}$$

This expression is similar to that for the one component van der Waals gas, but for the dependence of $\widetilde{\sigma}$ on v through $\widehat{\phi}$.

Several cases are possible, depending on α and σ. Consider for example the case $\alpha > \sigma$. Then, in the absence of segregation, the pressure is a strictly decreasing function of v. Since the pressure must be equal in the liquid and vapor phases, this precludes a condensation-evaporation transition in the absence of segregation. (This is quite different from the usual van der Waals case, or the case considered by Korteweg). The segregation permits a condensation-evaporation transition, and lowers the pressure in the liquid phase from what it would be without segregation. As the temperature is lowered further, the vapor phase may also undergo segregation. In this situation, the volume fractions are not determined. However, in the final section of the paper we shall see that because the local interaction model is the large volume limit of a model in which the minimizers must be symmetric decreasing, we can say more about this case. Thus when $\alpha > \sigma$, we have three critical inverse temperatures $\beta_1 < \beta_2 < \beta_3$. The first corresponds to a segregation transition. At the second there is a condensation-evaporation transition at which a segregated liquid phase is produced, and an unsegregated vapor phase. At the third, the vapor phase as well segregates. As a consequence of the above formula for the pressure, we prove the following:

Theorem 5.3. *Suppose that $\alpha \leq \sigma$. Then there is a critical inverse temperature β_c such that for $\beta > \beta_c$ a condensation-evaporation transition occurs, with or without segregation, depending on the values of β, h and mu, according to Theorem 5.1. On the other hand, if $\alpha > \sigma$, then either there is a unique minimizer or the condensation-evaporation happens at lower temperature than segregation.*

Proof: First we consider the case $\alpha < \sigma$. We differentiate (5.42) and get

$$\beta p'(v) = -\frac{1}{(v-b)^2} + \frac{\beta\widetilde{\sigma}(\widehat{\phi}(v^{-1}))}{2v^3} + \frac{\beta\widehat{\phi}(\sigma+\alpha)\widehat{\phi}'(v^{-1})}{2v^4}. \tag{5.43}$$

Since $\widehat{\phi}$ is non decreasing, the last two terms are both positive and at least linearly increasing with β. Hence, for any fixed v and sufficiently large β they dominate the first term. On the other hand, for any fixed β and v sufficiently close to b, the first term dominates. Therefore the pressure is not monotone.

Let us now consider the case $\alpha > \sigma$. In the absence of segregation, the pressure is a strictly decreasing function of v and this precludes a condensation-evaporation transition in the absence of segregation. (This is quite different from the usual van der Waals case, or the case considered by Korteweg). The segregation permits a condensation-evaporation transition, and lowers the pressure in the liquid phase from what it would be without segregation. As the temperature is lowered further, the vapor phase may also undergo segregation. ■

In conclusion we may have at most four phases, characterized by the following values of the density:

$$\rho_v^{\pm} = \rho_v \frac{1 \pm \widehat{\phi}(\rho_v)}{2}, \quad \rho_l^{\pm} = \rho_l \frac{1 \pm \widehat{\phi}(\rho_l)}{2}. \tag{5.44}$$

We now return to the search for the constrained minimizers which are density profiles $\rho_1(x), \rho_2(x)$ such that

$$\int_\Lambda dx\rho_1(x) = n_1|\Lambda|, \quad \int_\Lambda dx\rho_2(x) = n_2|\Lambda|. \tag{5.45}$$

Clearly if $\min\{n_1, n_2\} \leq \rho_v^-$ or $\max\{n_1, n_2\} \geq \rho_l^+$ the only possible solutions are $\rho_1 = n_1$ and $\rho_2 = n_2$. Otherwise, non homogeneous minimizers are possible. Because of the previous lemma it is clear that ρ_1 and ρ_2 can only take the above values in four regions A, B, C and D whose volumes are the only relevant properties because we are dealing with the local interaction case. The minimizer $(\rho_1^\star, \rho_2^\star)$ is given by

$$\rho_1^\star(x) = \begin{cases} \rho_l^+ & \text{if } x \in A, \\ \rho_l^- & \text{if } x \in B, \\ \rho_v^+ & \text{if } x \in C, \\ \rho_v^- & \text{if } x \in D, \end{cases} \qquad \rho_2^\star(x) = \begin{cases} \rho_l^- & \text{if } x \in A, \\ \rho_l^+ & \text{if } x \in B, \\ \rho_v^- & \text{if } x \in C, \\ \rho_v^+ & \text{if } x \in D. \end{cases} \tag{5.46}$$

or viceversa. Let a, b, c and d denote the ratios of above volumes with $|\Lambda|$. The constraints are then

$$a\rho_l^+ + b\rho_l^- + c\rho_v^+ + d\rho_v^- = n_1, \quad a\rho_l^- + b\rho_l^+ + c\rho_v^- + d\rho_v^+ = n_2, \tag{5.47}$$

which, together with the relation

$$a + b + c + d = 1 \tag{5.48}$$

do not suffice to determine the fractions occupied by each phases. Note that, in the case $\beta_s(\rho_l) < \beta < \beta_s(\rho_v)$ we have $\rho_v^+ = \rho_v^-$ and the relations available are sufficient to determine the volume fractions occupied by the three phases.

Now we return to the finite volume case where the rearrangement inequalities take over and ensure regularity of the minimizers. In fact because the minimizers must be symmetric monotone, not all of the phases can be in contact, and there are constraints on the ordering of the densities. Suppose for example that we have

$$\rho_\ell^+ > \rho_\ell^- > \rho_v^+ > \rho_v^- \ . \tag{5.49}$$

We shall show that this is impossible.

Pick numbers a, b and c separating these density values so that

$$\rho_\ell^+ > a > \rho_\ell^- > b > \rho_v^+ > c > \rho_v^- \ . \tag{5.50}$$

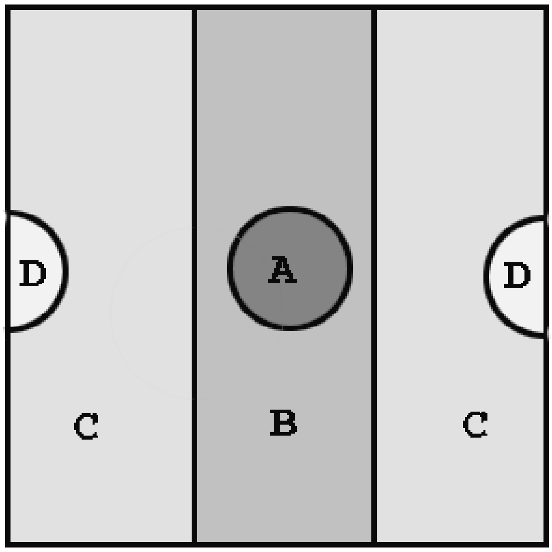
Define

$$A = \{\ x\ : \rho_1(x) > a\ \} \qquad B = \{\ x\ : a \geq \rho_1(x) > b\ \}$$
$$C = \{\ x\ : b \geq \rho_1(x) > c\ \} \qquad D = \{\ x\ : c \geq \rho_1(x)\ \}\ .$$

By the rearrangement inequalities, we see that ρ_2 must take its minimum in A, but we know from (5.46) that ρ_2 must take the value ρ_ℓ^- in A. So we must have instead that

$$\rho_\ell^+ > \rho_v^+ > \rho_v^- > \rho_\ell^- \ . \tag{5.51}$$

It is not evident how to deduce this in such generality by direct consideration of the local interaction model. The above regions are disjoint. To further determine the structure we need to consider the surface tension across their boundaries. In a later paper we will rigorously investigate the surface tension. Here we proceed on the assumption that because of it the phase boundaries will be so arranged as to have minimum surface area. Then, for a two dimensional torus, these regions would be arranged in the manner indicated in the diagram below:



Note that on a torus, the solutions of the isoperimetric problem are either disks, or bands depending on the size of the area to be enclosed.

Now we see that the liquid state rich in species one can only be in contact with the vapor state rich in species one, which in turn can only be in contact with the species one rich liquid state and the species two poor vapor state. In the case that all four phases are present, it is the surface tension of the domain boundaries that fixes the volume fractions in the finite range case.

6. The Microcanonical Minimization Problem.

In the previous sections we have considered a minimization problem related to a model in thermal contact with a reservoir. In this section we extend our results to a kinetic model preserving energy, but, for sake of simplicity, we confine ourselves to the ideal gas case with repulsion between different species and no attractive part. For example, consider the Boltzmann-Vlasov equations in [4], for the position and velocity distributions $f_i(x, v, t)$. The results on the minimization problem we have considered in the previous sections can be applied to determine the equilibrium solutions of these equations.

The equilibrium solutions of such a model are the minimizers of the entropy with energy and mass constraint. Therefore, the variational problem we want to deal with is the following: find the minimizers of

$$\mathcal{S}(f) = \int \mathrm{d}v \int_\Lambda \mathrm{d}x \sum_{i=1}^{2} f_i \log f_i \tag{6.1}$$

in the set

$$\mathcal{D}_{e,n_1,n_2} = \{f = (f_1, f_2) \,|\, |\Lambda|^{-1} \int \mathrm{d}v \int_\Lambda \mathrm{d}x f_i = n_i, \quad i = 1, 2; \quad E(f) = e\} \tag{6.2}$$

where f_1 and f_2 are probability densities in the phase space $\Lambda \times I\!R^3$ and

$$E(f) = |\Lambda|^{-1} \left[\int \mathrm{d}v \int_\Lambda \mathrm{d}x \frac{v^2}{2}(f_1 + f_2) + \int_\Lambda \mathrm{d}x \rho_{f_1} V * \rho_{f_2}\right], \quad \rho_{f_i} = \int \mathrm{d}v f_i. \tag{6.3}$$

This is equivalent to minimizing without constraints the functional

$$\mathcal{G}(f) = \mathcal{S}(f) + \beta E(f) - \sum_{i=1}^{2} \mu_i \int dv \int_\Lambda dv f_i \tag{6.4}$$

on

$$\mathcal{D} = \{f = (f_1, f_2) \,|\, |\, f_i \in L_1^+(\Lambda \times I\!R^3)\} \tag{6.5}$$

with β, μ_i, Lagrange multipliers to be determined by the constraints.

As is well known, the Euler-Lagrange equations for this functional force the v-dependence of f_i to be a Maxwellian at inverse temperature $\beta > 0$ and hence we are reduced to the following variational principle:

$$\mathcal{F}(\rho_1, \rho_2) = \int_\Lambda \mathrm{d}x \sum_{i=1}^{2} \rho_i[\log \rho_i - \bar{\mu}_i] + \beta \left[\int_\Lambda \mathrm{d}x \rho_1 V * \rho_2, \right] \tag{6.6}$$

on

$$\overline{\mathcal{D}} = \{(\rho_1, \rho_2) \,|\, |\rho_i \in L_1^+(\Lambda)\}, \tag{6.7}$$

with $\beta > 0$, $\bar{\mu}_i$ Lagrange multipliers to be determined by the constraints

$$\overline{E}(\rho_1, \rho_2) = |\Lambda|^{-1} \left[\int_\Lambda \mathrm{d}x \frac{3}{2} \beta^{-1} (\rho_1 + \rho_2) + \int_\Lambda \mathrm{d}x \rho_1 V * \rho_2 \right] = e, \quad |\Lambda|^{-1} \int_\Lambda \mathrm{d}x \rho_i = n_i. \tag{6.8}$$

Here $\bar{\mu}_i = \mu_i + \log(\sqrt{2\pi\beta^{-1}})^3$. This is equivalent to minimizing

$$\overline{\mathcal{S}}(\rho_1, \rho_2) = \sum_{i=1}^{2} \int_\lambda \mathrm{d}x \rho_i \log \rho_i \tag{6.9}$$

on the set

$$\overline{\mathcal{D}}_{e,n_1,n_2} = \{(\rho_1, \rho_2) \,|\, |\Lambda|^{-1} \int_\Lambda \mathrm{d}x \rho_i = n_i, \quad i = 1, 2; \quad \bar{E}(f) = e\}. \tag{6.10}$$

The discussion in previous sections obviously extends to the study of the minimizers of the functional (6.6). Therefore we now only deal with the solvability of the conditions on the Lagrange multipliers. This relies essentially on the local interaction case, because the arguments of section 3 apply.

Consider first the local interaction case, with unconstrained energy. For β sufficiently small, ($\alpha\beta n < 2$) there is only a homogeneous solution. The energy as a function of β is given by

$$E(\beta) = \frac{3}{2} \beta^{-1} n + \alpha n^2 |\Lambda|, \tag{6.11}$$

i.e. the usual linear behavior in $\beta^{-1} = T$. For sufficiently large β ($\alpha\beta n > 2$) the solution is of the form:

$$\rho_1(x) = \begin{cases} \rho_+, & x \in A, \\ \rho_- & x \in \Lambda - A, \end{cases} \quad \rho_2(x) = \begin{cases} \rho_-, & x \in A, \\ \rho_+ & x \in \Lambda - A, \end{cases} \tag{6.12}$$

with $\rho_\pm = \rho(1 \pm \phi(\beta))/2$, $\phi(\beta)$ the unique positive solution to

$$\tanh^{-1} \phi(\beta) = \frac{\alpha\beta\rho}{2} \phi(\beta), \tag{6.13}$$

and A a suitable subset of $|\Lambda|$, whose volume is determined to fulfill the mass constraints. Of course in the present situation $\rho = n = n_1 + n_2$ and hence the energy is given by

$$E(\beta) = \frac{3}{2}\beta^{-1}n + \alpha \int_\Lambda \mathrm{d}x \rho_1\rho_2 = \frac{3}{2}\beta^{-1}n + \alpha|\Lambda|\rho_+\rho_- = \frac{3}{2}\beta^{-1}n + \frac{\alpha|\Lambda|n^2}{4}(1-\phi(\beta)^2). \tag{6.14}$$

This is obviously continuous because $\phi(\beta) \to 0$ as $\beta \to 2/n\alpha$. Moreover by using arguments similar to those of section 5 one can show that $\phi(\beta)$ is increasing for $\beta > 2/n\alpha$. Therefore $E(\beta)$ is monotone decreasing, as sum of two decreasing functions, and hence invertible.

The function $\beta(e)$ is well defined and, with this definition, the phase space densities for the two species are given by

$$f_i(x,v) = \rho_i(x)\frac{\mathrm{e}^{-\beta(e)v^2/2}}{(2\pi\beta(e)^{-1})^{3/2}}, \tag{6.15}$$

where ρ_i are the minimizers of the free energy at temperature $\beta(e)$.

These densities give us the minimizers of the entropy $\mathcal{S}(f)$ under the constraints $E(f) = e$. Note that the determination of the function $\beta(e)$ in (6.15) require knowledge of the partition of e into its kinetic and interaction parts. This is provided by the results in the previous sections.

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Appendix A.

Here we prove the rearrangement inequalities used in the paper.

Proof of Lemma 2.2 We shall apply a sequence of symmetrization operations, and show that unless the minimizer is symmetric monotone, then one of the these operations would strictly lower the integrals in (2.5) and (2.6).

These symmetrization operations are "transplants" to the torus of the following symmetrization operation on the circle $\mathcal{S}^1 = \{(x_{1,2}) \mid x_1^2 + x_2^2 = 1\}$ in the x_1, x_2 plane. Fix any unit vector u on $\mathcal{S}^1$, and define the reflection R_u on $\mathcal{S}^1$ given by

$$R_u x = x - 2(u\cdot x)u \tag{A.1}$$

where $x = (x,y)$, and the dot denotes the usual inner product. It is also convenient to define $R_u\theta$ for $-\pi < \theta < \pi$ by

$$R_u(\cos(\theta),\sin(\theta)) = (\cos(R_u\theta),\sin(R_u\theta)) \ . \tag{A.2}$$

Next, define an operator, denoted R_u^+, on measurable functions on $\mathcal{S}^1$ by

$$R_u^+ g(x) = \begin{cases} \max\{g(x), g(R_u x)\} & \text{if } x\cdot u_0 \geq 0 \ , \\ \min\{g(x), g(R_u x)\} & \text{if } x\cdot u_0 < 0 \end{cases} \tag{A.3}$$

where u_0 denotes $(0,1)$. That is, the line through the origin perpendicular to u divides the circle in two, and the two halves are identified by the reflection fixing this line. The symmetrization operation swaps values at reflected pairs of points, if necessary, so that the large value is always on the side containing the "north pole", $(0,1)$. We also define an operator R_u^- in the same manner, except that we put the small values on the side with the "north pole". In other words, we interchange the minimum and maximum in (A.3)

We now state the lemma for which these definitions were made.

Lemma A.1 *Let F be a symmetric function on $\mathbb{R}_+ \times \mathbb{R}_+$ that satisfies (1.2) everywhere on its domain. Let K be any strictly decreasing non-negative function on $\mathbb{R}_+$. Then for any two bounded non–negative measurable functions g and h on $\mathcal{S}^1$ identified with $[-\pi,\pi)$,*

$$\int_{-\pi}^{\pi}\int_{-\pi}^{\pi} g(\theta)K(|\theta-\phi|)h(\phi)\mathrm{d}\theta\mathrm{d}\phi \geq \int_{-\pi}^{\pi}\int_{-\pi}^{\pi} R_u^+ g(\theta)K(|\theta-\phi|)R_u^- h(\phi))\mathrm{d}\theta\mathrm{d}\phi \tag{A.4}$$

and

$$\int_{-\pi}^{\pi} F(g(\theta),h(\theta)\mathrm{d}\theta \geq \int_{-\pi}^{\pi} F(R_u^+ g(\theta),R_u^- h(\theta))\mathrm{d}\theta\ . \tag{A.5}$$

Moreover, there is equality in inequality (A.4) if and only if for some fixed θ_0, $R_u^+ g(\theta-\theta_0) = g(\theta)$ and $R_u^- h(\theta-\theta_0) = h(\theta)$ for almost all θ.

Lemma A.1 will be proved after using it to prove Lemma 2.2. The argument is adapted from [3], which however does not consider cases of equality.

Consider first (2.5). Fix any index i, and fix values of x_j and y_j for $j \neq i$. Determine a sequence of unit vectors $\{u_j\}$ inductively as follows. Suppose that the u_i have been determined for $i < j$. For $i < j$, define g_i and h_i inductively by

$$g_i = R_{u_i}^+ g_{i-1}\ , \qquad g_0 = g \qquad \text{and} \qquad h_i = R_{u_i}^- h_{i-1}\ , \qquad h_0 = h\ . \tag{A.6}$$

Now choose u_j so that

$$\begin{aligned}&\int_{-\pi}^{\pi}\int_{-\pi}^{\pi} R_{u_j}^+ g_{j-1}(\theta)K(|\theta-\phi|)R_{u_j}^- h_{j-1}(\phi))\mathrm{d}\theta\mathrm{d}\phi - \int_{-\pi}^{\pi}\int_{-\pi}^{\pi} g_{j-1}(\theta)K(|\theta-\phi|)h_{j-1}(\phi))\mathrm{d}\theta\mathrm{d}\phi\\ &\leq \frac{1}{2}\sup_u\left\{\left(\int_{-\pi}^{\pi}\int_{-\pi}^{\pi} R_u^+ g(\theta)K(|\theta-\phi|)R_u^- h(\phi))\mathrm{d}\theta\mathrm{d}\phi - \int_{-\pi}^{\pi}\int_{-\pi}^{\pi} g_{j-1}(\theta)K(|\theta-\phi|)h_{j-1}(\phi))\mathrm{d}\theta\mathrm{d}\phi\right)\right\}\ .\end{aligned}$$

That is, we choose the direction u_j to get an effect that is at least half of the largest possible effect at that stage.

The operations $R_{u_j}^{\pm}$ preserve the modulus of continuity, so if g and h are continuous, the sequences $\{g_j\}$ and $\{h_j\}$ are strongly compact. Passing to a subsequence along which limits exist, define

$$g^\star = \lim_{j\to\infty} g_j \qquad \text{and} \qquad h_\star = \lim_{j\to\infty} h_j\ . \tag{A.7}$$

It follows from the choice of the sequence that for every u,

$$\int_{-\pi}^{\pi}\int_{-\pi}^{\pi} R_{u_j}^+ g^\star(\theta)K(|\theta-\phi|)R_{u_j}^- h_\star(\phi))\mathrm{d}\theta\mathrm{d}\phi = \int_{-\pi}^{\pi}\int_{-\pi}^{\pi} g^\star(\theta)K(|\theta-\phi|)h_\star(\phi))\mathrm{d}\theta\mathrm{d}\phi\ . \tag{A.8}$$

By Lemma 5.3, this is only possible if $g^\star$ is symmetric monotone about the "north pole", and $h_\star$ is symmetric monotone about the south pole, $-u_0$. Thus, since the $\{g_j\}$ are equimeasurable, as are the $\{h_j\}$, $g^\star$ and $h_\star$ are the symmetric monotone rearrangements of g and h respectively. Since

$$\int_{-\pi}^{\pi}\int_{-\pi}^{\pi} R_{u_j}^+ g_{j-1}(\theta)K(|\theta-\phi|)R_{u_j}^- h_{j-1}(\phi))\mathrm{d}\theta\mathrm{d}\phi - \int_{-\pi}^{\pi}\int_{-\pi}^{\pi} g_{j-1}(\theta)K(|\theta-\phi|)h_{j-1}(\phi))\mathrm{d}\theta\mathrm{d}\phi \leq 0 \tag{A.9}$$

for each j, it follows that

$$\int_{-\pi}^{\pi}\int_{-\pi}^{\pi} g^\star(\theta)K(|\theta-\phi|)h_\star(\phi))\mathrm{d}\theta\mathrm{d}\phi \le \int_{-\pi}^{\pi}\int_{-\pi}^{\pi} g(\theta)K(|\theta-\phi|)h(\phi))\mathrm{d}\theta\mathrm{d}\phi\ . \tag{A.10}$$

The requirement of continuity is easily removed by a density argument, exactly as in [3]. This proves the inequality in general. Now for equality to hold, it must be the case that

$$\int_{-\pi}^{\pi}\int_{-\pi}^{\pi} R^+_{u_j}g(\theta)K(|\theta-\phi|)R^-_{u_j}h(\phi))\mathrm{d}\theta\mathrm{d}\phi = \int_{-\pi}^{\pi}\int_{-\pi}^{\pi} g(\theta)K(|\theta-\phi|)h(\phi))\mathrm{d}\theta\mathrm{d}\phi \tag{A.11}$$

for all u. But according to Lemma 5.3, this is only possible if, up to a common rotation, $g = g^\star$ and $h = h_\star$. Now repeat the argument for each of the coordinates. A similar argument applies to (2.6). ■

Proof of Lemma A.1 Given a unit vector u, let $\mathcal{S}_+$ denote the set of points x in $\mathcal{S}^1$ such that

$$(x\cdot u)(u\cdot u_0) \ge 0\ , \tag{A.12}$$

where as before, u_0 is the "north pole" $(0,1)$. Let $\mathcal{S}_-$ be the complement of $\mathcal{S}_+$. That is, the line fixed by R_u slices S^1 in two, and $\mathcal{S}_+$ is the half containing the north pole. Then since R_u is a measure preserving transformation,

$$\begin{aligned}
&\int_{-\pi}^{\pi}\int_{-\pi}^{\pi} g(\theta)K(|\theta-\phi|)h(\phi)\mathrm{d}\theta\mathrm{d}\phi = \\
&\int_{\mathcal{S}_+}\int_{\mathcal{S}_+} g(\theta)K(|\theta-\phi|)h(\phi)\mathrm{d}\theta\mathrm{d}\phi + \int_{\mathcal{S}_-}\int_{\mathcal{S}_-} g(\theta)K(|\theta-\phi|)h(\phi)\mathrm{d}\theta\mathrm{d}\phi + \\
&\int_{\mathcal{S}_-}\int_{\mathcal{S}_+} g(\theta)K(|\theta-\phi|)h(\phi)\mathrm{d}\theta\mathrm{d}\phi + \int_{\mathcal{S}_+}\int_{\mathcal{S}_-} g(\theta)K(|\theta-\phi|)h(\phi)\mathrm{d}\theta\mathrm{d}\phi = \\
&\int_{\mathcal{S}_+}\int_{\mathcal{S}_+} g(\theta)K(|\theta-\phi|)h(\phi)\mathrm{d}\theta\mathrm{d}\phi + \int_{\mathcal{S}_+}\int_{\mathcal{S}_+} g(R_u\theta)K(|\theta-\phi|)h(R_u\phi)\mathrm{d}\theta\mathrm{d}\phi \\
&\int_{\mathcal{S}_+}\int_{\mathcal{S}_+} g(R_u\theta)K(|R_u\theta-\phi|)h(\phi)\mathrm{d}\theta\mathrm{d}\phi + \int_{\mathcal{S}_+}\int_{\mathcal{S}_+} g(\theta)K(|\theta-R_u\phi|)h(R_u\phi)\mathrm{d}\theta\mathrm{d}\phi
\end{aligned} \tag{A.13}$$

.

The desired inequality is then a consequence of the following inequality for paris of real numbers: Let a_1 and a_2 and b_1 and b_2 be any for positive real numbers. Rearrange a_1 and a_2 to decrease, and b_1 and b_2 to increase; i.e., let Let $a_1^\star = \max\{a_1,a_2\}$ and let $a_2^\star = \min\{a_1,a_2\}$, and let $b_1^\star = \min\{b_1,b_2\}$ and let $b_2^\star = \max\{b_1,b_2\}$. Then

$$a_1^\star b_1^\star + a_2^\star b_2^\star \le a_1b_1 + a_2b_2\ , \tag{A.14}$$

and there is equality if and only if $a_1 = a_1^\star$ and $b_1 = b_1^\star$ or $a_1^\star = a_2$, $b_1^\star = b_2$.

We now apply this with

$$a_1 = g(\theta) \quad a_2 = g(R_u\theta) \quad b_1 = h(\phi) \quad \text{and} \quad b_2 = h(R_u\phi)\ . \tag{A.15}$$

Then

$$a_1^\star = R_u^+g(\theta) \quad a_2^\star = R_u^+g(R_u\theta) \quad b_1^\star = R_u^-h(\phi) \quad \text{and} \quad b_2^\star = R_u^-h(R_u\phi)\ . \tag{A.16}$$

Since

$$K(|\theta - R_u\phi|) = K(|R_u\theta - \phi|) < K(|\theta - \phi|) \tag{A.17}$$

almost everywhere, we have that

$$\begin{aligned} &g(\theta)K(|\theta-\phi|)h(\phi) + g(R_u\theta)K(|\theta-\phi|)h(R_u\phi) + \\ &g(\theta)K(|R_u\theta-\phi|)h(\phi) + g(R_u\theta)K(|R_u\theta-\phi|)R_u^- h(R_u\phi) \ge \\ &R_u^+ g(\theta)K(|\theta-\phi|)R_u^- h(\phi) + R_u^+ g(R_u\theta)K(|\theta-\phi|)R_u^- h(R_u\phi) + \\ &R_u^+ g(\theta)K(|R_u\theta-\phi|)R_u^- h(\phi) + R_u^+ g(R_u\theta)K(|R_u\theta-\phi|)R_u^- h(R_u\phi) \end{aligned} \tag{A.18}$$

for almost every θ and ϕ in $\mathcal{S}_+$, with equality if and only if

$$g(R_u\theta) \le g(\theta) \qquad \text{and} \qquad h(R_u\phi) \ge g(\phi) \tag{A.19}$$

or

$$g(R_u\theta) \ge g(\theta) \qquad \text{and} \qquad h(R_u\phi) \le g(\phi) \tag{A.20}$$

for almost every θ and ϕ in $\mathcal{S}_+$. Now unless g is constant, we can find a θ and u so that either $g(R_u\theta) < g(\theta)$ or $g(R_u\theta) > g(\theta)$. Suppose it is the first case. Then (A.19) holds, and for almost every ϕ, we must have $h(R_u\phi) \ge g(\phi)$. Making a similar argument for h, we see that one of (A.19) or (A.20) must hold for almost every θ and ϕ. The only way that this can happen is if g and h are symmetric monotone.

The key here is the following pointwise inequality: If $a > b$ and $c > d$, then

$$F(a,d) + F(b,c) < F(a,c) + F(b,d) \ . \tag{A.21}$$

To see this, let $k = c - d$ and $h = a - b$. Then

$$(F(a,d) + F(b,c)) - (F(a,c) + F(b,d)) = -hk \int_0^1 \int_0^1 \frac{\partial^2 F}{\partial\rho_1 \partial\rho_2}(b + sh, d + tk) \mathrm{d}s\mathrm{d}t \ . \tag{A.22}$$

The second inequality of Lemma A.1 follows directly from this. ■

Remark As can be seen from the proof, the decrease upon rearrangement can be estimated quantitatively if we strengthen (1.2) so that the positive lower bound is uniform. This could be used to relax the conditions on the interactions.

Appendix B.

In this appendix we prove the following theorem which extends Theorem 2.3 to the case where no a priori bound on the densities is assumed, or the entropy term does not necessarily prevents vacuum, or both.

Theorem B.1 *Assume:*

i) G and D strictly convex functions

ii) $\lim_{r\to\infty} \frac{F(r)}{r} = \infty$,

iii) G and D satisfy the following doubling condition: there exists a constant K so that for all r,

$$G(2r) \le KG(r) \qquad \text{and} \qquad D(2r) \le KD(r) \ . \tag{B.1}$$

iv) there is an $L > -\infty$ so that for all $r \geq 0$,

$$G(r) \geq L \qquad \text{and} \qquad D(r) \geq L\ . \tag{B.2}$$

Then, under the same assumptions on the interactions of Theorem 2.3, the conclusions of Theorem 2.3 are still true.

Proof: We first observe that for any fixed admissible $\rho_1^{(0)}$, the functional $\mathcal{G}(\rho_2)$ defined by

$$\mathcal{G}(\rho_2) = \mathcal{F}(\rho_1^{(0)}, \rho_2) \tag{B.3}$$

is strictly convex on the set of densities satisfying the mean density constraint. By Fatou's lemma, it is also lower semicontinuous in the L^1 topology.

Now since we seek minimizers of $\mathcal{F}$, we may assume that $\rho_1^{(0)}$ is symmetric monotone about the point on the torus that is antipodal to the origin, and then we may restrict our consideration of $\rho_2 \mapsto \mathcal{F}(\rho_1^{(0)}, \rho_2)$ to densities that are symmetric monotone about the origin.

Now for any fixed constant M, the set of densities ρ_2 such that $\rho_2(x) \leq M$ for all x, and that ρ_2 is symmetric monotone is strongly compact in $L^1(\Lambda)$ by the Helly selection principle. It therefore follows that there is a unique minimizer $\widetilde{\rho}_2$ of $\rho_2 \mapsto \mathcal{F}(\rho_1^{(0)}, \rho_2)$ in this class. By Lemma 2.2, this is also the unique minimizer of $\mathcal{G}(\rho_2)$ in the class of densities satisfying:

(i) $\int_\Lambda \rho_2 \mathrm{d}x = n_2$.

(ii) ρ_2 is symmetric monotone with its maximum at the point antipodal to the origin.

(iii) $\|\rho_2\|_\infty \leq M$

We now show that for M large enough, this minimizer ceases to depend on M, so that the condition (iii) becomes superfluous.

Consider a variation $\widetilde{\rho}_2 + h$ of $\widetilde{\rho}_2$. Clearly, we must have $h \leq 0$ on

$$A_M = \{x\ |\widetilde{\rho}_2(x) = M\}\ , \tag{B.4}$$

and $h \geq 0$ on

$$A_Z = \{x\ |\widetilde{\rho}_2(x) = 0\}\ .$$

We must also have

$$\int_\Lambda h(x)\mathrm{d}x = 0 \tag{B.5}$$

in order to preserve the condition that $(\rho_1^{(0)}, \widetilde{\rho}_2 + h)$ belongs to $\mathcal{D}(n_1, n_2)$. The Euler–Lagrange condition then is

$$\int_\Lambda \left(F'(\widetilde{\rho}_2) + D'(\rho_1^{(0)} + \widetilde{\rho}_2) + U * \rho_1^{(0)}\right) h\mathrm{d}x \geq 0 \tag{B.6}$$

Let B denote $(A_M \cup A_Z)^c$. We first show that for M sufficiently large, $|B| \neq 0$. Indeed, if $|B| = 0$, then $\widetilde{\rho}_2 = M 1_{A_M}$. Since $\int_\Lambda \widetilde{\rho}_2(x)\mathrm{d}x = n_1|\Lambda|$, $|A_Z|$ then equals $n_2|\Lambda|/M$ and consequently,

$$\mathcal{G}(\widetilde{\rho}_2) \geq \frac{F(M)}{M} n_2|\Lambda|\ . \tag{B.7}$$

Since $F(M)/M$ tends to infinity with M, and since $\mathcal{G}(\widetilde{\rho}_2) \leq \mathcal{G}(n_2/|\Lambda|)$ whenever $M \geq n_2/|\Lambda|$ so that $\rho_2 = n_2/|\Lambda|$ is an admissible trial density, we obtain a contradiction. Henceforth take M large enough to ensure that $|B| \neq 0$. Note that on B that the Euler Lagrange equation

$$F'(\widetilde{\rho}_2) + D'(\rho_1^{(0)} + \widetilde{\rho}_2) + U * \rho_1^{(0)} = C \tag{B.8}$$

holds for some value of C, since on this set, if h is an admissible variations, so is $-h$.

We have the Chebyshev estimate

$$|A_Z| \leq \frac{n_2|\Lambda|}{M} \ . \tag{B.9}$$

Further increasing M, we may assume that $|A_Z| < |\Lambda|/3$. It then follows that

$$\text{either} \qquad |B| \geq \frac{|\Lambda|}{3} \qquad \text{or} \qquad |A_Z| \geq \frac{|\Lambda|}{3} \ . \tag{B.10}$$

Our next goal is to obtain an *a–priori* bound on C. We will obtain two such bounds: one that is valid, when $|B|$ is not too small, and one that is valid when $|A_Z|$ is not too small. By the above, at least one of these two must hold.

To obtain a bound that will be useful if $|B|$ is not too small, integrate the Euler Lagrange equation over B, and obtain

$$\int_B F'(\widetilde{\rho}_2)\mathrm{d}x + \int_B D'(\rho_1^{(0)} + \widetilde{\rho}_2)\mathrm{d}x + \int_B U * \rho_1^{(0)}\mathrm{d}x = C|B| \ . \tag{B.11}$$

Now, by convexity, for any a,

$$F'(a) \leq F(a+1) - F(a) \leq F(a+1) = F\left(\frac{2a+2}{2}\right) \leq \frac{1}{2}F(2a) + \frac{1}{2}F(2) \ . \tag{B.12}$$

Making use of the doubling condition, we finally have

$$F'(a) \leq \frac{K}{2}F(a) + \frac{1}{2}F(2) \ .$$

Replacing a by $\widetilde{\rho}_2$ and integrating over B,

$$\int_B F'(\widetilde{\rho}_2)\mathrm{d}x \leq \frac{K}{2}\int_\Lambda F(\widetilde{\rho}_2)\mathrm{d}x + \frac{1}{2}F(2)|\lambda| \ . \tag{B.13}$$

In the same way, we obtain

$$\int_B D'(\rho_1^{(0)} + \widetilde{\rho}_2)\mathrm{d}x \leq \frac{K}{2}\int_\Lambda D(\rho_1^{(0)} + \widetilde{\rho}_2)\mathrm{d}x + \frac{1}{2}D(2)|\lambda| \ . \tag{B.14}$$

Finally,

$$\int_B U * \rho_1^{(0)}\mathrm{d}x \leq \alpha|\Lambda|n_1 \ . \tag{B.15}$$

Combining estimates we have

$$C \leq \frac{1}{|B|}\left(\frac{K}{2}\mathcal{G}(n_2/|\Lambda|) + \frac{1}{2}\left(F(2) + D(2)\right) + \alpha n_1|\Lambda|\right) \ , \tag{B.16}$$

so that under the first alternative above, we have the *a–priori* bound

$$C \leq \frac{3}{|\Lambda|}\left(\frac{K}{2}\mathcal{G}(n_2/|\Lambda|) + \frac{1}{2}\left(F(2) + D(2)\right) + \alpha n_1|\Lambda|\right) \ . \tag{B.17}$$

Next, we obtain an estimate that will be useful is $|A_Z|$ is not too small. First, if $|A_Z| \neq 0$, we can consider a variation of the following type: Let h satisfying (B.5) with h_+ supported by A_Z, and with h_- supported in B. Then if we let $a = \int_\Lambda h_+ \mathrm{d}x$, we have from (B.6) that

$$F'(0) + \frac{1}{a}\int_\Lambda \left(D'(\rho_1^{(0)}) + U * \rho_1^{(0)} \right) h_+ \mathrm{d}x \geq C \ . \tag{B.18}$$

By this variational inequality,

$$C \leq F'(0) + \frac{1}{|A_Z|}\int_{A_Z} \left(D'(\rho_1^{(0)}) + U * \rho_1^{(0)} \right) \mathrm{d}x \ . \tag{B.19}$$

The same convexity arguments employed above now yield

$$C \leq F'(0) + \frac{1}{|A_Z|}\left(\frac{K}{2}\mathcal{G}(n_2/|\Lambda|) + \frac{1}{2}D(2) + \alpha n_1 |\Lambda| \right) \ , \tag{B.20}$$

which, under the second alternative above, becomes

$$C \leq F'(0) + \frac{3}{|\Lambda|}\left(\frac{K}{2}\mathcal{G}(n_2/|\Lambda|) + \frac{1}{2}D(2) + \alpha n_1 |\Lambda| \right) \ . \tag{B.21}$$

Now notice that in the case $F(t) = t\log t$, $F'(0) = -\infty$, so in this case, $|A_Z| > 0$ is precluded. We therefore have the *a–priori* estimate

$$C \leq \max\{F'(0), 0\} + \frac{3}{|\Lambda|}\left(\frac{K}{2}\mathcal{G}(n_2/|\Lambda|) + \frac{1}{2}\left(F(2) + D(2)\right) + \alpha n_1 |\Lambda| \right) \ . \tag{B.22}$$

To apply his, we suppose $|A_M| > 0$, and consider a variation of the following type: Let h satisfy (B.5) with h_-, the negative part of h, supported by A_M, and with h_+ supported in B. Then if we let $a = \int_\Lambda h_+ \mathrm{d}x$, we have from (B.6) that

$$-\int_\Lambda \left(F'(\widetilde{\rho}_2) + D'(\rho_1^{(0)} + \widetilde{\rho}_2) + U * \rho_1^{(0)} \right) h_- \mathrm{d}x + Ca \geq 0 \tag{B.23}$$

This means that

$$\frac{1}{a}\int_\Lambda \left(F'(\widetilde{\rho}_2) + D'(\widetilde{\rho}_2) + U * \rho_1^{(0)} \right) h_- \mathrm{d}x \leq C \ , \tag{B.24}$$

which in turn, since D' is an increasing function, means that

$$F'(M) \leq C \ . \tag{B.25}$$

Let G be the inverse function to F'; i.e.,

$$G(t) = \inf\{ r \geq 0 \mid F'(r) \geq t \ \} \ . \tag{B.26}$$

Then, we have the *a–priori* bound

$$M \leq G(C) \ ,$$

which, when combined with our *a–priori* bound on C implies that for sufficiently large M, $|A_M| = 0$.

Therefore, our minimizer $\widetilde{\rho}_2$ satisfies

$$\widetilde{\rho}_2 \leq G\left(\max\{F'(0), 0\} + \frac{3}{|\Lambda|}\left(\frac{K}{2}\mathcal{G}(n_2/|\Lambda|) + \frac{1}{2}\left(F(2) + D(2)\right) + \alpha n_1|\Lambda|\right)\right) . \tag{B.27}$$

Moreover, we see that wherever $\widetilde{\rho}_2 \neq 0$, it satisfies the Euler–Lagrange equation.

Now fix this minimizer $\widetilde{\rho}_2$ and consider the functional

$$\mathcal{G}_1(\rho_1) = \mathcal{F}(\rho_1, \widetilde{\rho}_2) . \tag{B.28}$$

Exactly the same argument shows that this has a minimizer $\widetilde{\rho}_1$ in the class of densities that are symmetric monotone with a maximum at the origin, and with $\int_\Lambda \rho_1 \mathrm{d}x = |\Lambda| n_1$. Moreover, we obtain in this way the *a–priori* bound

$$\widetilde{\rho}_1 \leq G\left(\max\{F'(0), 0\} + \frac{3}{|\Lambda|}\left(\frac{K}{2}\mathcal{G}(n_1/|\Lambda|) + \frac{1}{2}\left(F(2) + D(2)\right) + \alpha n_2|\Lambda|\right)\right) . \tag{B.29}$$

Now consider a minimizing sequence $(\rho_1^{(k)}, \rho_2^{(k)})$ in $\mathcal{D}(n_1, n_2)$ for $\mathcal{F}$. By the argument above, we can replace each pair $(\rho_1^{(k)}, \rho_2^{(k)})$ by another pair $(\widetilde{\rho}_1^{(k)}, \widetilde{\rho}_2^{(k)})$ in $\mathcal{D}(n_1, n_2)$ so that $\widetilde{\rho}_1^{(k)}$ and $\widetilde{\rho}_2^{(k)}$ are symmetric monotone about the origin and its antipodal point respectively, and so that $\widetilde{\rho}_1^{(k)}$ and $\widetilde{\rho}_2^{(k)}$ satisfy the *a–priori* L^∞ bounds above, and last but not least, so that

$$\mathcal{F}(\widetilde{\rho}_1^{(k)}, \widetilde{\rho}_2^{(k)}) \leq \mathcal{F}(\rho_1^{(k)}, \rho_2^{(k)}) . \tag{B.30}$$

Now by the Helly selection principle again, we have that for a subsequence,

$$\widetilde{\rho}_1 = \lim_{n\to\infty} \widetilde{\rho}_1^{(k_n)} \tag{B.31}$$

and

$$\widetilde{\rho}_2 = \lim_{n\to\infty} \widetilde{\rho}_2^{(k_n)} \tag{B.32}$$

exist almost everywhere. By the *a–priori* L^∞ bounds, and the dominated convergence theorem, these limits also hold in L^1, and hence $(\widetilde{\rho}_1, \widetilde{\rho}_2) \in \mathcal{D}(n_1, n_2)$. Moreover, by the lower semicontinuity of $\mathcal{F}$ discussed at the outset,

$$\mathcal{F}(\widetilde{\rho}_1, \widetilde{\rho}_2) \leq \liminf_{n\to\infty} \mathcal{F}(\widetilde{\rho}_1^{(k_n)}, \widetilde{\rho}_2^{(k_n)}) = \inf_{(\rho_1,\rho_2)\in\mathcal{D}(n_1,n_2)} \mathcal{F}(\rho_1, \rho_2) . \tag{B.33}$$

This proves the existence of our minimizers.

We now drop the tilde, and examine their properties. By the argument above, we clearly have

$$\rho_1(x) = G\left(C_1 - U * \rho_2(x) - D'(\rho_1(x) + \rho_2(x))\right) \tag{B.34}$$

on the set where $\rho_1(x) \neq 0$, and likewise

$$\rho_2(x) = G\left(C_2 - U * \rho_1(x) - D'(\rho_1(x) + \rho_2(x))\right) \tag{B.35}$$

on the set where $\rho_2(x) \neq 0$, and we have the asserted L^∞ bounds. Finally, since ρ_2 and ρ_1 are monotone symmetric, these L^∞ bounds imply L^1 bounds for the gradients of ρ_1 and ρ_2. ■

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