Weierstrass Institute for
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Leibniz
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Cluster Size Distributions in a Classical Many-Body System

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based on joint works with Collevecchio (Melbourne), **Jansen (Leiden)**, Metzger (Duisburg), Mörters (Bath), Sidorova (London).

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Background and Goals of this Talk

We consider a classical stable interacting many-particle system with attraction in continuous space.

- We study the transition between gaseous and solid phase in the thermodynamic limit.
- Very difficult at positive temperature and positive particle density.
- Most explicit results in the dilute low-temperature regime. Here, the particles organise themselves into small groups called clusters.
- We approximate the system with a well-known ideal-mixture of clusters (droplets) and prove that the difference vanishes exponentially with vanishing temperature.

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- We study
  - the free energy,
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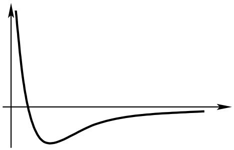
  - the optimal cluster-size distribution.
- We clarify the relation to existence and uniqueness of a Gibbs measure and its percolation properties.
- We prove crystallisation of the particles inside large clusters at positive temperature (in progress).

Energy

Energy of N particles in $\mathbb{R}^d$:

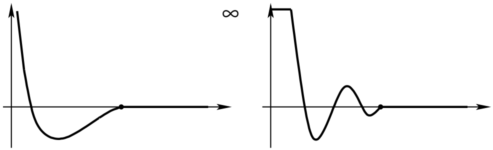
$$U_N(x) = U_N(x_1, \dots, x_N) = \sum_{\substack{i,j=1 \\ i \neq j}}^{N} v\big(|x_i - x_j|\big), \qquad \text{for } x = (x_1, \dots, x_N) \in (\mathbb{R}^d)^N.$$

Pair-interaction function $v\colon [0,\infty) \to (-\infty, \infty]$ of Lennard-Jones type:



Lennard-Jones potential

$v(r) = r^{-12} - r^{-6}$



examples of our potentials

- short-distance repulsion (possibly hard-core) implying stability,
- preference of a certain positive distance,
- bounded interaction length.

Random Clusters

inverse temperature $\beta \in (0,\infty)$

$$\text{Gibbs measure:} \qquad \mathbb{P}^{(N)}_{\beta,\Lambda}(\mathrm{d}x) = \frac{1}{Z_\Lambda(\beta,N)N!} \mathrm{e}^{-\beta U_N(x)}\,\mathrm{d}x, \qquad x \in \Lambda^N.$$

$$\text{Partition function:} \qquad Z_\Lambda(\beta,N) = \frac{1}{N!}\int_{\Lambda^N} \mathrm{e}^{-\beta U_N(x)}\,\mathrm{d}x.$$

Connectivity structure: Fix R larger than the interaction length of v.

Sites x and y are called connected if $|x-y| \leq R$.

clusters (droplets) $=$ the connected components

$N_k(x) =$number of k-clusters in $x = (x_1,\dots,x_N)$

$$k\text{-cluster density :} \qquad \rho_{k,\Lambda}(x) = \frac{N_k(x)}{|\Lambda|}$$

$$\text{cluster size distribution:} \qquad \rho_\Lambda = \big(\rho_{k,\Lambda}\big)_{k\in\mathbb{N}}$$

as an $M_{N/|\Lambda|}$-valued random variable, where

$$M_\rho := \Big\{(\rho_k)_{k\in\mathbb{N}} \in [0,\infty)^{\mathbb{N}} \,\Big|\, \sum_{k\in\mathbb{N}} k\rho_k \leq \rho\Big\}, \qquad \rho \in (0,\infty).$$

Regimes Considered

We study the cluster-size distribution in the box $\Lambda = [0, L]^d$

- in the thermodynamic limit

$$N \to \infty, \qquad L = L_N \to \infty, \qquad \text{such that } \frac{N}{L_N^d} \to \rho \in (0, \infty),$$

  followed by the dilute low-temperature limit

$$\beta \to \infty, \rho \downarrow 0 \qquad \text{such that } -\frac{1}{\beta} \log \rho \to \nu \in (0, \infty),$$

  (joint work with SABINE JANSEN (Leiden) and BERND METZGER [JKM11])

- and in the coupled dilute low-temperature limit

$$N \to \infty, \qquad \beta = \beta_N \to \infty, \qquad L = L_N \to \infty \qquad \text{such that } -\frac{1}{\beta_N} \log \frac{N}{L_N^d} \to \nu > 0.$$

  (joint work with A. COLLEVECCHIO (Venice), P. MÖRTERS (Bath) and N. SIDOROVA (London), [CKMS10])

Here,

- total entropy $\approx$ sum of the entropies of the clusters,
- excluded-volume effect between the clusters may be neglected,
- mixing entropy may be neglected.

LDP in the Thermodynamic Limit

Free energy per unit volume : $$f_\Lambda(\beta, \tfrac{N}{|\Lambda|}) := -\frac{1}{\beta|\Lambda|}\log Z_\Lambda(\beta, N).$$

limiting free energy : $$f(\beta,\rho) := \lim_{\substack{N,L\to\infty \\ N/L^d\to\rho}} f_{[0,L]^d}(\beta, \tfrac{N}{L^d}).$$

Goal: find $f(\beta,\rho,\cdot)\colon M_\rho \to [0,\infty]$ such that

$$\frac{1}{N!}\int_{\Lambda^N} \mathrm{e}^{-\beta U_N(x)}\,1\!\!1\Big\{(\rho_{k,\Lambda}(x))_{k\in\mathbb{N}} \approx (\rho_k)_{k\in\mathbb{N}}\Big\}\,\mathrm{d}x \approx \exp\Big(-\beta|\Lambda| f(\beta,\rho,(\rho_k)_{k\in\mathbb{N}})\Big),$$

and define the rate function as

$$J_{\beta,\rho}\big((\rho_k)_{k\in\mathbb{N}}\big) = \beta\big(f(\beta,\rho,(\rho_k)_{k\in\mathbb{N}}) - f(\beta,\rho)\big).$$

Large deviation principle with convex rate function, [JKM11]

In the thermodynamic limit $N\to\infty$, $L\to\infty$, $N/L^d\to\rho$, the distribution of ρ_Λ under $\mathbb{P}^{(N)}_{\beta,\Lambda}$ with $\Lambda=[0,L]^d$ satisfies a large deviation principle with speed $|\Lambda| = L^d$. The rate function $J_{\beta,\rho}\colon M_{\rho+\varepsilon}\to[0,\infty]$ is convex, and its effective domain $\{J_{\beta,\rho}(\cdot)<\infty\}$ is contained in M_ρ.

On the Proof of the LDP

Standard strategy, adapted to cluster-size distributions:

1. Projection: LDP for $(\rho_{k,\Lambda}(x))_{k=1,\dots,j}$ for fixed j with some rate function $J_{\beta,\rho,j}$.
   - Use subadditivity along special seqences of increasing cubes (having a separating margin) to define a densely defined preliminary rate function,
   - extend this rate function continuously and prove that it is finite on open sets,
   - fill the gaps for an arbitrary sequence of cubes,
   - show that the extended preliminary rate function gives an LDP.
2. Apply the Gärtner-Dawson theorem (projective limit LDP) to get full LDP with rate function

$$J_{\beta,\rho}\big((\rho_k)_{k\in\mathbb{N}}\big) = \sup_{j\in\mathbb{N}} J_{\beta,\rho,j}\big((\rho_k)_{k=1,\dots,j}\big).$$

Dilute Low-Temperature Limit

The ground state, i.e., zero temperature : $$E_N := \inf_{x \in (\mathbb{R}^d)^N} U_N(x).$$

stability & subadditivity $\Longrightarrow$ $e_\infty := \lim_{N\to\infty} \frac{E_N}{N} \in (-\infty, 0)$ exists.

Interpret $q_k = k\rho_k/\rho$ as the probability that a given particle lies in a k-cluster.

Approximate rate function: $$g_\nu\big((q_k)_k\big) := \sum_{k\in\mathbb{N}} q_k \frac{E_k - \nu}{k} + \Big(1 - \sum_{k\in\mathbb{N}} q_k\Big) e_\infty$$

on the set
$$\mathscr{Q} := \Big\{ (q_k)_{k\in\mathbb{N}} \in [0,1]^{\mathbb{N}} \,\Big|\, \sum_{k\in\mathbb{N}} q_k \le 1 \Big\}$$

Γ-convergence of the rate function, [JKM11]

In the limit $\beta \to \infty$, $\rho \to 0$ such that $-\beta^{-1}\log\rho \to \nu$, the function

$$\mathscr{Q} \to \mathbb{R} \cup \{\infty\}, \qquad (q_k)_k \mapsto \frac{1}{\rho} f\big(\beta, \rho, (\tfrac{\rho q_k}{k})_{k\in\mathbb{N}}\big)$$

Γ-converges to g_ν.

Explanation

Our Approximations:

- We approximate $f(\beta,\rho,(\rho_k)_k)$ by an ideal gas of clusters, neglecting the "excluded volume":

$$f^{\text{ideal}}(\beta,\rho,(\rho_k)_k) := \sum_{k\in\mathbb{N}} k\rho_k f_k^{\text{cl}}(\beta) + \Big(\rho - \sum_{k\in\mathbb{N}} k\rho_k\Big) f_\infty^{\text{cl}}(\beta) + \frac{1}{\beta}\sum_{k\in\mathbb{N}} \rho_k(\log\rho_k - 1).$$

  ($f_k^{\text{cl}}(\beta) =$ free energy per particle in a cluster of size k.)

- We approximate $f^{\text{ideal}}(\beta,\rho,(\frac{\rho q_k}{k})_{k\in\mathbb{N}})$ with $\rho g_\nu(q)$ using two simplifications:
  - cluster internal free energies $\approx$ ground state energies: $f_k^{\text{cl}}(\beta) \approx E_k$.
  - $$\frac{1}{\beta}\sum_{k\in\mathbb{N}} \rho_k(\log\rho_k - 1) = \sum_{k\in\mathbb{N}} \rho_k \frac{\log\rho}{\beta} + \frac{\rho}{\beta}\sum_{k\in\mathbb{N}} \frac{q_k}{k}\Big(\log\frac{q_k}{k} - 1\Big) \approx \sum_{k\in\mathbb{N}} \rho_k \frac{\log\rho}{\beta}$$
    $$\approx -\rho\sum_{k\in\mathbb{N}} q_k \frac{\nu}{k}.$$

In classical statistical physics: "Geometric (or droplet) picture of condensation".

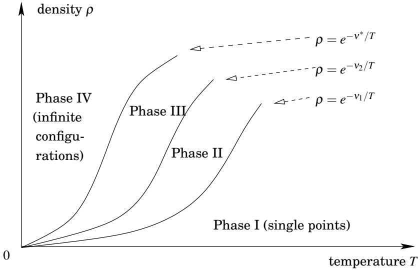
Closely related to the contour picture of the Ising model an lattice gases.

Corollary: Convergence of Minimisers

Consequences of Γ-convergence, [JKM11]

In the same limit $\beta \to \infty$, $\rho \downarrow 0$ such that $-\frac{1}{\beta}\log\rho \to \nu$,

- $$\frac{1}{\rho} f(\beta,\rho) \to \min_{\mathcal{Q}} g_\nu =: \mu(\nu),$$
- if ν is not a kink point of $\mu(\cdot)$, then any minimiser of $J_{\beta,\rho}$ converges to the minimiser of g_ν.



Properties of g_ν

- $\nu^* := \inf_{N\in\mathbb{N}}(E_N - Ne_\infty)$ lies in $(0,\infty)$.
- $\nu \mapsto \mu(\nu) = \inf_{\mathcal{Q}} g_\nu = \inf_{N\in\mathbb{N}} \frac{E_N-\nu}{N}$ is continuous, piecewise affine and concave.
- $\mu(\cdot)$ has at least one kink, and the kinks accumulate at most at ν^*.
- If $\nu \in (\nu^*,\infty)$ is not a kink point, then g_ν has the unique minimizer $\delta_{k(\nu)}$ (Dirac sequence) with $k(\nu)$ the unique minimizer of $k \mapsto (E_k - \nu)/k$.
- For $\nu < \nu^*$, the unique minimizer of g_ν is 0 (zero sequence).

Interpretation:

- There is at least one phase transition, possibly much more.
- In the high-temperature phase $\nu \gg 1$, all clusters are singletons.
- In any intermediate phase, all clusters have size $k(\nu)$.
- In the low-temperature phase $\nu \in (0,\nu^*)$, there are only infinite clusters.

Corollary: LLN for Cluster Sizes

The main consequence of the LDP, together with the Γ-convergence of the rate function, is:

Limiting distributions of cluster sizes, [JKM11]

Let $\nu \in (0,\infty)$ be not a kink point, and fix $\varepsilon > 0$. Then, if β is sufficiently large, ρ sufficiently small and $-\frac{1}{\beta}\log\rho$ is sufficiently close to ν, for boxes Λ_N with volume N/ρ,

$$\begin{aligned} \lim_{N\to\infty} \mathbb{P}^{(N)}_{\beta,\Lambda_N}\Big(\Big|\frac{k(\nu)}{\rho}\rho_{k(\nu),\Lambda} - 1\Big| > \varepsilon\Big) &= 0 \qquad \text{if } \nu > \nu^*, \\ \lim_{N\to\infty} \mathbb{P}^{(N)}_{\beta,\Lambda_N}\Big(\sum_{k\in\mathbb{N}} \rho_{k,\Lambda} > \varepsilon\Big) &= 0 \qquad \text{if } \nu < \nu^*. \end{aligned}$$

In other words, in this two-step limit, the model has only one cluster size, which is infinite for small ν.

Approximation with Ideal Mixture

- The approximation with g_ν is difficult to interpret physically, and g_ν has some "unphysical" properties: possibly many phase transitions of $\nu \mapsto \mu(\nu)$, and many minimisers of g_ν in the kinks. We think that just one of these phase transitions is "physical", the others correspond to cross-overs inside the gas phase.
- Much better is the approximation with the ideal mixture of droplets, f^{ideal}, which is known, under reasonable assumptions, to have only one phase transition.
- These assumptions are on the compactness of the shape of the relevant configurations at positive, but low temperature:
  - The main contribution to the cluster internal energy comes from compact (d-dimensional) configurations,
  - the correction term in the convergence $f_k^{\mathrm{cl}}(\beta) \to f_\infty^{\mathrm{cl}}(\beta)$ is of surface order: $k f_k^{\mathrm{cl}}(\beta) - k f_\infty^{\mathrm{cl}}(\beta) \geq C k^{1-1/d}$.

  (Verification seems out of reach yet.)
- We have rigorous bounds for the comparison of the original model with the ideal-mixture model, which are exponentially small in vanishing temperature, see next slides.

The ideal mixture

Recall:

$$f^{\rm ideal}(\beta,\rho,(\rho_k)_k) := \sum_{k\in\mathbb{N}} k\rho_k f_k^{\rm cl}(\beta) + \Big(\rho - \sum_{k\in\mathbb{N}} k\rho_k\Big) f_\infty^{\rm cl}(\beta) + \frac{1}{\beta}\sum_{k\in\mathbb{N}} \rho_k(\log\rho_k - 1).$$

- saturation density: Let

$$\rho_{\rm sat}^{\rm ideal}(\beta) := \sum_{k\in\mathbb{N}} k\,{\rm e}^{\beta k[f_\infty^{\rm cl}(\beta) - f_k^{\rm cl}(\beta)]} \in (0,\infty]$$

- chemical potential: For $\rho < \rho_{\rm sat}^{\rm ideal}(\beta)$, let $\mu^{\rm ideal}(\beta,\rho) \in (-\infty, f_\infty^{\rm cl}(\beta))$ be the unique solution of

$$\sum_{k=1}^{\infty} k\,{\rm e}^{\beta k[\mu^{\rm ideal}(\beta,\rho) - f_k^{\rm cl}(\beta)]} = \rho,$$

  and for $\rho \geq \rho_{\rm sat}^{\rm ideal}(\beta)$, let $\mu^{\rm ideal}(\beta,\rho) := f_\infty^{\rm cl}(\beta)$.
- Then, the minimiser $(\rho_k^{\rm ideal}(\beta,\rho))_k$ of $f^{\rm ideal}(\beta,\rho,\cdot)$ is given by

$$\rho_k^{\rm ideal}(\beta,\rho) = {\rm e}^{\beta k[\mu^{\rm ideal}(\beta,\rho) - f_k^{\rm cl}(\beta)]}.$$

- Under appropriate bounds on $f_k^{\rm cl}(\beta)$, the saturation density is finite at low temperature, and $f^{\rm ideal}(\beta,\rho,\cdot)$ has a phase transition.

Comparison with ideal mixture

Joint work with SABINE JANSEN (Leiden). Our hypotheses:

(1) Some Hölder continuity and uniform stability of v. (holds under general assumptions)

(2) Compact shape of ground states. (in $d \leq 2$ see [AU YEUNG, FRIESECKE, SCHMIDT (2011)])

(3) Compact shape of clusters at low temperature. (open)

(4) Surface-order correction: $kf_k^{\rm cl}(\beta) - kf_\infty^{\rm cl}(\beta) \geq Ck^{1-1/d}$. (open)

Let $H(a;b) = \sum_{k\in\mathbb{N}} (b_k - a_k + a_k \log \frac{a_k}{b_k})$ denote the entropy.

Approximation with ideal mixture

Under Hypotheses (1), (3) and (4), for any sufficiently large β and sufficiently small ρ,

$$0 \leq f(\beta,\rho) - f^{\rm ideal}(\beta,\rho) \leq \frac{C}{\beta} m^{\rm ideal}(\beta,\rho)\rho^{1/(d+1)},$$

and, for any minimiser $\rho = \rho^{(\beta,\rho)} = (\rho_k)_{k\in\mathbb{N}}$ of $f(\beta,\rho,\cdot)$, with $m := \sum_{k\in\mathbb{N}} \rho_k$,

$$\Big| \frac{m}{m^{\rm ideal}(\beta,\rho)} - 1 \Big|^2 \leq C'\rho^{1/(d+1)} \qquad \text{and} \qquad \frac{1}{2} H\Big(\frac{\rho}{m}; \frac{\rho^{\rm ideal}(\beta,\rho)}{m^{\rm ideal}(\beta,\rho)}\Big) \leq C'\rho^{1/(d+1)}.$$

If Hypotheses (3) and (4) are replaced by (2), this holds for $-\beta^{-1}\log\rho > v^* + \varepsilon$ with ε-dependent constants.

Coupled Limit

Idea: Couple inverse temperature $\beta = \beta_N \to \infty$ with particle density $N/L_N^d = \rho_N \to 0$ such that

$$-\frac{1}{\beta_N} \log \frac{N}{L_N^d} = \nu \in (0,\infty) \qquad \text{is constant.}$$

(Example: $\beta_N \asymp \log N$ and $|\Lambda_N| = |[0,L_N]^d = N^\alpha$ with $\alpha > 1$.)

Then energic and entropic forces compete on the same, critical scale, and determine the behaviour of the system.

Large $\nu \Longrightarrow$ entropy wins, i.e., typical inter-particle distance diverges,
Small $\nu \Longrightarrow$ interaction wins, i.e., crystalline structure in the particles emerges.

Free energy per particle in the coupled limit, [CKMS10]

$$-\mu(\nu) = \lim_{N\to\infty} \frac{1}{N\beta_N} \log Z_{[0,L_N]}(\beta_N, N).$$

The proof is a preliminary version of the proof of the above LDP.

This is taken from [JANSEN 2012]. Some further (natural) assumptions on v are made.

Introduce $\mathscr{P}_\theta$, the set of all shift-invariant distributions P of random point configurations $\omega = \sum_{x\in\xi} \delta_x$ with $\xi \subset \mathbb{R}^d$ locally finite. Denote

$$\begin{aligned}
\text{energy:} \quad & \mathscr{U}(P) &=& \frac{1}{2}\int P(\mathrm{d}\omega) \sum_{x\in\xi\cap[0,1]^d} \sum_{y\in\xi} v(|y-x|) \\
\text{entropy:} \quad & \mathscr{S}(P) &=& 1 - \lim_{\Lambda\to\mathbb{R}^d} H_\Lambda(P_\Lambda \mid Q_\Lambda) \\
k\text{-cluster number:} \quad & \rho_k(P) &=& \int P(\mathrm{d}\omega) \sum_{x\in\xi\cap[0,1]^d} 1\!\!1\{|C_\xi(x)| = k\},
\end{aligned}$$

where $C_\xi(x)$ is the cluster of ξ that contains x. By $\rho(P)$ we denote the P-expectation of $|\xi \cap [0,1]^d|$.

Percolation

For any $P \in \mathscr{P}_\theta$,

$$\sum_{k\in\mathbb{N}} k\rho_k(P) < \rho(P) \qquad \Longleftrightarrow \qquad P(\text{there is an infinite cluster}) > 0.$$

Identification of rate function

$$f\big(\beta,\rho,(\rho_k)_{k\in\mathbb{N}}\big) = \min\Big\{\mathscr{U}(P) - \frac{1}{\beta}\mathscr{S}(P)\colon P\in\mathscr{P}_\theta, \rho(P)=\rho, \rho_k(P)=\rho_k \forall k\Big\}.$$

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Gibbs variational principle

The minimizers $(\rho_k)_{k\in\mathbb{N}}$ of $f(\beta,\rho,\cdot)$ correspond with shift-invariant Gibbs measures P (with respect to a suitable chemical potential) satisfying $\rho(P)=\rho$ and $\rho_k(P)=\rho_k$ for all k.

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The following is a continuous version of what is called dependent percolation.

Bounds on (non-)percolation

- For $\nu>\nu^*$ and $\rho<\mathrm{e}^{-\beta\nu}$, for all large β, the Gibbs measure has no infinite cluster, and the cluster size distribution has exponentially decaying tails.
- For ρ large enough (up to a bound that does not depend on β) and all large β, the Gibbs measure has an infinite cluster with probability one.

Crystallisation at Positive Temperature

This is taken from [JANSEN, K., SCHMIDT, THEIL 2013+]: work in progress.
At zero temperature, [THEIL 06] proved crystallisation in $d=2$. That is, in the limit $N\to\infty$, the optimal particle configuration x, the minimiser in

$$e_\infty := \lim_{N\to\infty}\frac{1}{N}\inf_{x\in(\mathbb{R}^d)^N} U_N(x) = \min_{r\in(0,\infty)}\sum_{i\in\mathscr{L}} v(ir) = \sum_{i\in\mathscr{L}} v(i) \in (-\infty,0),$$

approaches the triangular lattice $\mathscr{L}$. (We normalised the potential v.) ($d=3$ is in preparation)

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Goal: Prove an analogous approximate probabilistic assertion for large β under $\mathbb{P}^{(N)}_{\beta,\Lambda}$.
More precisely, prove that, for $\rho \in (0,1)$ and large β, a macroscopic fraction of the N particles forms large grids with fluctuations around the grid sites, vanishing as $\beta \to \infty$.
We first concentrate on $d = 1$ (the two-dimensional case seems different) and approximate with the model that has Hamiltonian

$$U_N^{(W)}(x) = \sum_{i=1}^{N-1} W(|x_{i+1} - x_i|), \qquad \text{where} \quad W(r) = \sum_{i\in\mathbb{N}} v(ir).$$

where $x_1 < x_2 < \cdots < x_N$. This model has large, β-dependent grid-like clusters with large, β-dependent empty intervals inbetween.