Teilprojekt C6: **Fragmentation von Metallclustern**

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Summary of results in the period 1995–1998:

The main result of our research on phase transitions in small systems are:
The demonstration that microcanonical thermodynamics *MThD* is able

1. to define phase transitions of first order in non-extensive (here small) systems unambiguously,
2. to determine all relevant parameters of the transition as the transition temperature T_{tr}, the latent heat q_{lat} and the surface entropy s_{surf} respectively the surface tension σ_{surf},
3. to give insight into the details of the mechanism leading to the phase transition,
4. it is the *only way* to define a critical point of the liquid–gas transition in small systems
5. to approach with higher pressure the critical point,
6. so that it is able to determine the coexistence curve $T(\rho)$ for a small realistic mesoscopic cluster, and to analyse critical fluctuations in isolated small systems

The *MThD* can easily be extended to other non-extensive systems like astrophysical (see the contribution of V. Laliena /Lal99/) or to statistical fragmentation under strong inhomogeneous Coulomb field /BAG99/

Ergebnisse aus den Jahren 1995–1998

Phasenübergänge in Systemen ohne thermodynamischen Limes, vor allem in mesoskopischen Metallclustern:

Hier ist die Verwendung des mikrokanonischen Ensembles bzw. der mikrokanonischen Thermodynamik (*MThD*) entscheidend. Sie ist *die einzige Statistik in der für nicht-extensive (z. B. kleine) Systeme Phasenübergänge definierbar sind* siehe hierzu /ElY98/. Man erhält sehr detaillierten Einblick in die Mechanik des Phasenübergangs (hier und im Folgenden des Flüssigkeits–Gas Überganges). Wir haben mit der von uns entwickelten /Gro97/ mikrokanonischen Metropolis Monte Carlo Simulation *MMMC* die Fragmentation von Na-Clustern mit 200, 1000 und 3000 Atomen bei Drucken von 1 bis 250 Atmosphären untersucht. Das folgende Bild 1 zeigt die mikrokanonische calorische Zustandsgleichung bei 1 Atm. *T(h)* als Funktion der Gesamtenthalpie *h* pro Atom. Die Abbildung zeigt bereits alle wesentlichen Punkte /GKF98/ der mikrokanonischen Thermodynamik:
In der *MThD* wird ein Phasenübergang erster Ordnung in einem endlichen System durch das anomale Absinken der thermodynamischen Temperatur ($T = [\frac{\partial S}{\partial H}|_P]^{-1}$) mit steigender Anregungsenthalpie *h* signalisiert /GMS97/,/MGH97/,SKM97/,/GrM97/,/GrM98/,/Gro98/, /MaG98/. Die „Maxwellgerade“ zu $b(h) = 1/T(h)$ bestimmt:

1. die Übergangstemperatur T_{tr},
2. ihre Länge $h_3 - h_1 = q_{\mathrm{lat}}$ die spezifische latente Wärme des Übergangs
3. und die Fläche unter $b(h)$ zwischen h_1 und h_2 ist der Entropieverlust Δs_{surf} pro Atom, der durch die Ausbildung von Phasentrennflächen in den gemischten Konfigurationen mit der Enthalpie h_2 pro Atom entsteht. Die Oberflächenspannung ist dann gleich $\sigma_{\mathrm{surf}} = T_{\mathrm{tr}}\, N_0\, \Delta s_{\mathrm{surf}}/N_S$. Hier ist N_S gleich der Gesamtzahl der Oberflächenatome in den Fragmenten mit zwei oder mehr Atomen.

Die zwei Einschübe zeigen die Massenverteilungen der Fragmente einen Enthalpieschritt vor (1) dem Minimum und an dem Minimum (2) von *T*(*h*). Wie man sieht, besteht der Phasenübergang aus zwei konkurrierenden Prozessen: Zwischen h_1 und h_2 zerfällt der ursprüngliche Flüssigkeitstropfen durch Abdampfen von Monomeren und kleineren Fragmentclustern (siehe Einschub 1). Am Minimum ist es entropisch von Vorteil, wenn der Ursprungstropfen vollständig in kleinere Cluster multifragmentiert (Einschub 2). Bei noch höheren Anregungsenthalpien wird die Breite der Massenverteilung dieser kleinen Cluster immer monodisperser. Am Ende, bei hohen Enthalpien (*h*>1.2 eV/atom) liegt dann ein ideales einatomiges Gas vor.
Diese Einzelheiten des Phasenüberganges sind nur in der mikrokanonischen Thermodynamik sichtbar. In der konventionellen (kanonischen) Darstellung wird der ganze Phasenübergangsbereich h_1–h_3 in den einzigen Punkt $T = T_{\mathrm{tr}}$ abgebildet.
Das nächste Bild 2 zeigt die Koexistenzkurve T(r) für 200 bis 3000 Na-Atome bei steigenden Drucken bis zum kritischen Punkt Pcr, Tcr im Vergleich mit den experimentellen Bulkwerten

/FiL95/. Die kritische Temperatur Tcr ist deutlich geringer als im Bulk. Die Werte für den Druck und die Dichte am kritischen Punkt von 200 bzw. 1000 Na Atomen sind erstaunlicherweise sehr ähnlich den Bulkwerten (Pcr=253.06 atm. und ρcr=219kg/m3 /FiL95/).

Hier muss besonders betont werden, daß unsere Theorie es das erstemal ermöglicht, den kritischen Punkt in einem kleinen aber realistischen System zu bestimmen. Obendrein handelt es sich hier um ein metallisches System, das wegen der Leitungselektronen nicht von einer reinen Zweikörperkraft zusammengehalten wird. Hier würden übliche Molekulardynamik-rechnungen für 1000 Atome auf ganz erhebliche Schwierigkeiten stoßen.

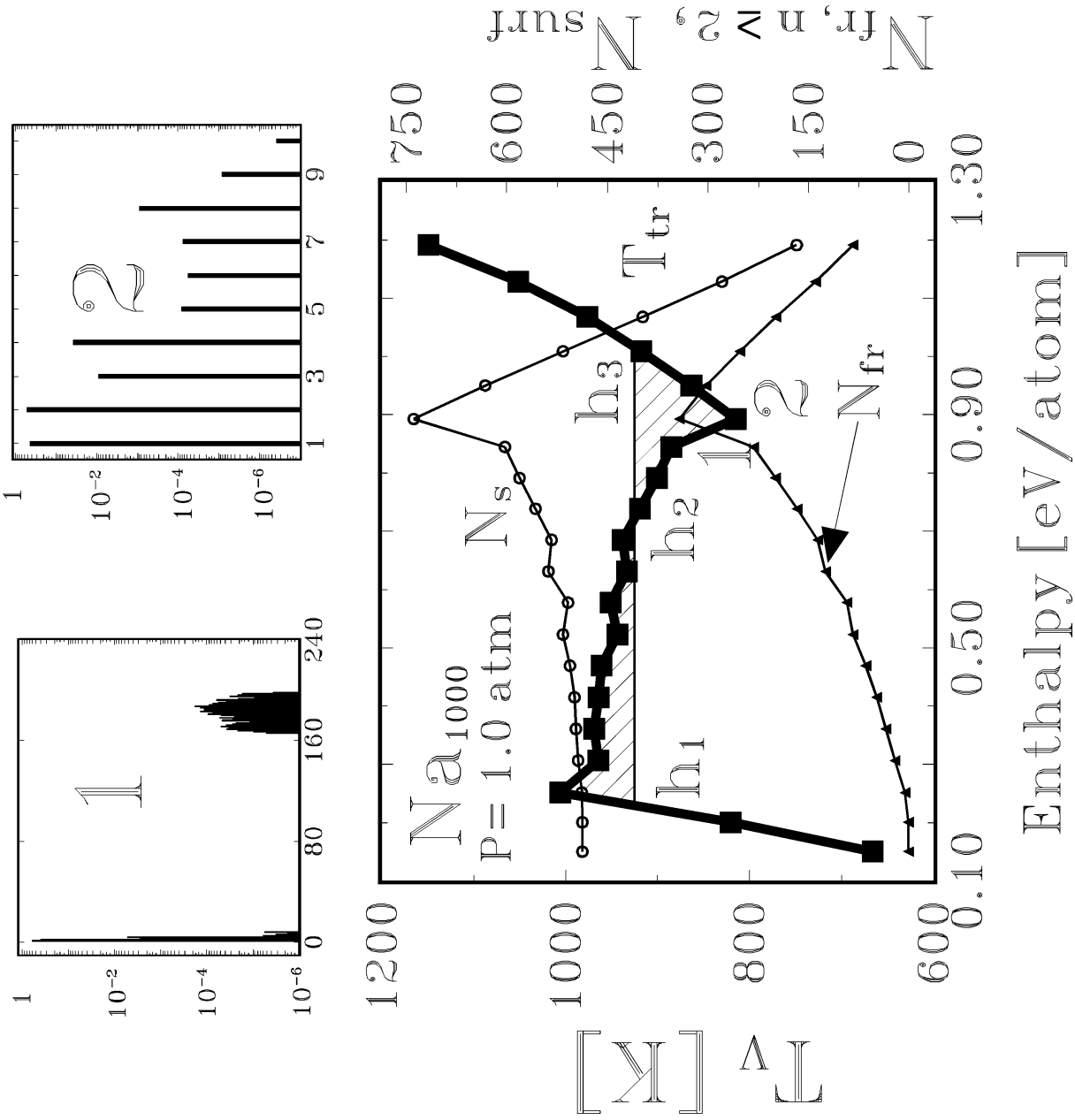

Abbildung 1: Calorische Kurve T(h) (fett), Zahl N_{fr} der Fragmente mit zwei oder mehr Atomen und die Gesamtzahl der Oberflächenatome in den Fragmentclustern (N_S) bei Normaldruck. Die Einsatzbilder geben die Massenverteilungen der Fragmente kurz vor (1) und an dem Minimum (2) von T(h) wieder. Offensichtlich gibt die *MThD* einen detaillierten Einblick in die Natur des Phasenübergangs von der reinen Verdampfung bei kleinen Anregungsenergien über die Multifragmentation bis zur Produktion eines idealen Gases aus reinen Na-Atomen.

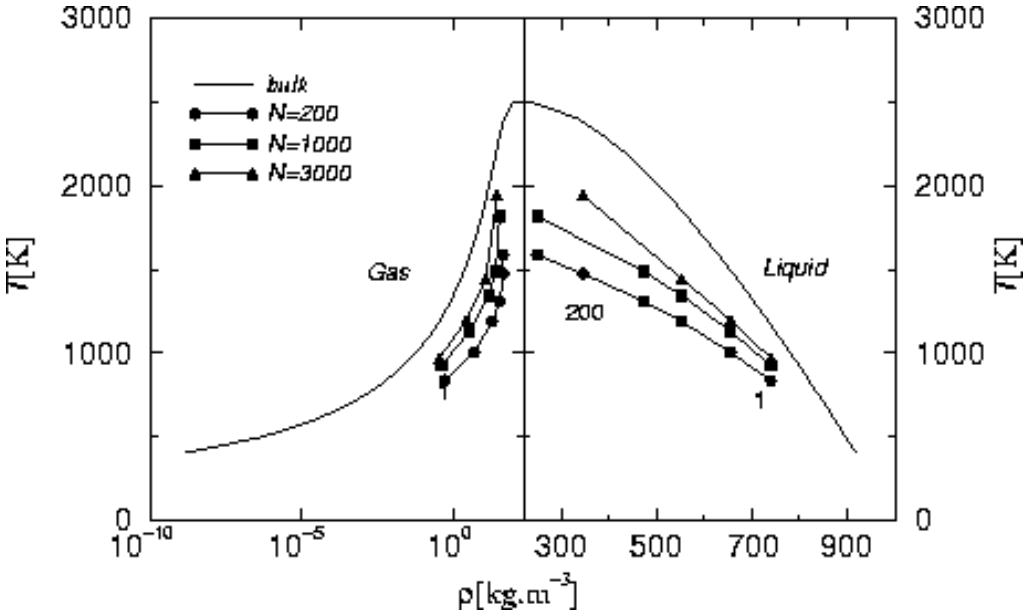

Abbildung 2: Koexistenzkurve $T(\rho)$ für Na für 200 und 1000 Atome bei wachsendem Druck von (1, 10, 50, 100, 200 und 250 atm.) im Vergleich zum Bulk.

Microcanonical Thermodynamics in systems with long range interactions and conserved angular momentum, by V. Laliena:

Conventional thermodynamics apply to systems with interactions sufficiently short-ranged and repulsively enough at short distances. The precise mathematical statement of this features for a two body potential is known as the *thermodynamical stability condition*. It has been rigorously proven that thermodynamical quantities computed from the principles of statistical mechanics have the required properties, like extensivity and convexity of the thermodynamical potential, in the thermodynamical limit. Moreover, in this limit all the statistical ensembles are equivalent in pure phases. The same is not true if the potential is unstable. In this case, the energy is not an extensive quantity and the usual thermodynamical limit cannot be taken. For finite systems, the statistical description is still possible, but different ensembles will give different results corresponding to the behaviour of the system under different physical situations /Gro97/.

Potentials of long range, like the Coulomb (when not screened) or newtonian gravitational ones, are unstable. In the second case, when, in addition, the forces are attractive, it is typical the appearance of a phase transition separating a low energy collapsing phase (CP), with a macroscopic part of the particles bound in a big cluster and the remaining forming a surrounding atmosphere, and a high energy homogeneous phase (HP), in which the system is

homogeneous [2,3]. These two regimes are separated by a phase transition where the microcanonical specific heat is negative, and the microcanonical and the canonical ensembles are not equivalent. It has been argued that self-gravitating systems must be described within the micro ensemble [1]. Conserved quantities, other than the energy, add further constraints to the microcanonical ensemble. In collapsing systems, angular momentum is relevant because it can notably affect the properties of the low energy phase.

The aim of our work is to understand the statistical mechanics of systems with thermodynamically unstable interactions, and, in particular, the effect of angular momentum in the collapsing phase transition. Details of this work can be found in ref. /Lal99/. We started studying the phase space microcanonical distribution function (PSMDF), which is uniform in the phase space shell defined by the additive conserved quantities. The PSMDF is not suitable neither for Monte Carlo nor for mean field computations, since it involves counting the number of states on a phase space shell, which is usually a very difficult task. To overcome this problem, we integrated out the momenta using Laplace and Fourier transform techniques, finding a microcanonical distribution function in configuration space (CSMDF). The CSMDF is not singular, unlike the PSMDF, which contains Dirac deltas, and can be used for Monte Carlo simulations, as well as for mean field computations.

Mean field techniques start introducing the particle density $r(\mathbf{r})$, and considering a large number of particles, *N*. In this case, we have shown /Lal99/ that the CSMDF can be written as the continuum limit of a functional integral. The only contribution to this functional integral comes from its saddle point if we take the $N\to\infty$ limit before the continuum limit. Since, as mentioned above, the thermodynamical limit does not exist, the $N\to\infty$ limit must be properly defined, sending the masses and coupling constant to zero as described in /Lal99/. Therefore, it is a hydrodynamical limit rather than a thermodynamical one. In this way, mean field equations arise naturally.

EXAMPLE: THE THIRRING MODEL.

Long ago Thirring proposed a very simple model for a star [1]. In spite of its extreme simplicity, it mimics the main features of self-gravitating systems with surprisingly good success [2]. The model can be described as follows: a set of N particles are confined in a spherical volume V. Inside this volume there is a spherical interaction region (core) V_0, concentric to V. Particles outside the core ("atmosphere") do not interact, and two particles inside the core have a constant attractive potential energy. The total potential energy is given by $\Phi = -(GM^2/2)a^2$, where $a = \int_{V_0} d^3r\, r(\mathbf{r})$ is the fraction of particles inside V_0; α is the order parameter for the collapsing transition. We solved the thermodynamics of this model with angular momentum. To simplify the computations, we worked in two dimensions. We introduced the dimensionless variables $k = V/V_0$, $e = E/(GM^2)$ and $\Omega = L^2/(2GM^3V^{2/3})$.

The behaviour of the ground state, which we defined as the macroscopic state with zero temperature and given angular momentum, is displayed in fig. 3. Its structure depends on Ω and the parameter κ.

For the discussion of the model at finite temperatures we consider $\kappa<1/2$ ("atmosphere" bigger than the core). The results displayed in figures 4 and 5 are for $k = 1/(e^3 - 1) \approx 0.0524$. There are three different phases, which can be distinguished by the behaviour of the order parameter α (see fig. 5a). Figs. 4a and 4b display the temperature and the entropy for values of Ω which are representative of each phase. The features of each phase are (a) $\Omega < \Omega_1$: complete collapse at low energies, with $\alpha \approx 1$. The HP at high energies is separated from the CP by an interval of energies with negative specific heat. The entropy shows a convex intruder in the energy interval where the two phases coexist. (b) $\Omega_1 < \Omega < \Omega_2$: collapse not complete at low energies with $\alpha < 1$. The entropy has a convex intruder. (c) $\Omega > \Omega_2$. There is no collapse at low energies. The entropy has no convex intruder.

From this analysis, the correlation between the collapsing phase transition and the anomalies in the caloric curve (T versus ε) and in the entropy become evident. The phase diagram in the plane (ε,Ω) is displayed in fig. 5b. There is a forbidden region, where the system cannot be since a minimal rotational energy is required to keep angular momentum constant. The boundary between the forbidden and allowed regions is the $T = 0$ isotherm. Inside the allowed region, there are two phases separated by a transition region. The transition region shrinks as Ω is increased, and disappears at the point $\Omega_c = \Omega_2$ on the zero temperature line.

The model considered here is unrealistic mainly due to the fact that the particles can only collapse in a fixed region. Therefore, there is no place here for more complicated regimes like multifragmentation. As discussed in /Lal99/, we expect a richer phase diagram in more realistic systems.

NUMERICAL COMPUTATIONS.

The CSMDF can also be studied by means of numerical simulations. A Monte Carlo algorithm, suitable for long range potentials, must be adapted to this problem. The most promising tool is the Hybrid Monte Carlo algorithm. With it, one can study system of particles with more realistic interactions. Work in this direction is in progress.

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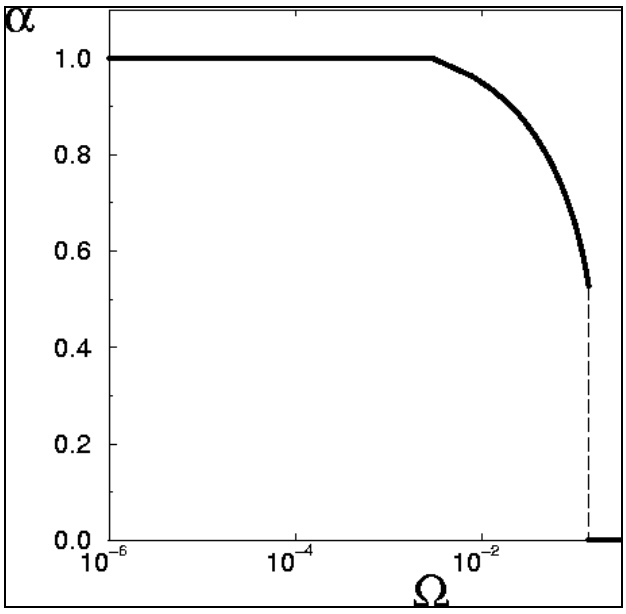
a)



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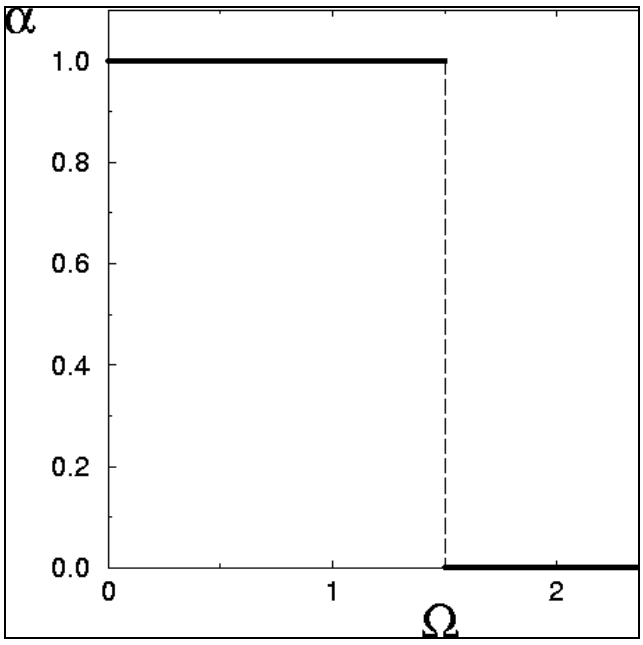
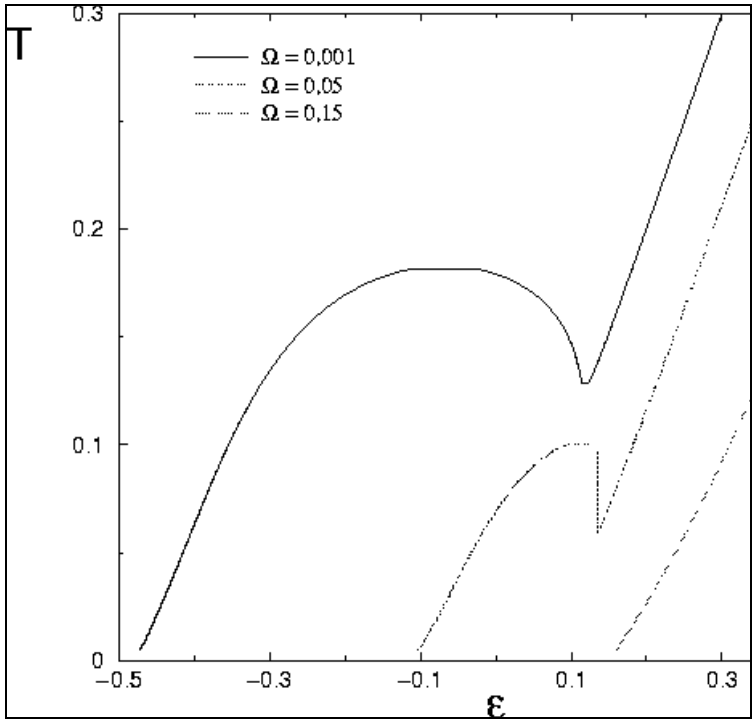


Figure 3: The structure of the ground state for a) $\kappa<1/2$ and b) $\kappa>1/2$.

a)



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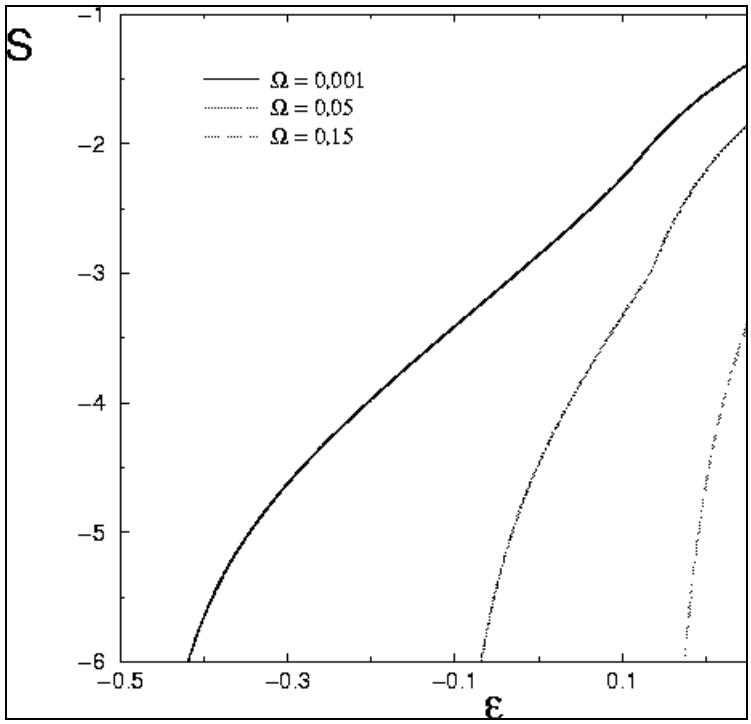
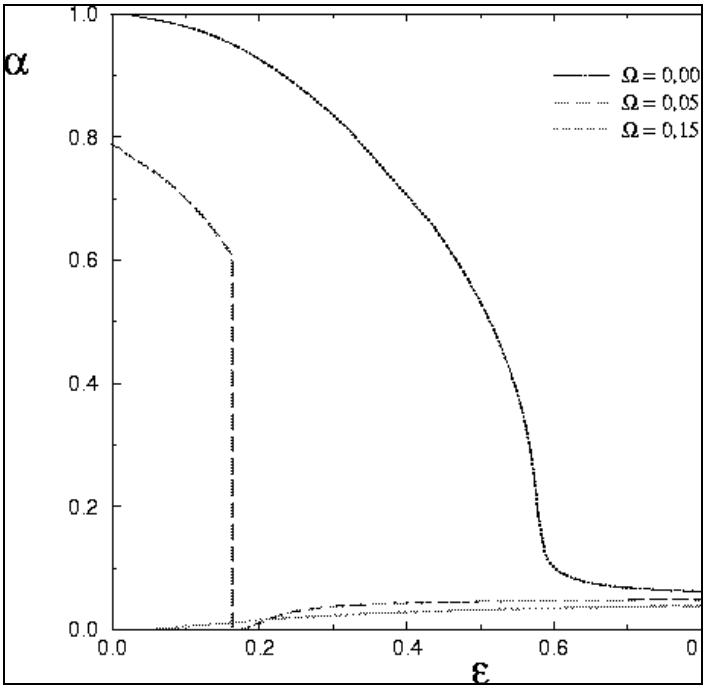


Figure 4: The microcanonical temperature T (a) and the entropy S; (b) versus the energy for three values of Ω, representative for each phase.

a)



b)

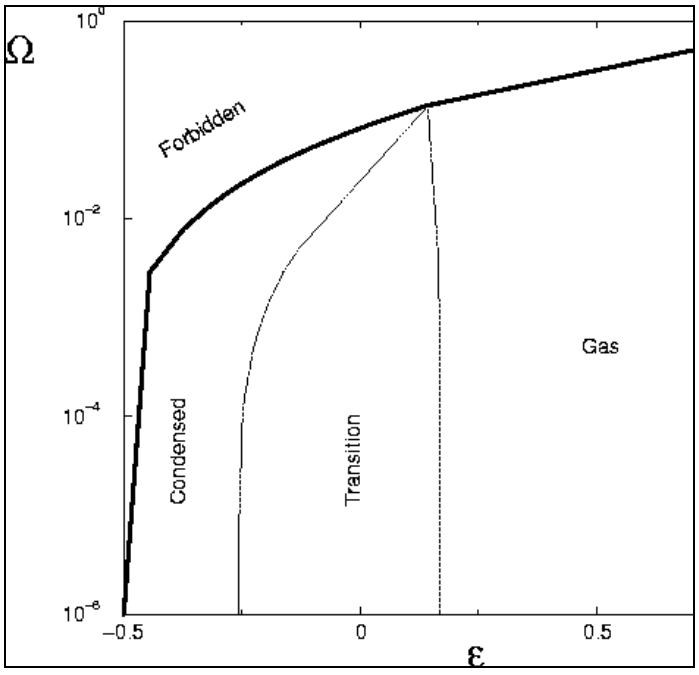


Figure 5: a) The order parameter α versus the energy for Ω on each of the three phases. b) The phase diagram of the model.

Material zum Teilprojekt C6

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