6. A.F. Volkov, M. Kogan: Sov.Phys.Usp. 11, 881 (1969)
7. V. de Giorgio, M.O. Scully: Phys.Rev. A 2, 1170 (1970)
8. C. Shannon, W. Weaver: The Mathematical Theory of Communication (University of Illinois Press, Urbana 1949)
9. E.T. Jaynes: Phys.Rev. 106, 620 (1957)
10. H. Mori: Phys.Rev. 115, 298 (1959)
11. J.A. McLennan: Advances in Chemical Physics (Ed.: I. Prigogine) (Interscience, New York 1963)
12. D.N. Zubarev: Sov.Phys.Dokl. 10, 526 (1965)
13. F. Schlögl: Z.Physik 253, 147 (1972)
14. H. Busch: Annalen d. Physik 64, 401 (1921)
15. B. Ross, J. Litster: Phys.Rev. A 15, 1246 (1977)
16. D. Bedeaux, P. Mazur, R.A. Pasmanter: Physica 86A, 355 (1977)
17. H. Haken: Synergetics, an Introduction (Springer, Berlin Heidelberg New York 1977)

On the Kinetics of Nucleation in Isochoric Gases

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1. Introduction

The process of phase separation in gases has been studied in terms of several different theoretical models (for review see refs. /1-3/). The classical nucleation theory /4-7/ is based on the droplet model, corresponding to finite and discrete fluctuations. Other approaches use theoretical methods with a continous description of the density fluctuations /8-13/. The classical nucleation theory /4-7/ is based on the assumption, that the density of free particles is constant and that the number of all particles in the system is not limited.

These assumptions correspond to a nucleation process in an infinite system. In the present work the problem will be considered how the finite system size will modify the nucleation process. Basing on the droplet model we propose the following model:
We consider a gas of N particles in a closed system with a constant volume V at a temperature T; it means:

$$N = \text{const.};\ V = \text{const.};\ T = \text{const.} \qquad (1.1)$$

Due to interactions between the particles (e.g. chemical reactions, collisions) the N particles will bond in clusters and a distribution of N_1 monomers (free particles), N_2 dimers (bound states of 2 particles), ..., N_N N-mers develops. This distribution will be described by

$$\underline{N} = \{N_1\ N_2 \ldots N_l \ldots N_N\} . \qquad (1.2)$$

Because of (1.1), the relation (1.3) holds:

$$N = \sum_{l=1}^{N} l\, N_l = \text{const.} \qquad (1.3)$$

l is the number of particles in the cluster; the number of clusters consisting of l particles is denoted by N_l.

From (1.3) it follows further that the maximal number of N_l is given by

$$0 \leqslant N_l \leqslant \frac{N}{l} \quad (l=1,\ldots,N) . \qquad (1.4)$$

The clusters are assumed to be an ideal mixture.

2. Equilibrium Distribution

With respect to the constraints (1.1) the equilibrium distribution of clusters is given by the canonical ensemble. We denote the probability distribution for the N particles in the phase space as $w(q_1 \ldots q_N, p_1 \ldots p_N)$, where q_l are the space coordinates and p_l are the momentum coordinates of the particles with the number $l = 1, \ldots, N$. In thermodynamic equilibrium the following relation is valid /14/:

$$w^o(q_1 \ldots p_N) = \frac{1}{Z} \exp\left\{-\frac{1}{k_B T} H(q_1 \ldots p_N)\right\} \quad . \tag{2.1}$$

$H(q_1 \ldots p_N)$ denotes the Hamiltonian of the N particle system, Z is the canonical partition function of the N particles in an atomic picture:

$$Z = \exp\left\{-\frac{1}{k_B T} F(T,V,N)\right\} \quad . \tag{2.2}$$

F(T,V,N) is the free energy of the N particles in the volume V with full interactions.

We consider now a certain configuration $C(\underline{N}) = C(N_1 \ldots N_N)$ of the N particles that corresponds to the assumed cluster distribution (1.2). We define the probability of the equilibrium distribution for this configuration by an integral of $w(q_1 \ldots p_N)$ over the subspace $C(\underline{N})$:

$$P^o(\underline{N}) = \int\limits_{C(\underline{N})} w^o(q_1 \ldots p_N) dq_1 \ldots dp_N \quad . \tag{2.3}$$

With (2.1) and (2.2) we get

$$P^o(\underline{N}) = \exp\left\{\frac{1}{k_B T} F(T,V,N)\right\} \int\limits_{C(\underline{N})} \exp\left\{-\frac{1}{k_B T} H(q_1 \ldots p_N)\right\} dq_1 \ldots dp_N \quad . \tag{2.4}$$

The partition function of the assumed cluster distribution (1.2) can be calculated by /15/:

$$Z(T,V,\underline{N}) = \int\limits_{C(\underline{N})} \exp\left\{-\frac{1}{k_B T} H(q_1 \ldots p_N)\right\} dq_1 \ldots dp_N \quad . \tag{2.5}$$

For a configuration $C(\underline{N})$ which in section 3 will be assumed as an ideal mixture of the clusters with the constraint (1.3) the free energy is introduced as follows:

$$F(T,V,\underline{N}) = -k_B T \ln Z(T,V,\underline{N}) \quad . \tag{2.6}$$

(Note, that $F(T,V,\underline{N})$ (2.6) and F(T,V,N) (2.2) are different functions.)

In this way we get the correct probability $P^o(\underline{N})$ of the equilibrium distribution:

$$P^o(N_1 N_2 \ldots N_N) = \exp\left\{\frac{1}{k_B T}[F(T,V,N) - F(T,V,N_1 N_2 \ldots N_N)]\right\} \quad . \tag{2.7}$$

3. The Free Energy

The free energy (2.6) of the ideal mixture of clusters with the distribution (1.2) is determined by the partition function (2.5). With the correct normalization it results:

$$Z(T,V,\underline{N}) = \prod_l \frac{1}{h^{3N_l} N_l!} \int \exp\left\{-\frac{1}{k_B T} H_l\right\} dq_1^{N_1} \ldots dp_N^{N_N} \quad . \tag{3.1}$$

H_l is the Hamiltonian of the cluster with the particle number l and the mass

$$M_l = \frac{M}{N_A} \; l \; ,$$

(M - molar mass, N_A - Avogadro-constant)

$$H_l = \frac{1}{2M_l}(p_{1l}^2 + p_{2l}^2 + p_{3l}^2) + f_l \; , \tag{3.2}$$

f_l denotes a potential contribution which depends only on the size l of the cluster.

Integration of (3.1) and use of (2.6) leads to the following expression for the free energy:

$$F(T,V,\underline{N}) = \sum_{l=1}^{N} N_l \left\{ f_l + k_B T \left(\ln \frac{N_l}{V} \lambda_l^3 - 1\right)\right\} \; , \tag{3.3}$$

with

$$\lambda_l = h \left[2\pi M_l k_B T\right]^{-1/2} \quad . \tag{3.4}$$

To determine the potential term f_l we choose a first approximation similar to the theory of atomic nuclei which includes only volume and surface effects:

$$f_l(T) = -Al + Bl^{2/3} \quad . \tag{3.5}$$

The second term of eq. (3.5) is proportional to the surface area and the surface tension σ. It follows

$$B = 4\pi \left(\frac{4\pi}{3} c N_A\right)^{-2/3} \sigma \; , \tag{3.6}$$

where c [mol/m^3] is the molar density in the cluster. The boundary conditions for the potential part are given by

$$f_1 = 0 \; ,$$
$$f_2 = \varphi_2 = \text{two-particle binding energy} \quad . \tag{3.7}$$

The free energy F = G-pV given by (3.3) includes the contribution of the partial pressures

$$-pV = -\sum_{l=1}^{N} p_l V = -\sum_{l=1}^{N} N_l k_B T \tag{3.8}$$

and the contribution of the Gibbs potential

$$G = \sum_{l=1}^{N} N_l \mu_l \tag{3.9}$$

with the chemical potential of a cluster with size l

$$\mu_l = f_l(T) + k_B T \ln \frac{N_l}{V} \lambda_l^3 . \qquad (3.10)$$

4. The Master Equation

To discuss the time evolution of the cluster size distribution we suppose the following assumptions:

(i) The growth and shrink of a cluster is due only to an addition or evaporation of monomers (Fig. 1). In terms of chemical kinetics this process can be represented by:

$$A_l + A_1 \underset{w^-}{\overset{w^+}{\rightleftharpoons}} A_{l+1} , \qquad (4.1)$$

where w^+ and w^- are the probabilities per unit time of the reaction in the given directions.

(ii) Interactions between the clusters with $l \geqslant 2$, like coagulation processes or collisions between two or more clusters are not taken into account. The probabilities of these events should be negligible in comparison with the probabilities of the reaction (4.1).

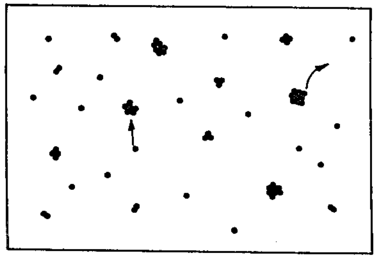

Fig. 1 Typical cluster distribution in a finite volume V and the elementary processes of attachment and evaporation of monomers

The interactions between the clusters are taken into account via their influence on the pressure in the system. This pressure is given as a sum of the partial pressures of the different kinds of clusters (3.8). We define the probability

$$P(N_1 N_2 \ldots N_N, t) = P(\underline{N}, t) \qquad (4.2)$$

that at a given time t there exist N_1 monomers, N_2 dimers ... etc. in the system. Then the time evolution of the cluster distribution (1.2) corresponds to the change of $P(\underline{N},t)$ with time. This evolution can be described by a Master equation

$$\frac{\partial P(\underline{N},t)}{\partial t} = \sum_{\underline{N}'} \{ w(\underline{N}|\underline{N}')P(\underline{N}',t) - w(\underline{N}'|\underline{N})P(\underline{N},t) \} . \qquad (4.3)$$

$\underline{N}'$ specifies those cluster distribution which are attainable from the assumed distribution $\underline{N}$ via the reaction (4.1). The quantities $w(\underline{N}|\underline{N}')$ are the transition probabilities per time unit for the transition from $\underline{N}'$ to $\underline{N}$.

Equation (4.3) allows us to introduce a probability flux

$$J(\underline{N}|\underline{N}') = w(\underline{N}|\underline{N}')P(\underline{N}',t) - w(\underline{N}'|\underline{N})P(\underline{N};t) . \qquad (4.4)$$

Substitution of (4.4) into (4.3) results in

$$\frac{\partial P(\underline{N},t)}{\partial t} = \sum_{\underline{N}'} J(\underline{N}|\underline{N}') . \qquad (4.5)$$

The stationary solution of (4.5) implies that

$$\sum_{\underline{N}'} J(\underline{N}|\underline{N}') = 0 . \qquad (4.6)$$

This is the so-called KIRCHHOFF solution /16/. With the condition of detailed balance follows a more restrictive form of (4.6):

$$J(\underline{N}|\underline{N}') = 0. \qquad (4.7)$$

This consistency condition (4.7) means:

$$w(\underline{N}|\underline{N}')P^0(\underline{N}') = w(\underline{N}'|\underline{N})P^0(\underline{N}) . \qquad (4.8)$$

In this equation $P^0(\underline{N})$ is the probability of the equilibrium distribution (2.7). To construct the transition probabilities my means of (4.8) we need one kinetic assumption with respect to (4.1). In agreement with earlier investigations /17/ we assume that the probability of an attachement of a monomer to a cluster of the size l increases with the surface of the cluster and with the density of clusters of size l and the density of monomers. In this way we get for the special process of attachment: $l \geqslant 2$

$$\begin{aligned} &W(N_1-1 \ldots N_l-1 N_{l+1}+1 \ldots N_N | N_1 \ldots N_l N_{l+1} \ldots N_N) \\ &\equiv w_l^+(N_1 N_l) = \alpha\, l^{2/3} N_l \frac{N_1}{V} . \end{aligned} \qquad (4.9)$$

We would like to underline that the probabilities of the growth process of clusters of different sizes l=1...N are correlated since the number of particles is conserved (1.3).

Note, that for l=1 the number of monomers changes from N_1 to N_1-2 by creating a dimer. The transition probability in this case reads:

$$w_1^+(N_1-2\ N_2+1\ N_3 \ldots N_N | N_1 N_2 N_3 \ldots N_N) \equiv w_1^+(N_1) = \alpha_1 \frac{N_1(N_1-1)}{V} . \qquad (4.10)$$

From (4.8) with (2.7) and (4.9) we get the transition probability for the oppositve process of (4.9) (evaporation) /18/:

$$\begin{aligned} &w(N_1 \ldots N_l N_{l+1} \ldots N_N | N_1-1 \ldots N_l-1 N_{l+1}+1 \ldots N_N) \\ &\cong w_{l+1}^-(N_{l+1}+1) = \alpha\, l^{2/3} N_l \frac{N_1}{V} \exp\Big\{ \frac{1}{k_B T} \\ &* \big[F(T,V,N_1-1 \ldots N_l-1\ N_{l+1}+1 \ldots N_N) - F(T,V,N_1 \ldots N_l\ N_{l+1} \ldots N_N)\big]\Big\} . \end{aligned} \qquad (4.11)$$

If we neglect in (4.11) a small term $(l/l+1)^{13/6}$, which is nearly equal to one, we get for the transition probability for the evaporation process $N_1 \ldots N_l \ldots N_N \longrightarrow N_1+1 \ldots N_{l-1}+1\ N_l-1 \ldots N_N$:

$$w_l^-(N_l) = \alpha\, l^{2/3} N_l \frac{1}{\lambda_1^3} \exp \frac{1}{k_B T}(f_l - f_{l-1}) , \qquad (4.12)$$

if $l = 2,\ \alpha \rightarrow \alpha_1$.

The transition probability of the evaporation of a monomer is not determined by the whole cluster distribution as in (4.9). Again, this transition probability is modified for the evaporation of a dimer, because the transition changes N_1, N_2 to (N_1+2), (N_2-1). We underline that in our model the transition probabilities for all processes different from (4.9) and (4.12) are assumed to be zero.

With the introduced transition probabilities the master equation (4.3) reads in an explicit form with $W_N = W_{N+1} = 0$:

$$\begin{aligned}\dot{P}(\underline{N},t) &= w_2^-(N_2+1)P(N_1-2N_2+1\ldots N_N,t)-w_1^+(N_1)P(\underline{N},t)+w_3^-(N_3+1)\\ &\cdot P(N_1-1,N_2-1N_3+1\ldots N_N,t)+w_1^+(N_1+2)\,P(N_1+2N_2-1\ldots N_N,t)\\ &-[w_2^+(N_1N_1)-w_2^-(N_2)]\,P(\underline{N},t)+\sum_{l=3}^{N} w_{l+1}^-(N_{l+1}+1)P(N_1-1\ldots N_{l-1}-1N_{l+1}+1\ldots N_N,t)\\ &+w_{l-1}^+(N_1+1N_{l-1}+1)P(N_1+1\ldots N_{l-1}+1N_l-1\ldots N_N,t)\\ &-[w_l^+(N_1N_l)+w_l^-(N_l)]\,P(\underline{N},t)\ . \end{aligned} \tag{4.13}$$

5. Discussion

(i) First we investigate the stationary probability distribution $P^0(N_1N_2\ldots N_l\ldots N_N)$ given by (2.7). The extremal probability we find from $\partial P^0/\partial N_l=0$. It yields the equilibrium distribution of clusters:

$$\frac{N_l^0\lambda_l^3}{V} \approx \exp\left\{l\ \ln\frac{N_1^0\lambda_1^3}{V} - \frac{f_l}{k_BT}\right\}\ , \qquad l=2,\ldots,N \tag{5.1}$$

where $N_1^0 = N - \sum\limits_{l=2}^{N} l\ N_l^0$.

Due to this boundary condition the solution of (5.1) is complicated. Inside the binodal region we generally expect multiple solutions in the $(N_1\ldots N_N)$ space. At least the system is a bistable one where two stable cluster distributions can exist separated by regions of a minimal probability. Figure 2 demonstrates that we find one regime with a large number of monomers and a second regime with a stable configuration of large clusters in coexistence with a small number of monomers.

The question which cluster distribution is realized depends on the absolute value of the probability distribution in the $(N_1\ldots N_N)$ space, which is determined by the thermodynamic parameters N,V,T. The problem of transitions between the two stable states of the cluster distribution can be turned out with the concept of the mean first passage time /19/ which allows the calculation of the mean transition time from the monomer phase to the cluster phase.

It can be estimated by the consideration of the eigenvalue problem of (2.7), by finding the smallest eigenvalue χ_1 which is different from zero. In bistable systems χ_1 is separated from the spectrum of the other eigenvalues $\chi_i(i=2,3,\ldots)$ /20/.

(ii) Let us derive the equations of the mean values /21/. The mean cluster number with size k

$$\langle N_k(t)\rangle = \sum_{\{N_i\}} N_k\ P(N_1\ldots N_N,t)$$

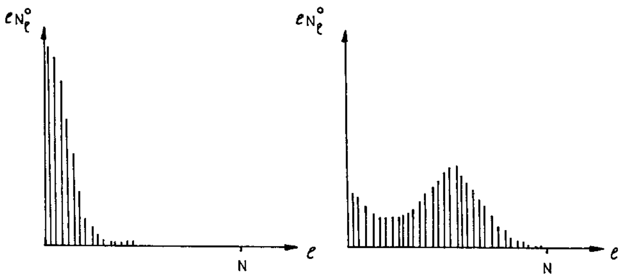

Fig. 2 Sketch of the equilibrium cluster distributions:
a) corresponds to a monomer phase, in b) the cluster phase is dominating

obeys the system of equations

$$\frac{\partial}{\partial t}\langle N_l\rangle = \left\langle\frac{\partial}{\partial l}(w_{l+1}^-(N_{l+1}) - w_l^+(N_lN_k))\right\rangle\ , \tag{5.2}$$

$$\frac{\partial}{\partial t}\langle N_1\rangle = -\left\langle w_1^+(N_1) - w_2^-(N_2)+\sum_{l=1}^{N-1}(w_l^+(N_1N_l)-w_{l+1}^-(N_{l+1}))\right\rangle . \tag{5.3}$$

The derivative in (5.2), which is a discrete one, expresses the conversion of the cluster number during the growth and shrinkage of clusters (l > 2). The mean cluster number changes only due to the formation or evaporation of dimers

$$\frac{\partial}{\partial t}\sum_{l=2}^{N}\langle N_l\rangle = \langle w_1^+(N_1) - w_2^-(N_2)\rangle\ . \tag{5.4}$$

The alteration of the actual pressure is defined by the equation for the mean numbers of clusters and monomers:

$$\dot{P}(t) = \frac{k_BT}{V}\frac{\partial}{\partial t}\sum_{l=1}^{N}\langle N_l\rangle = -\frac{k_BT}{V}\sum_{l=1}^{N-1}\langle(w_l^+(N_1N_l)-w_{l+1}^-(N_{l+1}))\rangle\ . \tag{5.5}$$

Obviously it follows for the whole number of particles in the considered case:

$$\frac{\partial}{\partial t}\left\{\langle N_1\rangle + \sum_{l=2}^{N}\langle l\ N_l\rangle\right\} = 0\ . \tag{5.6}$$

(iii) Treating N_l as a continuous function of l, we make use of a Kramers-Moyal expansion /22/ in (5.2). Approximating $\langle N_1\ N_l\rangle \approx \langle N_1\rangle\langle N_l\rangle$ we obtain a Fokker-Planck equation for the mean cluster distribution by neglecting higher than the second derivative:

$$\begin{aligned}\frac{\partial}{\partial t}\langle N_l\rangle &= -\frac{\partial}{\partial l}\left\{\langle N_l\rangle \propto l^{2/3}\left\{\frac{\langle N_1\rangle}{V} - \frac{1}{\lambda_1^3}\exp\left(\frac{1}{k_BT}\frac{\partial f_l}{\partial l}\right)\right\}\right\}\\ &+\frac{1}{2}\frac{\partial^2}{\partial l^2}\left\{\langle N_l\rangle \propto l^{2/3}\left\{\frac{\langle N_1\rangle}{V} + \frac{1}{\lambda_1^3}\exp\left(\frac{1}{k_BT}\frac{\partial f_l}{\partial l}\right)\right\}\right\} + \ldots \qquad l \geqslant 2\end{aligned} \tag{5.7}$$

If we further neglect the diffusion due to fluctuations we derive the deterministic case (Liouville equation)

$$\frac{\partial}{\partial t}\langle N_l\rangle = -\frac{\partial}{\partial l}\,\alpha\, l^{2/3}\langle N_l\rangle\left[\frac{\langle N_l\rangle}{V} - \frac{1}{\lambda_1^3}\exp\left(\frac{1}{k_BT}\,\frac{\partial f_l}{\partial l}\right)\right]. \qquad l \geqslant 2 \quad (5.8)$$

In both equations (5.7) and (5.8) the monomer distribution results from the boundary conditions (5.6).

Equation (5.8) is a balance equation. Therefore we find the deterministic kinetics

$$\dot{l} = \alpha\, l^{2/3}\left[\frac{\langle N_l\rangle}{V} - \frac{1}{\lambda_1^3}\exp\left(\frac{1}{k_BT}\,\frac{\partial f_l}{\partial l}\right)\right]. \qquad l \geqslant 2 \qquad (5.9)$$

By introduction of the cluster radius $r_l(t)$ and by a linear expansion of the exponential function in (5.9), results for a single cluster the known kinetic equation /23/:

$$\dot{r}_l(t) = \frac{\alpha}{\lambda_1^3}\,\frac{d_o}{\left(\frac{4\pi}{3}cN_A\right)^{1/3}}\left[\frac{1}{r_c(t)} - \frac{1}{r_l(t)}\right], \qquad (5.10)$$

where $r_c(t)$ is the time-dependent critical radius

$$r_c(t) = d_o\left(\ln\frac{\langle N_l\rangle}{V}\lambda_1^3 + \frac{A}{kT}\right)^{-1}, \qquad (5.11)$$

d_o being the capillarity length: $d_o = 2\sigma\,(c\,N_A k_B T)^{-1}$. The density of the free monomers $\langle N_1\rangle/V$ acts as an effective supersaturation.

(iv) The master equation (4.13) can be reduced to a kinetic equation for single clusters as well. If $P_{N_1+1}(l,t)$ is the probability that a l-mer is present in the environment of N_1 free monomers at time t, we propose the following ansatz:

$$\dot{P}(N_1 \ldots N_N, t) = \frac{G_N(t)}{N_2! \ldots N_N!}\prod_{l=2}^{N}\left(P_{N_1+1}(l,t)\right)^{N_l}\;;\quad N_1 = N - \sum_{l=2}^{N} l\, N_l\,. \qquad (5.12)$$

The index N_1+1 of $P_{N_1+1}(l,t)$ labels the number of free monomers at the very beginning of forming the present droplet. It is an invariant during the droplet growth. It yields

$$\begin{aligned}\dot{P}_{N_1+1}(l,t) = \alpha\Big\{&(l-1)^{2/3}\,\frac{N_1+1}{V}\,P_{(N_1+1)+(l-1)}(l-1,t)\\ &+ \frac{1}{\lambda_1^3}(l+1)^{2/3}\exp\left(\frac{f_{l+1}-f_l}{k_BT}\right)P_{(N_1-1)+(l+1)}(l+1,t)\Big\}\\ &- \alpha\, l^{2/3}\left\{\frac{N_1}{V} + \frac{1}{\lambda_1^3}\exp\left(\frac{f_l-f_{l+1}}{k_BT}\right)\right\}P_{N_1+1}(l,t)\,, \qquad (5.13)\end{aligned}$$

where

$$\dot{G}_N(t) = \alpha_1 G_N(t)\left[2^{2/3}\,\frac{1}{\lambda_1^3}\exp\left(\frac{f_2}{k_BT}\right)P_{(N_1-2)+2} - \frac{N_1(N_1-1)}{V}\right]. \qquad (5.14)$$

Equation (5.14) is the balance equation for the probability of the free monomers and realizes certain physical assumptions. In the considered case it implies the solution of the equations (5.13) and (5.14) under the constraints (1.3). Another possibility where only one single cluster performs the nucleation process is shown in ref. /17/. If we assume N_1 = const. it follows from (5.13) the classical Becker-Döring theory /5/.

References

1. A.C. Zettlemoyer (Ed.): Nucleation (Dekker, New York 1969); Nucleation Phenomena (Wiley, New York 1977)
2. F.F. Abraham: Homogeneous Nucleation Theory (Academic Press, New York 1974)
3. J.D. Gunton, M. San Miguel, P.S. Sahni, in: C. Domb (Ed.) Phase Transitions and Critical Phenomena (Academic Press, New York 1983)
4. M. Volmer: Kinetik der Phasenbildung (Steinkopf, Dresden 1939)
5. R. Becker, W. Döring: Ann.Phys. 24, 719 (1935)
6. J.L. Frenkel: Kinetic Theory of Liquids (Dover, New York 1955)
7. J.B. Zeldovich: J.Exp.Teor.Phys. UdSSR 12, 525 (1942)
8. J.W. Cahn, J.E. Hilliard: J.Chem.Phys. 28, 258 (1958); 30, 1121 (1959); 31, 688 (1959)
9. J.S. Langer: Kinetics of metastable states, in: Lecture Notes in Physics 132 (Springer, Berlin Heidelberg New York 1980)
10. J.S. Langer, A.J. Schwartz: Phys.Rev. A21, 948 (1980)
11. K. Binder, D. Stauffer: Adv. Phys. 25, 343 (1976)
12. K. Binder: Spinodal decomposition, in: Lecture Notes in Physics 132 (Springer, Berlin Heidelberg New York 1980)
13. A.S. Pataschinsky, B.I. Shumilo: JETP 77, 1417 (1979); FTT 22, 2609 (1980)
14. R. Becker: Theorie der Wärme (Springer, Berlin Heidelberg New York 1961)
15. W. Ebeling, D. Kremp, W.D. Kraeft: Theory of bound states (Akademie-Verlag, Berlin 1976; Isdatelstwo Mir, Moscow 1979 (in russ.))
16. H. Haken: Synergetics - An Introduction (Springer, Berlin Heidelberg New York 1978)
17. W. Ebeling: Sitzungsberichte der AdW der DDR, 22N, 33 (1981)
18. J.L. Lebowitz, D.G. Bergmann: Ann.Phys. (N.Y.) 1, 1 (1957)
19. S. Karlin, M. Taylor: Stochastic Processes (Mc Graw, New York 1974)
20. H.K. Janssen:Z.Phys. 270, 67 (1974)
21. W. Ebeling, R. Feistel: Physik der Selbstorganisation und Evolution (Akademie-Verlag, Berlin 1982)
22. C.W. Gardiner: Handbook of Stochastic Methods (Springer, Berlin Heidelberg New York 1984)
23. R.C. Cesai, S.W. Koch, F.F. Abraham: Physica 118A, 136 (1983)