

changes drastically. In other words, we could say that at the temperature where the relaxation time becomes $\tau \sim 10^{-10}$ s, which is caused not only by temperature but also by enzymes, the double helix would be melted out. Thus it has been found that the dynamical structure of the primary hydration shell plays an important role in the melting phenomena of the DNA double helix.

3. The Dynamics of First-order Phase Transition: *A Stochastic Approach with the Domain-size Distribution Function for the Late Stage*

(一次相転移の動力学後期過程の

ドメイン・サイズ分布関数による記述)

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Abstract

Many systems in nature make phase transitions of the first order. Such a system, if quenched from a single-phase thermal equilibrium state into a single-phase nonequilibrium state, evolves into another equilibrium thermodynamic state which consists of two coexisting phases. These phase separation processes are characterized by nucleation, spinodal decomposition, and late stage growth and coarsening.

In this thesis, we first review existing studies of the phase separation, and then attempt to treat the phase separation as a stochastic process. In both cases of the early stage and the late stage of phase transitions, all studies dealt with either semi-macroscopic dynamical variables, i.e. the order parameters or the time-dependent probability distribution functional. In their results, we place particular importance on their conclusions on the time dependence of the domain growth.

In the treatment of the early stage, an important aspect is how the nonlinearity is taken into account in the semi-phenomenological equations

of motion for the order parameters. Hillert (1961), Cahn (1961) and Cook (1970) analyzed the early stage of spinodal decomposition by a linearized theory, so that they could not of course describe the last stage behavior of phase separations. Then, Langer et al.(1975) handled both effects of thermal fluctuations and the nonlinearity by deriving an equation for the probability distribution functional of the order parameters. On the other hand, Saito (1976) emphasized the necessity of considering the coherency spread over a system as well as the nonlinearity in order to analyze the evolution of a quenched system from a nonequilibrium state to an equilibrium state.

In the treatment of the late stage, the characteristic aspect is how the nonlocality is considered. When two or more kinds of domains coexist, there are two ways of approach: the derivation of equations of motion for the interface and the investigation of the time dependence of domain growth. Kawasaki et al.(1982,1983) assumed that the interface thickness is thinner than the domain size, and transformed a nonlinear diffusion equation for the order parameter, i.e. the time-dependent Ginzburg-Landau equation, into an equation of motion for the interface. Concerning the domain size approach, Binder (1977) and Furukawa (1981,1984) assumed that the domain size distribution function or the observable, i.e. the scattering function, should be scaled in unit of averaged domain size, and obtained the domain growth rate. In all the theories mentioned above, the power law behavior for the domain growth, t^α , is predicted, and discussions for the exponent α are made. Shugard and Reiss (1976) proposed a stochastic model concerning the domain size distribution function and calculated the number of droplets with radii over the critical size at a time t .

In this thesis, we make an attempt to use a stochastic model in order to obtain the time dependence of the domain growth. The domain size is regarded as a coordinate in one-dimensional discrete "size space", and the process that a representative droplet grows or shrinks by the addition or loss of single element is thought of as a random walk on a lattice. The

domain size distribution function satisfies a differential equation or an integral equation. In both formulas, the time-dependent moving rate on the lattice determines two basic properties in the variation of domain size distribution function: the magnitude of a small change in domain size and the distribution of time interval necessary for the change. On the assumption of an adequate functional form on the moving rate, the power law behavior for the domain growth, $t^{\frac{1}{2}}$, is obtained. The domain size distribution function concerns originally with a one-dimensional characteristic length. In order to know the distribution of area or volume, the explicit shape of a domain must be considered. Therefore, we treat only one-dimensional case. The asymptotic logarithmic growth law, already predicted by one-dimensional equation of motion for the interface, is also confirmed by experiment. From these observed data, we try to infer the time dependence of the domain size distribution function.

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