

still both laws are equally well fitted. In order to determine which law is better, measurements by other technique with large  $f$  is required.

## 2. Study on Melting Process of DNA Double Helix by 5-pass Brillouin Spectroscopy

(ブリュアン散乱によるDNA二重らせんの  
融解現象の研究)

Hisako Ishido (石戸久子)

### ABSTRACT

It has been of great interest to clarify the dynamical mechanism of the melting process in the DNA double helix, because the melting phenomena have been found to relate closely with the basic molecular biological process, such as the replication and the transcription of genetic information. The melting phenomena can be caused not only by the interaction with enzymes but also by increasing the temperature. Numerous investigations on the melting phenomena of the DNA double helix in dilute solution have been carried out by means of absorption spectroscopy, circular dichroism and other methods with increasing temperature. But there have been only a few studies on the dynamical mechanism of the melting process in the condensed state.

In the present work it has been found that the dynamical structure of the primary hydration shell around the DNA double helix in the condensed state changes drastically during the melting process, which is observed by means of the Brillouin scattering spectroscopy using a 5-pass Fabry-Perot interferometer.

The lyophilized calf-thymus DNA was made into the gel state in the thin glass capillary by adding distilled water. It has been known that the 30 w/w% DNA gel has both the primary hydration shell and the secondary hydration shell as the hydrated water, but has no bulk water.

The relaxation time of the primary hydration shell has not yet been established experimentally but it has been estimated as about  $10^{-6}\sim^{-8}$  s at room temperature. Since the frequency range of Brillouin spectroscopy is near the reciprocal relaxation time of this primary hydration shell, the Brillouin spectra of the DNA gel are expected to be strongly influenced by the dynamical motions of the primary hydration shell.

The Brillouin scattering experiment of a non-crystalline sample such as DNA gel is very difficult, because there coexists a strong elastic scattering component which masks the weak Brillouin component. In order to overcome this difficulty the 5-pass Fabry-Perot interferometer has been constructed.

The temperature dependence of the Brillouin shift frequency  $\omega_B$  and the linewidth  $\Gamma_B$ , thus obtained, has the following anomalies. One is that there exists the maximum in the Brillouin shift frequency  $\omega_B$  at  $T_m \sim 80^\circ\text{C}$ , which is the melting temperature, and the other is that there exists the minimum in the linewidth  $\Gamma_B$  at  $\sim 75^\circ\text{C}$ , which is below the melting temperature. These anomalies cannot be explained by the characteristics of the DNA double helix itself, because the elastic constant of DNA double helix would decrease just before the melting. To explain the present experimental results, the coupled dynamics between the sound wave of the DNA gel and the relaxational motions of the primary hydration shell has been proposed. According to the coupled mode analysis, the Brillouin spectra have been strongly influenced around the frequency of  $\omega \sim 1/\tau$ , where  $\tau$  is the relaxation time of the primary hydration shell. The temperature dependence of  $\omega_B$  and  $\Gamma_B$  has been quantitatively explained by this coupled dynamics. The obtained relaxation time from this coupled mode analysis is  $\tau \sim 10^{-8}$  s at  $50^\circ\text{C}$  and  $\tau \sim 10^{-10}$  s at  $80^\circ\text{C}$ . This result means that during the melting process the relaxation time of the primary hydration shell

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)

Kazuyo Kaneko (金子一代)

#### 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