論文内容の要旨

論文題目: Thermophoresis, thermal force, and optothermal manipulation of single molecule and colloids

（訳 コロイド粒子と高分子の光熱捕捉と操作に関する研究）

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Thermal diffusion, also called thermophoresis or Soret effect, has been discovered for more than 100 years and has been found in different systems. This phenomenon describes the mass transfer caused from temperature gradient. It is generally believed that thermodiffusion can be described by linear nonequilibrium thermodynamic relations, given by the following phenomenological relation for concentration *C*:

$$j = j_D + j_{TD} = -D\nabla C - D_T C\nabla T ,$$

where j is the total mass flux, j_D is the mass flux due to diffusion, j_{TD} is the mass flux due to thermodiffusion, D is diffusion constant, and D_T is thermodiffusion constant. This phenomenon is studied from the concentration distribution in steady-state and not fully understood.

In this thesis, we study the thermophoresis from the single colloidal bead and single polymer. From the thermophoresis of beads, we find that the thermophoresis can be viewed as a

process driven by the temperature gradient induced force, thermal force. From the conformations of DNA during thermophoresis, we find the unique way of migration of DNA in temperature gradient which may help people to understand how the force is applied on the single polymer. Based on the results of thermophoresis, we experimentally for the first time measure the temperature induced force by optical tweezers. We also measure the thermal force of single DNA from its intrinsic elastic responses. We demonstrate that the conformation of polymer can be controlled by temperature gradient. The thermal force obtained in both cases shows the linear relation with temperature gradients.

We further use the temperature gradient to manipulate molecules. Single polymer can be trapped and colloidal particles can be assembled with spatial control of temperature. In study the interactions between different objects in temperature gradient, we find a new phenomenon relative to depletion force and the osmotic pressure in polymer solution. We propose a new general optical trapping method to control the entropic force by laser induced optothermal effects. We elucidate the mechanism from the entopic contribution of depletion layers. Various kinds of objects can be manipulated by this method. This entropic force induced phenomenon would bring advances in manipulation technology in many fields of science and engineering.

In chapter 2 thermophoresis:

We use surface heating method in a quasi-2D thin chamber to studying the thermophoresis of the single bead and single DNA. Our results suggest that the thermophoresis can be studied by the measuring the response of single particle. We show that the temperature dependence of thermodiffusion constant is same as the temperature dependence of viscosity of the background solution. This implies that the thermophoresis can be viewed as the result of the temperature gradient induced force. The properties of thermophoresis are determined by this force and the condition during migration.

In single DNA, we directly measure the conformation and orientation of single DNA during thermophoresis. The results suggest that the thermophoresis of DNA should be treated as thermophoresis of several linked segments without hydrodynamic interactions which has been long time anticipated but never been measured. We also find that DNA with different expansion ratios move with different speed in temperature gradient. This suggests that the conformations of DNA may bring additional nonlinear responses.

In chapter 3 thermal force:

We provide the new method to study the temperature gradient induced force, thermal force,

at single particle level. We experimentally measure the thermal force from the single bead and single DNA. We find that in both cases the relations between temperature gradient and thermal force relation are linear.

The thermal force of the particle measured from optical tweezers agrees with the drag force estimated in thermophoresis. This result not only shows that the thermophoresis is driven by thermal force but also indicates that even the particle does not move, there is still a force coming form the temperature gradient. This is further proved by the thermostretch of tethered DNA. The concept that thermophoresis is driven by this thermal force may directly lead a useful understanding about why temperature dependence of thermodiffusion constant is same as the temperature dependence of viscosity of the background solution.

This result also suggests that the linear range of thermal force is larger the linear range of thermophoresis because thermophoresis depends on details, like viscosity, interactions between particles, or the environment during migration. In this point of view, we may extend the thermodynamic linear relation on flux induced by temperature gradient, $J_{TD} = -D_T C \nabla T$, to linear relation on force induced by temperature gradient, $F = -f_T \nabla T$, in the single particle limitation, where f_T is the thermal force coefficient. This should be more appropriate for a single particle. This treatment may give an easy way to deal with thermal force in further study especially at single particle level.

We also show that controlling the temperature gradient is a powerful tool to study and to manipulate single molecule. Optical and magnetic tweezers use gradient of intensity in electric field and magnetic field respectively. Stretching DNA by shear flow uses a steep gradient of velocity. We demonstrated that gradient of temperature can be also used to apply force locally to single molecules. We stretch DNA by temperature gradient. This kind of manipulation may open possibilities for future applications of single DNA manipulation with temperature gradient, for instance in micro fluidic devices.

In chapter 4 optothermal manipulation:

We present two methods for optothermal manipulations, primary and secondary optothermal manipulation. In primary optothermal manipulation which depends on the responses of the object in temperature gradient, we show how to trap single DNA and how to assemble colloids by controlling the temperature profiles. This method is useful but it also needs more considerations of objects themselves and the optical setups. In the secondary optothermal manipulation which depends on the interactions between different components in temperature gradient, we discover a new way for all optical control of entropic force which overcomes the limitation of conventional laser tweezers. With this method, colloidal particles such as submicron sized beads, non-tethered

DNA molecules, and biological cells are easily trapped when only a little amount of polymer is added in solutions.

We also elucidate the mechanism of this new optical trap. It can be described as following. In the heating spot of laser focus, polymer density becomes lower than outer region, so the colloid which is dressed with the depletion layer allows more polymers to have their conformations when it is trapped in the center. Therefore the force directs toward the hot spot to increase overall entropy. This new trapping method can also become a method to control osmotic pressure. The osmotic pressure can be locally controlled with light may lead numerous applications since osmotic pressure is an important issue in cellular signal transductions and cell membrane function.