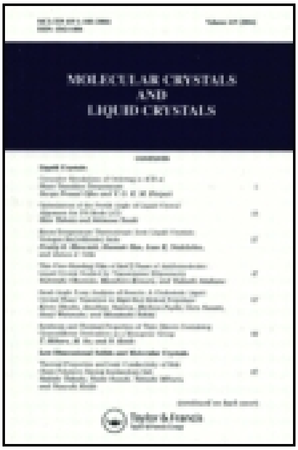

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl20

# Direct Measurement of Interaction Between Colloidal Particles in Nematic Liquid Crystal

Kenji Takahashi [a], Yoshihiko Fujiwara [a], Masatoshi Ichikawa [a] & Yasuyuki Kimura [a]

[a] Department of Physics, School of Sciences, Kyushu University, Hakozaki, Higashi-ku, Fukuoka, Japan

Version of record first published: 22 Sep 2010

*Mol. Cryst. Liq. Cryst.*, Vol. 475, pp. 183–192, 2007

ISSN: 1542-1406 print/1563-5287 online
DOI: 10.1080/15421400701732373



# Direct Measurement of Interaction Between Colloidal Particles in Nematic Liquid Crystal

**Kenji Takahashi**
**Yoshihiko Fujiwara**
**Masatoshi Ichikawa**
**Yasuyuki Kimura**
Department of Physics, School of Sciences, Kyushu University, Hakozaki, Higashi-ku, Fukuoka, Japan

*Interparticle force F between two colloidal particles accompanied by hyperbolic hedgehog defects has been studied in nematic liquid crystal by two methods. We have directly measured F as a function of interparticle distance R in the case of two "dipoles" composed of a particle and an accompanied defect aligning parallel to the far-field director. We have confirmed the theoretical prediction that F is proportional to* $R^{-4}$ *at longer distance but the magnitude of F is different in two methods. We have also observed that repulsive component emerges at shorter distance.*

**Keywords:** colloidal particle; dipole analogy; interparticle force; nematic liquid crystal; topological defect

## 1. INTRODUCTION

The interaction between colloidal particles in structural fluids such as liquid crystals and polymer solutions has recently attracted the attention of researchers from both fundamental and application points of view. A colloidal particle dispersed in liquid crystals breaks the continuous rotational symmetry of the medium. For sufficiently

This work is fully supported by Grant-in-Aid for Scientific Research form Japan Society for the Promotion Science and from Ministry of Education, Culture, Sports, Science and Technology of Japan. M.I. is also financially supported by the Sumitomo foundation and the Sasakawa Scientific Research Grant from The Japan Science Society.

Address correspondence to Kenji Takahashi, Department of Physics, School of Sciences, Kyushu University, 6-10-1 Hakozaki, Higashi-ku, Fukuoka 812-8581, Japan. E-mail: ken8scp@mbox.nc.kyushu-u.ac.jp

strong surface anchoring, a particle will generate an orientational (topological) defect. At the same time, another defect emerges near the particle to restore the orientational order far from the particle. Furthermore, the specific interaction mediated by orientational elasticity of the surrounding liquid crystal appears between these particles even if there is no direct interaction between them. The interaction between the particles in liquid crystal depends on the types of the defects and is determined by orientation of the liquid crystal at surface of the particles. In this paper, we report specific interactions between spherical colloidal particles with normal anchoring condition in nematic liquid crystal.

In our case, a particle by itself becomes a topological defect called a radial hedgehog and another defect called a hyperbolic hedgehog emerges near the particle to conserve the far-field director. Therefore a particle always accompanies a defect in this case (Fig. 1) [1].

Lubensky *et al.* [2] showed a particle in nematic liquid crystal can be regarded as "dipole" by its electrostatic analogy. The interparticle force $F$ takes a form similar to that between dipole-moments, and $F$ is proportional to $R^{-4}$, where $R$ is the interparticle distance. In particular, when two pairs of a particle and a defect align parallel to a far-field director as shown in Figure 2, $F$ is predicted to be attractive and proportional to $R^{-4}$. This theoretical prediction has been confirmed by various experiments [3,4].

One of the previous experiments is the direct observation of the motion of droplets filled with ferrofluid in nematic solvent [3]. Another experiment with optical tweezers has been reported [4]. Both these experiments show that $F$ is proportional to $R^{-4}$. However,



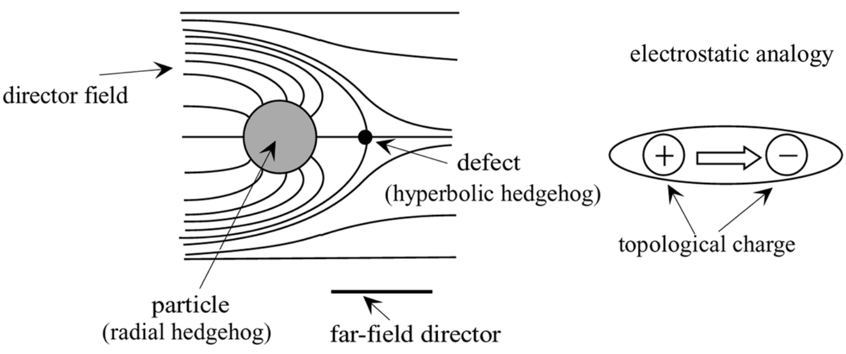


**FIGURE 1** Configuration of a topological dipole-moment composed of a particle and its accompanied defect, and its electrostatic analogy. The vector of the topological dipole is parallel to the far-field director.

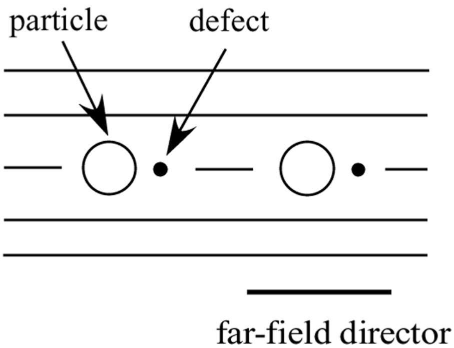


**FIGURE 2** Configuration of two topological dipoles. Two dipoles align along the far-field director.



there is difference between them. In the case of direct observation, the maximum force is as large as several pN. On the other hand, in the case of the optical tweezers the maximum force is several tens pN. In this study, we have measured the force $F$ in the configuration shown in Figure 2 by direct observation and by optical tweezers.

## 2. EXPERIMENT

In our experiments, we used a polystyrene latex particle (radius $a = 1.5\,\mu m$) as a colloidal particle and MJ032358 (MERCK) as nematic liquid crystal. The refractive index of the particle is 1.6 and that of liquid crystal is $n_e = 1.5$ and $n_o = 1.46$. The particles were coated with DMAOP to promote homeotropic anchoring at their surface. The particles and the liquid crystal were simply mixed and sandwiched between glass plates. These surfaces were coated with thin polyimide layers and were rubbed in one direction to attain homogeneous alignment of liquid crystal. The thickness of a sample was fixed to $10\,\mu m$ by using spacer films. We used dual beam optical tweezers [5–7] to manipulate particles within a two dimensional plane and to measure the force directly. The beam of the Nd-YAG laser (spectra physics, wavelength: 1064 nm) is introduced to a inverted fluorescence microscope (TE200) and focused by a 100x oil immersion objective lense (N.A. = 1.4). A laser spot of one tweezers was controlled by two galvano mirrors and the spot of the other tweezers is fixed. Galvano mirrors are controlled by a function generator (NF1946), and we can move the spot as we want. We also used video microscope to capture motion of the particles.

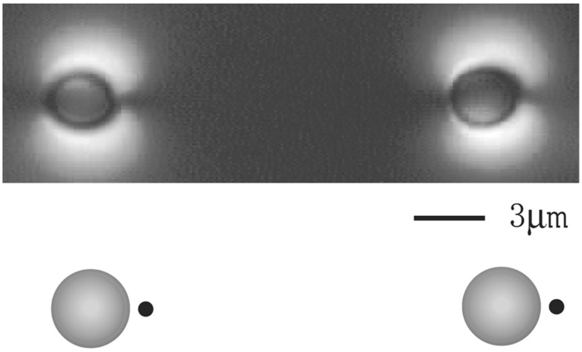


**FIGURE 3** Parallel configuration of two particles under a polarizing microscope.

## 2.1. Release Experiment

We can evaluate the force between two particles in a nematic host by simply observing the particles' motion under a polarizing microscope, as was done in ref [3]. We selected two particles in the parallel configuration in a sample cell (Fig. 3), and forced them to align parallel to the orientational axis of liquid crystal by the optical tweezers. After turning off the optical tweezers, we recorded the motion of the two particles till they attach and stop their motion. The force is calculated by the same way as ref [3].

## 2.2. Dual Beam Optical Tweezing Measurement

We also used a dual beam optical tweezers to measure the interparticle force. Though the force has already been measured by optical tweezers in ref [4], we measured it by a different method. We selected two particles, and trapped each particle in a different tweezers. By moving galvano mirrors, we change the position of one of the particles to vary the interparticle distance. If the particle doesn't experience any external force, the particle stays at the bottom of its potential. But when the particle experiences any external force, it shifts its position and stays at the place where the external force balances the force originated from the trapping potential of the tweezers. Therefore we can obtain the interparticle force $F$ by observing a displacement of the particle trapped by the fixed tweezers. In this experiment, we moved the laser spot slowly to change the interparticle distance $R$, and observed the displacement of the fixed particle.

# 3. RESULTS

## 3.1. Release Experiment

From Brownian motion of a single particle, we can measure the local viscosity in following way. We captured the motion of the particle by a CCD camera, and we observed the time evolution of the mean-square displacement of the particle from the snap-shots. The self-diffusion constant of a Brownian particle along director $D_{\|}$ is given by Stokes-Einstein relation $D_{\|} = k_{\mathrm{B}}T/6\pi\eta_{\|}a$ [8], where $\eta_{\|}$ is viscosity along the director, $k_{\mathrm{B}}$ is the Boltzmann constant and $T$ is the temperature. Figure 4 shows the relation between square displacement of a particle $\Delta_{\|}^2$ along the director and the elapsed time $t$. Since $\Delta_{\|}^2$ is theoretically given as $\Delta_{\|}^2 = 2D_{\|}t$, the slope of best fitted line in Figure 4 is defined as $2D_{\|}$. From Figure 4, we have obtained the viscosity $\eta_{\|} = 3.04 \times 10^{-2}$ [Pa $\cdot$ sec]. In the equilibrium condition, the frictional force exerted on a particle balances the interparticle force. Therefore, we can obtain the interparticle force $F$ from the viscosity and the approaching speed of the two particles.

Except for random displacements caused by Brownian motion, the particles approached each other along to the far-field director. The observed time evolution of the interparticle distance $R$ is shown in Figure 5. The distance $R$ monotonously decreases as time passes until $R$ reaches about 3.7 μm. After that, $R$ no longer changes. From Figure 5, we can calculate the approaching speed $v$.

Assuming that $F$ is equal to the frictional force, we can calculate the interparticle force $F = 3\pi\eta_{\|}av$ (Fig. 6(a)), where $\eta_{\|}$ is the viscosity parallel to the director, $v$ is the approaching speed between the two particles. The force $F$ is also plotted in Figure 6(b) as a function of $R$ in logarithmic

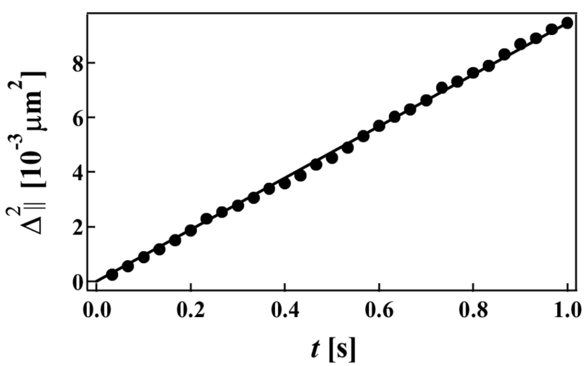


**FIGURE 4** The relation between the square displacement $\Delta_{\|}^2$ and elapse time $t$. $\Delta_{\|}^2$ is proportional to $t(\Delta_{\|}^2 = 2D_{\|}t)$ and the slope of best-fitted line corresponds to the diffusion constant $D_{\|}$.

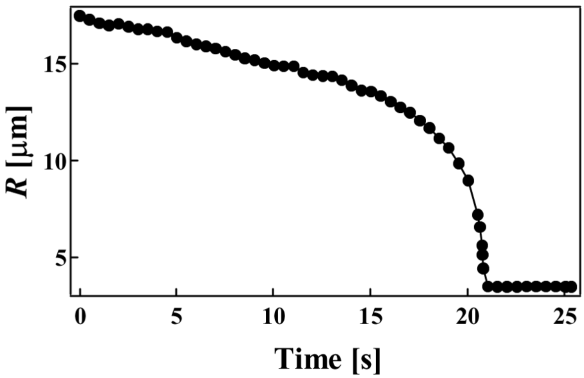


**FIGURE 5** Time evolution of interparticle distance $R$. The interparticle distance decreases as the time passes. Once the distance reaches about 3.7 μm, the motion of particles stops.

scale. The force is attractive at all distance where we measured the force, and is nearly proportional to $R^{-4}$ except at short distance.

At this range, we can find that repulsive component emerges strongly. In Figure 6(a), we show the best-fitted curve of $\alpha R^{-4} + \beta R^{-\gamma}$, where $\gamma$ is found to be 6.06.

In this experiment, we assumed that the viscosity is independent of the interparticle distance $R$ and is equal to the one obtained from Brownian motion. At large $R$, this assumption is approximately right. But at small $R$, the orientation of the liquid crystal around the particle differs from one at the large $R$. Therefore the viscosity at the short distance should be different from one at the long distance.

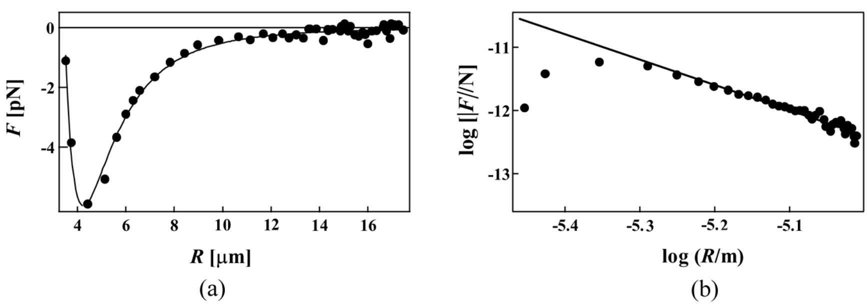


**FIGURE 6** Dependence of interparticle force $F$ on the interparticle distance $R$. (a) Linear scale. The negative sign shows the attractive force and the solid line is the best-fitted curve of $\alpha R^{-4} + \beta R^{-\gamma}$, where $\gamma$ is 6.06. (b) Logarithmic scale. Slope of the solid line is $-4$.

We also assumed that the interparticle force equals the frictional force. But at the short distance, the nematic host can't flow quickly due to hydrodynamic effect and this will affect the interparticle force. Therefore, in order to obtain the interparticle force alone, we need to consider the effect discussed above or to measure the force with the method not depending on the viscosity and the hydrodynamic effect.

## 3.2. Dual Beam Optical Tweezing Measurement

As the method not depending on the viscosity and the hydrodynamic effect, we adopted the method of dual beam optical tweezers and measured the interparticle force $F$. At first, we measured the trap potential of the optical tweezers. The distribution of the particle's position gives direct information about the potential profile. Since the obtained distribution is well described by Gaussian, we regard the trapping potential by a tweezers as harmonic one. From Boltzman relation, the probability density $P$ is given as

$$P = A\exp\left\{-\frac{1}{2}k(x-x_0)^2/k_BT\right\}, \tag{1}$$

where $k$ is the spring constant which characterizes the harmonic potential, $A$ is the normalizing factor [9]. We fit Eq. (1) to the experimental result (Fig. 7), and we obtained $k = 2.83 \times 10^{-5}$.

By use of the obtained profile of the laser potential, we can calculate the dependence of the interparticle force $F$ on the interparticle distance $R$. From the images under a microscope, we obtained the time evolution of the displacement of the particle trapped by the fixed

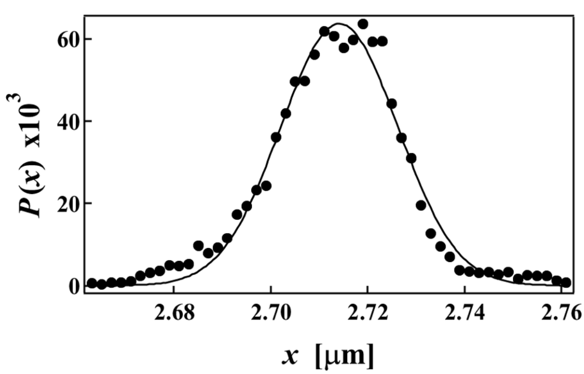


**FIGURE 7** Probability density of a trapped particle's position $P(x)$. The solid line is the best-fit curve of Gaussian.

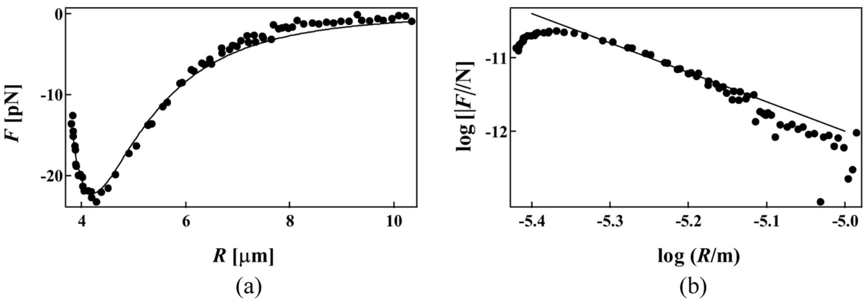


**FIGURE 8** Dependence of interparticle force $F$ on the interparticle distance $R$. (a) Linear scale. The negative sign indicates the attractive force and the solid line is the best fitted curve of $\alpha R^{-4} + \beta R^{-\gamma}$, where $\alpha$, $\beta$ and $\gamma$ are the fitting parameters. In this case, $\gamma$ is 10.556. (b) Logarithmic scale. The slope of the solid line is $-4$.



tweezers while the other particle was moved at the constant speed of 60 nm/s by another tweezers. From the displacement relative to the center of the fixed tweezers, we obtained the interparticle force $F$. The dependence of $F$ on $R$ is shown in Figure 8(a). Since the negative sign indicates the attractive force, therefore the force is attractive over the entire range we measured. We find again that the force is nearly proportional to $R^{-4}$ except the short distance (Fig. 8(b)). At this short distance, repulsive component also emerges strongly. These results look similar to that obtained by the release experiment.

We measured the force in both cases of approaching and going away the two particles. But we can't find any difference between the results in these two cases. We also made the speed of the laser spot more slowly (12 nm/s), but there are little differences between two experimental results.

## 4. DISCUSSION

In both experiments, we find that $F$ is nearly proportional to $R^{-4}$. On this point, these results make good agreement with the reported ones [3,4]. But, there is difference between the two experiments about the magnitude of the force. We can also confirm this difference between [3] and [4].

The origin of the difference is due to the viscosity used in the calculation of $F$ in the first experiment, which is obtained simply by Brownian motion. When the interparticle distance changes, the orientation of the liquid crystal also has to change and the frictional force exerted on moving particle strongly depends on orientation of the liquid crystal around it. Therefore, the assumption that viscosity is constant independent of the interparticle distance is not so good approximation in the first experiment.

We can also observe interesting phenomena in both experiments when the two particles approach closely. As we can confirm from our experiments, at short distance, a repulsive component emerges strongly. In recent numerical analysis [10,11], this kind of component is also confirmed. Actually, in the electrostatic analogy [2], the quadrupolar component is considered as repulsive component in calculating the interparticle force. But, in this theory, the repulsive component is much smaller than the attractive one and we can neglect this repulsive one. In this point, there is difference between our results and the theory [2].

There are some possible origins for the repulsive component. One possible origin is the hydrodynamic effect at the short distance. But this effect can be eliminated in the experiment with optical tweezers. The other possible origin is the existence of the defect between the two particles. In parallel configuration, the attractive force acts to extinguish the defects and to minimize the free energy of the liquid crystal. But when two particles approach considerably, the hyperbolic hedgehog defect between the two particles is deformed and repulsive force emerges.

## CONCLUSION

We measured the force between the particles accompanied with hyperbolic hedgehog in nematic liquid crystal. Similar to the reported research, the force depends on the interparticle distance and is nearly proportional to $R^{-4}$. This dependence is well described at the longer distance. But when the particles approach considerably, we observed repulsive component. Although some possible origins of this component have been discussed, the structure of the hyperbolic defect between the particles is most likely origin.

## REFERENCES

[1] Chaikin, P. M. & Lubensky, T. C. (1995). *Principles of Condensed Matter Physics*, Cambridge University Press.
[2] Lubensky, T. C., Pettey, D., Currier, N., & Stark, H. (1998). *Phys. Rev. E*, *57*, 610.
[3] Poulin, P., Weitz, D. A., & Cabuli, V. (1997). *Phys. Rev. Lett.*, *79*, 4862.
[4] Yada, M., Yamamoto, J., & Yokoyama, H. (2004). *Phys. Rev. Lett.*, *92*, 185501.
[5] Askin, A., Dziedzic, J. M., Bjyourkholm, J. E., & Chu, S. (1986). *Opt. Lett.*, *11*, 288.
[6] Svoboda, K. & Block, S. M. (1994). *Annu. Rev. Biophys. Struct.*, *23*, 247.
[7] Smalyukh, I. I., Lavrentovich, O. D., Kazumin, A. N., Kachynski, A. V., & Prasad, P. N. (2005). *Phys. Rev. Lett.*, *95*, 157801.

[8] Loudet, J. C., Hanusse, P., & Poulin, P. (2004). *Science*, *306*, 1525.
[9] Muševič, I., Škarabot, M., Babič, D., Osterman, N., Poberaj, I., Nazarenko, V., & Nych, A. (2004). *Phys. Rev. Lett.*, *93*, 187801.
[10] Fukuda, J.-I., Stark, H., Yoneya, M., & Yokoyama, H. (2004). *Phys. Rev. E.*, *69*, 041706.
[11] Fukuda, J.-I., Stark, H., Yoneya, M., & Yokoyama, H. (2005). *Mol. Cryst. Liq. Cryst.*, *435*, 63.