

LASER BEAM INDUCED ORIENTATION CONTROL OF A NANOSHEET LIQUID CRYSTAL BY EMPLOYING AN OBJECTIVE LENS WITH A LOW NUMERICAL APERTURE

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ABSTRACT

Optical manipulation of nanosheet liquid crystals prepared by exfoliation of inorganic layered crystals realizes local and on-demand orientation control of nanosheets. Although a tightly focused laser beam by using an objective lens with a high numerical aperture (NA) is typically employed for the optical manipulation to enhance the gradient force of an irradiated laser beam, the dominant driving force of the optical orientational manipulation of nanosheets has recently been attributed to the scattering force, suggesting that an objective lens with a high NA is not indispensable for the nanosheet manipulation. In this study, we carried out optical orientational manipulation of nanosheets using an objective lens with low NA, 0.4, and compared the results with those obtained by a lens with high NA, 1.2. Orientational manipulation was realized with both of the lenses, and the size of the manipulated domain using a lens with NA=0.4 was smaller than that obtained by using a lens with NA=1.2. The domain size increased with the irradiated laser power, reflecting the dominant contribution of scattering force to the optical manipulation of nanosheet liquid crystals.

Key words: metal oxide nanosheet, liquid crystal, radiation pressure

INTRODUCTION

Inorganic nanosheets prepared by exfoliation of layered compounds, such as nontronite, fluorohectorite, and metal oxides, are known to exhibit lyotropic liquid crystallinity (Nakato and Miyamoto, 2009; Miyamoto et al., 2010; Michot et al., 2006; Gabriel et al., 2001). One of the authors has studied liquid crystalline nanosheet colloids of a layered niobate (Miyamoto and Nakato, 2004). Nanosheets are aligned in a lamellar manner with a basal spacing of several tens of nanometers in the liquid crystalline state (Yamaguchi et al., 2012). Orientation control of nanosheets in liquid crystalline state is

important challenge for realizing devices utilizing their large anisotropy. Liquid crystalline domains grow through self-assembly by incubating liquid crystalline nanosheet colloid at room temperature to reach a sub-millimeter size (Nakato et al., 2014). The nanosheets respond to an external field, such as a shear force (Miyamoto and Nakato, 2004) and an electric field (Nakato et al., 2011). Hierarchical macroscopic structures can be formed in the liquid crystalline nanosheet colloids by combining the domain growth and an external field (Nakato et al., 2017). However, with these methods, a colloid sample is organized in the same manner in the entire sample. The local and on-demand orientation by optical manipulation should expand the flexibility and arbitrary structural control of the nanosheet liquid crystals.

Thus, we have attempted to control the orientation of the colloidal nanosheets in the liquid crystalline state by utilizing a radiation pressure of a focused laser beam. In general, irradiation of a focused laser beam provides two types of forces as a radiation pressure on an object (Ashkin, 1992; Harada and Asakura, 1996). One is a scattering force which is applied toward the propagation direction of the incident laser beam

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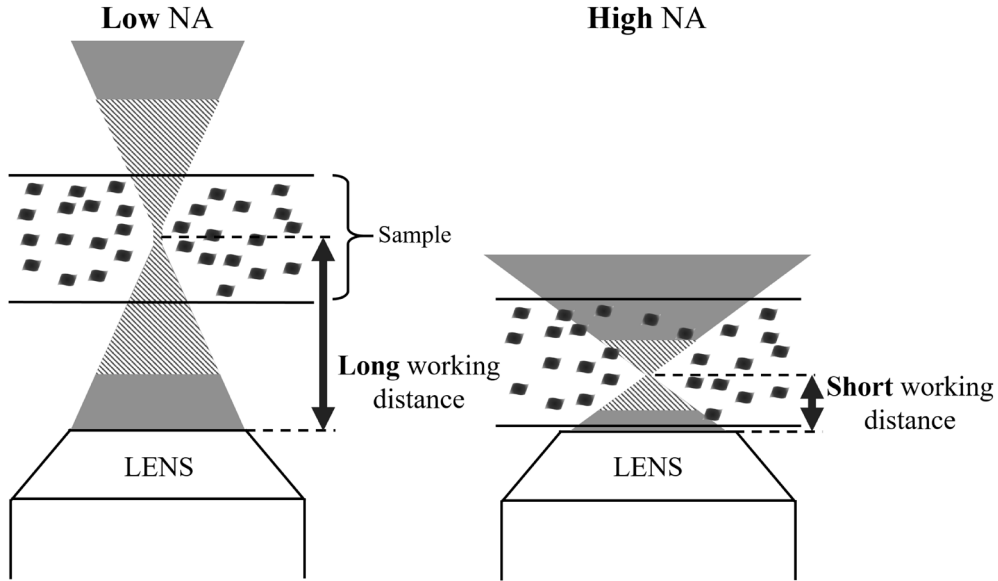


Fig. 1. Schematic representation of working distances expected to objective lenses with a high (1.2) and a low (0.4) numerical apertures.

to the object. Thus, when the scattering force is applied to an object, the object moves along the propagation direction of the incident laser beam. The other is a gradient force which is applied perpendicular to the propagation direction of the incident laser beam. While the scattering force transfer an object to the direction of light propagation, the gradient force assists trapping of the object located within a focal plane to the focal point. We have realized on-demand control of local orientation of niobate nanosheets (Tominaga et al., 2018a). In addition, we have found that niobate nanosheets oriented parallel to the propagation direction of an incident laser beam are assembled to form a tree-ring-like domains at the periphery of a focal point when the nanosheets are in the liquid crystalline state (Tominaga et al., 2018b). Such optical manipulation of nanosheets has been attributed mainly to the contribution of scattering force.

For optical manipulation, a laser beam has generally been focused by using a liquid-immersion objective lens with a high numerical aperture (NA). This is because the gradient force applied to a trapped object can be increased when an objective lens with a higher NA is employed. However, employment of a liquid-immersion lens with a high NA often limits freedom of the optical setup. As shown in Figure 1, due to the short working distance of the high NA lens, distance between surface of the sample cell and objective lens was less than a few μm in our previous studies (Tominaga et al., 2018a, 2018b). Furthermore, the use of a liquid-immersion lens limits the spatial arrangement of the optical setup and prevents the insertion of any other optical parts in between sample and lens.

Considering that the optical manipulation of niobate nanosheet liquid crystal giving the tree-ring-like alignment has been ascribed mainly to the scattering force, a lens with a high NA should not be indispensable for the manipulation. Thus, in this study, we irradiated a laser beam to the liquid crystalline niobate nanosheets by employing an objective lens with NA of 0.4. The orientational change obtained by laser irradiation was compared with those obtained by using

an objective lens with NA of 1.2. We have found a tree-ring-like arraignment of nanosheets was formed even when an objective lens with an NA of 0.4 was employed, as in the case employing an objective lens with NA of 1.2.

MATERIALS AND METHODS

As for a nanosheet to be manipulated, we employed niobate nanosheet which has already been manipulated in our previous studies (Tominaga et al., 2018a, 2018b). A niobate nanosheet colloid was prepared by exfoliation of layered $\text{K}_4\text{Nb}_6\text{O}_{17}$ according to the previously reported method (Miyamoto and Nakato, 2004; Nakato et al., 2014). The nanosheet concentration was 5 gL^{-1} and the colloid sample exhibited biphasic mixture consisting of isotropic and liquid crystalline phases at room temperature. The lateral lengths of the nanosheets obtained from transmission electron microscopy (TEM) observations exhibited a size distribution that obeyed a log-normal distribution to give an average size of $1.6\mu\text{m}$. The colloid sample was injected into a thin-layer glass cell with a $100\mu\text{m}$ thickness. The cell was set on a stage of an inverted microscope (IX70, Olympus, Japan).

A linearly polarized CW laser beam emitting at 532 nm (Millennia Pro, Spectra Physics, United States) was focused on the sample using an objective lens at room temperature. The power after the objective lens was 20 mW unless otherwise noted. As for the lens, we used a dry objective lens, ($25\times$, $\text{NA}=0.4$, Olympus, Japan) or a water-immersion objective lens ($60\times$, $\text{NA}=1.2$, Olympus, Japan). The beam diameters at focal points were calculated to be $0.7\mu\text{m}$ and $0.3\mu\text{m}$ for objective lenses with $\text{NA}=0.4$ and 1.2 , respectively. The focal depths in our experimental condition were $1.4\mu\text{m}$ and $0.3\mu\text{m}$ for objective lenses with $\text{NA}=0.4$ and 1.2 , respectively. For polarized optical microscope observations under laser irradiation, the sample was illuminated by a halogen lamp, and the image was monitored with a digital CMOS camera (ORCA-Flash 4.0 V3, Hamamatsu Photonics, Japan). A pair of crossed polarizers (polarizer and analyzer) was inserted

out of the beam path of the laser beam. Bright-field optical microscope images were observed using the same optical setup without crossed polarizers.

RESULTS AND DISCUSSION

Irradiation of the laser beam (20mW) to the niobate nanosheet liquid crystal induced essentially the same orientational change of nanosheets irrespective of the NA of objective lens employed for focusing the laser beam. Optical microscope images under laser irradiation with crossed polarizers are shown in Figure 2. The images before laser irradiation were dark, which indicates the homogeneous orientation of niobate nanosheets with their in-plane direction perpendicular to the propagation direction of the incident laser beam (hereafter, perpendicular orientation) in the thin-layer cell (Nakato et al., 2014). After irradiating the laser beam for 30 s, a circularly textured birefringence appeared. The brightness of the circularly textured area increased gradually until 120 s.

The birefringence indicates the orientation of nanosheets

with their in-plane direction parallel to the propagation direction of the incident laser beam (hereafter, parallel orientation) (Nakato et al., 2014). Therefore, the appearance of birefringence supports that the orientation of niobate nanosheets was changed from perpendicular to parallel by laser irradiation. The brightness of the birefringence area gradually increased accompanied by the continuous laser irradiation. Nanosheets locating in the beam path but out of the focal depths are known to change their orientation gradually from perpendicular to parallel under the continuous laser irradiation (Tominaga et al., 2018b). Thus, the gradual increase of the birefringence area is attributed to the increase of the ratio of the niobate nanosheets in parallel orientation at outside the focal point.

The sizes of textured areas were somewhat larger for NA=1.2. After irradiating the laser beam for 120 s, the domain diameter reached $120\mu\text{m}$, which was 400 times larger than the focal-point diameter when a lens with NA of 1.2 was employed. However, the domain diameter after irradiating the laser beam for 120 s was only $55\mu\text{m}$, which was 80 times larger than the focal-point diameter when a lens with NA of 0.4 was employed. As already described, the beam diameters at the focal points were $0.7\mu\text{m}$ and $0.3\mu\text{m}$ for objective lenses possessing NA=0.4 and 1.2, respectively. Therefore, the irradiance at the focal point focused by using an objective lens with NA=0.4 is about 20% that focused by an objective lens with NA=1.2. This means, when an objective lens with NA=0.4 is used, scattering force applied to nanosheets existing close to focal point is one-fifth of that provided by an objective lens with NA=1.2. Owing to the small scattering force at the focal point, when an objective lens with NA=0.4 is used, the diameter of the birefringence area was thought to be smaller than the case when an objective lens with NA=1.2 is used.

Although the size of the birefringence area was small when the laser beam was irradiated using an objective lens with NA=0.4, the circular texture was somewhat clearer and brighter than the case employing an objective lens with the NA of 1.2. This fact suggests that orientational order of nanosheets in parallel orientation becomes high when a lens with a low NA was employed. The focal depths are $1.4\mu\text{m}$ and $0.3\mu\text{m}$ for objective lenses with the NA of 0.4 and 1.2, respectively. The reason for the improvement of the orientational order is attributed to a longer focal depth and therefore larger high irradiance zone of the low NA lens.

As shown in Figure 3, bright-field optical microscope images are in accordance with those observed by the polarized microscope. The images before laser irradiation is featureless. This is consistent with the perpendicular orientation of nanosheets. When the laser beam was irradiated for 30 s, domains, which corresponded to the circularly textured birefringence areas, appeared gradually. In these domains, nanosheets in parallel orientation formed tree-ring-like textures. The diameters of the tree-ring-like textures after continuous laser irradiation for 120 s were $110\mu\text{m}$ for NA=1.2 and $50\mu\text{m}$ for NA=0.4, respectively. The sizes of the tree-ring-like textures were similar to the birefringence areas seen in polarized microscope images. Although the size of the textures obtained by employing an objective lens with

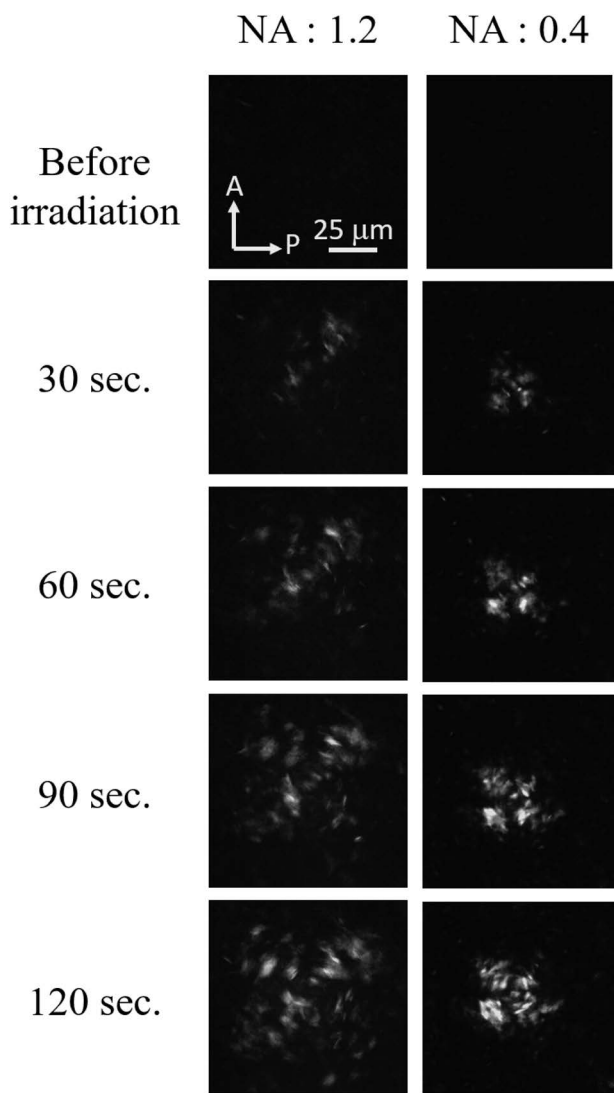


Fig. 2. Polarized optical microscope images of a liquid crystalline niobate nanosheet colloid (5gL^{-1}) under 20mW of laser irradiation.

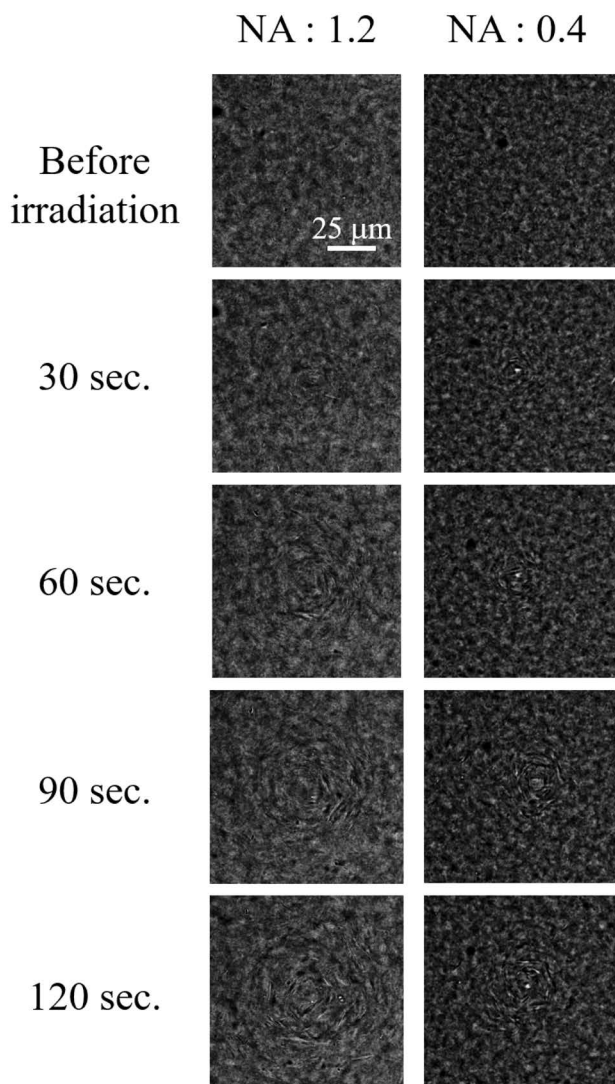


Fig. 3. Bright-field optical microscope images of a liquid crystalline niobate nanosheet colloid (5 g L^{-1}) under 20 mW of laser irradiation.

NA of 0.4 was smaller than those obtained by employing an objective lens with NA of 1.2, similar tree-ring-like textures were indeed appeared even when a lens with a NA of 0.4 was employed. These results demonstrate that the size of domain obtained using a lens with NA of 0.4 was smaller than that obtained by a lens with NA of 1.2 even when the irradiated laser power was the same. The difference of the size of the obtained domains is quite natural, considering the difference of scattering force depending on the NA of the lenses employed.

Finally, we examined power dependence of the tree-ring-like domain size. As shown in Figure 4, the size of the tree-ring-like domain increased as the consequence of increment of the power of irradiated laser beam, and thus the scattering force. Although the size of the domain obtained using a lens with a NA of 0.4 was smaller than that obtained by a lens with a NA of 1.2 at the same power, we have found the size of domain obtained using a lens with a NA of 0.4 could be increased by increasing the power of the incident laser beam. Therefore, it is concluded here that the employment of a lens with a low NA is simple and effective means for realizing orientational control of nanosheet liquid crystals.

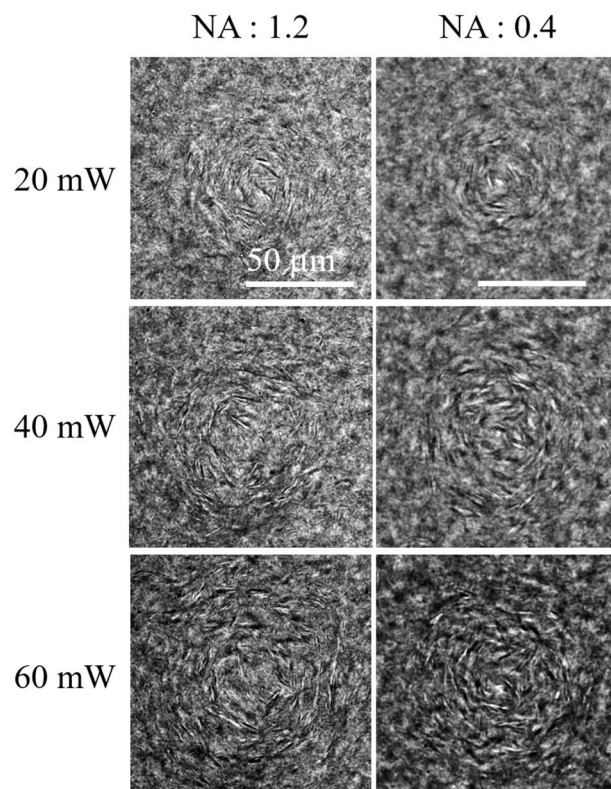


Fig. 4. Bright-field optical microscope images of the domain of nanosheets in tree-ring-like orientation when a laser beam with various laser power was irradiated for 120 s to a liquid crystalline niobate nanosheet colloid (5 g L^{-1}).

CONCLUSIONS

In this study, we have demonstrated that the orientation control of nanosheets in liquid crystalline states can be realized even by employing an objective lens with a low NA. The use of low NA lens should make it possible for us to manipulate nanosheets by laser irradiation under a long working distance. The freedom obtained by the long working distance for the optical setup should expand the application of optical orientation control of nanosheet. Thus, it should be concluded that the use of low NA lens is a simple and a convenient means for manipulating nanosheets in colloids.

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