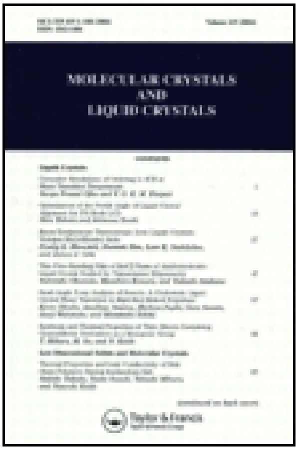

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Helical Tubular Filaments of a Smectic-A Phase in an Isotropic Phase

Masayoshi Todorokihara [a], Koki Fujihara [a] & Hiroyoshi Naito [a]

[a] Department of Physics and Electronics, Osaka Prefecture University, Sakai, Osaka, Japan

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HELICAL TUBULAR FILAMENTS OF A SMECTIC-A PHASE IN AN ISOTROPIC PHASE

Masayoshi Todorokihara, Koki Fujihara, and Hiroyoshi Naito
Department of Physics and Electronics, Osaka Prefecture University, 1-1 Gakuen-cho, Sakai, Osaka 599-8531, Japan

Smectic-A helical filaments grown in an isotropic phase have been observed in the binary mixture of octyloxycyanobiphenyl and dodecyl alcohol. The helical structures are made from reeling of the tubules, and are formed by the liquid crystalline molecules without chirality. The temperature sequence has been established for the observation of smectic-A helical filaments. It is found from the observation that the stability of the helical filaments depends essentially on the cooling rate.

Keywords: binary mixture; smectic-A helical filaments; tubules

INTRODUCTION

Thermo-temporal evolution of smectic-A phase is a geometrically interesting example of self-organization processes. A smectic-A phase grown from an isotropic phase first appears in the form of a collection of filaments. These filaments have a fixed radius and buckle continuously as they grow. The filaments exhibit spectacular dynamics, and hence a study of smectic-A filaments has attracted much attention [1–6].

In the last few decades, various helical structures of chiral lipid bilayers have been found by many researchers [7]. These structures are made from twisting or rolling of planar bilayer sheets from the corners or the edges. We find that smectic-A helical filaments in an isotropic phase are observed experimentally. We note that the smectic-A helical filaments observed here are made from reeling the tubes instead of rolling the sheets. Moreover the molecular arrangement for the smectic-A phase is a layer structure and the

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Address correspondence to Hiroyoshi Naito, Department of Physics and Electronics, Osaka Prefecture University, 1-1 Gakuen-cho, Sakai, Osaka 599-8531, Japan.

molecules exhibiting smectic-A phase have no chirality. Helical myelin figures in egg-yolk lecithin/water system are similar to those of smectic-A filaments in the sense that both molecules have no chirality and both helical structures are made from reeling tubes [8]. However, the myelin figures exhibit a double helical structure instead of a single helical structure as found in smectic-A phase.

In this paper, we report observations of the pattern formation of smectic-A helical filaments grown from an isotropic phase. We show the specific temperature sequence for observation of smectic-A helical filaments whose stability depends essentially on the cooling rate.

EXPERIMENTAL SETUP

Dodecyl alcohol (DODA) was mixed with octyloxycyanobiphenyl (8OCB) (a molar ratio of DODA to 8OCB was 6 to 4) to observe smectic-A filaments in an isotropic phase [1]. The mixture was filled into the liquid crystal (LC) cell with dimensions of $10\,mm \times 10\,mm$ and of thickness $\sim 100\,\mu m$. The LC cells were cooled from the isotropic phase to the coexisting region of the smectic-A and isotropic phases to observe smectic-A helical filaments. The sample temperature was controlled in a hot stage (Instec HS1-i). The pattern formation of smectic-A filaments was observed with a polarizing microscope (Nikon X2TP-11) equipped with a color digital camera (Olympus DP-11).

RESULTS AND DISCUSSION

As the LC cells were cooled from the isotropic phase at $-0.1^\circ C/min$, at $\sim 40^\circ C$ the smectic-A phase initially appears in the form of a number of spherical droplets which grow in size and then begin elongating into cylindrical structures [4]. The cylinders grow rapidly in length but not in diameter and become long and entangled filaments at $\sim 38^\circ C$ (Fig. 1(a)). Some of the entangled filaments suddenly collapse forming compact domains to lower the surface energy of the smectic-A/isotropic interface (Fig. 1(b)). The helical filaments can be found in the entangled filaments on rare occasions but the observation of an isolated helical filament is rather difficult. In order to improve the reproduciblity and frequency of the observation of the helical filaments, we have found the following temperature sequence essential for observing the helical filaments.

The cell temperature is raise $39^\circ C$, and then the remaining filaments become unstable and compulsorily collapse into compact domains on the substrates (Fig. 1(c)). Smectic-A straight filaments are grown from the domains when the LC cells are cooled from $39^\circ C$ at $-0.05^\circ C/min$

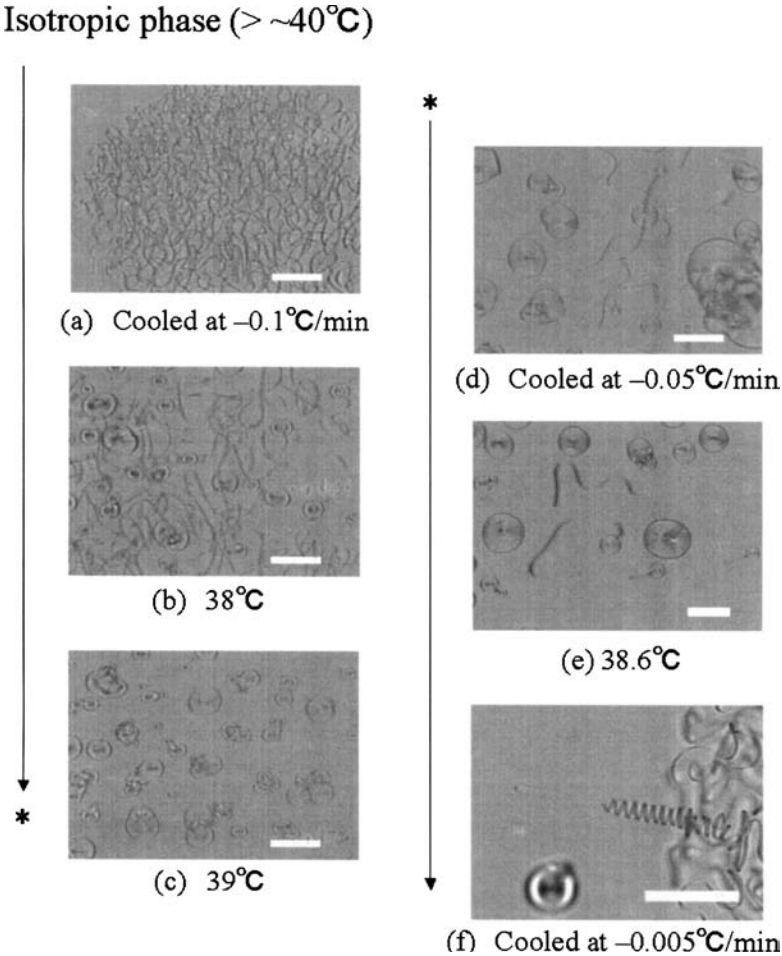

FIGURE 1 Temperature sequence essential for observing smectic-A helical filaments grown from an isotropic phase. The bars indicate 100 μm.

(Fig. 1(d)). Cooling is stopped at 38.6°C and the LC cells are maintained at this temperature for 10 minutes. Some of the domains have a cylindrical structure that has a radius $\sim 5\,\mu m$ and the length of the cylindrical domains become shorter (Fig. 1(e)). The cylindrical domains become an equilibrium shape in 10 minutes. The LC cells are cooled once again at $-0.005^\circ C/min$. Filaments with radii of $2.5 \pm 0.3\,\mu m$ grow from the one end of the cylindrical domains. We note that the growth takes place at the one end of each cylindrical domain, rather than by local elongation,

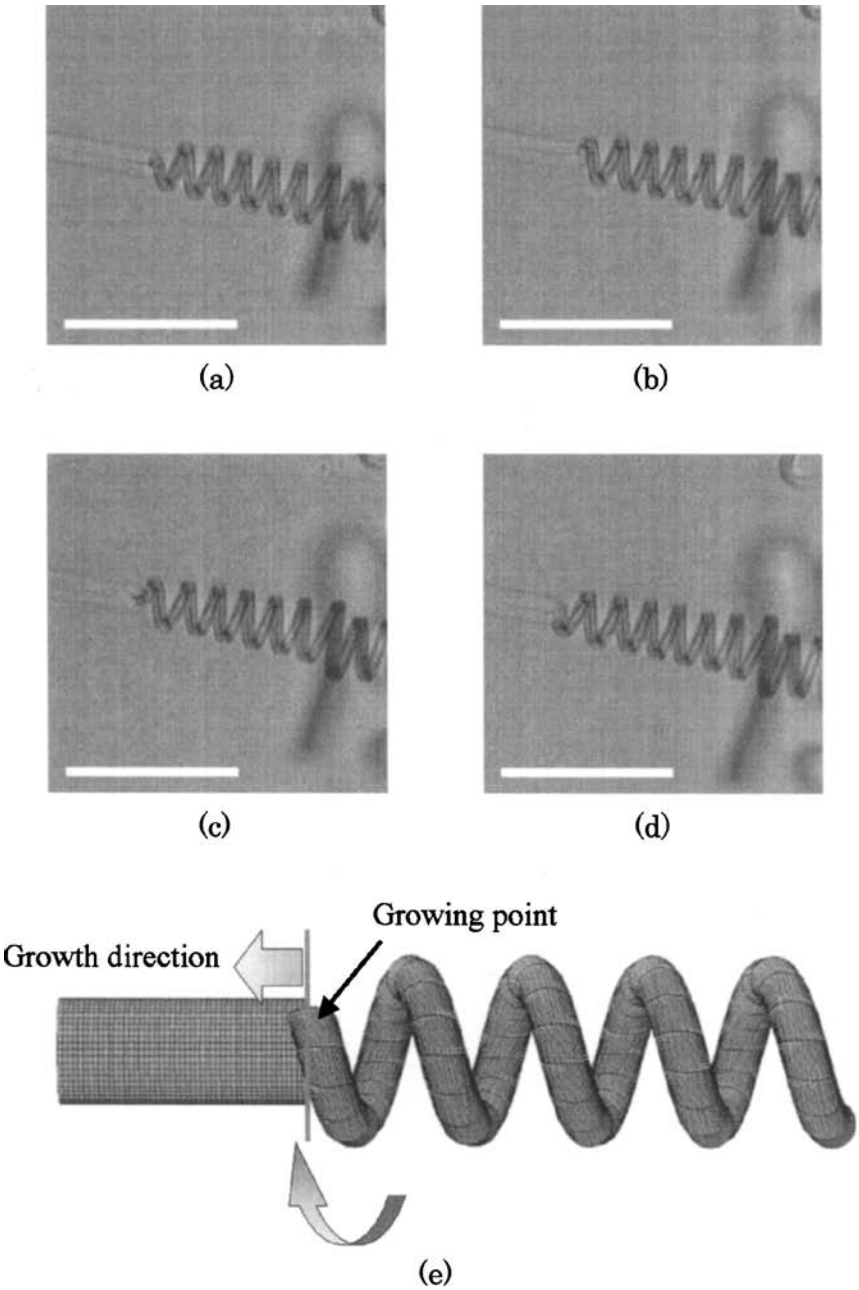

FIGURE 2 (a)–(d) growth process of the smectic-A helical filament, and (e) schematic illustrations of the growth process. The bars indicate 50 μm.

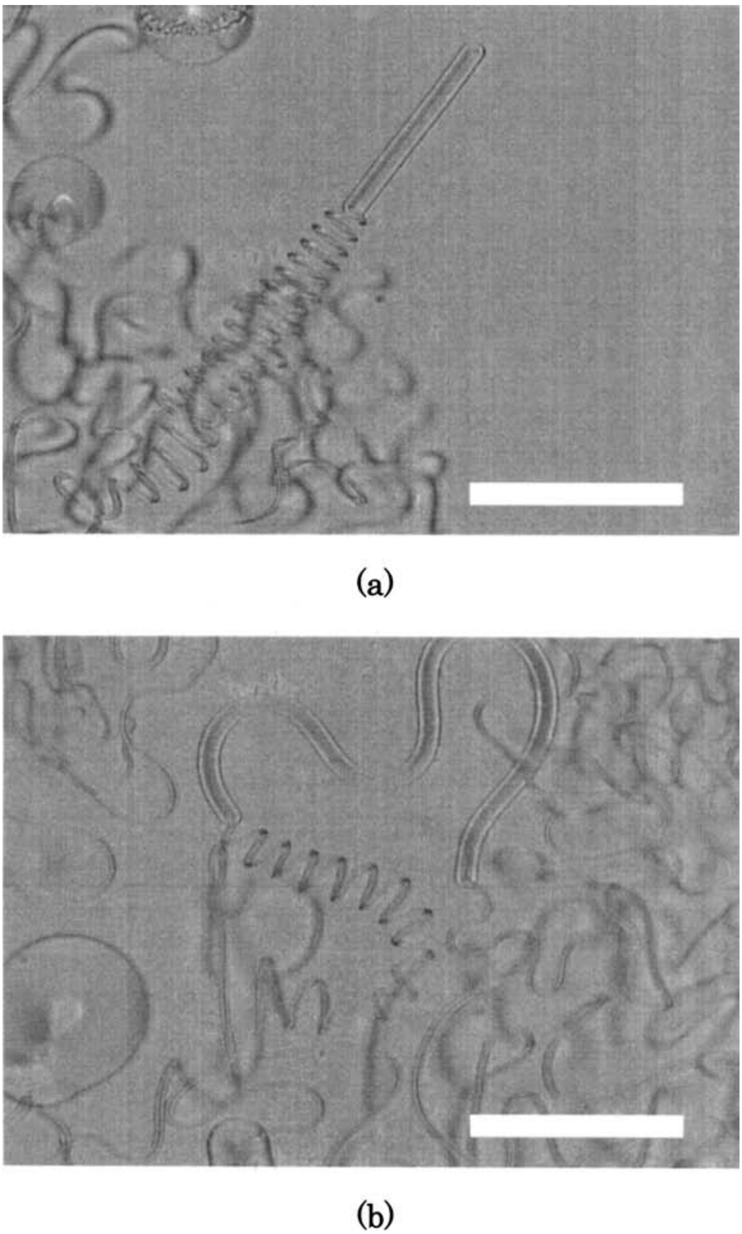

FIGURE 3 (a) left- and (b) right-handed smectic-A helical filaments at a cooling rate of -0.005°C/min. The bars indicate 100 μm.

and that each cylindrical domain retain its size and volume during the growth of the filaments. After attaining the length of $\sim 100\,\mu m$ the straight filaments begin to buckle at about 38.5°C; the form of the filaments gradually changes from straight filaments to wavy filaments in most cases and the filaments buckle continuously. Some of the straight filaments change to helical filaments (Fig. 1(f)). Once the helical filaments are formed, no changes in their spatial pattern are observed during the growth of the helical structure (Fig. 2(a)–(d)), whereas other filaments are continuously changed during the observation (for instance, the spatial pattern of smectic-A filaments observed in the background of Figure 3(a) has been changed as the cell temperature was lowered). Figure 2(e) shows the growth process of the smectic-A helical filament from a cylindrical domain with a larger radius, in which we illustrate the growth direction and the growth of the helical filament by rotating the growing point.

Both the right- and left-handed helices (Fig. 3(a) and 3(b), respectively) were observed in equal probability and no significant differences existed between the left- and right-handed helical filaments in their structures or stabilities (helical filaments observed here have a helical pitch of $9.4 \pm 1.8\,\mu m$ and a radius of $12.1 \pm 1.2\,\mu m$, and are stable for $\sim 60\,s$). These results are qualitatively reasonable since the molecules that exhibit a smectic-A phase have no chirality. Smectic-A helical filaments can also be observed at the cooling rate being either higher or lower than -0.005°C/min but the helical filaments are unstable; the filaments do not retain their shapes and eventually become a three-dimensional entangled pattern. These results indicate that the stability of the helical filaments depends essentially on the cooling rate.

The interpretation of the shapes of helical filaments is an interesting theoretical issue. We consider that the stability of the helical filaments can be explained in terms of minimizing the free energy of smectic-A filaments consisting of the Gibbs free energy difference between the smectic-A and isotropic phases, the surface energy of the smectic-A/isotropic interfaces and the curvature elastic energy of a smectic-A phase [6]. A study along this line is now in progress.

CONCLUSIONS

We have observed the pattern formation of smectic-A helical filaments in an isotropic phase. The features of the smectic-A helical filaments are unique in the sense that the helical filaments are made from reeling of tubules and that the smectic-A molecules have no chirality. We have established the temperature sequence essential to the reproducible observation of smectic-A helical filaments. We have found no significant difference in the helical structures and in their observed frequency between left- and right-handed helical filaments. We have also found from the observations that the stability of the helical filaments depends essentially on the cooling rate.

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