Shrink-Induced Striped Pattern on a Thin Gold Film and Switching of Its Orientation

Masashi Watanabe

Faculty of Textile Science and Technology, Shinshu University, Ueda, Nagano 386-8567, Japan

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ABSTRACT: A well-ordered striped pattern was formed on a thin gold film, which had been deposited using an ion sputtering technique onto an elastic silicone substrate prestretched by 20%. We also prepared a two-layer (silicone/conducting rubber) substrate onto which a thin gold film with a striped pattern was produced using a similar method. By applying a voltage (24 V) between both ends of the conducting rubber to heat the substrate, I succeeded in reversibly changing the orientation of the striped pattern. © 2006 Wiley Periodicals, Inc. J Appl Polym Sci 101: 2040–2044, 2006

Key words: thin films; silicones; self-organization

INTRODUCTION

The spontaneous formations of ordered surface patterns have attracted interest for many years.[1] They are scientifically interesting and there are many potential applications for them. Such patterns are, for example, produced using the wetting instability of a Langmuir-Blodgett film,[2] the evaporation of a polymer solution on a substrate,[3–5] and the photoisomerization of azobenzene-functionalized polymers.[6] Another example uses the ordered pattern that appeared on a thin gold film deposited onto an elastic substrate.[7] Applications of these patterns can be for cell culture substrates,[8] two-dimensional photonic crystals,[9] templates for patterning by electroplating,[10] and diffraction gratings.[7]

These applications are 'static' ones. If one can change the patterns by some stimulus, other 'dynamic' applications will be possible. I also think that this stimulus should be electrically controllable so that one can easily control it. Therefore, in this study, I propose a method to change the patterns using an electrically controllable stimulus.

In an earlier article,[11] I reported the spontaneous formation of a well-ordered striped pattern on a thin gold film. This pattern was produced by depositing gold onto a prestretched silicone rubber, using an ion sputtering technique. This method was very simple, but the obtained pattern was highly ordered. Some other groups also reported patterns on thin metal films deposited onto an elastic substrate.[12–17] Their methods required a precreated pattern (e.g., a low relief pattern[7] and a patterned mold[15]) to obtain ordered patterns. However, the method discussed here does not require such a previously created pattern. This allows for a very simple procedure for producing a striped pattern on a thin metal film.

In this study, I investigated a method to change the orientation of the striped pattern by a stimulus that can be electrically controlled. I succeeded in reversibly switching the orientation. This switching method may find applications for optical devices.

EXPERIMENTAL

I prepared single- and two-layer substrates. First, the single-layer substrate, which consisted of a silicone sheet, was prepared from a two-liquid type room temperature vulcanizing (RTV) silicone (KE-109-A and B, Shin-Etsu Chemical, Tokyo, Japan). The KE-109-A (5.0 g) and KE-109-B (5.0 g) were mixed and degassed under vacuum to remove the foam. The mixture was spread on a Teflon plate, which was then placed in an oven controlled at 100°C for 1 h. The vulcanized silicone was obtained as a sheet (~0.7 mm thick) and then removed from the Teflon plate. The silicone sheet was then cut into a rectangular strip 75 mm long and 15 mm wide.

The two-layer substrate consisted of a silicone upper layer and a conducting rubber lower layer. The silicone was the two-liquid type RTV silicone mentioned earlier. The conducting rubber was purchased from Kinugawa Rubber Industrial, Chiba, Japan, (No. S60; volume resistivity, 1 Ω/cm) as a 0.5-mm thick sheet. A strip of the conducting rubber (75×15 mm^2) was stuck on a Teflon tray using a double-faced ad-

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Correspondence to: M. Watanabe (mwatana@giptc.shinshu-u.ac.jp).

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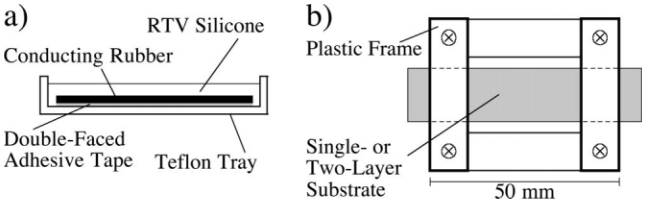

Figure 1 (a) Preparation of the two-layer substrate. (b) Frame to hold the stretched substrate.

hesive tape [Fig. 1(a)]. A mixture (1.5 g) of the RTV silicone (KE-109-A/B = 50/50 wt %) was poured into the tray, which was then placed in an oven controlled at 100°C for 1 h to vulcanize the silicone. The obtained two-layer substrate was removed from the Teflon tray. It was then cut into a rectangular strip 75 mm long and 15 mm wide. The thicknesses of the upper and lower layers were about 0.8 and 0.5 mm, respectively.

Gold deposition onto each substrate was carried out as follows. A homemade plastic frame [Fig. 1(b)] held the above single- or two-layer substrate that was prestretched by 20%. A thin gold film (~30 nm thick) was deposited onto the substrate by an ion-sputtering technique using an ion coater (IB-3; Eiko Engineering, Hitachinaka, Japan).

The surface of the gold film was then observed using a light microscope (BH-2; Olympus, Tokyo, Japan) without relaxing the stretched substrate. I also observed the surface while heating it by applying a DC voltage (24 V) to both ends of the conducting rubber (i.e., the lower layer).

RESULTS AND DISCUSSION

Gold deposition onto the single-layer substrate

Gold was deposited using an ion-sputtering technique, onto the single-layer substrate that was prestretched by 20%. The thickness of the deposited gold film was about 30 nm, which was estimated based on the data provided by the manufacturer of the sputtering apparatus. I observed the gold film surface, using a light microscope without releasing the stretching of the substrate. A well-ordered striped pattern was observed (Fig. 2).

Although I have already reported the details of such a striped pattern formation, its mechanism is as follows [Figs. 3(a–c)]. During the deposition by ion sputtering, the surface of the substrate (silicone) is automatically heated and expanded [Fig. 3(b)]. The surface temperature during the deposition was estimated to be about 68°C, using the method reported by Ohzono et al.[18] (I placed the substrate held on the frame in an oven and measured the temperature at which the surface clouded because of the striped pattern on it and became a mirror surface.) After the deposition, the surface is cooled to room temperature and shrinks [Fig. 3(c)]. Because the thermal expansion coefficient of gold (1.4×10^{-5} K^{-1}) is much smaller than that of silicone (9.6×10^{-4} K^{-1}),[19] the surface becomes wavy. However, because the substrate is stretched and fixed on the frame, it shrinks only in the direction perpendicular to the elongation axis. As a result, a striped pattern parallel to the axis forms [Fig. 3(c)].

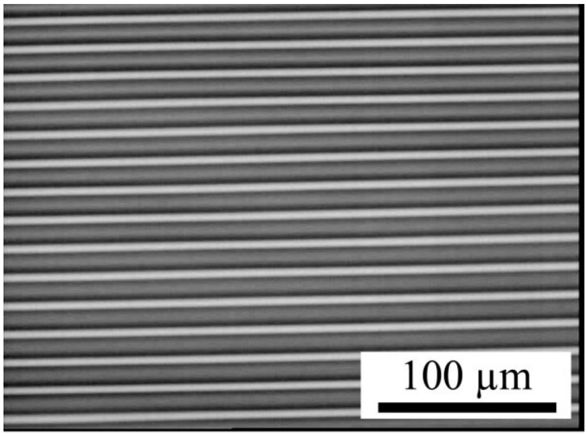

Figure 2 Optical micrograph of the stripe-patterned gold film.

I took care of the following two points to obtain the well-ordered pattern.[1] The single-layer substrate was prepared by vulcanizing the RTV-silicone liquid spread on a Teflon plate. I deposited gold onto its upper surface because the backside, which had been in contact with the Teflon plate during the preparation, was not sufficiently smooth.[2] During the gold deposi-

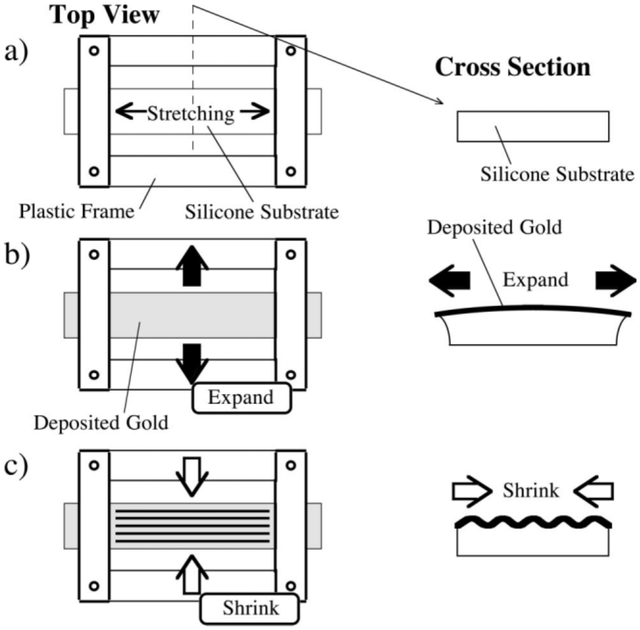

Figure 3 Mechanism of the striped-pattern formation.

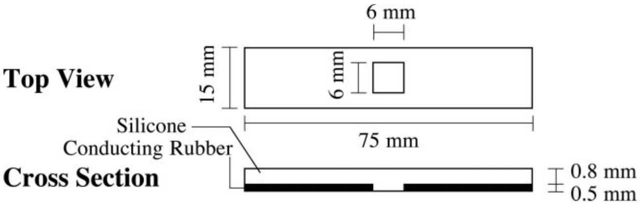

Figure 4 Two-layer substrate having a square hole in the lower layer.

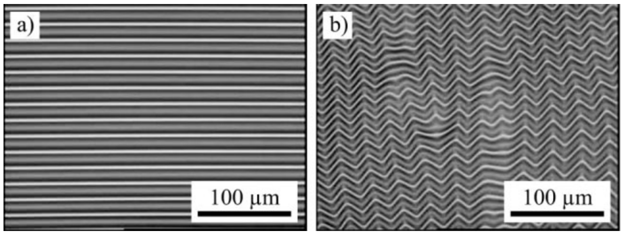

Figure 5 Gold film (a) in and (b) out of the hole.

tion by the ion sputtering, I did not allow the backside of the substrate to be in contact with the sample stage of the sputtering apparatus. When the backside was in contact with the stage, this contact hindered the freedom of the thermal expansion of the substrate. As a result, the obtained pattern was not ordered.

Gold deposition onto the conducting rubber

I deposited gold onto the conducting rubber, intending that a voltage applied to the rubber might generate heat and stimulate the deposited gold film. However, I failed to obtain a well-ordered striped pattern on the gold film. The conducting rubber was a silicone that contained carbon black as conducting filler. Therefore, its surface was not sufficiently smooth, and its thermal expansion was not homogeneous on a micrometer scale. These could be the reasons for the unordered pattern.

Conducting rubber/silicone two-layer substrate

I prepared a two-layer substrate, which consisted of silicone (the upper layer) and conducting rubber (the lower layer). I expected to obtain a well-ordered pattern on the deposited gold film because the upper layer was smooth and homogeneous. However, the gold deposition onto the substrate while stretched did not give a well-ordered striped pattern. Although the upper surface was smooth, the nonhomogeneity of the lower layer seemed to affect the pattern formation.

To remove such an undesirable effect by the lower layer, I made a square hole in the center of this layer (Fig. 4). I deposited gold onto this substrate while stretched by 20%. As a result, a well-ordered striped pattern was successfully obtained in the square region over the central hole of the underlying conducting rubber [Fig. 5(a)]. However, an unordered zigzag pattern was observed outside the square region [Fig. 5(b)].

Heating the substrate by applying a voltage to the conducting rubber

The two-layer substrate (Fig. 4) was heated by applying a DC voltage (24 V) between both ends of the underlying conducting rubber. I then observed the gold film on the substrate, using a light microscope. A major part of the initial horizontal striped-pattern changed to a vertical one within 90 s by the voltage application (Fig. 6).

The following descriptions are details of this changing process (Fig. 7). The striped pattern was initially oriented in the horizontal direction, which was parallel to the elongation axis of the substrate [Fig. 7(a)]. Each line of this pattern then had slight constrictions

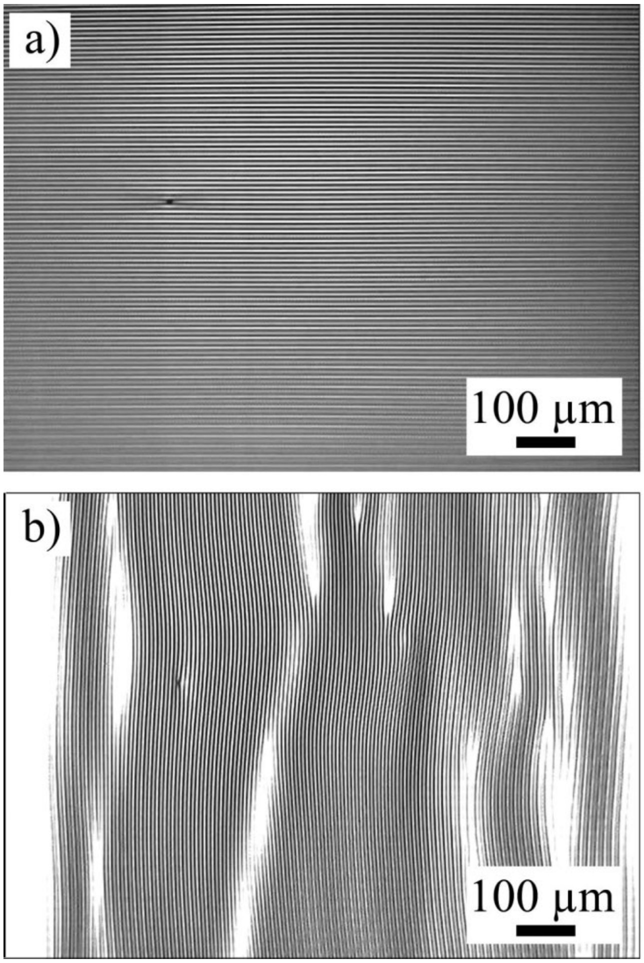

Figure 6 Striped pattern of the gold film (a) before and (b) during voltage application, across the conducting rubber layer.

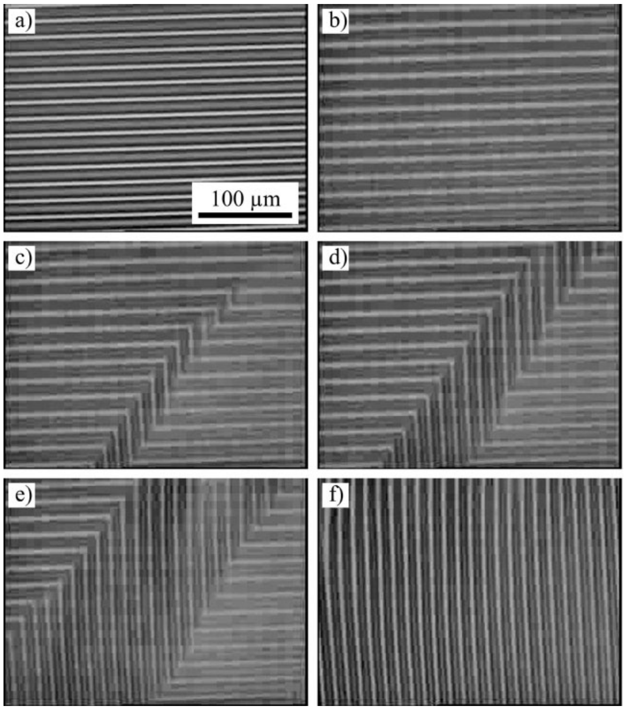

Figure 7 Change in orientation of the striped pattern at (a) 0, (b) 49, (c) 50, (d) 51, (e) 52, and (f) 90 s from the beginning of the voltage application.

[Fig. 7(b)]. Suddenly, vertical short lines appeared in part of the visual field [Fig. 7(c)], which increased in length and number as if dominoes had fallen down, [Figs. 7(d) and 7(e)] finally, all lines in the visual field of the microscope (0.32 × 0.24 mm^2) were oriented in the vertical direction [Fig. 7(f)]. By interrupting the voltage, these vertical lines returned to the horizontal ones within 70 s (Fig. 8). Thus, the orientation of the striped pattern could be reversibly switched. This switching could be repeated several times, though cracks appeared on the gold film by repeating the switching (Fig. 9).

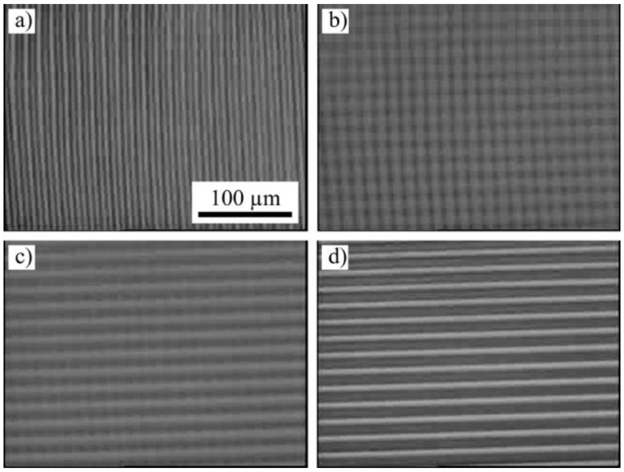

Figure 8 Change in orientation of the striped pattern at (a) 8, (b) 39, (c) 46, and (d) 68 s after the voltage application was interrupted.

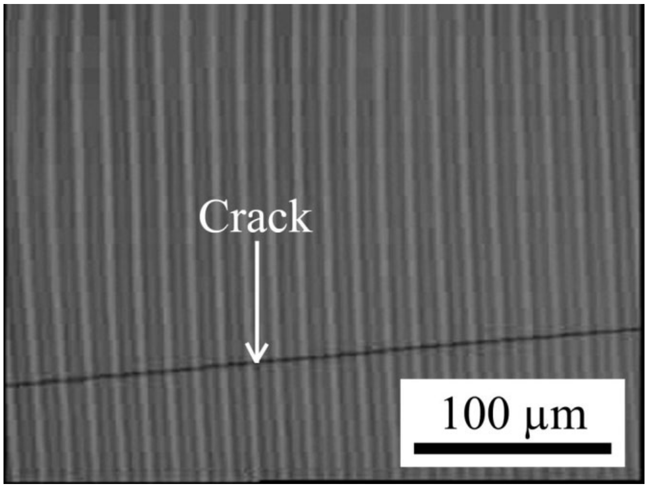

Figure 9 A crack appeared on the gold film during the voltage application.

As shown in Figure 8, when the heating was interrupted by the cessation of the voltage application, I observed a uniform change in the orientation of the striped pattern on the gold film. However, the change during the heating process did not proceed homogeneously (Fig. 7). Because the gold film was heated only at the edge of the square area in the center of the sample (see Fig. 4), this heating was not homogeneous. However, the film was homogeneously cooled in air after the heating was ceased. Therefore, the change in the orientation of the pattern uniformly took place.

The temperature of the substrate was measured while the voltage was applied. Using an infrared thermometer, I measured the temperature of the backside (i.e., the conducting rubber side) of the substrate. The temperature reached about 75°C by application of 24 V for 90 s; this was comparable to the response time of the orientation switching of the striped pattern. In addition, this temperature (75°C) was slightly greater than that during the gold deposition (~68°C) as described earlier. This increase in temperature could cause the substrate to expand only in the direction perpendicular to the elongation axis, because the substrate was stretched along this axis and was fixed on the frame. Therefore, I considered that such an expansion changed the orientation of the striped pattern from a horizontal one to a vertical one.

CONCLUSIONS

A well-ordered striped pattern forms on a thin gold film, which is deposited on a stretched silicone substrate. By heating the substrate, using a conducting rubber as the heating element, the orientation of the striped pattern can be reversibly switched.

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