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Laser-induced wavy pattern formation in metal thin films

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Laser-induced well-ordered and controllable wavy patterns are constructed in the deposited metal thin film. The micrometer-sized structure and orientation of the wavy patterns can be controlled via scanning a different size of rectangle laser spot on the films. Ordered patterns such as aligned, crossed, and whirled wave structures were designed over large areas. This patterning technique may find applications in both exploring the reliability and mechanical properties of thin films, and fabricating microfluidic devices. © *2004 American Institute of Physics*. [DOI: 10.1063/1.1787891]

There is much current interest in the induced micropattern formation technique for their potential applications in microdevices.[1–5] Various patterns with interesting morphologies such as hillocks,[6] straight blisters,[7] and wavy patterns,[8–12] have been reported formation in compressed thin film-resulted from the thermal-induced mechanism. Some of these morphologies, such as the wavy patterns, have been ubiquitously observed in many different film-substrate systems, including deposited metal,[13–15] covalent-ionic compounds,[16] and diamondlike carbon films[17–19] on Si, glass, and steel substrates, respectively, which hint at a certain universality of underlying mechanics. The complexity of the blister shapes and the patterns has both fascinated and befuddled investigators, especially when the experimental and theoretical knowledge of the detailed mechanism is still unclear and the patterns are still unpredictable and uncontrollable.[20–26] Here, a laser-induced micropattern technique is reported to realize the controllability and be very helpful to the explanation of this phenomenon.

Controllable wavy pattern formation in thin films in this study is presented in a schematic diagram [Figs. 1(a)–1(d)]. Aluminum films with typical thickness of 300 nm were deposited onto a glass substrate by an ac magnetron sputtering technique. During sputtering deposition, the substrate temperature was kept at 120 °C, and then the system was cooled down slowly to room temperature (25 °C) [Fig. 1(a)]. With that, a rectangular laser beam focused on the area of smooth surface [Fig. 1(b)]. The instantaneous injection of the thermal flux separated the irradiated region of the film from the substrate immediately due to the abrupt thermal expansion of the film and the large elastic mismatch between the film and the substrate. Then the film, as shown schematically in Fig. 1(c), delaminated from the substrate and deformed into wrinkles right away through the release of the internal compressive stress. The formation of the wavy pattern in the delaminated region is due to the existence of the isotropic biaxial compression in the film [Fig. 1(d)]. This phenomenon is very sensitive to the laser flux with a certain range of energy when the compressive stress existed in thin film-substrate combinations.

The process of the wavy structure formation is recorded by the Figs. 1(e)–1(g). The laser pulse, with a pulse energy of 600 J/m^2 and a spot size of 12.5×7.5 μm^2, was applied on the region marked by the dashed rectangular frame [Fig. 1(e)]. Here, the "M"-marked region was used for a point of reference. Figure 1(f) shows the formation of the initial wrinkle. With the movement of the laser beam along the film

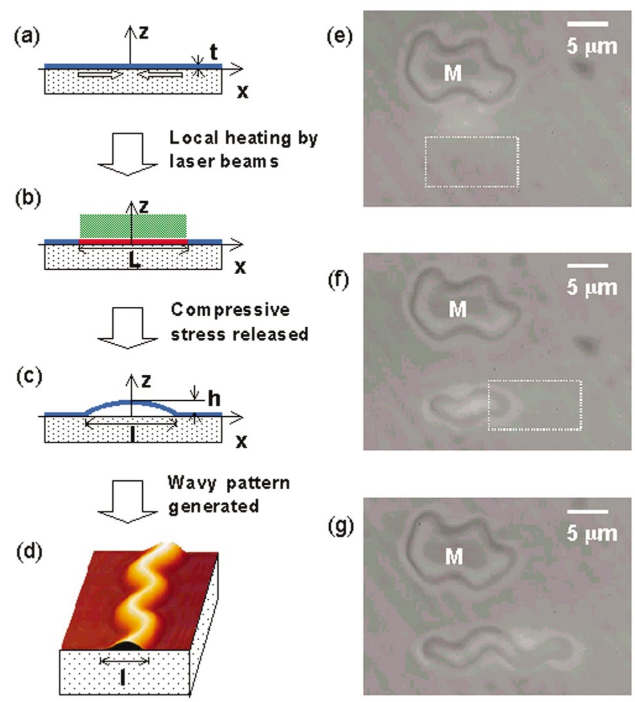

FIG. 1. (Color) The sketch for the process of the wavy structure formation. Aluminum films, with a typical thickness of 300 nm, were deposited onto a glass substrate using an ac magnetron sputtering technique (a). The isotropic biaxial compressive stresses existed in the well-bonded films. A laser pulse with the width *L* heated the film and increased the film stresses transitorily to overcome the bonding force (b). As a result, the film delaminated from the substrate (c) and the wavy pattern generated under the biaxial compressive stress (d). The wrinkle width *l* and the maximum height *h* are given in the sketch. The practices are recorded by the micrographs (e)–(g), where "M" is marked on an accidental damaged region of the smooth film as a reference. A Nd:YAG laser (New Wave Research QuikLaze™) with wavelength of 532 nm, energy of 600 J/m^2, and spot size of 12.5×7.5 μm^2 was used here. The dashed rectangular frame in (e) and (f) marked the irradiated area. Then the wavy patterns induced by the first and the following second laser pulse, respectively, are shown in (f) and (g).

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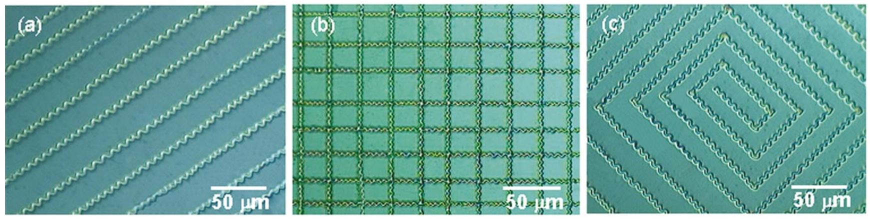

FIG. 2. (Color) Optical micrographs of the ordered patterns induced by the different laser-scanning mode: (a), aligned structures parallel to each other, (b), crosslinked net, and (c), whirled wavy patterns.

surface [dashed rectangular frame in Fig. 1(f)], a single wrinkle developed into a wavy structure, as shown in Fig. 1(g).

More interestingly, various wavy patterns can be constructed on the film surface by controlling the scanning regime of the laser beam. When the laser spot scanned the metal film in one direction, a preferred direction of growth was obtained by arrays of cords growing parallel to each other [Fig. 2(a)]. These arrays covered the entire irradiated part of the film. Similarly, if the rectangle spot was controlled to scan the film surface alternatively in the longitudinal and transversal directions, the crosslinked pattern could be well constructed [Fig. 2(b)]. Via controlling the irradiated area by a computer-assisted design system, complicated wavy patterns could be generated arbitrarily, such as whirled wavy patterns [Fig. 2(c)].

To study the main factors and mechanism dominated the formation of the delaminated wavy structure, the laser beam spots of different sizes were employed to induce the pattern formation. The gradually changed wavy structures were constructed on the same metal film with the different experimental conditions. Figures 3(a)–3(c) displayed a series of wrinkle widths l, increased with the laser spot width L.

It was found that there is an ultimate value of wrinkle

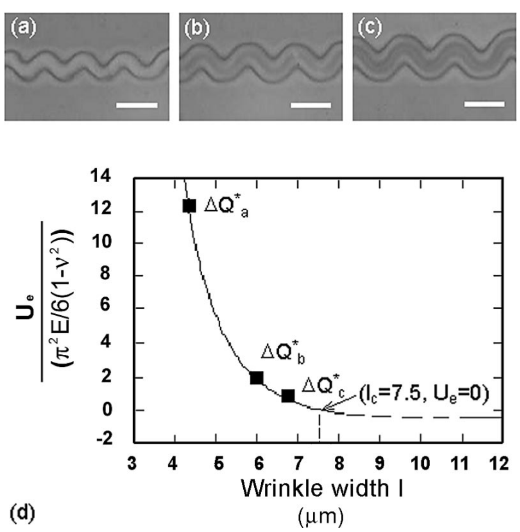

FIG. 3. The influence of the wrinkle width l on elastic energy U_e. The width l is measured from optical micrographs, (a)–(c), which exhibit the different widths of wavy wrinkles induced by different laser spot. Here all scale bars are 10 μm. (d) Model simulation of the dependence between U_e and the width l. This theoretical curve is calculated by substituting $t=0.3$ μm and $\varepsilon=-7.7\times10^{-3}$ into Eq. (1b). The vertical dashed line indicates the wavy pattern with critical width $l_c=7.5$ μm and minimum residual internal elastic energy $U_e=0$. When the estimated Young's modulus, $E=140$ GPa, was applied to Eq. (3), the experimental data points with different ΔQ_x^*, labeled with ■, fitted the simulated results well.

width l for a given specimen, when the laser spot width L increased over a threshold value. To describe the change trend of the wrinkle width l, a concept of internal energy was introduced.[27] $\Delta U=U_0-U_e$ is the internal energy release per unit area for the delamination of the film, where U_0 and U_e is the internal energy per unit area before and after the formation of the wavy patterns, respectively. Generally, the unit-area internal energy can be formulated as follows:[27]

$$U_0 = \frac{Et\varepsilon^2}{2(1-\nu^2)}, \tag{1a}$$

$$U_e = \frac{Et\pi^2\varepsilon^2}{6(1-\nu^2)}\left[\left\{\frac{2}{\varepsilon}\left(\frac{t}{l}\right)^2 - \frac{\pi^2}{\varepsilon^2}\left(\frac{t}{l}\right)^4\right\}\right], \tag{1b}$$

where E is the Young's modulus of the film, ν is the Poisson's ratio, and ε is the strain under a given film thickness, t.

In the viewpoint of the energy balance, the film is delaminated when

$$\Delta U + Q_{eff}^* = 2\gamma. \tag{2}$$

Here, γ is the bonding energy, and Q_{eff}^* can be regarded as the minimum heat flux applied on the film that can help the irradiated area to overcome the bonding force and initiate the onset of the film delamination. For the critical width l_c, $Q_{\rm eff}^*$ reached its minimum value $Q_{\rm c}^*$ due to the balance between the thermal energy and the intrinsic energy, and the energy difference ΔQ^* can be expressed by following relation:

$$\Delta Q^* = U_e - U_c = U_e \approx 3.3E\left[59.3\left(\frac{1}{l}\right)^4 - \left(\frac{1}{l}\right)^2\right]. \tag{3}$$

This equation indicated the relation between ΔQ^* and the wrinkle width l, if the Young's modulus E was given.

The experimental results, ΔQ_a^*, ΔQ_b^*, and ΔQ_c^*, are in satisfactory agreement with the calculated U_e vs l curve depicted in Fig. 3(d), when the Young's modulus $E=140$ GPa is adopted in Eq. (3). Another important factor of the bonding energy γ can be obtained by substituting E, l, and Q_{eff}^* into Eq. (1a), (1b), and (2). Here, the calculated value of 20 J/m^2 was obtained for γ. The values for E and γ estimated here are successfully in the same order of magnitude reported before.[27,28]

This pattern-formation technique could be used in microdevices, as the metal films are detached from substrates and locally formed well-shaped hermetic tunnels between the films and the substrates. When a laser beam scanned over an existed wavy wrinkle, the metal thin films can be constructed to trirouteway and quadrirouteway patterns (see EPAPS Ref. 29). More significantly, the trirouteway and quadrirouteway patterns can be converted to each other with laser-induced treatment, which may be used as laser-controlled valve. So, laser-controlled wavy structures on a thin films-substrate system can be designed in intricate order patterns, and especially, can be constructed to a trirouteway or quadrirouteway pattern, which may be potentially used in microfluidic reactors.

In summary, a technique has been shown to fabricate the well-ordered delaminated pattern on a thin film. Several kinds of the delaminated wavy structures were successfully constructed on an aluminium film, and the basic mechanical properties of the film, such as E and γ, were determined. This controllable patterning technique may open a way to construct the complicated structures for microdevices research.

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[29]See EPAPS Document No. E-APPLAB-85-013435 for the micrographs of trirouteway and quadrirouteway patterns. A direct link to this document may be found in the online article's HTML reference section. The document may also be reached via the EPAPS homepage (http://www.aip.org/pubservs/epaps.html) or from ftp.aip.org in the directory /epaps/. See the EPAPS homepage for more information.