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Expansion effect of liquid substrates on the ordered structures in the Al films

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Abstract

An aluminum (Al) film system deposited on silicone oil drop surfaces by thermal evaporation method has been fabricated. A characteristic ordered pattern, namely, band, is observed in the continuous Al film system. Each band is composed of a large number of parallel key-shaped domains, which formed naturally during the deposition. It is found that these bands develop along the circumference of the silicone oil substrates and the length of the key-shaped domains show a marked dependence on the liquid expanding speed and other deposition conditions.

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Keywords: Metallic thin films; Ordered structure; Substrate expansion

1. Introduction

Most recent research effort has been directed towards the first stage of the mechanical failure of thin films. Stress distribution, stress origin, edge effects, grain boundaries, plastic flow and phase changes under stress are some of the topics treated in the recent literatures [1,2]. Films and coatings fabricated by vapor deposition, sputtering, etc., often develop residual compressive stresses during the deposition process itself [3,4]. Additional residual stresses may be induced during cooling when a thermal expansion mismatch exists between the films and the substrates [5,6]. Thin films containing such large residual compressive stress are susceptible to delamination and spalling or, when exposed to atmospheric conditions [7,8], deformation, which may result in very interesting topographical patterns [9–12]. Recently, it is reported that some characteristic microstructures may exist in the metallic film systems owing to the expansive and mobile nature of the silicone oil substrates [13–15]. Therefore, it is naturally speculated that various characteristic patterns, mirroring the internal stress distribution and evolution in the films, may occur apparently in these nearly free sustained films [16].

In this Letter, we report a large scale ordered pattern existing in a continuous aluminum (Al) film system deposited on silicone oil drop surfaces. The most interesting phenomenon is that the ordered pattern, namely band, develops along the circumferences of the silicone oil drops. The formation mechanism of the ordered pattern can be understood with the aid of experiments comparing different film thickness, deposition rates, and the expansion speed of the oil drops.

2. Experiment

The samples were fabricated by thermal evaporation of 99.99% pure Al in a vacuum chamber of 6×10^{-4} Pa at room temperature. A drop of pure commercial silicone oil (Dow Corning 705 Diffusion Pump Fluid, saturated vapor pressure $<10^{-8}$ Pa) with a diameter $\phi = 2$–4 mm was dripped on a piece of glass surface, which was fixed 190 mm above the evaporating filament (tungsten). The deposition rate f and the nominal film thickness d were determined by a quartz-crystal thickness monitor (a step-200 profilometer, TENCOR), which was located just beside the substrate. After the samples were removed from the vacuum system, all images for the surface morphologies of the

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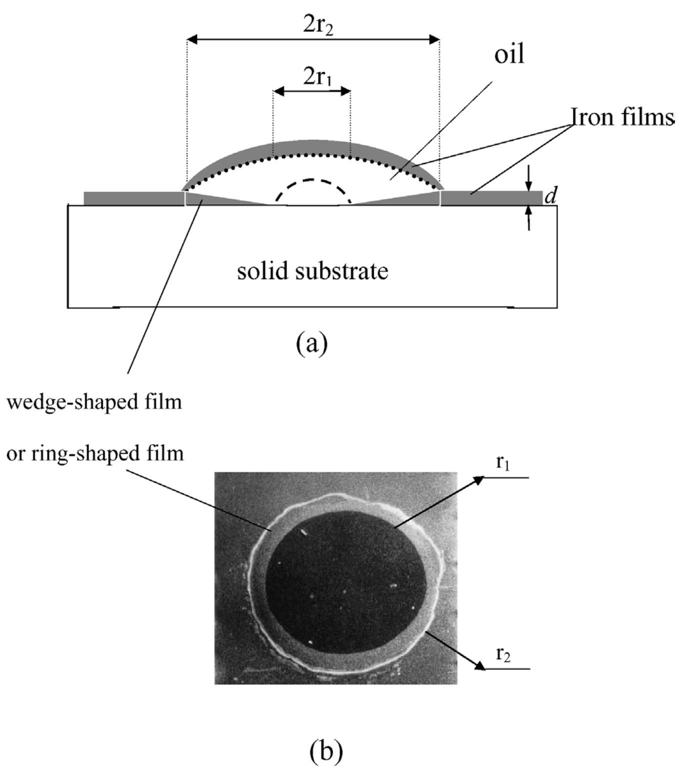

Fig. 1. (a) Schematic view of the Al films deposited on both the glass and oil drop surfaces. The shaped parts are the Al films. The dashed and dotted curves denote the outlines of the oil drop before and after deposition. (b) Photograph of the ring-shaped film on a glass surface (1.0 cm × 1.1 cm).

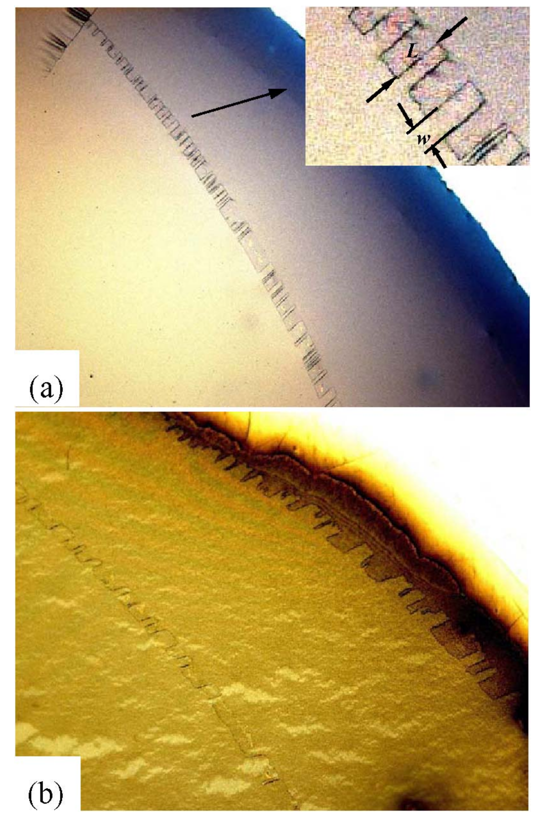

Fig. 2. Typical optical micrographs (reflection mode) of the bands existing in the Al films deposited on the oil drop surfaces. (a) $f = 0.05$ nm/s, $d = 70.0$ nm with image size of 1440×1065 μm^2; the inset is the higher-resolution image of the bands with image size of 180×130 μm^2; (b) $f = 0.10$ nm/s, $d = 40.0$ nm with image size of 30×15 μm^2.

samples were taken immediately with an optical microscope, equipped with a CCD camera (Leica DMLM and Leica DC 300).

3. Results and discussion

The schematic view of the Al film deposited on the oil drop surface is shown in Fig. 1. An important phenomenon in the experiment is that the radius of the oil drop expanded steadily during deposition mainly due to the heat radiation from the filament and the bombardment of the deposition atoms [13,14], which is the key property of the liquid drops used in our experiment. For each sample, there is a ring-shaped Al film on the glass surface, as shown in Fig. 1(b). The film thickness in the ring-shaped film increases linearly from the inner radius r_1 to the outer radius r_2 approximately, indicating that the oil drop expands and its radius r (or its radius increment $\Delta r = r_2 - r_1$) increases uniformly during the deposition [14].

The typical ordered patterns, i.e., the bands, in the Al films deposited on the silicone oil drop surfaces are shown in Fig. 2. The bands are consisted of a large number of parallel key-shaped domains, namely keys, and generally the neighboring keys possess different width w but nearly uniform length L (see Fig. 2). The most interesting observation is that the bands almost develop along the circumferences of the silicon oil drops. In Fig. 2(b) we can see that two bands exist together along the film edge, which is approximately perpendicular to the expansion direction of the oil drop. In the experiment, we find that the average length of the keys in the bands, i.e., the average length of the bands, near the oil drop edges are usually larger than that of the bands which are located far away from the substrate edges (see Fig. 2(b)). The total length of each band in our experiment may be more than 10 mm. When the bands extend, they may undergo a gradual and slight change in their propagation directions. Depending on the nominal film thickness d, deposition rate f and the locations, the size of the keys varies widely from a few micrometers to several hundred micrometers. The maximum length and width of the keys observed in our experiment are $L_m \approx 180$ μm and $w_m \approx 400$ μm, respectively.

Many previous works have reported that during stress relief the film materials always orderly organize in some regions driven by the internal stress and form characteristic bucking patterns such as sinusoidal shapes [17,18]. We propose here that the ordered patterns shown in Fig. 2 also originate from spontaneous material organization owing to the stress relief. According to the previous studies [19–21], strong and detectable residual internal stress always exists in metallic films deposited on liquid substrates and characteristic stress-induced surface morphologies can be observed in these nearly free sustained films. We believe that the Al films and the silicon oil substrates both expand during deposition owing to the heat radiation from the