Analysis of laser-assisted chemical etching processes of a pinhole by monitoring diffraction patterns of reflected beams

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The detailed processes of laser-assisted chemical etching are studied by using an optical in situ monitoring method. Various Al thin-film specimens are processed in aqueous H₃PO₄ solutions by shining focused Ar⁺ laser beams. As etching progresses, it is observed in the backreflected laser beam that a concentric circular fringe pattern appears and shrinks. The development of the etching process is explained by analyzing the diffraction patterns with an etched shape function of a Gaussian nature.

Key words: Diffraction pattern, laser-assisted chemical etching.

1. Introduction

Laser-assisted chemical etching of metallic thin films has been attracting continuous attention as a laser microfabrication technique. An appropriate laser beam is focused onto a metallic thin film that is immersed in an aqueous solution or gas environment of an etchant. In this case the laser beam is simply a heat source for elevating the temperature of a region of a material in order to accelerate pyrolytic reaction.

Both reflected and transmitted laser beams have been used in obtaining information on the surface morphology or the process dynamics.

In the present experiment we also use both reflected and transmitted laser beams simultaneously to obtain detailed information on the etching processes taking place within the beam waist of the focused Ar⁺ laser beam. The reflected laser beam is projected onto a screen and concentric circular fringes are observed. A similar ring fringe pattern was observed previously by Bloch and Zeiri in a laser-induced hole formation on Cu thin film, but a detailed theoretical explanation was not given. We attempt to explain systematically the ring fringe pattern by using a diffraction theory applied to the reflected laser beam.

2. Diffraction of the Reflected Laser Beam

In Fig. 1 the P₀ plane denotes the surface of the film, P₁ is the boundary of an etchant with a refractive index nE, T is the focusing lens with a focal length of f, and P₃ is the screen. In the case of pinhole formation by laser-assisted chemical etching of thin films, the focused laser beam passing through the focusing lens T is backreflected from the surface P₀, where the reflected complex light field is denoted by g₀(ro) (which has cylindrical symmetry). The complex light distribution g₃(r₃) at the image plane P₃ is written as

\[
g₃(r₃) = Cₒ \int r₂dr₂ \int r₁dr₁ \int r₀dr₀g₀(r₀) \times \exp \left[ \frac{ik₀nE}{2l₀₁}(r₀^2 + r₁^2) \right] J₀ \left( \frac{k₀nEro₁}{l₀₁} \right) \times \exp \left[ \frac{ik₀}{2l₁₂}(r₁^2 + r₂^2) \right] J₀ \left( \frac{k₀ro₂}{l₁₂} \right) \times \exp \left( - \frac{ik₀}{2f} r₂^2 \right) \exp \left[ \frac{ik₀}{2l₂₃}(r₂^2 + r₃^2) \right] J₀ \left( \frac{k₀ro₃}{l₂₃} \right), \tag{1} \]

where Cₒ is a proportionality complex constant, k₀ is the wave number in free space, and J₀ is the zero-order Bessel function of the first kind. Since the integrations over P₁ and T are assumed to be taken from 0 to ∞ for r₁ and r₂, Eq. (1) may be written as

\[
g₃(r₃) = C₁ \int r₀dr₀g₀(r₀) \exp \left[ \frac{ik₀}{2l₀₂}(1 - \frac{L₂}{l₀₂})r₀^2 \right] \times J₀ \left( \frac{k₀L₂ro₃}{l₀₂l₂₃} \right), \tag{2} \]
where $C_1$ is another complex constant and

$$l_{02} = l_{01}/n_E + l_{12},$$

$$\frac{1}{L_2} = \frac{1}{l_{02}} - \frac{1}{f} + \frac{1}{l_{23}}.$$  \hfill (3)

If we know the complex light field $g_0(r_0)$ at the $P_0$ plane, we can calculate numerically the complex light field $g_3(r_3)$ at the $P_3$ plane.

Here the laser beam that is focused onto the film surface has a Gaussian intensity profile and so we assume a Gaussian shape for the etched film, as shown in Figs. 2(a) and 2(b). This assumption is supported by the micrographs discussed below in Section 4. Thus we introduce the etched shape function $S(r_0)$ as

$$S(r_0) = A_E \exp(-r_0^2/\omega_E^2),$$ \hfill (4)

where $A_E$ and $\omega_E$ are the parameters representing the etching depth and etching width, respectively. Then the reflected complex light field $g_0(r_0)$ at the $P_0$ plane can be written as

$$g_0(r_0) = C_2 R_F \exp(-r_0^2/\omega_0^2) \exp[-2ik_0n_ES(r_0)],$$ \hfill (5)

where $C_2$ is a complex constant, $\omega_0$ is the beam waist radius ($1/e^2$ intensity point) of the incident laser beam at the thin-film surface, and $R_F$ is the amplitude reflection coefficient of the thin film, which depends on the remaining film thickness $d(r_0)$ and $N_F(=n_F + ik_F)$, $n_S$, and $n_E$, the refractive indices of the thin film, the substrate, and the etchant, respectively. When the thin film is not pierced yet ($A_E < \omega_0$, the initial film thickness), the remaining film thickness $d(r_0)$ is simply

$$d(r_0) = \omega_0 - S(r_0),$$ \hfill (6)
as shown in Fig. 2(a). But when the thin film is pierced \( A_E > d_0 \), \( d(r_0) \) is given by

\[
d(r_0) = \begin{cases} 
  d_0 - S(r_0) & \text{if } r_0 > r_E \\
  0 & \text{otherwise}
\end{cases}
\]

where \( r_E = \frac{w_E}{2} \ln(A_E/d_0) \) is the radius of an etched pinhole, as shown in Fig. 2(b). If we know \( A_E \) and \( w_E \) in \( S(r_0) \), we can calculate the reflected amplitude \( g_3(r_3) \) from Eq. (2), which gives the reflected intensity \( I_3(r_3) = |g_3(r_3)|^2 \).

Figure 3(b) shows the intensity profiles \( I_3(r_3) \) calculated from Eq. (2) for different etched shape functions (A), (B), and (C), as shown in Fig. 3(a). In these calculations, we use the same parameters \( \omega_0 = 4.1 \mu\text{m}, f = 55 \text{mm}, l_{02} = 56.3 \text{mm}, l_{23} = 5 \text{m}, d_0 = 0.25 \mu\text{m}, n_E = 1.33, N_F = 0.65 + i5.3 \) for Al thin film, and \( n_S = 1.52 \) except \( A_E \) and \( w_E \). They are normalized with respect to the center peak of the intensity profile for \( A_E = 0 \) (i.e., no etching has occurred). In the case of \( A_E = 0 \), the intensity profile is just a Gaussian.

As etching progresses \([A \rightarrow C]\) in Fig. 3, the diffraction fringes appear and shrink. The etched shape function \( S(r_0) \) could be found by analyzing the experimental diffraction fringes.

### 3. Experiment

The Al thin films prepared by vacuum deposition upon a glass substrate \( (n_S = 1.52) \) are of thicknesses ranging from 100 to 650 nm. Etching solutions are prepared by mixing an 85.6 wt.% solution of \( \text{H}_3\text{PO}_4 \) and deionized \( \text{H}_2\text{O} \) at a volume ratio of 1 to 9.\(^9,^{10}\) The refractive index of the etchant \( (n_E = 1.33) \) is

\[7613\]
LSER POWER = 82 mW (3.1x10^-5 W/cm^2) 

d = 0.25 μm

Fig. 6. Time dependence of the transmitted laser intensity as pinhole formation progresses. During this experiment the diffraction patterns in Fig. 7 are also taken. At \( t_i = 5.0 \) min, the Al thin film is pierced.

assumed to be the same as that of \( \text{H}_2\text{O} \). The volume of etchant used is far larger than the volume of the specimens.

The experimental setup is described in Fig. 4. A cw \( \text{Ar}^+ \) laser beam with a wavelength of 514.5 nm is used to irradiate the Al thin films. The incident laser beam power on the specimen is in the range of 50 ~ 200 mW. The illumination time \( t_i \) of the \( \text{Ar}^+ \) laser is controlled by a mechanical shutter. A Kepler-type beam expander (expansion ratio 1:2.5) collimates the laser beam to a radius of 2.3 mm at the focusing lens position with a beam divergence of 2.1 mrad. An attenuator is used to control the incident laser beam power. A beam splitter is used to split the laser beam reflected from the surface of the Al thin film. The focal length of the focusing lens \( T \) is 55 mm. Since the input beam has a beam divergence of 2.1 mrad, the beam waist is located at 56.4 mm (=102), 1.4 mm off the focus of the lens \( f = 55 \) mm. At the focal plane, the radius of the beam waist \( w_0 \) is measured by the scanning knife edge method as 4.1 μm in air, which is the same as in the etchant.

The sample is immersed in the etchant placed on the computer-controlled x-y translation stage. A photodetector is set in the center of the x-y translation stage behind the sample to detect the transmitted laser beam. The patterns of the reflected laser beam projected onto the screen are photographed with a camera, or monitored by a scanning photodetector (with an aperture with a diameter of 0.1 mm) to obtain the diffraction intensity profiles. The distance between the focusing lens and the screen \( l_{20} \) is 5 m.

4. Results and Discussion

Figure 5(a) shows an optical micrograph of etched pinholes of an Al thin film with various illumination times \( t_i = 10 ~ 160 \) s. In this case, the incident laser power at the film surface is fixed to 200 mW. Scanning-electron micrographs (SEM's) of this etched thin film are shown in Fig. 5(b) \( t_i = 80 \) s; the film is not pierced yet) and Fig. 5(c) \( t_i = 110 \) s; the film is pierced). As shown in these SEM pictures, we can see that the assumption of the Gaussian etched shape function is reasonable.

Fig. 7. Photographs of the diffraction patterns: (a) \( t_i = 0 \), (b) \( t_i = 2 \) min, (c) \( t_i = 4 \) min, (d) \( t_i = 6 \) min, (e) \( t_i = 8 \) min, (f) \( t_i = 10 \) min, (g) \( t_i = 12 \) min, (h) \( t_i = 14 \) min. The Al film thickness is 250 nm, and the incident laser power is 82 mW.

As illumination proceeds, we observe both the transmitted and the reflected laser beam simultaneously. Figure 6 shows the intensities of the transmitted laser beam measured by the photodetector beneath the specimen. At \( t_i = 5.0 \) min, which is shown on the illumination time axis, the transmitted laser beam starts to appear, which implies that the Al thin film is pierced and that the radius of the etched pinhole starts to increase. The curve indicates that the radius of the etched pinhole approaches saturation value. Photographs of the reflected ring fringe patterns are shown in Fig. 7. They were taken at the times (a)–(h) indicated in Fig. 6. From these, the etching process of the thin films can be classified into a sequence of three stages:

1. An initial stage \( 0 \leq t_i \leq 5.0 \) min in Fig. 6), in which the transmitted laser beam is not detected and the film is not pierced yet. But, in this stage, the ring fringe pattern appears gradually and begins to
Fig. 8. The radii of the fringes as functions of illumination time.

shrink as etching takes place. Note that the diameter of the bright disk decreases from (a) to (c) in Fig. 7.

(2) An intermediate stage, in which the transmitted laser beam is detected. The thin film is pierced, followed by an increase in the radius of the etched pinhole \( r_E \). In this stage the ring fringe pattern keeps on shrinking [(d), (e), and (f) in Fig. 7].

(3) A final stage, in which the increase of the transmitted laser beam is slow, so that the radius of the etched pinhole \( r_E \) no longer increases (see Fig. 11 below). In this stage the ring fringe pattern stops shrinking [(g) and (h) in Fig. 7].

To deduce the etched shape function \( S(r_0) \) from the behavior of the reflected ring fringe pattern, we have measured the decreasing fringe radius \( r_f \) as a function of the illumination time \( t_i \), which is shown in Fig. 8. From this and Eq. (5) (Fig. 2), we can determine the appropriate value of \( A_E \) and \( r_E \) in \( S(r_0) \), which gives the proper fringe radius \( r_f \). Typically, the parameters are as follows:

\[
\begin{align*}
A_E &= 0.13 \, \mu\text{m}, \, w_E = 0.89 \, \mu\text{m}, \text{ at } 2 \, \text{min}; \\
A_E &= 0.22 \, \mu\text{m}, \, w_E = 1.34 \, \mu\text{m}, \text{ at } 4 \, \text{min}; \\
A_E &= 1.05 \, \mu\text{m}, \, w_E = 1.65 \, \mu\text{m}, \text{ at } 5.5 \, \text{min}; \\
A_E &= 300 \, \mu\text{m}, \, w_E = 1.39 \, \mu\text{m}, \text{ at } 8 \, \text{min}; \\
A_E &= 10^5 \, \mu\text{m}, \, w_E = 1.19 \, \mu\text{m}, \text{ at } 12 \, \text{min}.
\end{align*}
\]

For example, the calculated intensity profile of the reflected ring fringe pattern from Eq. (2) at 12 min (i.e., \( A_E = 10^5 \, \mu\text{m} \) and \( w_E = 1.19 \, \mu\text{m} \)) is shown in Fig. 9 and is compared with the experimental data (denoted by ●) measured by the scanning photodetector. In this figure the center intensity of the experimental value is set to be the same as that of the calculated one, and is normalized with respect to the center peak of the intensity profile for \( A_E = 0 \). Figure 10 shows the incident Gaussian intensity profile and the change of etched shape as the illumination time increases. As the illumination time increases, the slope of the etched pinhole edge becomes steeper. From Eq. (6), we can obtain the etched pinhole radius \( r_E \) as a function of the illumination time \( t_i \), which is compared with the experimental data, as shown in Fig. 11.

From the change of the etched shape of the thin film, as shown in Fig. 10, we calculate the etched volume as a function of the illumination time and plot this in Fig. 12. Before the thin film is pierced [in stage (1)], the volume etching rate is low and increases slowly. But the volume etching rate increases rapidly when the film is pierced, and this state...
is continued until the radius of etched pinhole \( r_E \) becomes approximately equal to the input beam waist \( w_0 \) [stage (2)]. In stage (3), the volume etching rate decreases, and the etching process will be terminated finally.

In stage (3), since the etched edge is approximately vertical, as shown in Fig. 9, we may assume that the etched profile is rectangular, as shown in Fig. 2(c). In this case, the etched shape function \( S(r_0) \) becomes the circle function\( ^3 \) such that

\[
S(r_0) = \text{circ}(r_0/r_E) = \begin{cases} 
0 & r_0 < r_E \\
1 & r_0 > r_E
\end{cases}
\]

instead of Eq. (7). From the numerical calculation of Eq. (5), we have established the relation between the fringe radius \( r_f \) and the etched pinhole radius \( r_E \), as in the case of the Gaussian etched shape function. Then, from this relation \( r_f \) versus \( r_E \) and Fig. 8 \( r_f \) versus \( t_i \), we can finally obtain the etched pinhole radius \( r_E \) as a function of the illumination time \( t_i \), as shown in Fig. 11 (denoted by dashed curve), which is compared with experimental data and the calculation by the Gaussian etched shape function (denoted by a solid curve).

5. Conclusions

In order to obtain information on the pinhole etching process from the reflected laser beam projected onto a screen, we have used the diffraction theory to explain the time-dependent and pinhole radius-dependent behavior of the diffraction rings. We have assumed the etched shape of a thin film to be a Gaussian following the incident laser beam intensity profile. Our experimental data agrees well with the theory, i.e., the intensity distribution of the diffraction pattern (Fig. 10) and the radius of the etched pinhole (Fig. 11). The process of pinhole formation on Al thin film can be classified into three stages: (1) the initial stage, in which etching starts taking place primarily in the normal direction until the film is pierced; (2) the intermediate stage, in which the radius of the etched pinhole increases up to the radius of the laser beam waist; and (3) the final stage, in which the etching process is in the radial direction but basically ceases. The final stage may be described by the circular top-hat etched shape function, and the Gaussian function is not needed. The diffraction theory is applied successfully to the investigation of a laser-assisted pinhole etching process, and the whole process of pinhole etching can be monitored accurately from the Fresnel diffraction pattern of the reflected beam.

References

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