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Experimental study on microdeposition of the copper thin film by femtosecond laser-induced forward transfer*

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The morphologies of the deposited dots on the 40 nm-thick copper film by the femtosecond laser-induced forward transfer that depend on the irradiated laser fluence have been studied, and the variations of orderliness of the diameter of deposited dots on the quartz substrate and forward ablated dot on the donor substrate with increasing pulse fluence have been obtained experimentally. The experimental results show that a thinner copper film would generate larger-sized ablated dot and deposited dot at the threshold fluence for transfer. By x-ray diffraction measurement, it is demonstrated that the crystal form of the transferred copper films is unaltered and the size of the crystallites is diminished.

Keywords: femtosecond laser, laser-induced forward transfer, thin film, ablation, deposition
PACC: 4262A, 4280W, 7360D

1. Introduction

The laser-induced forward transfer (LIFT) is a new technology, which utilizes short or ultrashort laser pulses to selectively remove the thin film (donor) material with micrometer size on a transparent support substrate and eventually forward transfer it onto a receiver substrate, as shown in Fig.1. Under the control of computer, the process enables fabrication of dots, lines and pattern. Due to the advantages of simplicity, firm sticking to receiver substrates, contamination-free and versatility for a variety of thin films even composed of complicated ingredients, LIFT technique has presented its huge potential applications in fields of micro-optics,[1,2] micro-electronics[3] and micro-biology,[4,5] etc. Recent studies have revealed that the sub-micrometer scale of the deposited dot can be obtained with the femtosecond LIFT.[6−8] However the fundamental physical mechanism of femtosecond LIFT is still not completely understood. In this paper, we present the morphologies of the deposited dots by femtosecond LIFT of 40-nm-thick copper film, which depends on the irradiated laser energy. It was found experimentally the variations of orderliness of the diameter of deposited dots on quartz substrate and the forward ablated dot on donor substrate with increasing pulse fluence. We also found that at threshold fluence, a thinner copper film can generate larger-sized ablated dot and deposited dot. And by x-ray diffraction (XRD) measurement, it has been demonstrated that the crystal form of the transferred copper films is unaltered and the size of the crystallites is diminished.

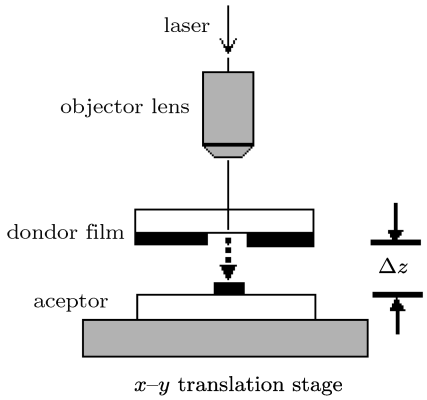


Fig.1. Schematic diagram of LIFT.

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2. Experimental setup

The experiment was performed on a commercial femtosecond laser processing platform (UMW-2110i, Clark-MXR Inc.). The apparatus was composed of three fully computer-controlled components: a femtosecond laser amplifier system, a three-dimensional moving stage with a resolution of 10 nm and a CCD monitoring system. The femtosecond laser system generated pulse trains of 775 nm central wavelength, 148 fs pulse duration at 1 kHz repetition rate. The sample was mounted on the x–y plane of the stage. The source film (donor) was a copper thin film of ∼40 nm in thickness deposited on a quartz substrate of 3 mm thick. Another blank quartz substrate with the same dimensions was placed in contact with the source film ($\Delta z = 0$) served as a receiver. The laser beam was focused onto the inside surface of source film (interface between copper film and support quartz substrate) through the transparent quartz substrate by a 5× objective mounted on the z-axis of the stage. The radius of the focus spot was estimated as ∼8 μm. In order to investigate the effect on material transfer the laser pulse energy was varied over a wide range of values. The experiment was performed in atmosphere at room temperature.

3. Experimental results and discussion

3.1. Influence of the pulse fluence on the morphology of the deposited film

The effect of the laser fluence on the morphology of the deposited material is shown in Fig.2.

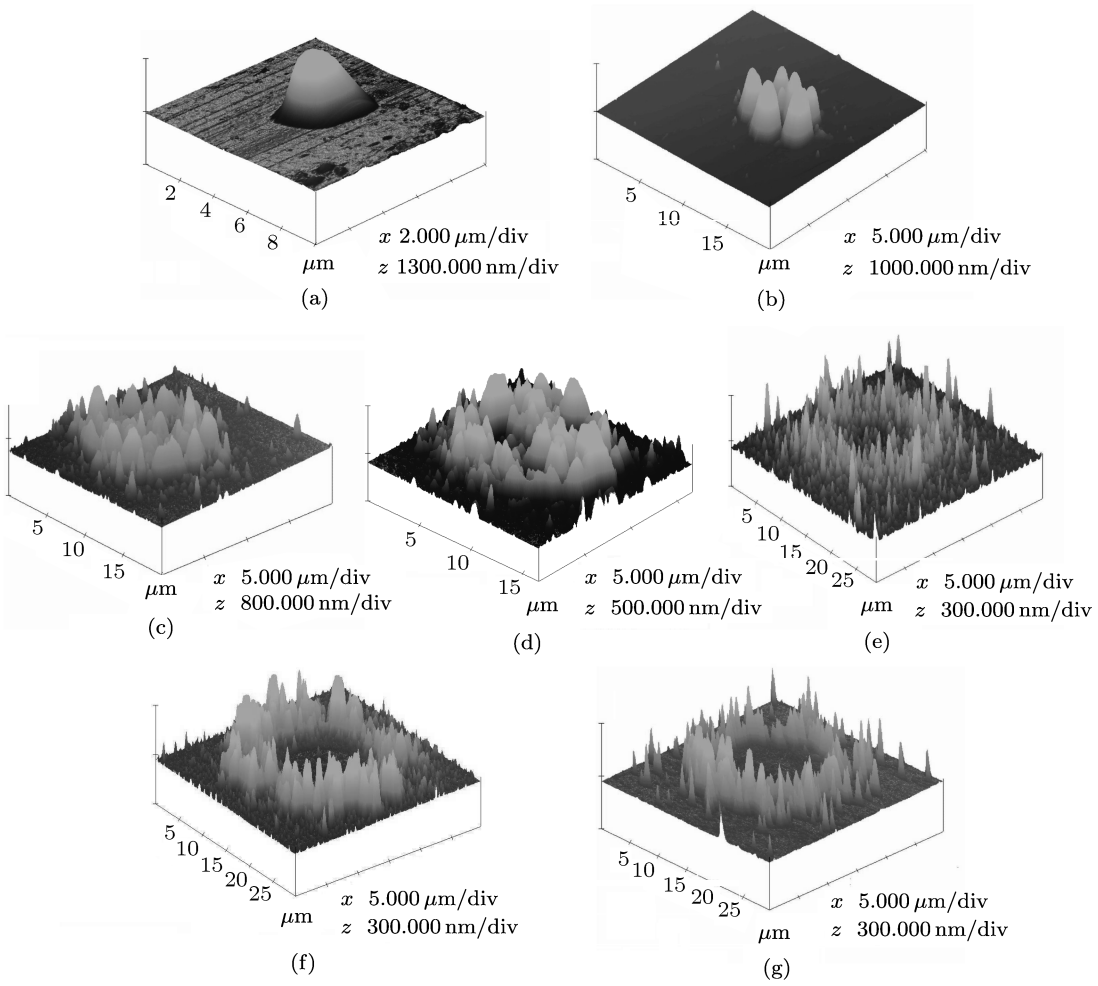


Fig.2. AFM image of the deposited dots by the fs-LIFT. The laser fluence is (a) 1.74 J/cm^2, (b) 2.24 J/cm^2 (c) 2.73 J/cm^2, (d) 3.23 J/cm^2, (e) 3.73 J/cm^2, (f) 4.23 J/cm^2, and (g) 4.98 J/cm^2.

We found that the copper film could not be peeled from support quartz substrate when the incident pulse fluence was ∼1.24 J/cm^2. When the incident pulse fluence increased to 1.74 J/cm^2, a small droplet was transferred to the receiver, as shown in Fig.2(a), i.e., the threshold fluence ($E_{\rm th}$) for transfer was 1.74 J/cm^2 for a copper thin film of ∼40 nm-thick. From Fig.2(a), we can see that the deposit is circular with a diameter around ∼2.5 μm, and has a symmetrical structure and highly smooth surface. A dome profile is observed with a peak height of ∼ 1.3 μm, i.e. a small droplet of ∼2.5 μm in diameter has been transferred. The transfer of droplets is repeatable as shown in Fig.3. Moreover, the diameter of the copper droplet was much smaller than that of the film ablated, and the latter height was much bigger than the thickness of the copper film. Through observation of the droplet's and ablated crater's shape, we consider that the following three points can be affirmed: 1) At $E_{\rm th}$, the ablated film material is mainly in the liquid state; 2) because of the Gaussian beam, the central section of the ablated film on the free surface would be melted first, and the molten material rapidly expands; 3) Because metal in the liquid phase has very high surface tension and the push of the plasma is moderate at a constrained interface as shown in Fig.4, the deposit such as that shown in Fig.2(a) is believed to arise from the moderate pressure of the plasma generated at the interface and a droplet grows on the free surface of the source film during the rapid expansion of the molten material.[9−12] On the receiver, the momentum of droplets has been used up exactly.

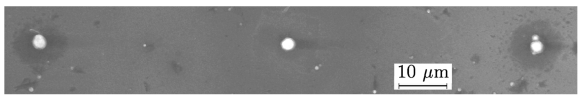


Fig.3. SEM images of the deposited dots by the fs-LIFT. The laser fluence is 1.74 J/cm^2.

The mass and the kinetic energy of the peeled film material would be expected to determine the size and morphology of the resulting deposit with the laser fluence increased. These parameters depend on the temperature of the peeled source film and the compressive stress of the plasma, which are a function of the laser fluence. At about 2.24 J/cm^2, a multiple of deposits were obtained per single laser shot as shown in Fig.2(b). The morphology of the multiple deposit became much more outstanding with the increasing of energy as shown in Figs.2(c) and 2(d). These deposits exhibit the break up of a droplet during transfer, the diameter of which is increased and the height of which is decreased with the applied fluence. This feature may be attributed to a larger impact momentum of the droplet on the receiver, which hailed from the pressure of the plasma generated at the interface between the film and the support substrate, and the lower surface tension of larger mass droplet.

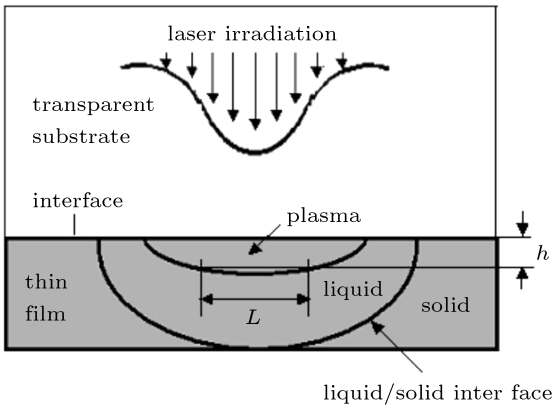
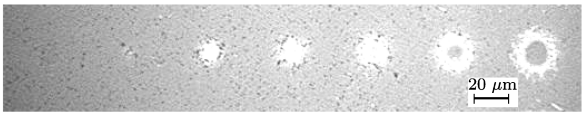


Fig.4. Schematic diagram of heating of the donor film by the femtosecond laser pulse at $E_{\rm th}$.

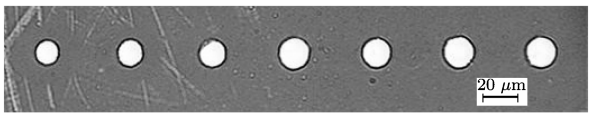
At about 3.73 J/cm^2 the deposited film becomes an island-like geomorphology as shown in Fig.2(e). We attribute this phenomenon to phase explosion[13,14] occurring almost in the free surface of the source film. Phase explosion involves the nucleation of gas bubbles throughout a superheated liquid in which the number of bubbles is an exponential function of the liquid temperature. In this situation the material may be transferred in the mixture of liquid and gas phases. It is believed that the superheated liquid was scattered by a gas bubble gaining sufficient internal pressure into the state of the nanometer grain during transfer. So that the deposited film on the receiver shows a symmetrical nanometer fine structure. It is also observed that the center part of the dot has been thinned down, in which a crater-like morphology is formed. This feature may be attributed to the inhomogeneous intensity distribution of the Gaussian beam, the sticking coefficient becomes minimal at the center of the irradiation.[15] A very big shock can cause a wide range of spread of the deposited material when the collision happens between the transferred particles and the acceptor substrate, in which a ring morphology is formed as shown in Figs.2(f) and 2(g).

3.2. Influence of the pulse fluence on the size of deposited film

In the experiment, we obtained the orderliness of the diameter of deposited dots on quartz substrate and a forward ablated dot on donor substrate with the pulse fluence increased as shown in Fig.5. By optical microscope images, we can see that the deposited dots are not clear at the threshold fluence, in which a center dark dot and a faintness light ring around have been present. This feature may be attributed to film interference phenomena of the light. Figure 6 presents the plot of the diameter of deposited dots and ablated dot on donor substrate as a function of the laser fluence.



(a) catoptric light imaging



(b) transmission light imaging

Fig.5. Optical microscope images of dots (a): deposited on acceptor quartz substrate and (b): forward ablated on donor substrate. The laser fluence = 1.74, 2.24, 2.73, 3.23, 3.73, 4.23 and 4.98 J/cm^2, from left to right.

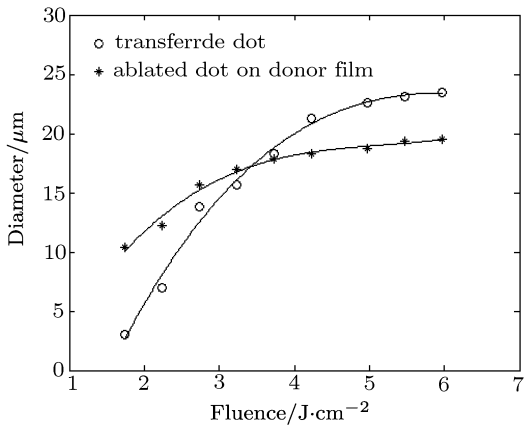


Fig.6. Plot of the diameter of the deposited dots and ablated dot as a function of the laser fluence.

From Figs.5 and 6, we can see that the diameters of the deposited dots and the ablated dot are all the lowest at the threshold fluence, and the diameter of deposited dots is much less than that of ablated dot. The dimensions of deposited dots and ablated dot are gradually increasing along with the increase of the laser fluence. But the increase in the diameter of deposited dots is much more rapid than that of the ablated dot, until it exceeds it and finally becomes changeless. These features can be attributed to as follows: On the one hand, bigger area material is peeled from source film contributed to the larger dimension on the receiver substrate with the increase of the laser fluence. Alternatively, the kinetic energy of the peeled film material is gradually increasing along with the increase of the laser fluence, which can cause a wide range of spread of the deposited material when the collision happens between the transferred material and the acceptor substrate.

However, the increase of the dimensions becomes slower for higher energies. This may be explained as follows: Due to the inhomogeneous intensity distribution of the Gaussian beam, the removed area is confined in the middle of the laser spot as shown in Fig.7, where the laser beam is most intense and sufficient to blow off copper film from the quartz substrate. When the fluence at the laser beam centre reaches the threshold, a smaller area is removed. With increasing pulse energy, the fluence at the beam centre is far above the threshold, while just above the threshold at the edges, creating a larger ablated spot. Finally, the laser pulse removes the whole film within the laser spot.

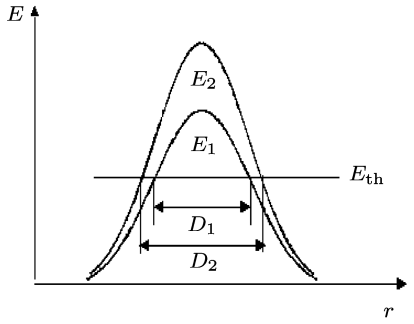


Fig.7. Schematic diagram of the Gaussian pulse ablation.

Moreover, we also found that the size of microdroplet from the thinner film was relatively larger at E_{th} for transfer as shown in Fig.8, which could be understood by larger size of ablated dot on source film as shown in Fig.9. The relatively larger area of transferred material in the liquid phase from source film contributed to the larger dimension on the receiver substrate.

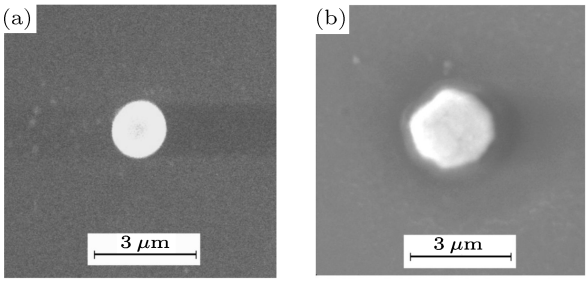


Fig.8. SEM image of the deposited dot by the fs-LIFT at $E_{\rm th}$ for transfer, (a) ∼80 nm, 3.23 J/cm^2, (b) ∼40 nm, 1.74 J/cm^2.

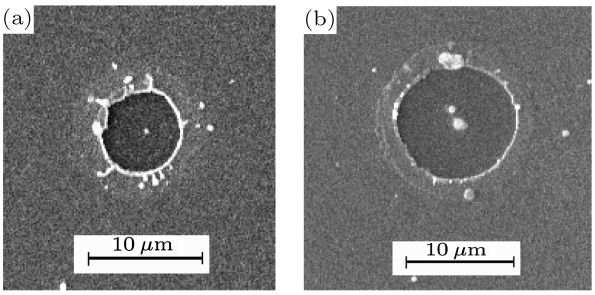


Fig.9. SEM image of the ablated dot on donor film by the fs-LIFT at $E_{\rm th}$ for transfer, (a) ∼80 nm, 3.23 J/cm^2, (b) ∼40 nm, 1.74 J/cm^2.

Considering that the material is fully peeled off from the donor by inside-surface-induced plasma, the pushing force of plasma dominates the whole process. This feature of the relatively larger size of the ablated dot on thinner source film may be explained as follows: The laser pulse with spatially Gaussian distribution induces the plasma that is Gaussian-distributed-dimensional. The central area of focusing spot induces a thicker plasma at a certain single pulse energy, which can bring a stronger plasma pushing force. The minimal pushing force which can fully peel the copper thin film is determined by the thickness of the film. A thinner film requires a smaller pushing force, in which a lesser thickness (h) of the plasma is required correspondingly as shown in Fig.4. While irradiated by laser pulses, the dimension of a transverse zone (L), in which the pushing force reaches the minimal value (corresponding to a minimal thickness (h) of the plasma) for peeling thin film, may be inversely proportional to the thickness of the source film.

3.3. Influence of pulse fluence on inner structure of deposited film

Furthermore, we investigate the crystal form of transferred dots by XRD measurement. The crystal form of the donor film is a polycrystalline form as shown in Fig.10 (curve 1), that is consistent with the powder diffraction file 04-0386, i.e. the crystal structure of the donor film is a face-centered cube. At the threshold fluence, the XRD signal of the transferred droplets has no peak. It is evident that the transferred droplet by the femtosecond LIFT is converted into the amorphous metal, i.e. the liquid droplet is abruptly cooled down to be amorphous on the receiver. This mechanism would be similar to the melt-quenching generally used as the fabrication method of amorphous metals.[16] At about 3.23 and 3.73 J/cm^2 the crystal form of the transferred film is still a polycrystalline form as shown in Fig.10 (curves 2 and 3), in which the crystal form of the transferred copper films is unaltered. But it can be known that the size of the crystallites has been diminished due to the increase of the strong line extent. This phenomenon can be attributed to the recrystallization of the copper plasma.

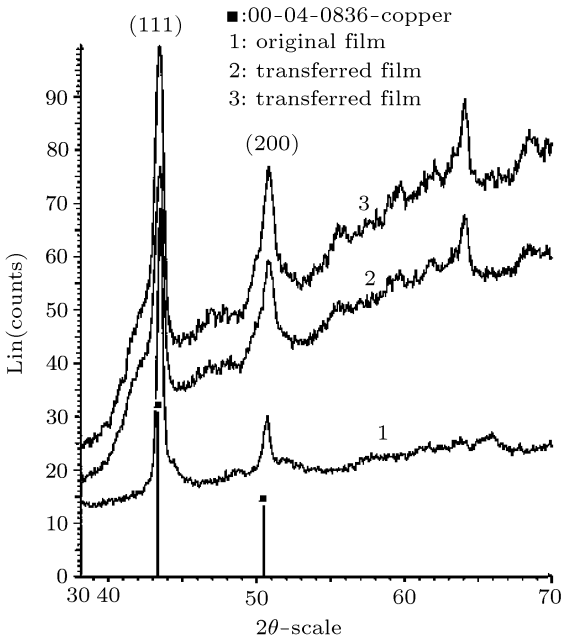


Fig.10. XRD signals of the copper films.

4. Conclusions

In conclusion, we have presented the multiform morphologies of the deposited dots by the femtosecond LIFT for the 40 nm-thick copper film, and have acquired experimentally the orderliness of the diameter of the deposited dots on quartz substrate and forward ablated dot on donor substrate with the pulse fluence increased. It is found that at the threshold

fluence for transfer, a thinner copper film generates a larger-size ablated and deposited dot. It may be attributed to the fact that the transverse dimension (L) of the induced plasma in the inside surface is relatively larger for thinner films than that for thicker ones. By XRD measurement, we have also found that the crystal form of the transferred copper films was unaltered and the size of the crystallites has been diminished.

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