Using direct nanoimprinting to study extraordinary transmission in textured metal films

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Abstract: In this paper, we describe a thermal embossing imprint method, which we name "nano-imprinting in metal" (NIM), for patterning metal films with a variety of profiles. Metal films exhibiting either perforated hole-arrays or corrugated structures with various surface morphologies can be fabricated rapidly. The SPR phenomenon allowed energy coupling to the other side of the textured metal film, causing a dramatic increase in the transmission. As a technique for readily controlling the working wavelength and transmittance, the NIM method has great potential for application in various optoelectronic devices.

OCIS codes: (220.4000) Microstructure fabrication; (240.6680) surface plasmons; (310.6628) subwavelength structures.

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1. Introduction

Many sub-wavelength metal hole-array structures exhibiting surface plasmons phenomena and extraordinary transmission have been reported recently [1–3]. It is believed that the transmission in such systems is caused by incident light of certain wavelengths passing through the metal hole-arrays having specific hole-diameter and periods. This phenomenon is generally attributed to the coupling of light to the surface plasmons excitation on the surface of the periodic metal films [3]. For light coupling into a surface plasmons wave, periodic structures always play an important role, providing an extra wave-vector between the surface plasmons wave and the light, as indicated in the following equation:

$$k_{sp} = k_0 \sin\theta \pm mG_x \pm nG_y \tag{1}$$

where k_{sp} is the wave vector of the surface plasmons wave, k_0 is the wave vector of the incident light, $k_0 \sin\theta$ is the projection of k_0 onto the xy plane, G_x and G_y are grating momentum, and m and n are integers. Therefore, for a square array of period P, the location of λ_{max} in the normal incidence transmittance spectra can be identified approximately from the following dispersion relationship [2]:

$$\lambda_{max} = \frac{P}{\sqrt{i^2 + j^2}} \cdot \sqrt{\frac{\varepsilon_m \cdot \varepsilon_d}{\varepsilon_m + \varepsilon_d}} \tag{2}$$

where the indices i and j are scattering orders from the periodic structure, ε_m is the real part of the metal's dielectric constant, and ε_d is the dielectric constant of the neighboring dielectric material. It was reported that the metal holes was not necessary and that continuous metallic films with appropriate corrugation could also permit enhanced transmission [4-8]. When the metal film is sufficiently thin, the surface plasmons modes of the two surfaces can overlap and interact on both sides of the patterned metal film. Therefore, extraordinary transmission phenomena can also be found in continuously corrugated metal films that do not feature perforated hole-arrays. Metal films having sinusoidal profiles can be fabricated using conventional holographic techniques. Recently, Bai *et al*. prepared sinusoidal profiles in metal films without various profiles by using a complicated double-exposure holographic technique [8]. Generally, it is difficult to control the depth and shape of sinusoidal metal profiles when using holographic lithography because of nonlinearity of the photoresist sensitivity.

We reported a thermal embossing imprint method, which we name "nano-imprinting in metal" (NIM), for patterning metal films with various profiles [9]. Conventional nanoimprint lithography defines patterns through physical deformation of deformable polymer materials under a suitable pressure and temperature [10-14]. The etching step that is required to transfer the structure to the underlying substrate tends to limit the technique's throughput and increase the cost of fabrication. Recently, a direct imprinting process for gold films was reported using an ultra-high imprinting pressure (several hundred megapascals) and an oil-press imprint instrument [15]. Generally, such processes utilizing ultra-high pressures would, however, damage the underlying substrates or devices. In the earlier work, we improved upon the direct-imprint processes by using a sharp mold and a soft pad layer to reduce the imprint pressure [9]. When using suitable processes, the imprint pressure in the NIM method could be reduced to become compatible with conventional imprinting processes. The imprint pressure of NIM was only ca. 1% of that used in the previous process of directly imprinting in the metal [15]. Using NIM, metal films with various profiles can be obtained readily when using

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variously shaped molds and imprinting pressures. In this study, the patterned metals we formed not only possessed the inverse shape of the mold but also exhibited corrugated structures of various depths. Furthermore, we extend the NIM technique to perforating metal thin-films, creating 2D metallic hole arrays, and experimentally verifying plasmonic behavior in patterned devices.

Generally, metallic hole arrays have been patterned using focused ion beam (FIB) or electron beam lithography combing lift-off or etching processes. In this study, we used a sharp mold with a suitable pressure to perforate the hole arrays on the metal films. Therefore, we could rapidly fabricate metal structures possessing either corrugated or perforated structures using the NIM method, which allowed us to study the special surface plasmons phenomena of metals exhibiting various surface morphologies. In this study, we observed that the localized maximum of the electric field at the tip of a corrugated metal film could allow the energy to readily couple to the other side of the metal film, increasing the transmission dramatically.

2. Simulation and Experiment Setup

Figure 1 displays a schematic representation of the NIM method. We applied commercial NEB-22 (Sumitomo) and SU-8 (Microchem) resists as the soft pad layer on glass substrates. A thermal evaporator system was used to deposit gold films on the resist-coated substrates. The imprint process was performed using a conventional imprint instrument. The imprint pressure and heating temperature of the imprint processes were 2–16 MPa and 40–60 °C, respectively. Further details on the relationship between imprint temperature, pressure, and effect of release layers can be found in Ref. [9]. The silicon mold employed in our experiments was fabricated using electron beam lithography (Leica, Weprint-200) followed by reactive ion etching. A high-density-plasma reactive ion etching (HDP-RIE) system (Duratek, Mutiplex Cluster) equipped with an inductively coupled plasma (ICP) source was used to fabricate molds having various profiles. The bias and RF power of the HDP-RIE were the critical parameters for controlling the surface profiles of the molds. As indicated in Figure 1, two-dimensional corrugated gold structures were readily fabricated by twice using the direct-imprinting metal method. Images of the patterned metal films were measured using an atomic force microscope (AFM) and a scanning electron microscope (Hitachi, S-4000). Transmission spectra of the patterned metal films were measured using an optical spectrometer (Hitachi, U-4100). Furthermore, the finite-difference time domains (FDTD) method was used to analyze the optical behavior of transmission light on the corrugated gold film structures within the near-field regime.

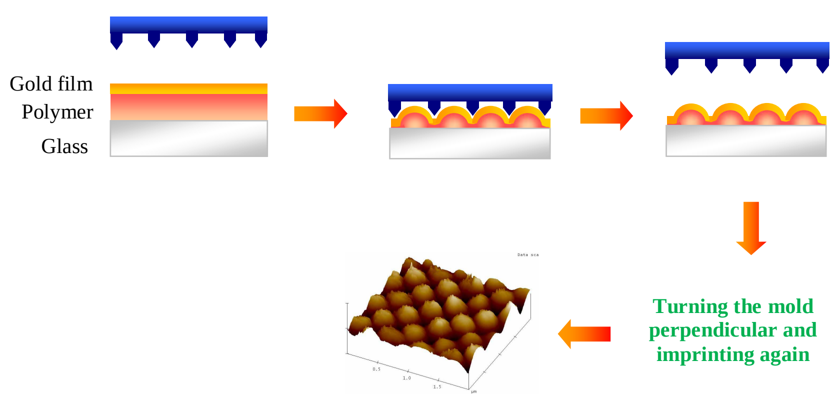

Fig. 1. Schematic representations of the NIM and double-imprinting processes.

3. Results and Discussion

Figure 2 displays microscopy images of the molds and their imprints. Figure 2(a) presents the image of a grating mold having a period of 400 nm. After imprinting under a pressure of 12

MPa, we obtained a corrugated gold film having a period of 400 nm and a depth of 110 nm [Fig. 2(c)]. Figure 2(b) displays an ultra-sharp hexagonal mold having a high aspect ratio (period: 400 nm; depth: 1100 nm) that we fabricated by increasing the RF power and bias voltage in the HDP-RIE system. The same imprint pressure (12 MPa) was highly effective for allowing the tip of the ultra-sharp mold to transpierce gold films having a thickness of 45 nm. As indicated in Figure 2(d), large-area metal hole arrays were readily obtained through the use of this one-step imprinting process without the need for any expensive lithography or etching instrumentation. As illustrated in Figure 1, two-dimensional periodic structures could also be prepared through imprinting with a one-dimensional grating mold, by turning the mold perpendicular and imprinting again, in a process that we name "double-imprinting." Figure 2(e) presents an image of a metal film that had been double-imprinted using the grating pattern mold displayed in Figure 2(a). Therefore, it is clear that with suitable choice of the imprinting pressure and mold, we could use the NIM method to pattern either continuously corrugated metal films or perforated metal hole arrays.

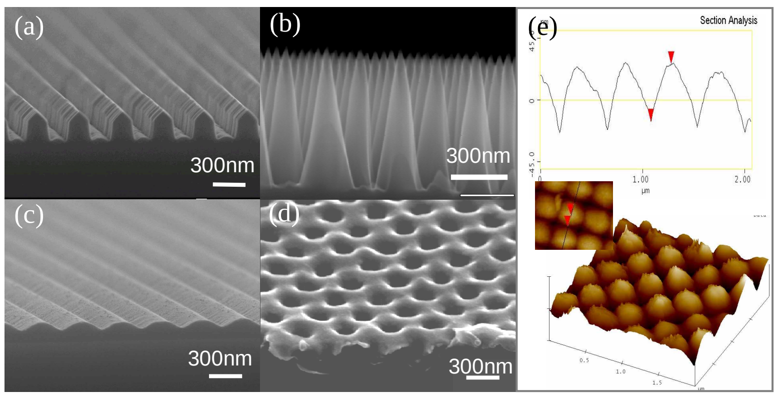

Fig. 2. SEM images of (a) a grating mold, (b) an hexagonal ultra-sharp mold, (c) a corrugated gold film patterned using the NIM method, (d) hole arrays in a gold film patterned using the NIM method. (e) AFM image of a corrugated gold film that had been double-imprinted using a mold possessing a grating pattern.

To study these extraordinary transmission phenomena, we used an optical spectrometer to measure transmission spectra of the textured metal films. Figure 3(a) displays the transmission spectrum of a flat gold film having a thickness of 45 nm; we observe only an intrinsic transmission peak for gold at 510 nm. The transmission of the gold intrinsic peak varied only with respect to the thickness of the gold film. After performing the imprinting process, Figure 3(a) indicates that the transmission spectrum of the corrugated gold film displayed weak transmission peaks at 530 and 670 nm that arose from the surface plasmons resonance modes at the air–gold and resist–gold interfaces, respectively. The weak transmission enhancement was due to the mismatch of the refractive indices on both sides of the asymmetrically corrugated metal film (i.e., air/corrugated gold film/resist) [16]. This refractive index mismatch in the asymmetric structure caused weak surface plasmons coupling between the two sides of the textured gold film. After coating a resist film as an index-matching layer above the corrugated gold film, Figure 3(a) indicates that the transmission of the symmetric structure (i.e., resist/corrugated gold film/resist) increased dramatically, presumably because of the surface plasmons excited at the two interfaces had the same wave vector and resonated with one another. In this symmetric structure, we observed a strong transmission peak at 680 nm having a transmittance of 45%, much higher than that of the flat gold film. The enhanced transmission near 680 nm arose from the fundamental mode of the surface plasmons resonance at the resist–gold interface in the corrugated gold film having a

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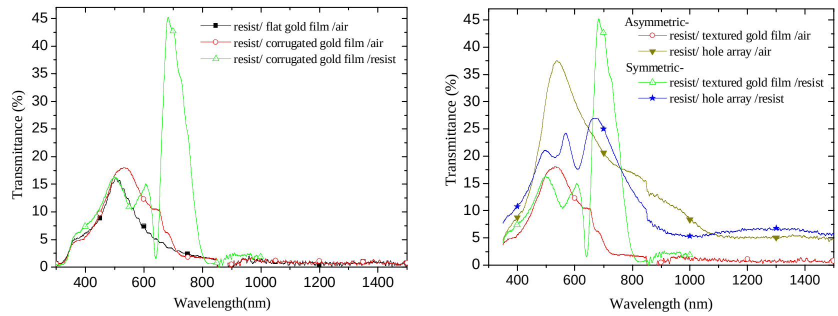
period of 400 nm [5, 8]. The transmittance increased further to 50% after increasing the imprint pressure to 16 MPa.



(a) (b)

Fig. 3. (a) Transmission spectra of gold films before and after patterning with corrugated structures, with and without index-matching layers. (b) Transmission spectra of gold films possessing hole array and corrugated structures.

We also used an sharp mold to perforate hole arrays into metal films under suitable pressures. Figure 2(d) indicates that metal films having hexagonal hole arrays were readily obtained through a one-step imprinting process. Figure 3(b) displays transmission spectra of the continuously corrugated and perforated metal films before and after adding an index-matching resist. In the asymmetric hole-array structure (air/perforated gold film/resist), we observe the transmission increase around 530 nm for the surface plasmons resonance mode of the air–gold interface. Because some of the incident light was transmitted directly through the metal hole arrays, we found that the signals in the transmission spectrum increased after perforating the hole arrays into the thin gold film. After coating a resist film as an index-matching layer above the perforated gold film, Figure 3(b) indicates that the surface plasmons resonance mode of the resist–gold at 680 nm in the symmetric structure (resist/perforated gold film/resist) increased dramatically to 27%. We also observed that the continuously corrugated gold films had narrower and stronger transmission peaks than did the hole-array gold film, consistent with plasmon assisted studies performed by Avrutsky *et al.* [5].

Next, we investigated the optical propagation mechanism using the FDTD method. Because the FDTD method calculates the electromagnetic fields over the entire computational domain as they evolve over time, it lends itself to providing animated displays of the electromagnetic field's movement through the model. We used the FDTD method to analyze the propagation of light within the near-field regime of the continuously corrugated metal film structures. As presented in Figure 4, a plane wave having a wavelength of 680 nm was propagated from 1 μm above the symmetric structure (resist/gold film/resist) in the presence or absence of a corrugated pattern in the gold film having a thickness of 45 nm. Figure 4(a) displays the plane wave that propagated to the thin gold film in the absence of a textured structure. Because the gold film absorbed most of the incident light, we observed that no apparent transmission electric field existed below the thin gold film. Similarly, Figure 4(b) displays the plane wave that propagated to the gold film having a continuously corrugated structure with a period of 400 nm and a depth of 40nm; we find that the transmission field had increased significantly. After increasing the depth of the corrugated structure to 90 nm, a higher transmission field was found [Fig. 4(c)].

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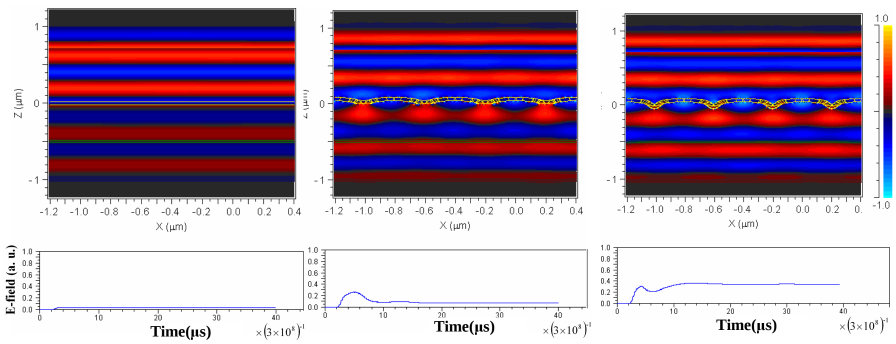
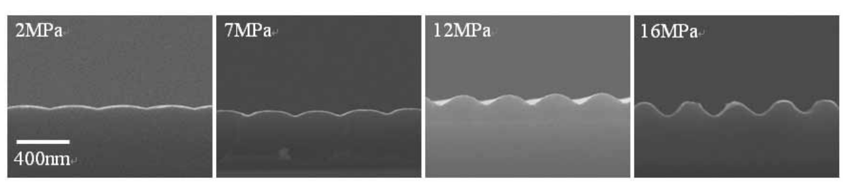


(a) (b) (c)

Fig. 4. FDTD diagrams of (a) a flat gold film and (b) a gold film possessing a continuous corrugated structure having a period of 400 nm and a depth of 40nm (c) a depth of 90nm

After monitoring the propagation of the transmission electric field through the continuously corrugated film, we found that the transmission electric field exhibited a localized maximum at the tip of the corrugated gold film. Because of its large curvature, the electric field was concentrated at the tips of the corrugated gold film after performing the NIM process. We found that the localized maximum of the electric field allowed the energy to readily couple to the other side of the corrugated gold film, causing the increased transmission. Because a more deeply corrugated structure has a larger curvature, it is predicted that the degree of extraordinary transmission is a function of the depth of continuously corrugated metal films.

As displayed in Figure 5, we used imprinting pressures ranging from 2 to 16 MPa to fabricate symmetric structures (resist/textured gold film/resist) having groove depths ranging from 20 to 130 nm. Using this approach, we could modify the curvature of the metal films by varying the imprint pressure.



(a) (b) (c) (d)

Fig. 5. The depth of surface-profile in metal films can be tuned by using different imprint pressure (a) 2MPa, (b) 7 MPa, (c) 12 MPa, (d) 16 MPa.

Figure 6 indicates that the transmittance at 680 nm increased dramatically from 3 to 50% after increasing the imprinting pressure. We also found that the transmittance near 510 nm and the appearance of transmission spectra recorded from 800 to 1000 nm did not change for the corrugated gold films having different groove depths. The transmission of the gold intrinsic band near 510 nm varied only with respect to the effective thickness of the gold film. Therefore, we conclude that no change in the thickness of the corrugated gold films occurred during the NIM processing under the various imprinting pressures. The subtle changes in the transmission spectra may be due to small changes in metal profile. The further work will understand the degree of distortions in the metal thin films.

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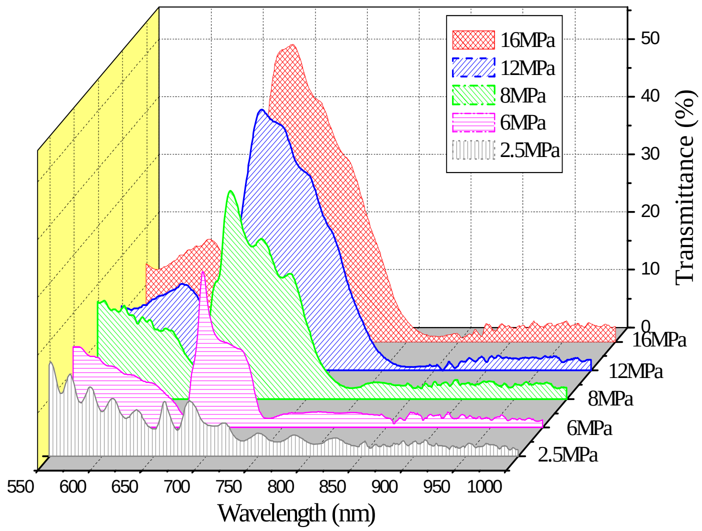
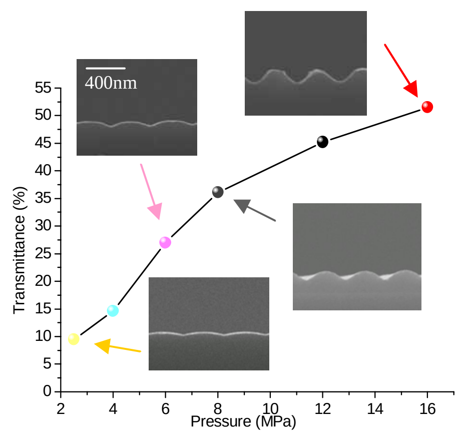


(a)



(b)

Fig. 6. (a) Transmission spectra of gold films patterned with corrugated structures through the use of imprinting pressures ranging from 2 to 16 MPa. (b) Transmittance at 680 nm of corrugated structure (with SEM images of the corresponding groove depths) plotted as a function of the imprinting pressure.

The NIM method has great potential for use in the fabrication of optical band-pass filters, color filters, and beam splitters without the need for complicated optical multilayer thin film-based filters [17]. The preparation of optical filters exhibiting various working wavelength regimes and transmissions can be controlled by selecting suitable imprinting molds and pressures, respectively. Because it allows the rapid fabrication of metallic gratings, this method is also suitable for fabricating grating couplers that could be used to replace the prism couplers in SPR-based biosensors. This NIM technique could also be used for the preparation of various optoelectronic devices exhibiting increased external quantum efficiencies through surface plasmons phenomena.

4. Conclusion

We have developed an imprinting method—so-called "nano-imprinting in metal" (NIM)—for patterning metal films with a variety of profiles. We improved upon direct imprinting processes by using a sharp mold and a soft pad layer to reduce the imprinting pressure so that it would be compatible with conventional imprinting processes. Metal structures possessing both corrugated and perforated topographies were rapidly fabricated using a range of mold shapes and imprinting pressures. Using the NIM method allowed us to study the special surface plasmons phenomena in a series of metal structures possessing different surface morphologies. We found that the transmission electric field through corrugated gold films exhibited localized maxima at the tips of the corrugations. Because of their large degrees of curvature, the electrical fields were concentrated at the tips of the corrugated gold films prepared through NIM processing. The localized maxima of the electric field allowed the energy to readily couple to the other side of the corrugated gold film, causing the increased transmission. The NIM method has great potential for use in the fabrication of optical band-pass filters and beam splitters operating over various working wavelength regimes and transmissions. For the rapid fabrication of textured metal structures, this NIM method is suitable for fabricating SPR-based couplers for biosensors and other optoelectronic devices.

Acknowledgments

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