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Ultra-smooth metal surfaces generated by pressure-induced surface deformation of thin metal films

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ABSTRACT We present a mechanical pressing technique for generating ultra-smooth surfaces on thin metal films by flattening the bumps, asperities, rough grains and spikes of a freshly vacuum deposited metal film. The method was implemented by varying the applied pressure from 100 MPa to 600 MPa on an e-beam evaporated silver film of thickness 1000 Å deposited on double-polished (100)-oriented silicon surfaces, resulting in a varying degree of film smoothness. The surface morphology of the thin film was studied using atomic force microscopy. Notably, at a pressure of ~ 600 MPa an initial silver surface with 13-nm RMS roughness was plastically deformed and transformed to an ultra-flat plane with better than 0.1 nm RMS. Our demonstration with the e-beam evaporated silver thin film exhibits the potential for applications in decreasing the scattering-induced losses in optical metamaterials, plasmonic nanodevices and electrical shorts in molecular-scale electronic devices.

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1 Introduction

Potential new approaches for constructing revolutionary nanoelectronic circuits involve unconventional building blocks such as molecules, DNA, carbon nanotubes or nanowires. Unlike the metals commonly used for interconnecting conventional integrated circuit (IC) devices, innovative devices and circuits employing these new building blocks may require the use of noble metals such as silver (Ag), platinum (Pt) and gold (Au) [1–3]. Another area of scientific research that employs noble metals is in photonic metamaterials, which have recently been experimentally demonstrated [4, 5]. Novel properties such as artificial plasmonic response, synthetic magnetism at terahertz frequencies and negative refractive index have been observed. Imaging far below the diffraction limit is one of the most remarkable properties of negative-index materials, and Fang et al. have successfully demonstrated this phenomenon for the first time using a Ag thin film superlens [6].

The need for atomically smooth uniform metal surfaces in modern electronic and photonic systems cannot be overemphasized. Many of the future nanodevices are expected to be smaller than the typical grain size of freshly vacuum deposited metals. The thin film deposition techniques most commonly pursued are evaporation, ion-beam-assisted deposition and rf/dc sputtering. An analysis of the surfaces of most deposited metal surfaces using characterization techniques such as atomic force microscopy reveals a rough surface topography – often rougher than the size of the desired nanoscale circuit elements [7]. This topography contributes to plummeting circuit yield as well as decreased device reliability. In metamaterial-based devices, surface roughness of the metal films has been identified as a significant source of loss at optical frequencies [8, 9]. For all these reasons, a smooth metal surface is a vital prerequisite for future devices and systems.

In this paper, we describe a novel mechanical imprint-based technique for generating ultra-flat surfaces from a freshly deposited e-beam evaporated thin Ag film. The basic technique is to apply pressure to the thin Ag film with a mechanical pneumatic press. The contacting surface of the press is an atomically flat silicon (100-oriented) mold with mesas patterned by reactive ion etching (RIE). Upon contact with the Ag surface, the mold smooths the film via plastic deformation of the surface features. The method was implemented by varying the applied pressure from 100 MPa to 600 MPa on the Ag film of thickness 1000 Å deposited on double-polished silicon(100), resulting in a varying degree of film smoothness. This planarization procedure also utilizes a molecular monolayer (1H,1H,2H,2H-perfluorodecyltrichlorosilane) on the silicon mold surface, which prevents adhesion of the metal to the mold and subsequent delamination of the film from the substrate, especially for large contact pressures.

2 Experimental procedure

We examined several thin film metal surfaces, such as Pt, Ag and Au, deposited on (100)-oriented silicon substrates using conventional techniques such as e-beam evaporation. Atomic force microscopy (AFM) revealed a very rough surface topography on all of the deposited metal surfaces, which led us to examine a number of techniques for producing smoother films.

The semiconductor industry has developed chemical mechanical polishing (CMP) of metal and inter-layer dielectric (ILD) layers to enable the fabrication of metal interconnects in semiconductor-based ICs. A substantial CMP development

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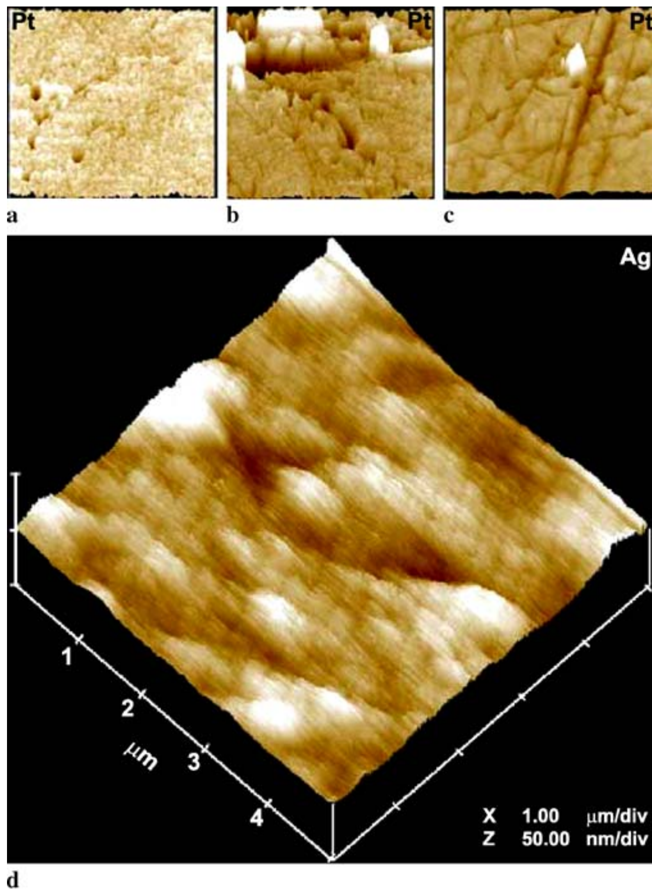


FIGURE 1 Typical micro/nano pits, scratches and slurry trapping on CMP polished Pt surfaces. Low-pressure polishing generates low density and shallow scratches and pits (**a**). Scratches, in general, follow a specific orientation during polishing under low pressure. At high pressure, the density of scratches increases and their orientation is random (**b**). A combination of pits and scratches generated by CMP is shown in (**c**) for a polished Pt surface. The issues escalate when soft surfaces such as Ag or Au are polished. (**d**) shows a Ag thin film surface after a 10-s CMP process was applied with very low pressure (less than 1 PSI). The Ag metal is partially delaminated during the polishing process

effort over several years in the microelectronics industry has resulted in noteworthy precision in controlling the material-removal rates, maintaining a high degree of planarity and keeping excellent local and global uniformity of the final surface. However, the nanoscale smoothness of polished metal surfaces is still rather poor.

A recent demonstration of polishing Pt surfaces with CMP showed some remarkably flat surfaces, but the issues of pits, scratches and slurry trapping are still major limitations [10]. Figure 1 presents some of these issues that we experienced while using CMP to generate smooth metal surfaces. The mechanism for material removal in CMP is highly dependent on a number of factors, which can be described by the Preston equation [11]

$$\frac{\mathrm{d}h}{\mathrm{d}t} = k_\mathrm{p} P v \,, \qquad (1)$$

where h is the thickness of the material removed, t the polishing time, P the applied pressure, v the relative velocity of the two surfaces in contact and k_p the Preston constant. The Preston constant is a function of the material hardness, the size of the abrasive as well as the chemical reaction kinetics [12]. The pressure distribution often depends on the feature size, shape and density [13].

A more generalized form of (1) typically simplifies the Preston constant to a ratio of the wear coefficient, k_w, to the hardness of the material being polished, H:

$$\frac{\mathrm{d}h}{\mathrm{d}t} = \frac{k_\mathrm{w}}{H} P v \,. \qquad (2)$$

Therefore, softer metals such as Ag with a hardness of around 500 MPa [14] allow a higher removal rate. Often, this results in the creation of surface defects, such as nano- or microscratches, phenomena such as dishing and overpolishing and even delamination of the film. CMP processes are also not well developed for many metals, especially those with a low modulus of elasticity such as Ag (75.8 GPa) and Au (69.5 GPa). Currently, the work on developing new slurries and pads for CMP of silver is still in the nascent stage [15, 16]. The role of the various chemical factors and their impact on the CMP process and on the surface roughness of the resultant Ag surface material are still poorly understood [17]. Moreover, CMP requires multi-step processes with different slurries, making it a tedious and expensive process to achieve surfaces with sub-nm roughness.

Figure 1d shows a typical surface of a Ag thin film deposited on a Si wafer after we polished it for only 10 s with virtually no applied pressure. We used a slurry that was developed for Pt (modulus of elasticity of Pt is 168 GPa), since there is no commercially available slurry for Ag. Similar issues of delamination were observed when Cu (modulus of elasticity of Cu is 130 GPa) polishing slurry was used for polishing Ag films. Hence, an alternative technique is needed for generating ultra-smooth surfaces on soft metals such as Ag and Au.

We designed our experimental setup to prevent delamination and suppress the creation of high-density micro- and nanoscratches during the smoothing process. Figure 2 is a schematic representation of the experimental apparatus, which consists of two parallel plates and an assembly to press them together. The bottom plate was held firmly on the stage of an imprinting tool while the top plate was constrained to translate in a vertical direction, allowing a variable separation between the plates. A flexible thin force sensor (Tekscan Inc.) was placed at the bottom plate to quantify the load acting on the samples. Silver-coated samples were then placed on the force sensor (face up), while a patterned Si mold was attached to the top plate (face down). In some experiments, the Ag samples were not constrained in-plane and hence were free to move laterally.

A customized pneumatic-pressure system was used to quasi-statically apply a variable pressure between 100 MPa to 700 MPa between the two plates to plastically deform the Ag film. Prior to pressing, the surfaces were ensured to be free of debris and dust particles, as they are known to create critical local stress concentration sites leading to substrate or mold breakage. The two surfaces were then pressed against each other along the vertical axis with varying controlled pressure and time, ensuring that sufficient contact has been achieved without any tilting. Once released, the samples were then characterized using AFM and scanning elec-

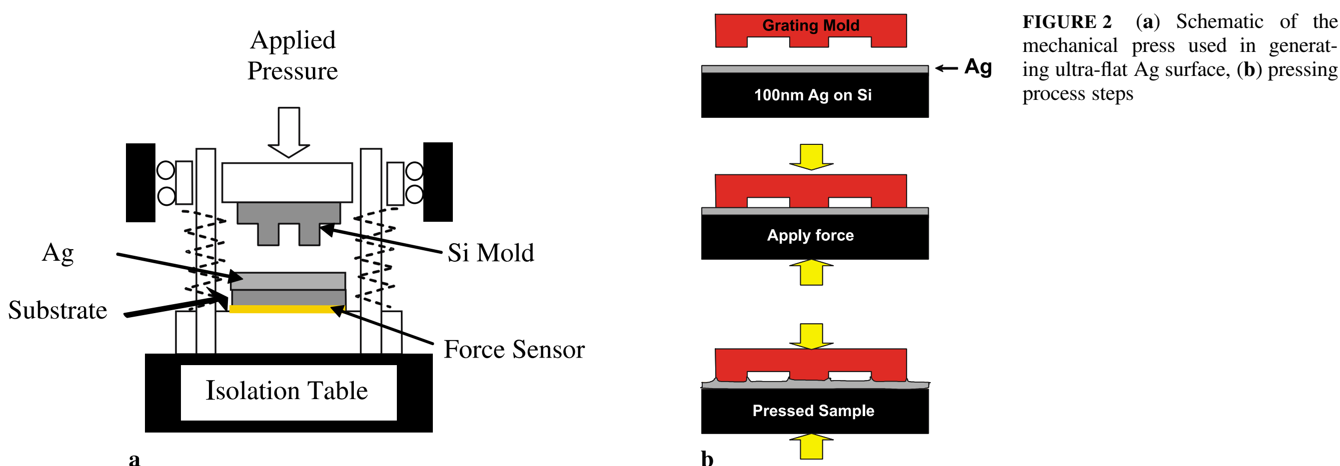
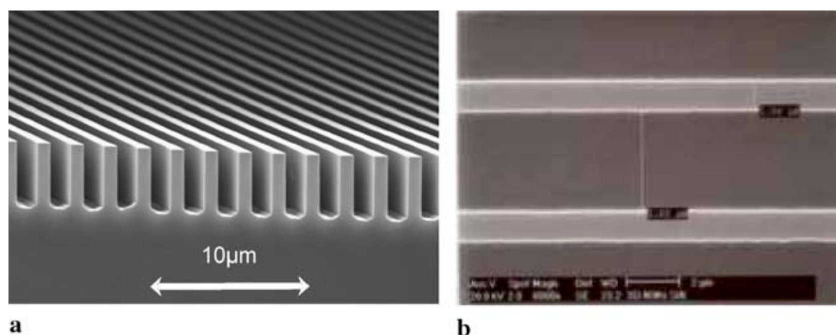


FIGURE 2 (**a**) Schematic of the mechanical press used in generating ultra-flat Ag surface, (**b**) pressing process steps



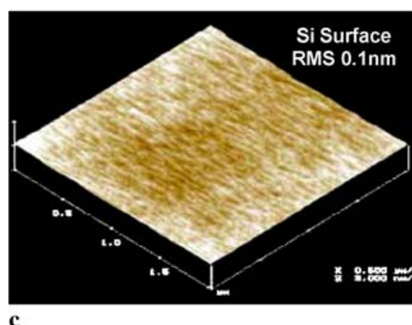


FIGURE 3 (**a**) A Si surface patterned with linear grating for pressing Ag film; (**b**) linear grating pitch of valley 4 µm and mesa 2 µm; (**c**) AFM characteristics of ultra-smooth Si surface with ∼ 1-Å RMS roughness

tron microscopy (SEM). Some of the etched Si mold samples were coated with a polymer releasing layer (1H,1H,2H,2H-perfluorodecyltrichlorosilane) to minimize the adhesion of Ag to the Si mold surface.

Figure 3a and b show the SEM image of the patterned Si grating mold surface and Fig. 3c shows a typical AFM surface profile of the Si mold. Due to the difficulty of processing an entire wafer using our current setup, we cleaved a (100)-oriented Si wafer (4″ diameter) into small pieces of ∼ 1 cm × 1 cm size. Chemical cleaning (acetone, IPA, methanol in ultrasonic bath) before lithography or after any imprint process was strictly observed to ensure highly clean surfaces. A thin Ag layer of ∼ 100 nm was deposited using an e-beam evaporator at a rate of ∼ 1 Å/s. It is important to keep the Ag samples in a nitrogen environment as oxidation further degrades the surface roughness. Another Si wafer with the same orientation and an ultra-smooth surface was patterned with grating features composed of ∼ 2-µm-wide lines and 4-µm spaces. These features were defined using photoresist as mask and removing Si on the exposed surface using a RIE process. The RIE etched sample was then characterized with AFM for surface roughness and was found be ∼ 0.1 nm in RMS roughness on a scan area of ∼ 2 µm × 2 µm.

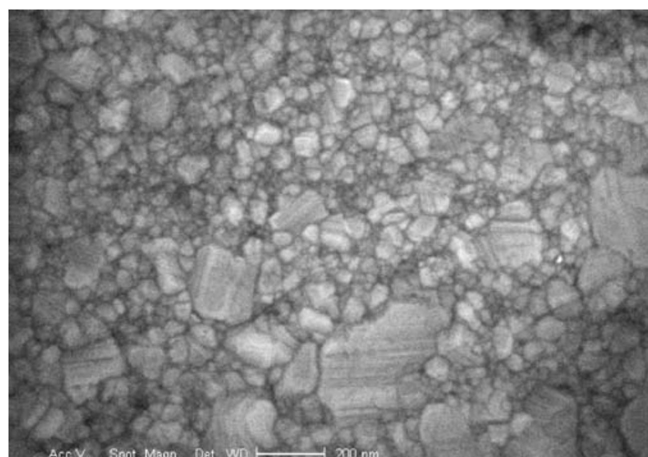


FIGURE 4 Freshly deposited 1000-Å Ag film on silicon(100) substrate

3 Results and discussion

A representative SEM surface morphology of 1000 Å freshly deposited Ag on a silicon(100) substrate is shown in Fig. 4. The large and random sized grains are clearly visible and they contribute to high surface roughness and hence directly to material losses in the optical regime for plasmonic nanodevices, photonic metamaterials,

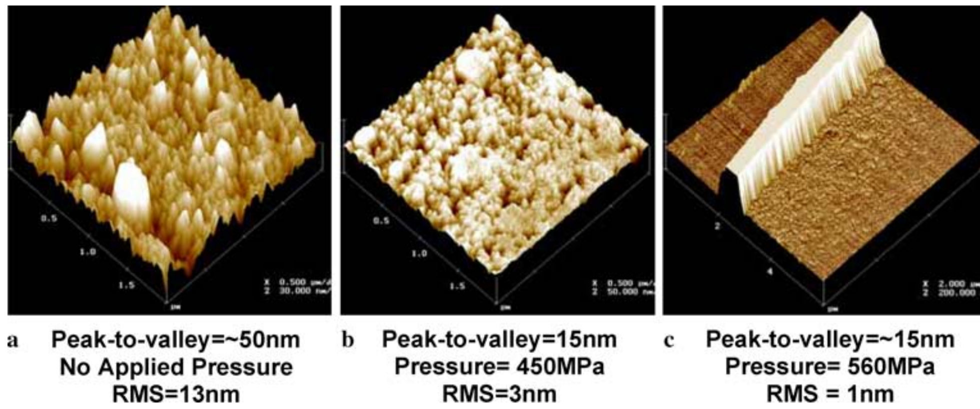


FIGURE 5 (**a**) As-deposited 1000-Å Ag film with ∼ 13-nm RMS roughness. (**b**) A half-smooth Ag surface with improved RMS roughness to 3 nm with applied pressure of 450 MPa. (**c**) Upon further increase of pressure to 560 MPa, material flow was achieved and mesas formed. The RMS was down to 1 nm

FIGURE 6 (**a**) SEM image of a large-area imprinted Ag grating on the sample with an applied pressure ∼ 600 MPa due to flattening of the grains on the Ag surface. (**b**–**d**) Magnified images of the grating areas indicating the mesas and the valleys. (**e**) AFM image of the Ag grating. (**f**) The *dotted line* in (**e**) was scanned with an AFM and the cross-sectional analysis shows ∼ 100-nm depth in the grating with ∼ 250-nm height above the Ag metal surface

superlens and electrical shorts in molecular-scale electronic devices.

In the first set of experiments, the Ag sample was not constrained and hence was free to undergo planar motion while the Si grating mold was fully constrained. Figure 5 shows Ag surface morphology before, in-between and after the completion of the pressing process with an ultra-flat Si grating mold surface. The pressing of a hard and flat Si surface (Fig. 3a) and a rough Ag thin metal surface (Fig. 4) has successfully generated a flat surface with reduced roughness. The roughness has changed from ~ 13 nm as-deposited to ~ 3 nm in a sample with 450 MPa applied pressure (Fig. 5b). Gradually increasing the pressure to 560 MPa induced the Ag material to plastically flow and thus the grating (mesa) pattern transfer was achieved. The RMS roughness was about 1 nm (Fig. 5c).

Over a larger pressed area of an unconstrained Ag sample, the grating mold was successfully imprinted onto the Ag film, as depicted in Fig. 6a–f. The periodicity and fidelity of the imprinted Ag grating pattern have been carefully preserved and are in good agreement with the original Si grating mold geometry. The Ag grating valley was 4.11 μm and the mesa 0.8 μm compared to 3.88 μm and 1.08 μm on the Si grating mold.

The high surface roughness contrast region is shown in Fig. 6d. The Ag grating mesa delineates the un-pressed Ag and the pressed plastically deformed smooth Ag surfaces. The Ag imprinting pattern transfer is a result of the plastic deformation that involves the breaking of a small number of atomic bonds by the movement of dislocations. While the force needed to break the bonds of all the atoms in a crystal plane all at once is enormous, the movement of dislocations requires little energy and allows atoms in crystal planes to slip past one another at much lower stress levels. This, as a result, helps to flatten any uneven and jagged metal surface. Heat treatment is likely to enhance the movement of dislocations and a low modulus of elasticity expedites this process.

In order to highlight a gradual change in roughness with an applied pressure gradient, both the Ag sample and the Si mold were completely constrained from moving. The Si mold used was now a square mesa 100 μm × 100 μm with a height of 2.6 μm instead of a grating pattern. At a pressure of ~ 600 MPa, the imprinted Ag surface results in approximately 1-nm surface roughness across the imprinted surface while in the smoothest regions a roughness below 0.1 nm is achieved (Fig. 7a–c). This deformation can be observed by strictly confining the relative lateral movement of the square imprint mold and Ag-coated substrate.

Figure 7a (SEM) and b (AFM) show both un-pressed (A) and pressed (B) regions that were on either side of a square

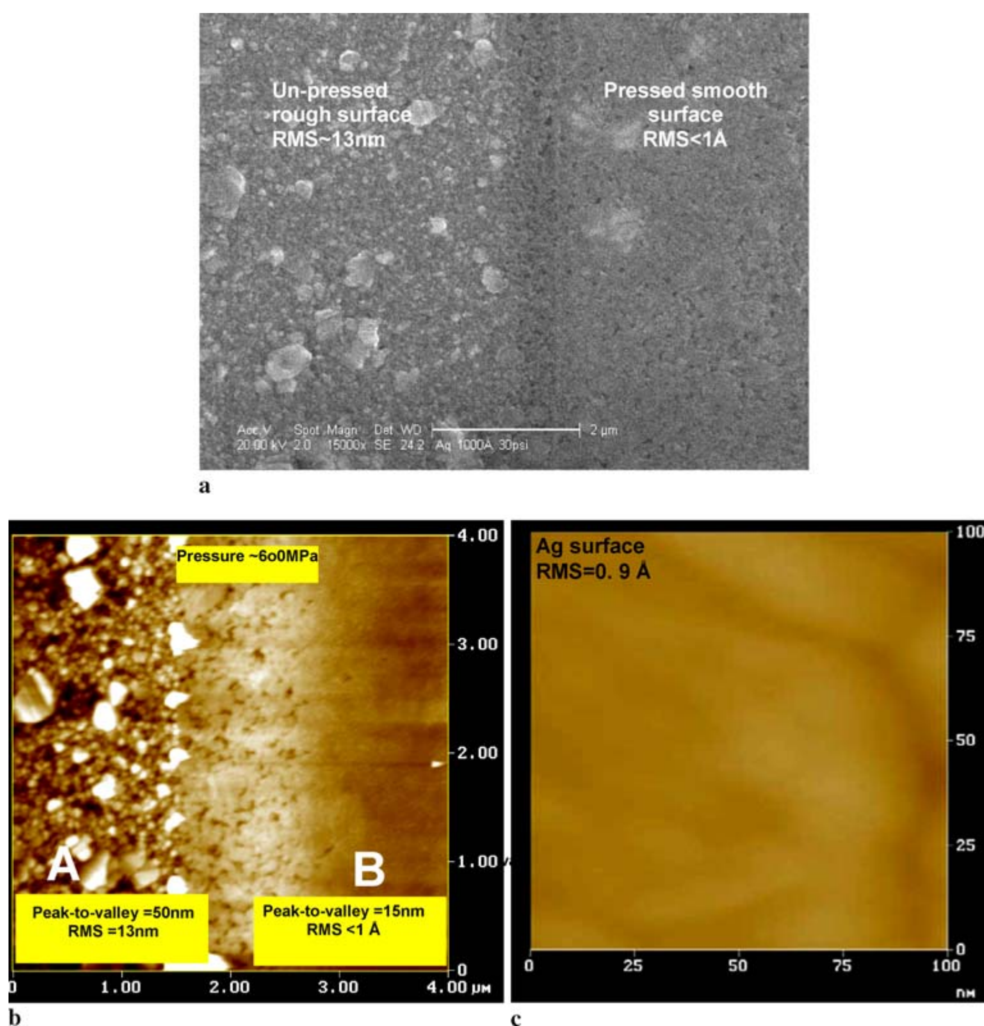


FIGURE 7 (**a**) SEM image showing the gradual change in the Ag surface roughness. (**b**) The RMS roughnesses of the unpressed (region A) and pressed (region B) surfaces are shown. More than two orders of magnitude improvement in the smoothness of the Ag surface is observed. (**c**) An ultra-flat region with a surface roughness less than 0.1 nm

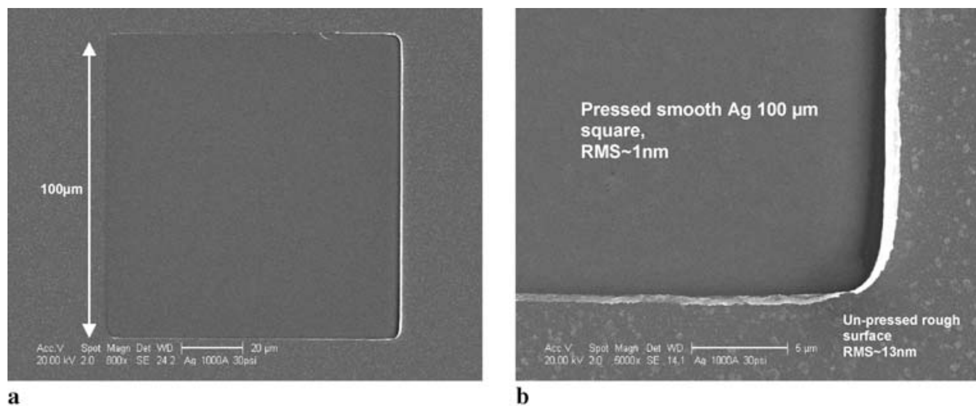


FIGURE 8 (**a**) SEM image showing the 100 µm by 100 µm square pattern imprinted on a Ag sample; (**b**) a magnified image of the boundaries between the rough and smooth surfaces. Due to the nature of the mold–sample constraint and plastic deformation there is a non-uniform variation in the depth of the transferred pattern

mesa on the Si mold. A distinctive difference can be observed from the AFM image. The surface roughness of side B clearly demonstrates the potential of our pressing technique for creating a smooth metal surface.

Consequently, a combination of transverse and lateral movement of the mold relative to the substrate may open opportunities for Ag direct patterning using a hard mold. Hence, a direct patterning and simultaneous smoothing of a Ag surface is feasible and is evident in Fig. 8a and b. By restricting the Ag smoothing process to a fixed area using simple mold geometry, specific locations on a 1 cm × 1 cm sample may be imprinted. An immediate device application would be, for example, flattening of a superlens or optical metamaterial that uses Ag as the plasmonic metal [18].

4 Conclusion

We developed a novel technique to generate an ultra-flat surface on soft metals such as Ag and Au. A sub-0.1-nm roughness was achieved using this pressure-induced mechanical deformation. Currently we are addressing the issue of adhesion of Ag to the mold by using an improved releasing layer between the Ag film and the Si mold. Further studies and thorough optical and electrical characterization will shed more light on this technique. Understanding of heat treatment during the pressing process will also be investigated. Our demonstrated technique is mass-manufacturable and can be executed using a nanoimprint lithography system. The results could potentially decrease scattering losses in optical plasmonic nanodevices and metamaterial-based devices operating at optical frequencies and inhibit short-induced failure in all nanoscale devices that interface with metal surfaces.

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