# A smooth optical superlens

Pratik Chaturvedi,[1] Wei Wu,[2] VJ Logeeswaran,[3] Zhaoning Yu,[2] M. Saif Islam,[3] S. Y. Wang,[2] R. Stanley Williams,[2] and Nicholas X. Fang[1,a)]
[1]*Department of Mechanical Science and Engineering, University of Illinois at Urbana-Champaign, 1206 W. Green St., Urbana, Illinois 61801, USA*
[2]*Information and Quantum Systems Lab, Hewlett-Packard Laboratories, 1501 Page Mill Rd, MS 1123, Palo Alto, California 94304, USA*
[3]*Department of Electrical and Computer Engineering, University of California at Davis, One Shields Ave, Kemper Hall, Davis, California 95616, USA*

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We demonstrate a smooth and low loss silver (Ag) optical superlens capable of resolving features at $1/12^{\text{th}}$ of the illumination wavelength with high fidelity. This is made possible by utilizing state-of-the-art nanoimprint technology and intermediate wetting layer of germanium (Ge) for the growth of flat silver films with surface roughness at subnanometer scales. Our measurement of the resolved lines of 30 nm half-pitch shows a full-width at half-maximum better than 37 nm, in excellent agreement with theoretical predictions. The development of this unique optical superlens leads promise to parallel imaging and nanofabrication in a single snapshot.  [doi:10.1063/1.3293448]

The resolution of optical images has historically been constrained by the wavelength of light, a well known physical law which is termed as the diffraction limit. Recent theory, however, suggested that such a limitation can be overcome by a thin negative index (simultaneous negative permittivity and permeability) film which allows recovery of evanescent waves that carry the subdiffraction information.[1] This planar slab of negative index film, termed as "superlens," derives this ability by excitation of surface plasmons. It has been shown that in the electrostatic near-field limit, having only negative permittivity suffices for near-field superlensing effect for transverse magnetic (TM) polarization.[1] This makes metals like silver which allow the recovery of evanescent waves,[2] a natural candidate for superlens at optical frequencies. It has been demonstrated experimentally[3] that a silver superlens allows to resolve features well below the working wavelength. Resolution as high as 60 nm half-pitch or $1/6^{\text{th}}$ of wavelength has been achieved.

Theoretically, it was predicted that a resolution as high as $\lambda/20$ (where $\lambda$ is the illumination wavelength) is feasible with careful design of silver superlens.[4] However, challenges remain to realize such a high resolution imaging system, such as minimizing the information loss due to evanescent decay, absorption or scattering. Particularly, the surface morphology of silver film plays a significant role in determining the image resolution capability. Below a critical thickness silver is known to form rough islandized films.[5] Rougher films perturb the surface plasmon modes causing loss of subwavelength details and hence diminished resolution.[6] Some recent research efforts directed toward smoothing silver films have utilized template stripping[7] and other postprocessing steps such as chemical polishing or mechanical pressing.[8] Template stripping is however limited to thicker films (>30 nm), as thinner films have poor wettability on glass or silicon substrates. Mechanical polishing technique, on the other hand, suffers from issues common to contact processes such as creation of surface defects, scratches, and delamination of Ag films.

In this work, we utilize a unique approach to grow ultrasmooth silver films characterized by much smaller root mean square (RMS) surface roughness. An intermediate ultrathin Ge layer (~1 nm) is introduced before depositing Ag.[9] Utilizing Ag–Ge surface interactions, smooth superlens down to 15 nm Ag thickness was fabricated. Roughness measurements of thin silver films (15 nm) deposited with and without Ge layer (1 nm) were performed using atomic force microscopy (AFM) and x-ray reflectivity techniques. These measurements indicate drastic improvement in Ag surface morphology with roughness below 0.8 nm with introduction of Ge layer.[10] It is postulated that Ge acts as a wetting layer for Ag and helps a layer by layer growth. A detailed study of the growth of Ag on Ge has been described elsewhere.[9] Some earlier works have indicated that thin and smooth Ag films can also be prepared epitaxially with metal oxides such as magnesium oxide and nickel oxide.[11]

The configuration of the smooth silver superlens is illustrated in Fig. 1. An array of chrome (Cr) gratings 40 nm thick with 30 nm half-pitch, which serve as the object, was

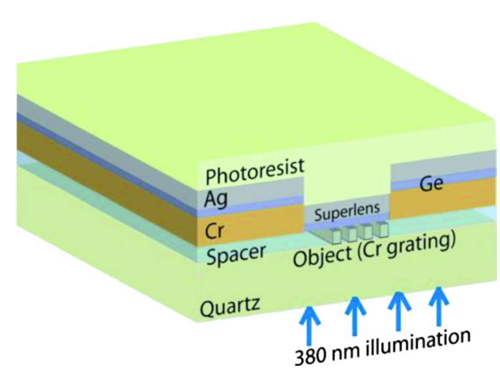


FIG. 1. (Color online) Schematic drawing of a smooth silver superlens with embedded 30 nm chrome gratings on a quartz window, operating at 380 nm wavelength. To prepare the smooth superlens, a thin germanium layer is seeded.

[a)]Author to whom correspondence should be addressed. Electronic mail: nicfang@illinois.edu.

 

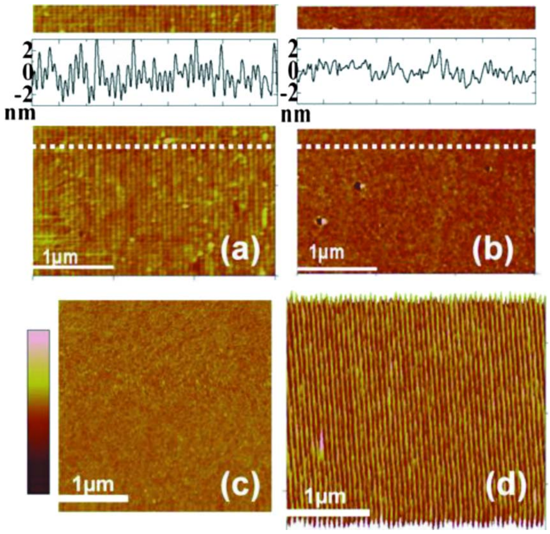


FIG. 2. (Color online) Step by step surface characterization of the prepared smooth silver superlens sample with embedded gratings using AFM. (a) Close-up image ($3\times3\ \mu m^2$) of the nanoimprinted chrome gratings of 30 nm half-pitch prepared on quartz windows. Inset presents the line section plot at the marked dotted line. (b) Surface profile of the sample after planarization with 6 nm spacer layer onto chrome gratings, showing an RMS roughness of 1.3 nm. (c) Surface profile of the sample after the deposition of Cr window, Ge and Ag layer. (d) The image of the 30 nm half-pitch Cr grating area recorded on the photoresist layer after exposure and development. (Color scale for all images: 0 to 20 nm).

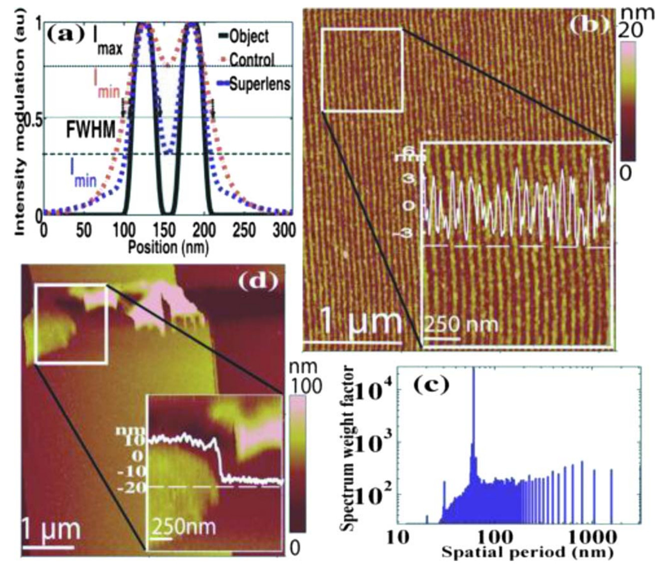


FIG. 3. (Color online) (a) Computed image modulation with superlens FWHM=37 nm (blue dashed curve) and without superlens FWHM =113 nm (red dotted curve). (b) Subdiffraction optical imaging with superlens: AFM of developed photoresist with zoom in panel showing section analysis. (c) Fourier analysis of the developed image grating. (d) Near-field optical imaging without superlens.

patterned using a nanoimprint process developed by Hewlett-Packard Laboratory. In Fig. 2, we present a step-by-step surface characterization of the prepared smooth silver superlens with embedded chrome gratings using AFM. In order to prepare a flat superlens on top of the objects [Fig. 2(a)], it is necessary to deposit a planarization layer to reduce the surface modulations. Surface modulations can alter the dispersion characteristics of the plasmons and it smears out the image details. Also, the planarization layer should be thin to prevent a significant loss of evanescent components from the object. In our process, a planarization procedure using nanoimprint technique is developed to reduce the surface modulations below 1.3 nm [Fig. 2(b)]. This is achieved by flood-exposure of 66 nm thick UV spacer layer over a flat quartz window under pressure, followed by subsequent reactive ion etching to back etch the spacer to 6 nm thickness on top of the chrome gratings. A 35 nm thick Cr window layer is photolithographically patterned on top of the spacer layer to enhance the contrast with dark-field imaging. Subsequently, 1 nm Ge and 15 nm Ag layer (superlens) is evaporated over the window layer [Fig. 2(c)], followed by coating with a thick layer of optical adhesive (NOA-73) which serves as the photoresist. The substrate is exposed with a collimated 380 nm UV light for 120 s (Nichia UV-LED, 80 mW). The optical image recorded on the photoresist is developed and imaged with AFM [Fig. 2(d)].

For a qualitative comparison, we theoretically computed the resolving power of a thin ultrasmooth Ag–Ge superlens. Using transfer matrix approach,[12] we computed the point spread function (PSF) of the multilayer lens system comprising of the spacer (6 nm), Ge (1 nm) and Ag layers for TM polarization at incident wavelength of $\lambda=380$ nm. Adding Ge is generally unfavorable at UV wavelengths, as it is absorptive. However with only 1 nm thick Ge in Ag–Ge superlens, the evanescent decay is only significant for feature sizes below $\lambda/12$. Computed PSF of such a superlens with optimal thickness of Ag (20 nm) has full-width at half-maximum (FWHM) of 23 nm. An object grating constructed with FWHM of 30 nm at 60 nm pitch when convoluted with this PSF gives an image grating with FWHM=37 nm [Fig. 3(a), dashed blue curve]. Moreover, the intensity contrast appearing in the image ($r=I_{max}/I_{min}\sim3$) is sufficient to resolve this object with most commercial photoresists using superlens photolithography. In contrast, a near-field lens without Ag layer (e.g., spacer 27 nm thick) gives a PSF with FWHM of 45 nm. Constructed image of the object grating with this lens gives a FWHM of 113 nm ($\sim\lambda/3$) [Fig. 3(a), dotted red curve]. The resulting image contrast ($r\sim1.3$) is not sufficient to resolve the grating using photolithography.[13] We experimentally verify our findings by imaging Cr gratings with 30 nm width at 60 nm pitch using Ag–Ge superlens and near-field control lithography experiments without Ag.

Figure 3(b) shows the image of the Cr grating area recorded on the photoresist layer after exposure and development. It is evident from section analysis of the recorded image (zoom in panel) that with careful control of surface morphology, the recorded image has ~6 nm height modulations. The Fourier-transformed spectrum shows clear peaks up to third harmonic of the 60 nm pitch Cr gratings recorded on the resist layer [Fig. 3(c)]. In a control experiment, when the Ag–Ge layers are replaced by equally thick spacer layer, we observe that only a portion of grating area is developed [Fig. 3(d)]. Moreover, the developed wires are much thicker (~47 nm) and the poor contrast suggests loss of resolution as predicted by the PSF calculation. This confirms that near-field imaging alone without evanescent enhancement is not capable of resolving high-frequency spatial features ($\sim\lambda/12$) located just 27 nm($=\lambda/14$) away from the surface.

In conclusion, we have demonstrated a unique approach to realize ultrasmooth Ag superlenses with an unprecedented $\lambda/12$ resolution capability. Incorporating few monolayers of Ge drastically improves Ag film quality and minimizes the subwavelength information loss due to scattering. Our theoretical and experimental results clearly indicate subdiffraction imaging down to 30 nm half-pitch resolution with 380 nm illumination. This ultrahigh image resolution capability can also be extended to far-field[14] by incorporating a corrugated silver surface on top of Ag–Ge superlens. The development of such unique superlenses would enable real-time dynamic imaging at the molecular level.

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