## Extraordinary Optical Transmission of Ultra-Thin Freestanding Plasmonic Membranes

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**Abstract:** We demonstrate an ultra-thin freestanding plasmonic membrane that supports surface plasmon resonances. The 30 nm-thick membrane is perforated with an array of holes using the imprint-and-transfer approach. The fabricated plasmonic membrane exhibits extraordinary optical transmissions in the mid-wave infrared wavelength range and can be used as an optical sensor to measure the absorption of a thin polymer film.

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Nanophotonic devices that support plasmon resonances have been widely studied for their applications in solar cell, solid-state lighting, biological and chemical detection, and lasers [1, 2]. In particular, the metal film with periodic patterns supports a phenomenon known as extraordinary optical transmission (EOT), where the percentage of light transmitted through the metal film patterns can exceed the percentage of open area at the EOT resonant wavelengths [3, 4]. The EOT structures have been studied as potential optical filters, and they are of significant interest as biosensors to measure biomolecule interactions. Here, we demonstrate an ultra-thin freestanding plasmonic membrane with the thickness of 30 nm. The plasmonic membrane with an array of holes was fabricated by the imprint-and-transfer approach. By leveraging the plasmonic resonances and the chemical specificity of vibrational spectroscopy, we performed real-time and label-free detection of thin polymer films.



Fig. 1. (a) - (f) Fabrication process flowchart for the freestanding plasmonic membrane. The imprint-and-transfer approach was used to generate the patterned membranes inexpensively. (f) Schematic of the freestanding plasmonic membrane.

Fig. 1(a) shows a schematic illustration of the ultra-thin plasmonic membrane comprised of a freestanding gold film with a two dimension (2D) square lattice of sub-wavelength holes. The membrane is suspended over the 100  $\mu$ m-diameter apertures of a nickel mesh. The geometry of this membrane, including period (A), hole diameter (d), and membrane thickness (t), can be designed to manifest plasmon resonances in a desired wavelength range. As summarized in Fig. 1(b), the fabrication process of the plasmonic membrane consists of three major steps: imprint, transfer, and thin-film coating. Before imprint, a PDMS stamp was prepared by molding from a silicon wafer bearing the 2D array of nano-wells [5]. The array pattern was first transferred to a 50 nm-thick polymer film by squeezing a low viscosity copolymer of 0.5% poly(vinyl formal) between the PDMS stamp and a glass slide. The copolymer was at room temperature under a pressure of 8.3 kPa. After removing the PDMS stamp, the patterned polymer film was transferred from the glass slide to the surface of water in a beaker. The surface tension of water helped to release and supports the polymer film. We placed the nickel mesh on the polymer film and carefully picked up the mesh. The sample was baked (at 100 °C) to assure that the polymer membrane was fully perforated. Finally, a 30 nm-thick film of gold was deposited over the polymer membrane and the polymer membrane was removed by soaking the sample in chloroform.



Fig. 2. SEM images of the fabricated plasmonic membrane. (a) Membrane on a mesh. (b) Plasmonic membrane with  $\Lambda = 2.5 \mu m$ , d = 1.25  $\mu m$ , and t = 30 nm. (c) SEM of the cross-section of the gold membrane. Scale bar: (a) 100  $\mu m$ ; (b) and (c) 5  $\mu m$ .

Fig. 2(a)-(c) is the scanning electron microscopy (SEM) images of the plasmonic membrane. The plasmon resonances of the device were modeled using finite-difference time-domain (FDTD) simulation and characterized experimentally by a Fourier transform infrared spectrometer. The simulation and measurement results are compared in Fig. 3(a) and (b) as a function of incident angle ( $\theta_i$ ). The measured transmission spectra show very good spectral agreement with simulation results. The plasmonic membrane exhibits significantly enhanced peak transmission at the resonant wavelength ( $\lambda_r$ ). When normalized to the same mesh without the metal membrane, the peak transmission reaches 70% for the normal incidence case. The resonant wavelength increases with the change of the incident angle. When  $\theta_i = 0^\circ$ , 10°, and 20°, the resonant wavelength are 2.94 µm 3.21 µm, and 3.60 µm respectively. The angular dispersion of the EOT structure is approximately 33 nm/°. As the incident angle moves away from normal, we observe a decrease in the transmission coefficients, which is expected for incident light with a wave vector in the direction of SPP propagation. During the tests, the incident light was TM polarized with the electric field component parallel to the incident plane.



**Fig. 3**. (a) Measured transmission spectra at different incident angle ( $\theta_i = 0^\circ$ , 10°, and 20°). (b) FDTD simulation results for the plasmonic membrane.

**Fig. 4**. Transmission spectra of the plasmonic membrane coated with a thin polymer film.

The infrared plasmonic membrane offers remarkable opportunities for exploration of surface-based sensors. As an example, we adopted the plasmonic membrane to characterize the coating of a thin polymer film. The 50 nmthick film was coated by dipping the device in Formvar solution (0.1%) and air-dried. As shown in Fig. 4, the coating results in a shift of the EOT peak by 220 nm. The phenomena can be exploited for a refractive index-based label-free detection. In addition, the plasmonic membrane is capable of enhancing the chemical specific absorption in infrared, owing to the strengthened near field around the membrane. By comparing the measurements at  $\theta_i = 0^{\circ}$ and 30° shown in Fig 4, it can be seen that the absorption C–H stretching modes (2950 cm<sup>-1</sup> and 2994 cm<sup>-1</sup>) is significantly enhanced when vibrational modes of analyte fall in the plasmonic resonance.

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