Thickness Dependent Optical Properties of Transdimensional TiN

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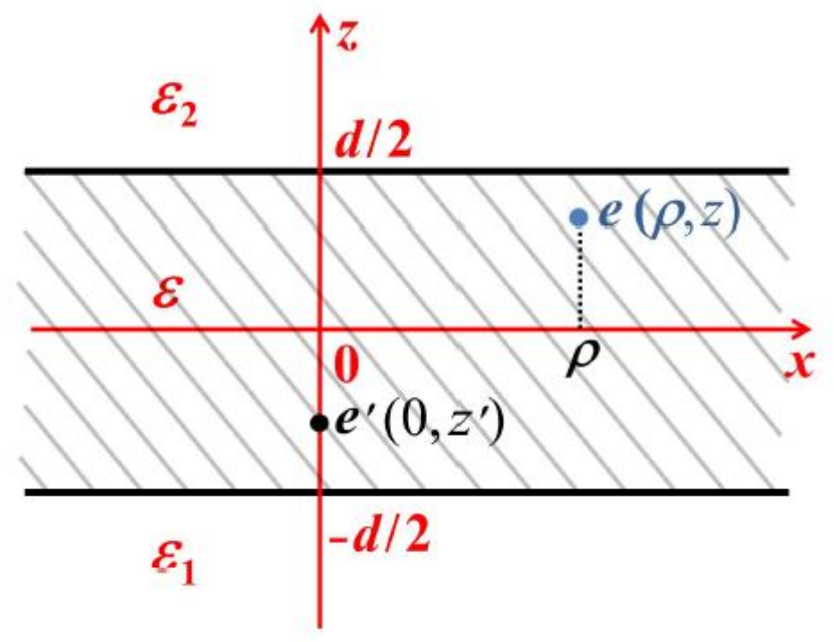
Introduction

Transdimensional materials (TDMs) are atomically thin metals with thicknesses below 10 nm. Many unique light matter interactions and material functionalities in this thickness regime[1].

- Enhanced nonlinear optical properties[2]
- High sensitivity to external electrical and optical perturbations[3]
- Engineering the optical properties by adjusting, thickness, surface termination, and strain[4]

Growing continuous, ultrathin metal films is a challenge, particularly for noble metals. On the other hand, epitaxial, continuous TiN films with thicknesses down to 1-2 nm can be grown[5], making them an attractive material platform for tailorable and dynamically switchable metasurfaces[6].

Nonlocal Drude Model



As the film thickness *d* decreases, the Coulomb potential between two charges increases due to spatial confinement.

When the inter-charge distance is larger than *d,* the Coulomb potential turns into the in plane 2D potential called the *Keldysh-Rytova (KR) potential*

The thickness dependent plasma frequency is derived based on the KR potential as[7]

$$\omega_p = \omega_p(k) = \frac{\omega_p^{3D}}{\sqrt{1 + (\varepsilon_1 + \varepsilon_2)/\varepsilon k d}}$$

where *k* is the plasmon wave vector and ω_p^{3D} is the bulk plasmon frequency.

The plasma frequency acquires a spatial dispersion.

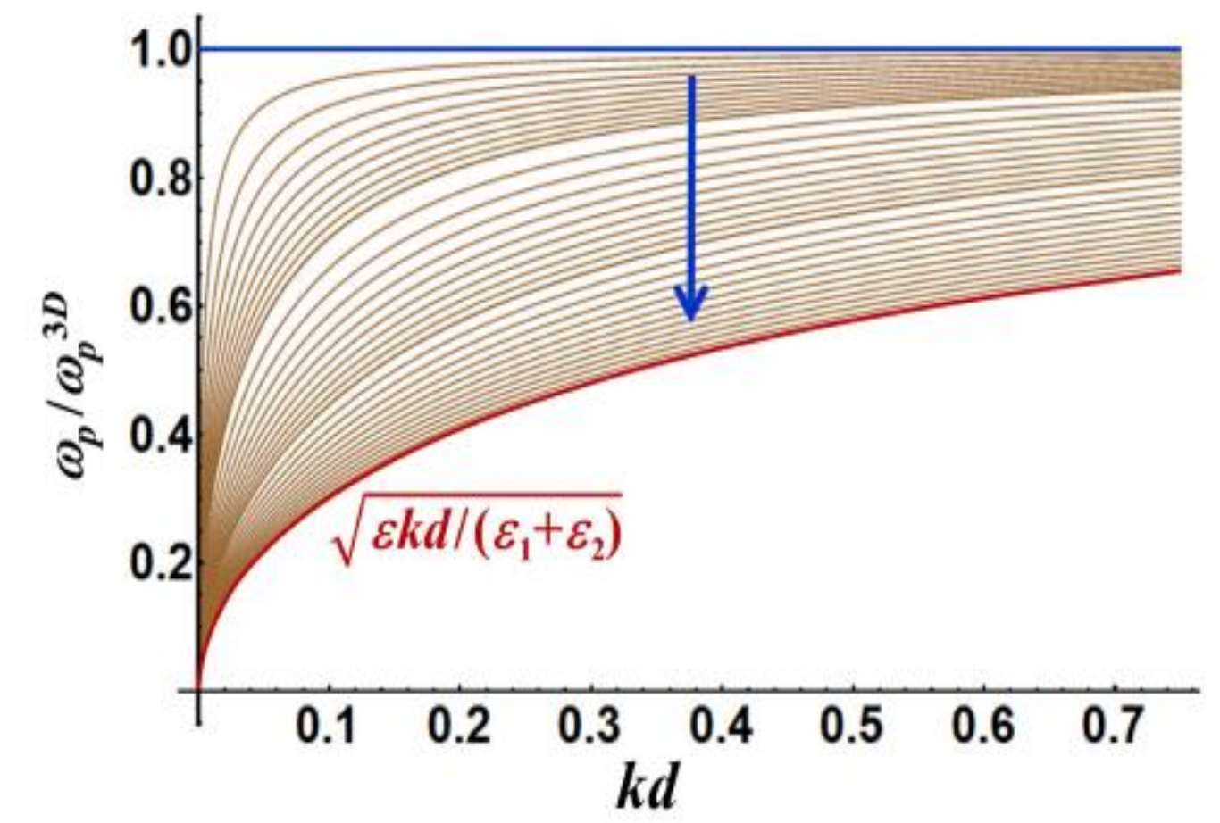


Fig. 1. Normalized plasma frequency as a function of *kd*

Material Growth

Atomically flat, epitaxial ultrathin TiN films (1 – 10 nm thickness) were grown on MgO using reactive magnetron sputtering with the substrate heated to 800 C. A 10 nm in-situ passivation layer of AlScN was grown on each TiN film to prevent oxidation.

Fig. 2. AFM of passivated 2 nm TiN

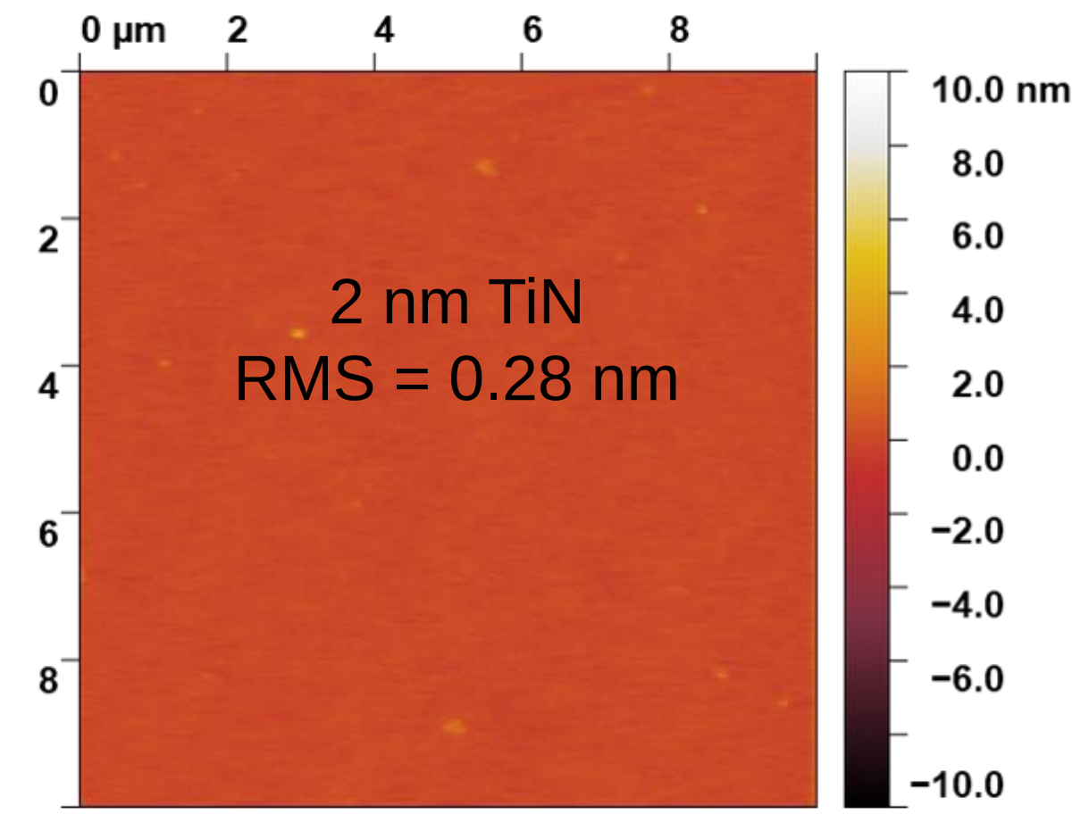
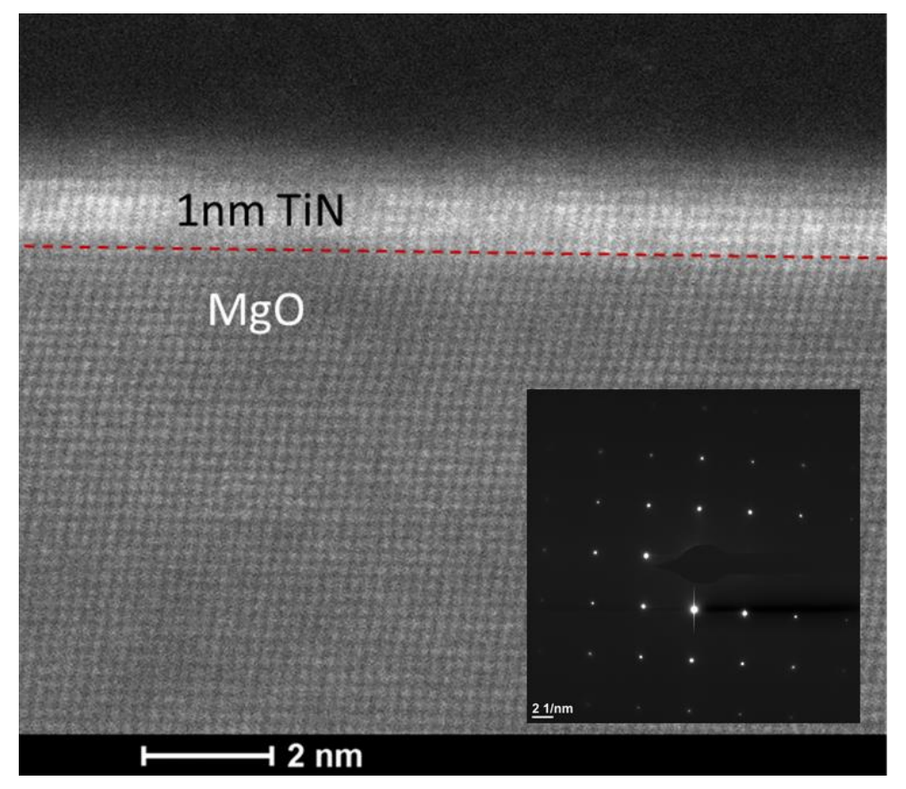


Fig. 3. TEM of epitaxial 1 nm TiN



Optical Characterization

Spectroscopic Ellipsometry
A Drude-Lorentz model was used to fit the ellipsometry measurements,

$$\varepsilon(\omega) = \varepsilon_b - \frac{\omega_{pD}^2}{\omega^2 + i\Gamma_D \omega} + \sum_L \frac{f_L \omega_L^2}{\omega_L^2 - \omega^2 - i\Gamma_L \omega}$$

where ε_b is the background permittivity, ω_{pD} is the Drude plasma frequency, Γ_D is the Drude damping, f_L, ω_L, and Γ_L are the strength, resonant frequency and broadening of the Lorentz oscillator, respectively.

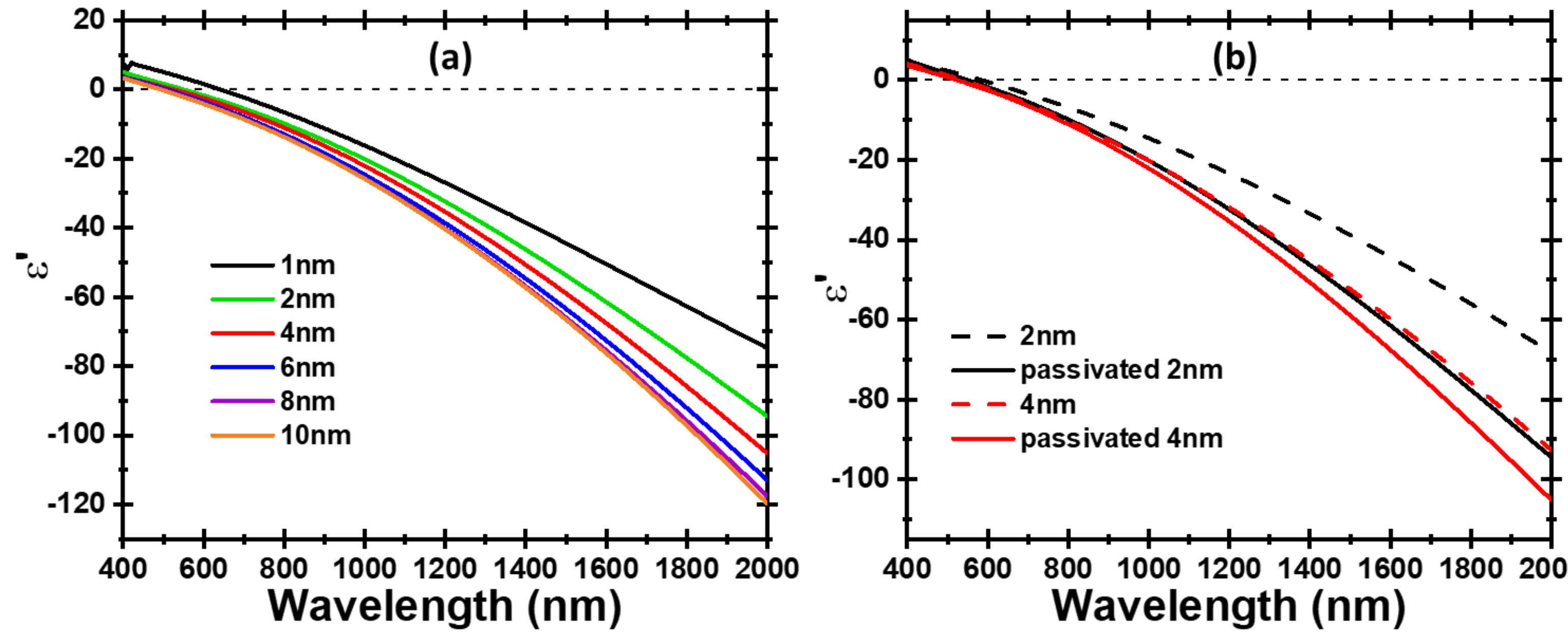


Fig. 4. (a) Real part of the permittivity, *ε'*, for different thicknesses. (b) ε' of unpassivated and passivated films. Passivated films have a larger |*ε'*|, indicating a stronger metallic character.

- Metallicity decreases and crossover wavelength red shifts in thinner films
- 1-2 nm films are still plasmonic at visible and NIR wavelengths

Thickness Dependent Drude Plasma Frequency

The thermal averaging of ω_p from the nonlocal Drude model gives[8]:

$$\overline{\omega}_p(d) = \frac{2C^2 d^2\, \omega_p^{3D}}{(1+2Cd)\sqrt{Cd(1+Cd)} - \sinh^{-1}(\sqrt{Cd})} \qquad C = \varepsilon_b k_c / (\varepsilon_1 + \varepsilon_2)$$

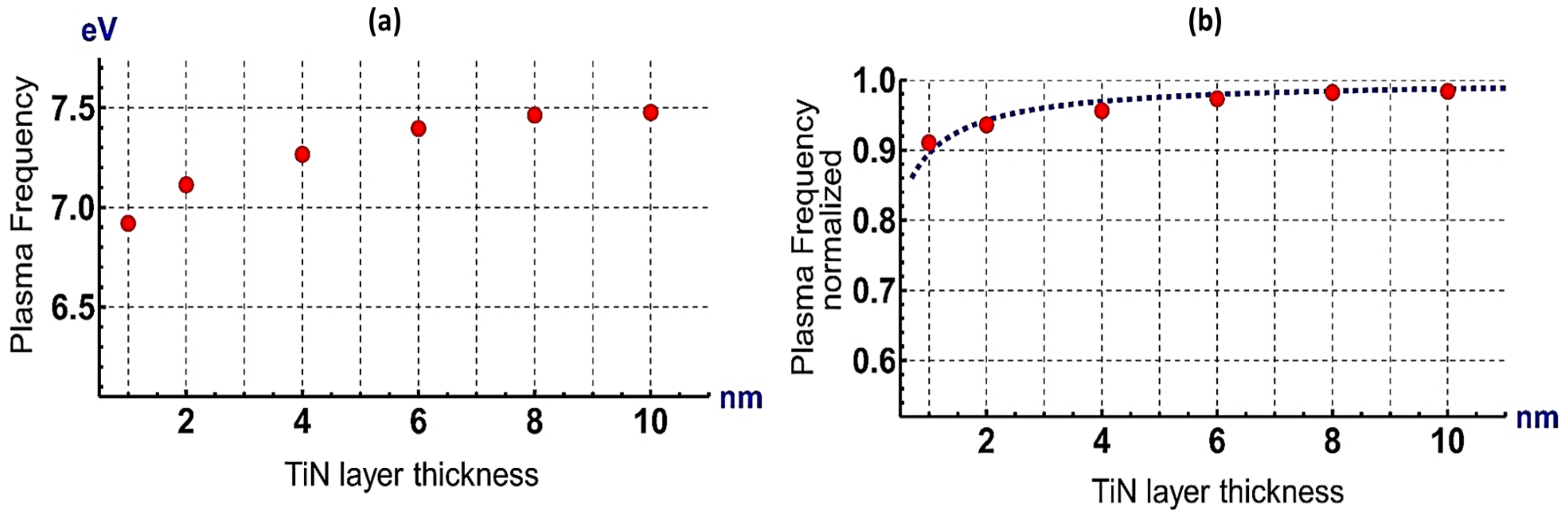


Fig. 5. (a) Thickness dependent plasma frequency extracted from ellipsometry. (b) The data point set of (a) normalized. The black dashed line shows the ratio $\bar{\omega}_p(d)/\omega_p^{3D}$ as given by the fitting with parameter *C* = 7.7 nm^{-1} (ε_b = *9.1)*.

The experimentally measured thickness-dependent plasma frequency agrees with the predictions of the nonlocal Drude model

Conclusion

- We are able to grow continuous, epitaxial films of TiN down to 1 nm with low roughness, while maintaining metallic properties
- A decrease in the metallicity and Drude plasma frequency in thinner films is observed due to spatial confinement.
- We demonstrate confinement induced optical properties in plasmonic transdimensional TiN as predicted by the nonlocal Drude model based on the Keldysh-Rytova potential

References

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