

# Supporting information for: Optical dielectric function of silver

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In this work three template stripped (TS) silver samples are measured with spectroscopic ellipsometry. The resulting dielectric data are corrected for Kramers-Kronig consistency and are shown in the main text together with data before correction. The dielectric data of silver before correction for Kramers-Kronig consistency are directly inverted from the ellipsometry measurements, and retains the instrumental errors. For consistency check with our previous work [1] and for comparison with silver, we also measured the dielectric function of three TS gold samples as described in the main text. We recommend ‘Ag\_C\_corrected.csv’ being used as a standard reference for silver dielectric function.

## I. ELLIPSOMETRY DATA OF SILVER BEFORE AND AFTER CORRECTION

Fig. S1 shows the direct inverted dielectric data from ellipsometry measurements and the Kramers-Kronig consistent fit with good agreement. Photon energy in linear scale (a), and in logarithmic scale (b) are plotted to emphasize the Vis/UV and IR spectral range, respectively. Due to residual instrumental errors the direct inverted dielectric data are not perfectly Kramers-Kronig consistent. We therefore fit the dielectric data of silver with an analytic formula consisting a Drude and three Gaussian functions, a procedure commonly used for metals [2, 3].

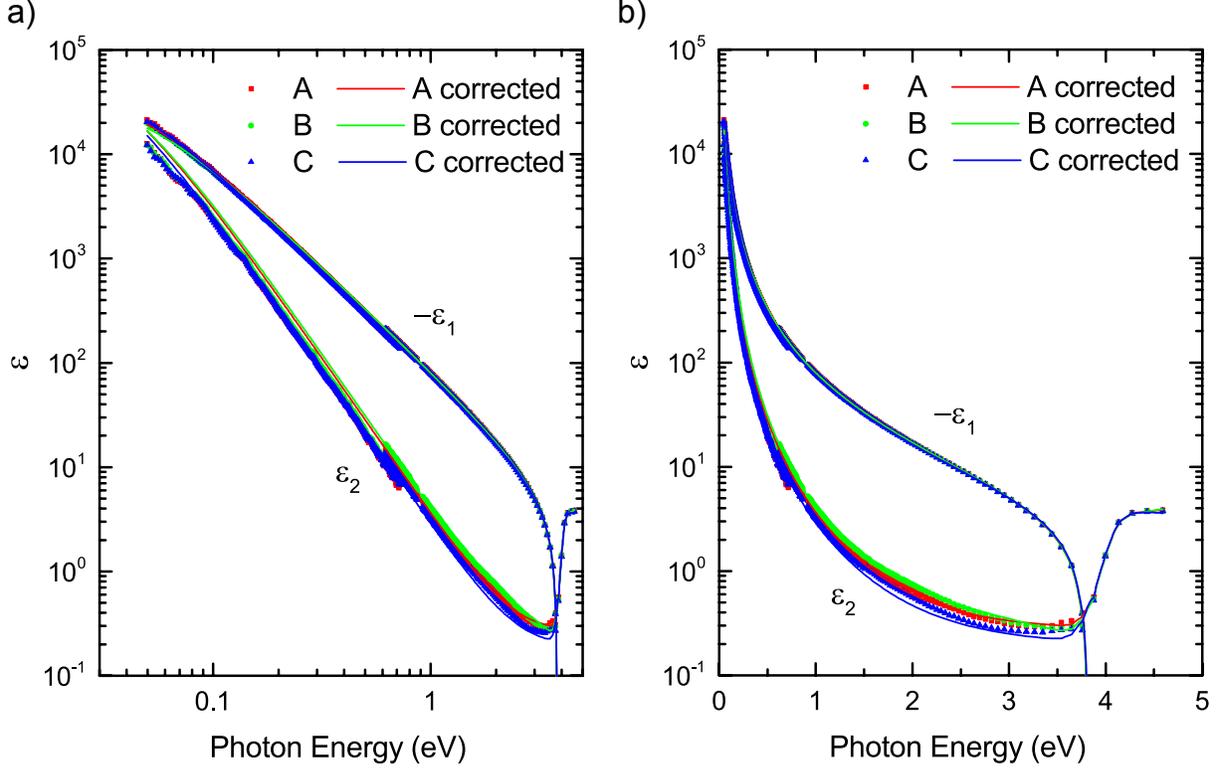


FIG. S1. Dielectric function of silver from direct inverted ellipsometry measurement (dot), and a Kramers-Kronig consistent fit (line). The three samples are shown in red, green, and blue. Photon energy in linear scale (a), and in logarithmic scale (b) are plotted to emphasize the Vis/UV and IR spectral range, respectively.

As noted in the main text, the deviation of the direct inverted dielectric function  $\epsilon_2$  from the Drude fit at energies below 0.1 eV is the cause of a frequency dependent  $\tau(\omega)$ .

## II. DIELECTRIC FUNCTION OF GOLD

Fig. S2 shows the direct inverted dielectric data of TS gold measured in this work in comparison with fitted dielectric function presented in our previous paper [1]. The relative differences in measured dielectric data between the three TS gold samples are less than the differences between the three TS silver samples.

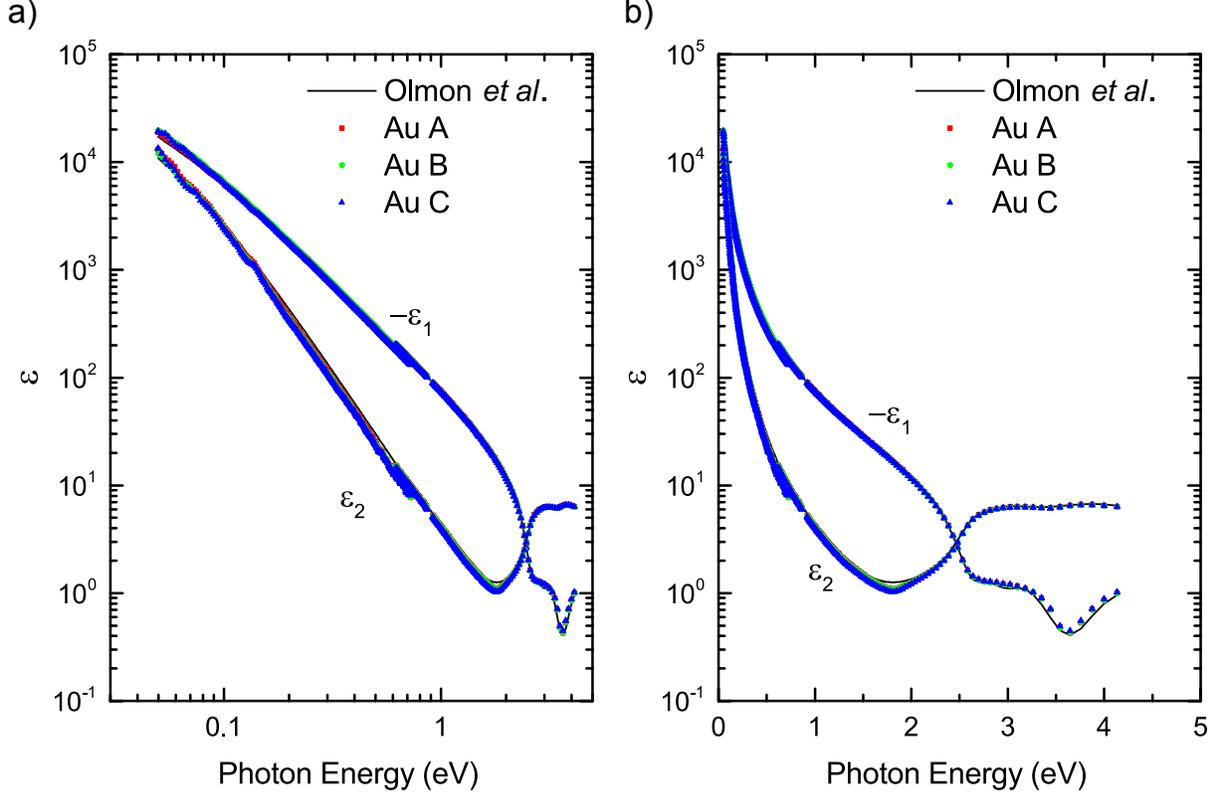


FIG. S2. Dielectric function of three TS gold from direct inverted ellipsometry measurement (red, green, and blue), in comparison with the fitted dielectric function presented in our previous paper (black) [1]. Plots with phonon energy in linear (a) and in logarithmic scale (b).

### III. INSTRUMENT ERROR VS SAMPLE DIFFERENCE

As shown in Fig. S1 and S2, in the overlapped spectral range of the two instruments of  $0.62 - 0.73$  eV ( $1.7 - 2 \mu\text{m}$ ), the measured dielectric functions between the two instruments have an offset. This offset is fundamentally limited by the instruments, and is likely due to different light sources, difference in illumination spot sizes, and signal detection methods. The offset imposes a systematic error on our data. As the difference between the dielectric function of the three samples is less than this offset, we can conclude our data is dominated by the instrumental error.

#### IV. SAMPLE DEGRADING OVER TIME

Comparing the measurements to gold, we see larger variations from sample to sample in silver. Existence of large variations even among nominally identical samples indicates silver is very sensitive to environmental conditions, such as surface oxidation, impurity, and crystallite size. These variations also affect the electron relaxation time  $\tau$ .

To test the influence of surface oxidation, we measured the three samples in a day later for comparison. Fig. S3 shows the comparison of dielectric function for fresh samples (line) and the same samples one day later (dot).  $\epsilon_1$  of three samples only vary slightly. However,  $\epsilon_2$  of samples measured one day later are consistently smaller than fresh samples, indicating sample degrading. As the relaxation time  $\tau$  can serve as a characteristic parameter for sample quality, we apply extended Drude model analysis to extract  $\tau(\omega)$ . The resulting  $\tau(\omega)$  is shown in Fig. S4, with decreased relaxation time  $\tau$  after one day for the same samples. We also measured the same samples after 3 days and 7 days, but these results do not have a consistent trend for  $\tau$ . This could be due to oxidation/contamination of the samples in an uncontrolled fashion over time when stored in ambient conditions.

#### V. TABLE OF DIELECTRIC FUNCTION: SILVER AND GOLD

Measured Dielectric functions of silver and gold shown in Fig. S1 and S2 are given in the online material in tabular form (in .pdf and .csv format), which are post online separately.

- Ag\_A.pdf, Ag\_A.csv
- Ag\_B.pdf, Ag\_B.csv
- Ag\_C.pdf, Ag\_C.csv
- Ag\_A\_corrected.pdf, Ag\_A\_corrected.csv
- Ag\_B\_corrected.pdf, Ag\_B\_corrected.csv
- Ag\_C\_corrected.pdf, Ag\_C\_corrected.csv
- Au\_A.pdf, Au\_A.csv
- Au\_B.pdf, Au\_B.csv

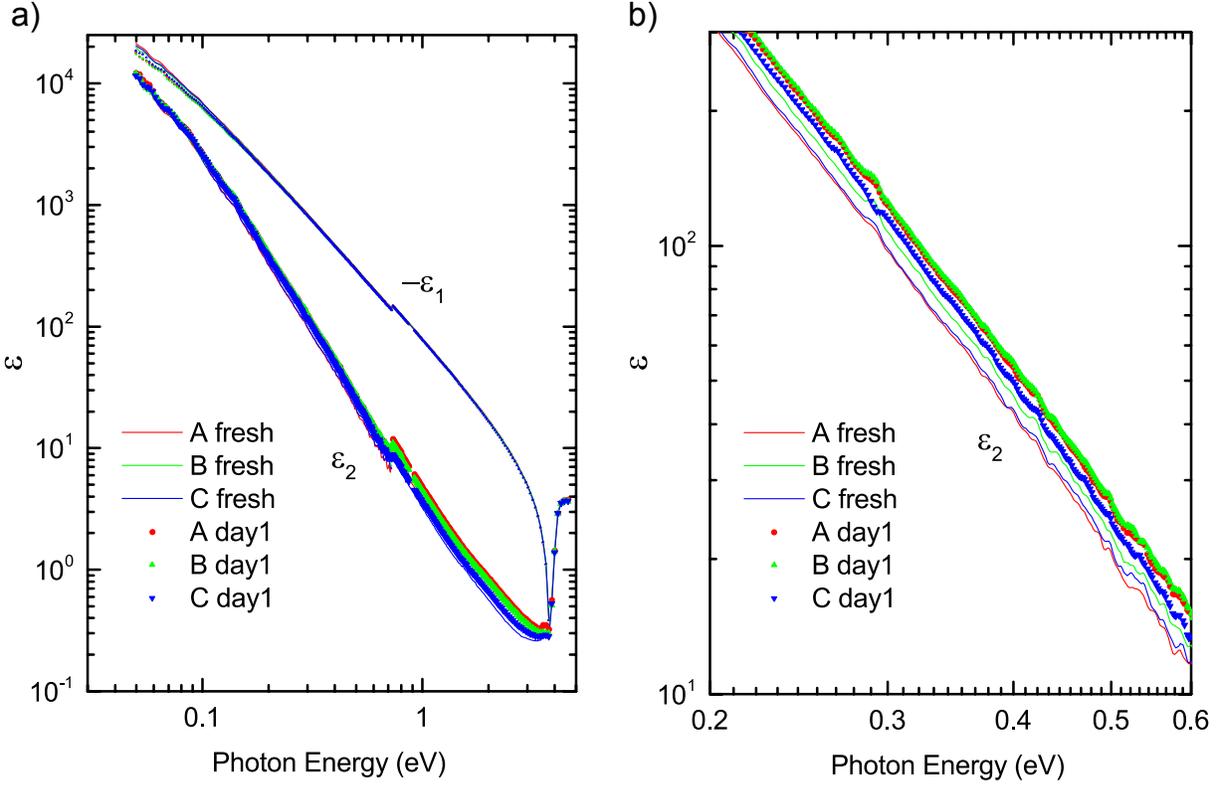


FIG. S3. (a)  $-\epsilon_1$  and  $\epsilon_2$  for fresh silver samples (line) and one day later (dot) of sample A – C in red, green, and blue. (b) Zoom-in of  $\epsilon_2$  in (a) to show the increase in  $\epsilon_2$  of samples measured one day later compared to fresh samples.  $\epsilon_2$  of samples measured one day later are consistently larger than fresh samples, indicating sample degrading.  $\epsilon_1$  only vary slightly.

• Au\_C.pdf, Au\_C.csv

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- [2] A. D. Rakic, A. B. Djuricic, J. M. Elazar, and M. L. Majewski, *Applied optics* **37**, 5271 (1998).
- [3] A. O. Melikyan and B. V. Kryzhanovsky, *Optical Memory and Neural Networks* **23**, 1 (2014).

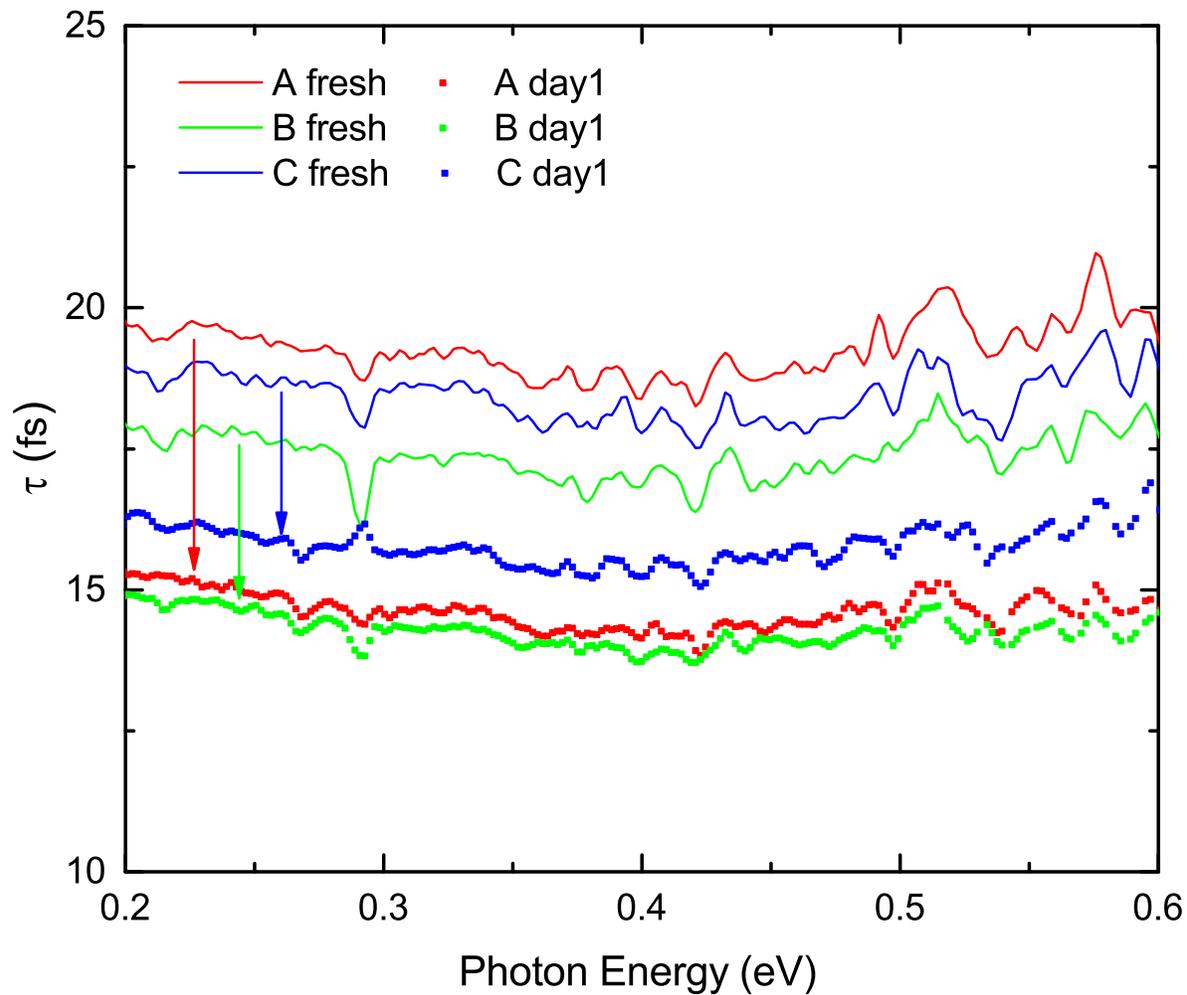


FIG. S4. Extracted relaxation time  $\tau$  from extended Drude analysis. Fresh samples have a larger  $\tau(\omega)$  compared to samples measured one day later, indicating sample degrading over time.