**Electronic Supplementary Information (ESI)**

# Electrodeposition of Nanometer-Thick Epitaxial Films of Silver onto Single-Crystal Silicon Wafers

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**Laue oscillation calculations for Ag on Si(111)**

The film thickness can be determined by the satellite peak positions from the Laue oscillation using the equation below:

$$t = \frac{(L_2 - L_1)\lambda}{2(\sin\theta_1 - \sin\theta_2)} \qquad (1)$$

where $t$ is the film thickness, $L$ is the satellite peak order, $\lambda$ is the X-ray wavelength, and $\theta$ is the satellite angle [1].

**Twinning relationship analysis for Ag on Si(110)**

The (411) twin observed in the (110) silver on silicon could be described either as a 180° rotational twin along the [111] direction or a reflection twin. This could be written as the transformation: [1]

$$(PQR) = T_{(111)}(pqr) \qquad (2)$$

where ($PQR$) is a column matrix for the lattice plane after the twinning, described by the twinning matrix $T_{(111)}$ which happens on the (111) slip plane. ($pqr$) is the original crystalline plan. For face-centered cubic (fcc) system such as Ag in this case, the twin matrix can be expressed as

$$T_{(111)} = \frac{1}{3}\begin{pmatrix} -1 & 2 & 2 \\ 2 & -1 & 2 \\ 2 & 2 & -1 \end{pmatrix} \qquad (3)$$

Thus, by applying this matrix on (110) plane using equation (2), the twinned plan is (114).

**Supplementary Figures**

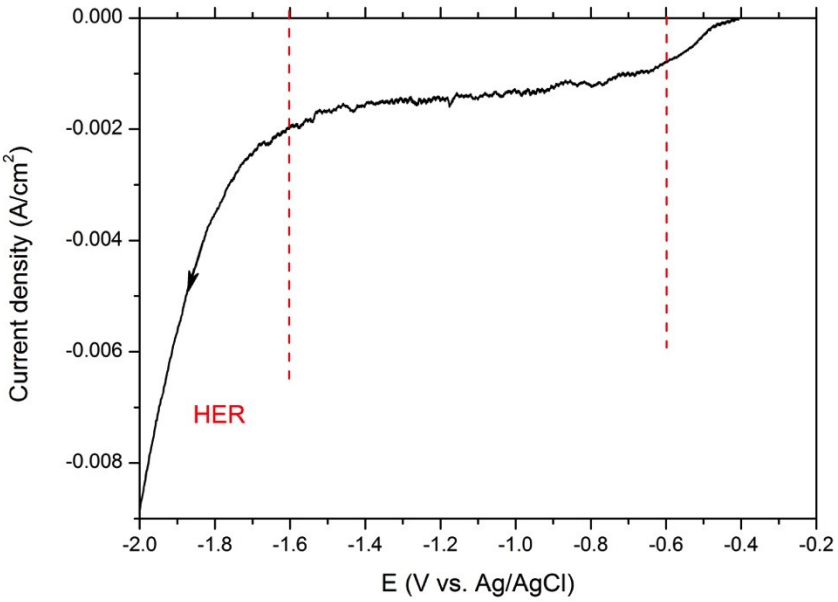


**Fig S1.** LSV curve of the Au electrode in the stirred cyanide bath showing the Ag deposition range and the hydrogen evolution range. The arrow in the plot indicates the scan direction. The scan rate was 10 mV $s^{-1}$.

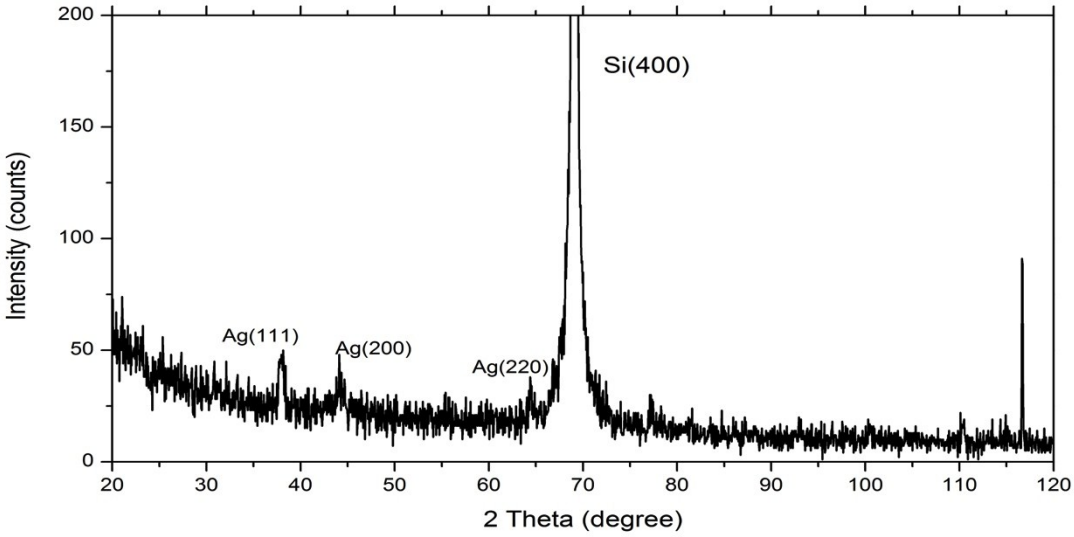

**Fig S2.** The 2 theta scan of the Ag film deposited for 1 min on Si(100) in the cyanide bath; showing Ag (111), (200) and (220) crystalline planes.

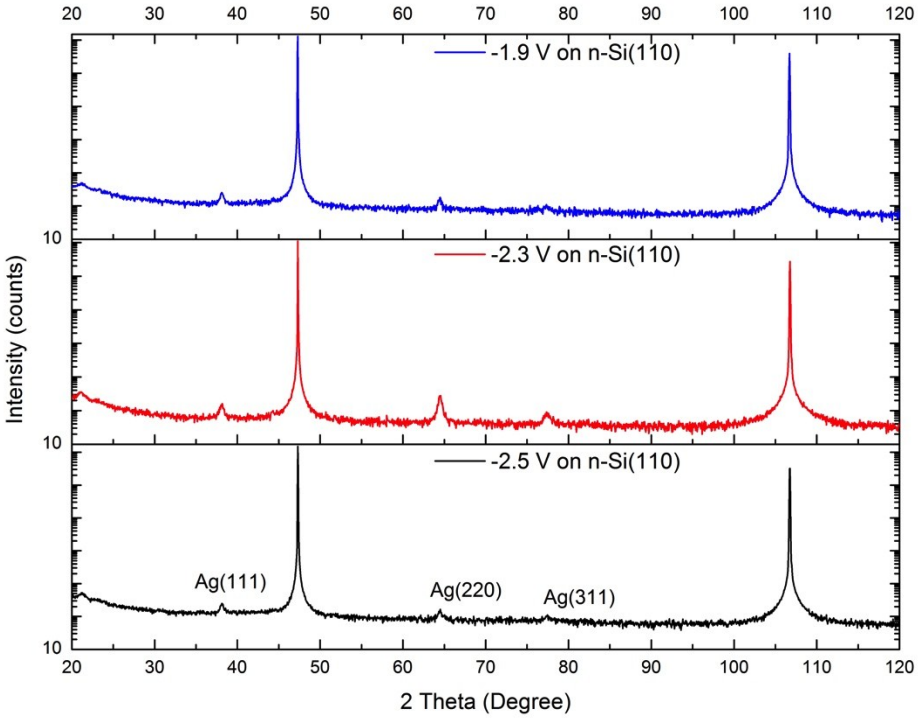


**Fig S3.** The 2 theta scan of the Ag film deposited for 1 min on Si(110) in the cyanide bath; showing Ag (111), (200) and (220) crystalline planes. Note that three different potentials were applied when depositing and that all three films are disoriented.

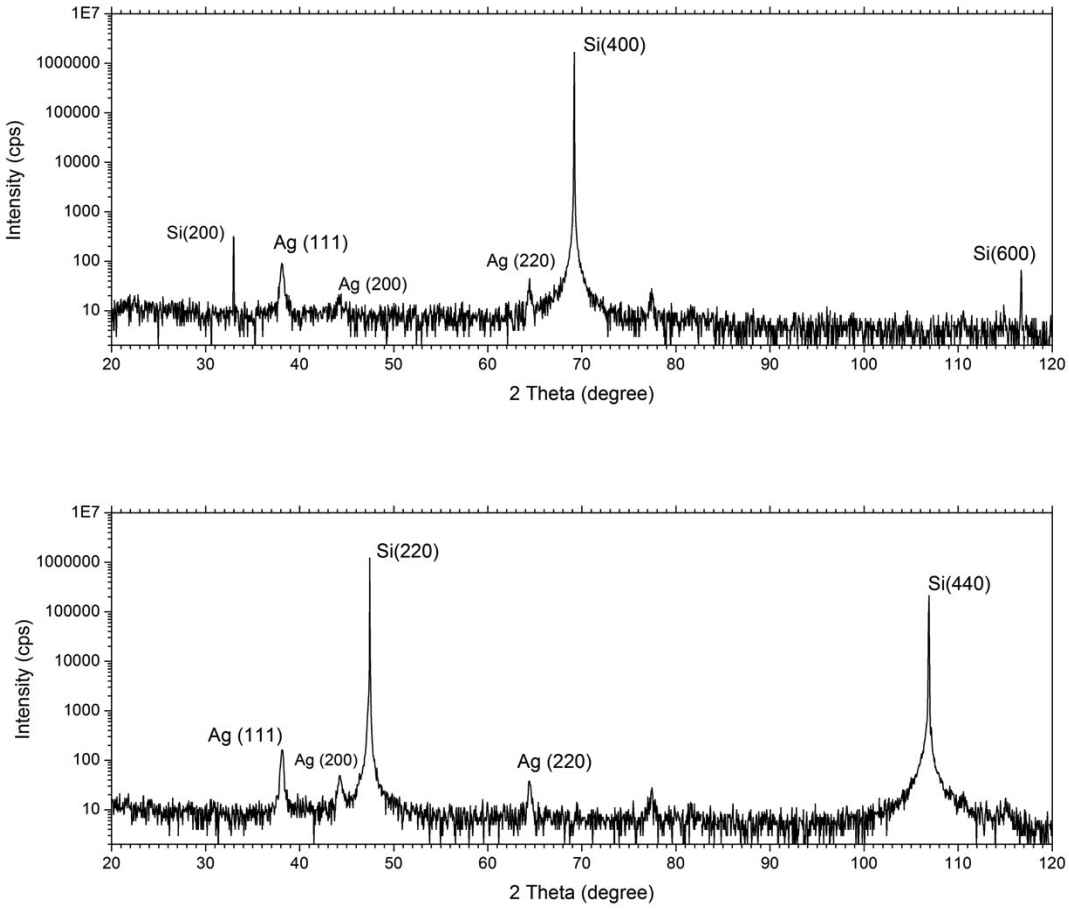


**Fig S4.** The 2 theta scan of the Ag film deposited for 3 min at -1.9 V vs. Ag/AgCl on Si(100) and Si(110) in the cyanide bath; showing increased intensity of Ag (111) peaks in both cases.

## References

1 J. A. Switzer, J. C. Hill, N. K. Mahenderkar and Y. C. Liu, *ACS Appl. Mater. Interfaces*, 2016, **8**, 15828–15837