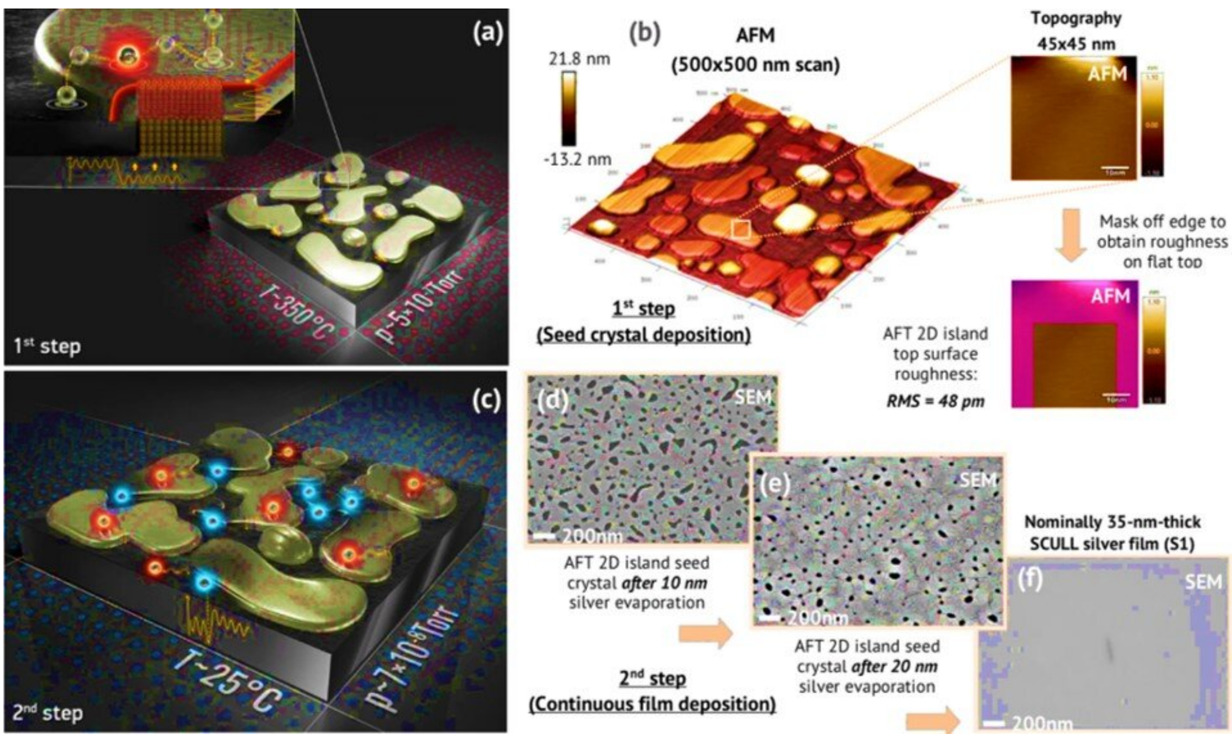
Quantum engineering atomically smooth single-crystalline silver films

August 30 2019, by Thamarasee Jeewandara



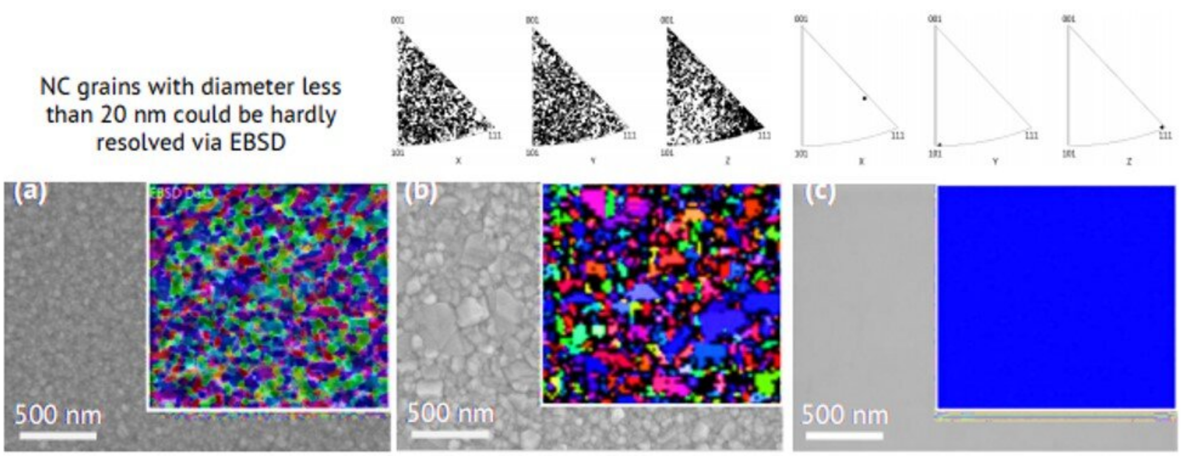
The SCULL (Single-crystalline Continuous Ultra-Smooth Low-loss Low-cost) process: Two-step deposition of single-crystalline silver films. (a) In the first step, an AFT 2D. Ag (111) seed crystal is deposited under 350 °C temperature. (b) Atomic force microscopy (AFM) scan of AFT 2D Ag (111) islands (silver islands) deposited on a Si (111) substrate. Most of the AFT 2D Ag(111) islands have atomically flat top surface with an RMS (root mean square) roughness less than 50 pm. In the second step, the process is stopped, and the substrate is cooled down to 25 °C followed by additional silver evaporation until continuous silver film is formed. (c) SEM images illustrate film morphology evolution during the second step after nominally 10 nm (d) and 20 nm (e) silver evaporation on an

AFT 2D seed crystal at 25 °C. (f) Scanning electron microscopy (SEM) image of nominally 35-nm-thick single-crystalline film. The defect on the film surface is purposefully created (by electron beam burning) to facilitate focusing on the atomically smooth surface. Credit: Scientific Reports, doi: 10.1038/s41598-019-48508-3

Ultra-low-loss metal films with high-quality single crystals are in demand as the perfect surface for nanophotonics and quantum information processing applications. Silver is by far the most preferred material due to low-loss at optical and near infrared (near-IR) frequencies. In a recent study now published on *Scientific Reports*, Ilya A. Rodionov and an interdisciplinary research team in Germany and Russia reported a two-step approach for electronic beam evaporation of atomically smooth single crystalline metal films. They proposed a method to establish thermodynamic control of the film growth kinetics at the atomic level in order to deposit state-of-the-art metal films.

The researchers deposited 35 to 100 nm thick, single-crystalline silver films with sub-100 picometer (pm) surface roughness with theoretically limited optical losses to form ultrahigh-Q nanophotonic devices. They experimentally estimated the contribution of material purity, material grain boundaries, surface roughness and crystallinity to the optical properties of metal films. The team demonstrated a fundamental two-step approach for single-crystalline growth of silver, gold and aluminum films to open new possibilities in nanophotonics, biotechnology and superconductive quantum technologies. The research team intends to adopt the method to synthesize other extremely low-loss single-crystalline metal films.

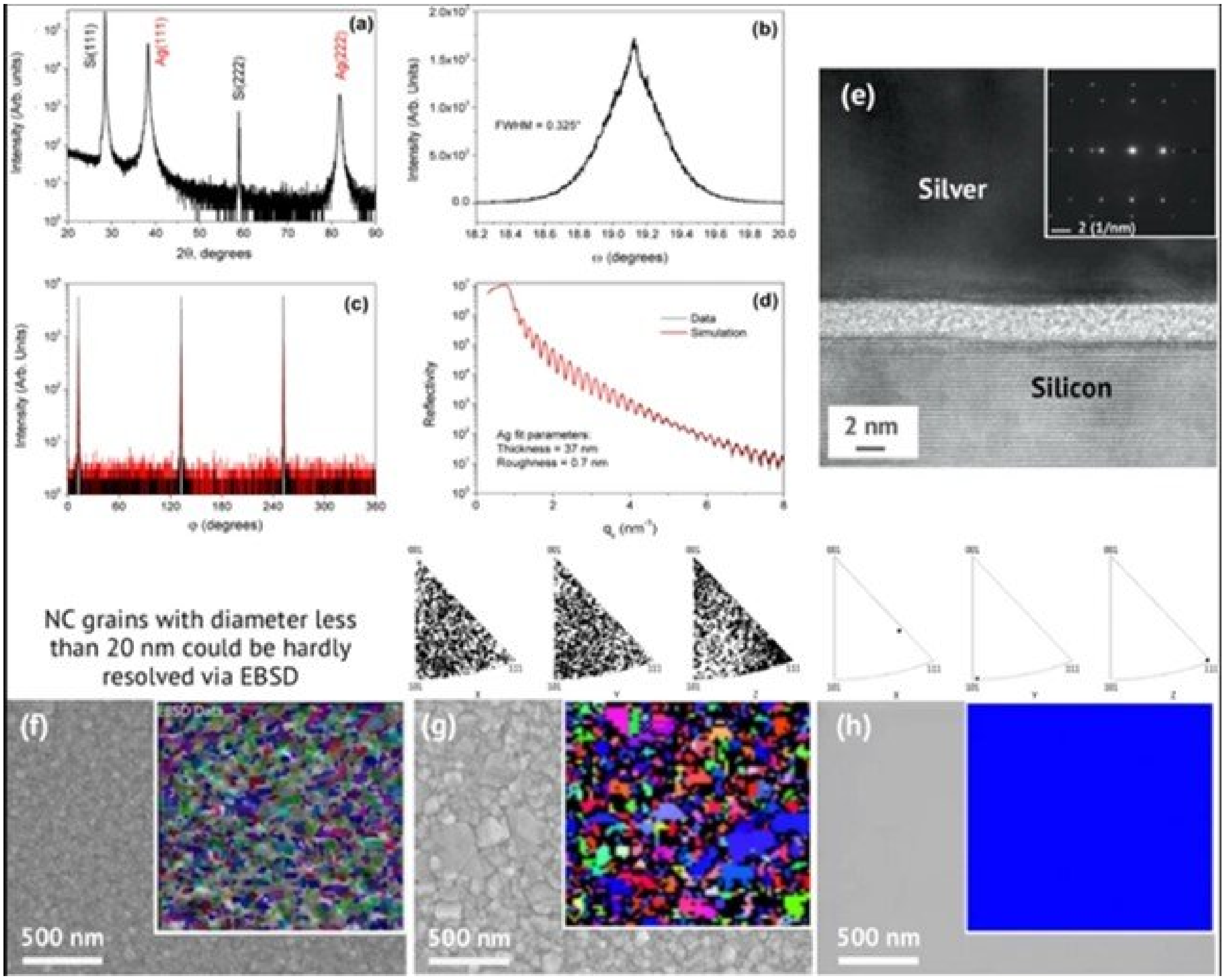
Optoelectronic devices with plasmonic effects for near-field manipulation, amplification and sub-wavelength integration can open new frontiers in nanophotonics, quantum optics and in quantum information. Yet, the ohmic losses associated in metals are a considerable challenge to develop a variety of useful plasmonic devices. Materials scientists have devoted research efforts to clarify the influence of metal film properties to develop high performance material platforms. Single-crystalline platforms and nanoscale structural alterations can prevent this problem by eliminating material-induced scattering losses. While silver is one of the best known plasmonic metals at optical and near-IR frequencies, the metal can be challenging for single-crystalline film growth.



Scanning electron microscopy (SEM) images with electron backscatter diffraction (EBSD) insets. Nanocrystalline (NC) (a), Polycrystalline (PC) (b) and single-crystalline (S1) (c) silver films highlighting film grains. EBSD inverse pole figures are shown above the SEM images, to demonstrate very tight crystal orientation density of the S1 film (c) along all the normal directions. Only a single domain is observed in the S1 film, confirming the high quality and single-crystalline nature without grain boundaries over a large length scale. Credit: Scientific Reports, doi: 10.1038/s41598-019-48508-3

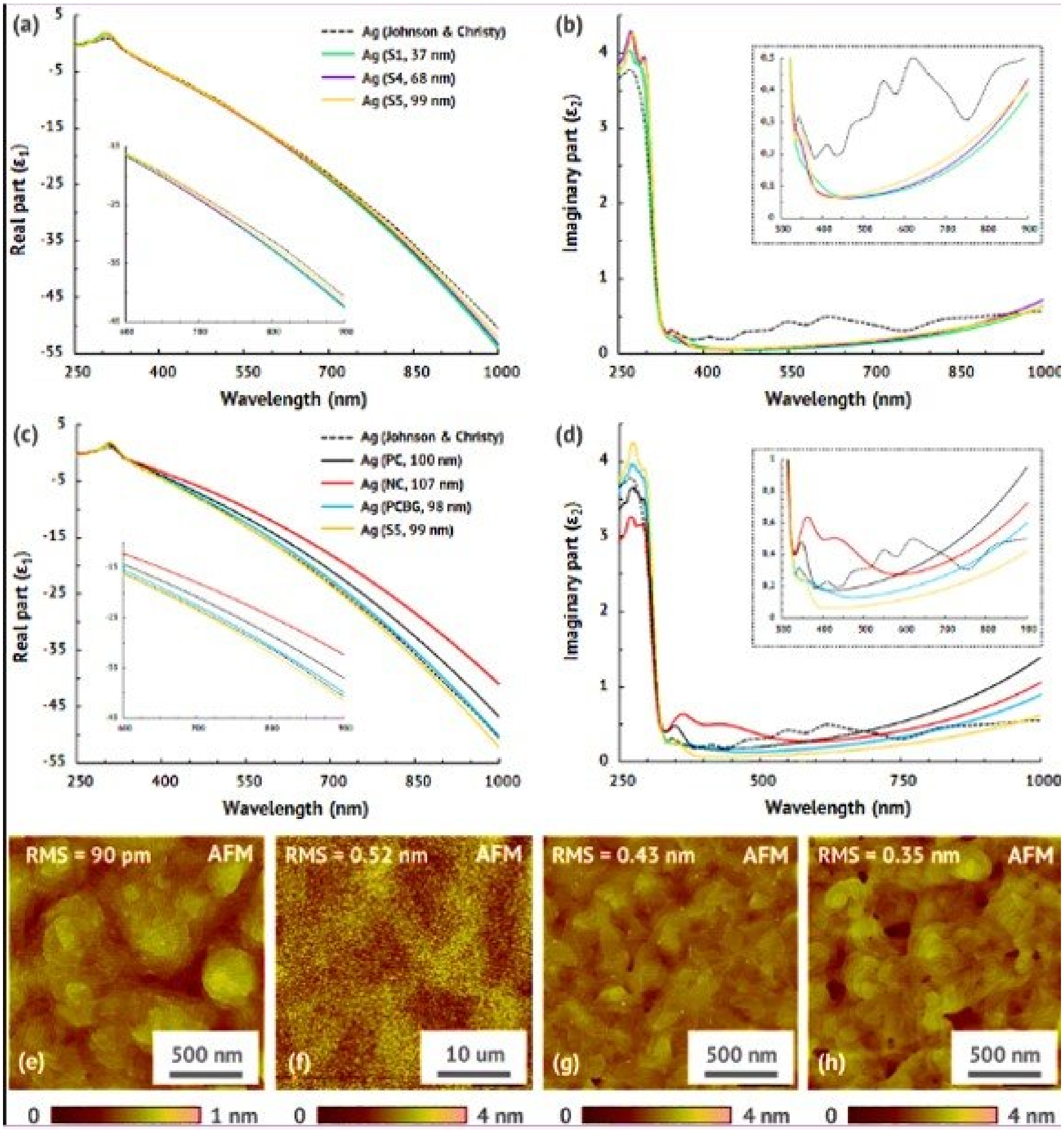
Previous reports on single-crystalline silver film growth methods relied on molecular beam epitaxy (MBE) or physical vapor deposition (PVD) with atomic smoothness and significantly lower optical losses. In the present study, Rodionov et al. used a two-step PVD growth approach previously developed by the same research team to obtain atomically smooth single-crystalline metal films using a high-vacuum electron-beam evaporator. The method facilitated high crystallinity and purity across an atomically smooth surface with unique optical properties and thermodynamic stability. The process is flexible, inexpensive and fast with a high deposition rate compared to the MBE technique. The team can replicate the method with a variety of metals including silver, gold and aluminum—widely used in quantum optics and quantum information.

During the two-step deposition process for materials development, Rodionov et al. first grew a seed crystal containing strained two-dimensional silver islands (atomic features) with atomically flat top surfaces (AFT 2-D islands) on a substrate at 350 degrees C. According to the electronic growth model, silver islands are an electron gas confined to a 2-D quantum well (energy barriers that confine an electron). Then, the researchers cooled the substrate to 25 degrees C in the same vacuum cycle to prevent a dewetting effect. They evaporated the silver on the AFT 2-D seed to form a continuous single-crystal film to completion. Subsequently they annealed the silver film at higher temperatures (320-480 degrees C), which improved the crystalline structure and surface roughness of the resulting film. The scientists named their deposition process the SCULL—for "single-crystalline Continuous Ultra-Smooth Low-loss Low-cost"—thin-film production.



Microstructure characterization of a 37-nm-thick Si (111)/Ag (111) film (S1) and SEM images with EBSD insets of (NC), (PC) and (S1) films. (a) XRD (θ–2θ) pattern indicating only Ag (111) and Si (111) substrate peaks. (b) Measured transverse scan (rocking curve, ω-scan) through the Ag (111) diffraction peak. (c) Grazing incidence of the in-plane X-ray diffraction scan (phi-scans) of the Ag(111) plane. (d) X-ray reflectivity curve. (e) HRTEM image and the electron diffraction pattern (inset in the right corner), the growth direction is bottom-up. SEM images with EBSD insets of NC (f), PC (g) and S1 (h) silver films highlighting film grains. EBSD inverse pole figures are shown above the SEM images, demonstrating very tight crystal orientation density of the S1 film (h) along all the normal directions. Only a single domain is observed for S1 film in both small-scale 2 µm (h). Credit: Scientific Reports, doi: 10.1038/s41598-019-48508-3

The research team developed materials using SCULL and compared the results for six representative films, which included three SCULL single crystalline films of varying thickness (35 nm, 70 nm and 100 nm) and three 100 nm thick polycrystalline films. The scientists used high-resolution wide-angle X-ray diffraction (XRD) to view the high quality of films with minimal levels of defect. Then using high-resolution transmission electron microscopy (HRTEM), the research team demonstrated the single-crystalline nature of the silver film. They used electron backscatter diffraction (EBSD) to analyze the domain structures and extract the average grain size of the single-crystalline and polycrystalline films.



Optical properties and surface characterization. Real (a) and imaginary (b) part of the dielectric permittivity of the single-crystalline films (S1, S4, S5). Dielectric permittivity (c,d) of nominally 100-nm-thick single-crystalline (S5) and polycrystalline (PC, NC, PCBG) films. AFM scans of S1 (e), S4 (g) and M1 (h) films measured over a 2.5 × 2.5 μm2 area, and S1 (f) film, measured over a 50 × 50 μm2 area. All the films surfaces are continuous without pinholes and we observe no grain boundaries for single-crystalline films (e–h). The S1 film is extremely smooth with an atomic level of root mean square (RMS) roughness equal to 90 pm (e), which is the smoothest reported single-crystalline silver film. The RMS roughness of thicker films S4 and M1 are slightly larger, but still extremely smooth of 0.43 nm (c) and 0.35 nm (d). Credit: Scientific Reports, doi: 10.1038/s41598-019-48508-3

Rodionov et al. characterized the optical properties and surface topography of the single-crystalline films using atomic force microscopy. They then extensively demonstrated material purity and surface roughness to indicate a much purer silver film in the study. The SCULL silver films introduced in the work will have potential applications in the evolving field of quantum plasmonics and atomically smooth single-crystalline films that require low optical absorption and high conductivity. Rodionov et al. observed a theoretically predicted surface plasmon polariton propagation length for silver and exceptional performance of experimental plasmonic devices with the SCULL silver films.

In this way, Ilya A. Rodionov and co-workers developed a two-step approach for electronic beam evaporation to form continuous atomically smooth, single-crystalline metal films across a wider range of thickness from 35-100 nm. The researchers envision their proposed SCULL process will be used to deposit a variety of atomically smooth single crystalline thin films using an easy, top-down fabrication device in the future. The unique physical and optical properties of the resulting SCULL films can open new possibilities in diverse fields of technology.

More information: Ilya A. Rodionov et al. Quantum Engineering of Atomically Smooth Single-Crystalline Silver Films, *Scientific Reports* (2019). DOI: 10.1038/s41598-019-48508-3

P. Melentiev et al. Plasmonic nanolaser for intracavity spectroscopy and sensorics, *Applied Physics Letters* (2017). DOI: 10.1063/1.5003655

Harry A. Atwater et al. Plasmonics for improved photovoltaic devices, *Nature Materials* (2010). DOI: 10.1038/nmat2629