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Supporting Information

Mussel-inspired Polydopamine Coatings for Large-scale and Angle- independent Structural Colors

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Experimental

Materials: Silicon wafer was cleaned by piranha solution (98% H_2SO_4/30% H_2O_2, v/v = 7:3) for 30 min, rinsed by deionized water, and blew dried by nitrogen gas before use. Other substrates such as thin foil, aluminum sheet, coin and stainless steel were washed by acetone overnight to remove impurities and then blew dried by nitrogen gas before use. Dopamine hydrochloride and N-isopropylacrylamide were purchased from Sigma-Aldrich. Other reagents such as tris(hydroxymethyl) aminomenthane, copper(II) sulfate pentahydrate, Hydrogen peroxide, ethanol, hydrochloric acid, acetone, ethanol, heptane and tetrahydrofuran were purchased from Sinopharm Chemical Reagent Co., Ltd and used without further purification. Water used in all experiments was deionized and filtrated to18.2 MΩ with an ELGA LabWater system (France).

Fabraication of PDA coatings on various substrates triggered by $CuSO_4$/H_2O_2: Dopamine hydrochloride (2 mg/mL) was dissolved in Tris buffer solution (pH = 8.5, 50 mM), and $CuSO_4$ (5 mM) and H_2O_2 (19.6 mM) were immediately added into above soutions. Typically, silicon wafer was prewetted by ethanol and immersed in the above deposition solutions for 40 min. Then, the samples were washed by water and instantly blew dried by nitrogen gas. Subsequently, deposited silicon wafer immersed in fresh as-prepared deposition

solutions and repeated aforementioned process for certain times. The other substrates were dealt with the same method.

Deposition of PDA on silicon wafer with different metal salts: Dopamine hydrochloride (2 mg/mL) was dissolved in Tris buffer solution (pH = 8.5, 50 mM) with different metal salts. Typically, silicon wafer was prewetted by ethanol and immersed in the above deposition solutions for 40 min. Then, the samples were washed by water and instantly blew dried by nitrogen gas. Subsequently, deposited silicon wafer immersed in fresh as-prepared deposition solutions and repeated aforementioned process for certain times.

Synthesis of PNIPAM: *N*-isopropylacrylamide (0.5 g, 4.42 mmol) and 10 mL tetrahydrofuran were added into a 50 mL round-bottomed flask with a magnetic stirrer. The flask was sealed and subjected to three freeze-pump-thaw cycles in order to remove dissolved oxygen. Then, 2,2'-azobisisobutyronitrile (10 mg, 0.06 mmol) was added under the atmosphere of nitrogen. After that the flask was immersed in an oil bath at 60 °C. The polymerization was stopped after 24 h. The mixture was then precipitated in hexane. The final pure product was dried under reduced pressure overnight.

Co-deposition of PDA and PNIPAM on silicon wafer triggered by $CuSO_4/H_2O_2$: Dopamine hydrochloride (2 mg/mL) and PNIPAM (10 mg/mL) were dissolved in Tris buffer solution (pH = 8.5, 50 mM), and $CuSO_4$ (5 mM) and H_2O_2 (19.6 mM) were immediately added into above soutions. Typically, silicon wafer was prewetted by ethanol and immersed in the deposition solutions for 40 min. Then, the samples were washed by water and instantly blew dried by nitrogen gas. Subsequently, deposited silicon wafer immersed in fresh as-prepared deposition solutions and repeated aforementioned process for certain times.

Characterization: Morphology of the samples was characterized by field emission scanning electron microscope (Hitachi S4800, Japan). X-ray photoelectron spectra were collected by a spectrometer (Perkin Elmer, USA) with Al Kα excitation radiation (1486.6 eV). The thickness and refractive index were detected by spectroscopic ellipsometer (Semilab Sopra, GSE-5E) at

incident angel of 70°, the light spot size is 360 × 360 μm^2. The surface morphologies and roughness of PDA-coated silicon wafer were measured by scanning probe microscopy (VEECO, Multimodel). The colors of different samples were observed by optical microscope with a reflected light imageing system (ZEISS, Germany). The order of samples was conducted by SAXS (Bruker-AXS, Germany) with a angle range from 0.044° to 4.4°. Reflectance meter (Olympus USPM-Ru, Perkin-Elmer, USA) was used to detect the reflectance spectra of different samples under different view angles.

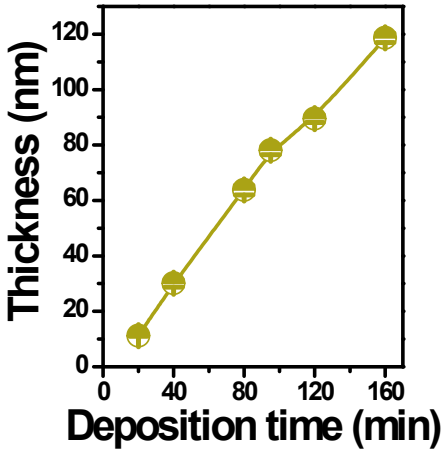

Figure S1. Time-dependence of thickness for the $CuSO_4/H_2O_2$-triggered PDA coatings deposited on silicon wafers, as determined by ellipsometry.

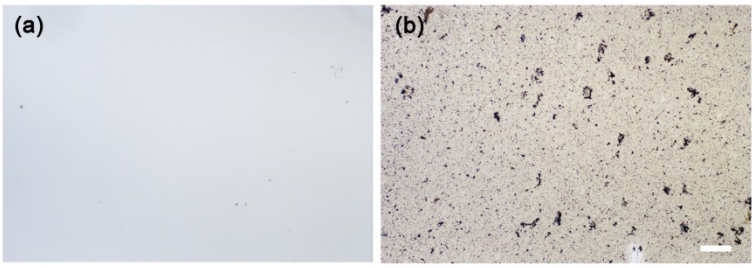

Figure S2. Optical micrographs of (a) nascent silicon wafers and (b) PDA coatings deposited on silicon wafers oxidized by air. The scale bar and exposure time is 100 μm and 80 ms, respectively. The thickness of PDA coatings is 70 nm.

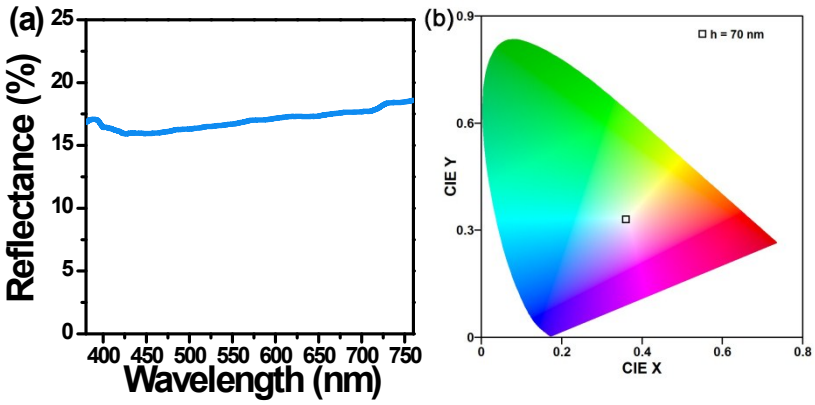

Figure S3. (a) Reflectance spectrum and (b) according CIE 1931 chromaticity coordinates of PDA coatings on silicon wafers oxidized by air. The scale bar is 100 μm.

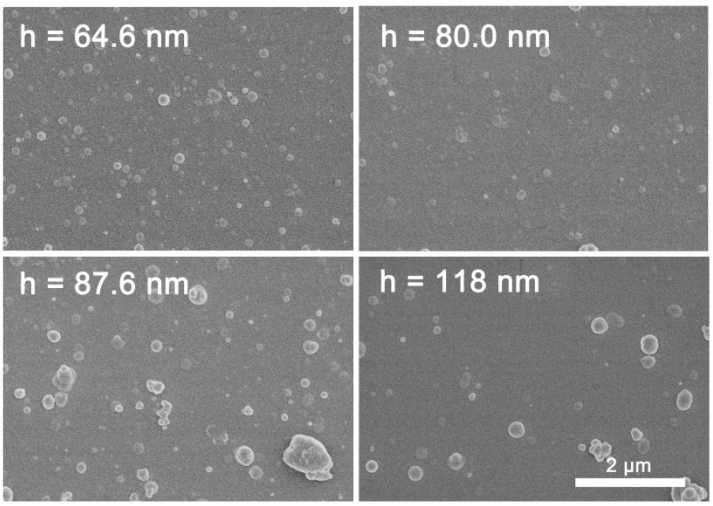

Figure S4. SEM images of PDA coatings deposited on silicon wafers with different thicknesses using $CuSO_4/H_2O_2$ as a trigger.

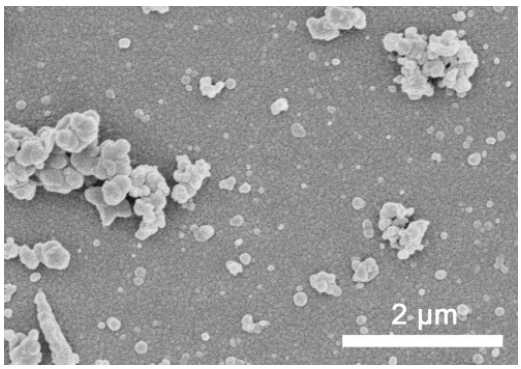

Figure S5. SEM image of PDA coatings on silicon wafers oxidized by air. The scale bar is 2 μm.

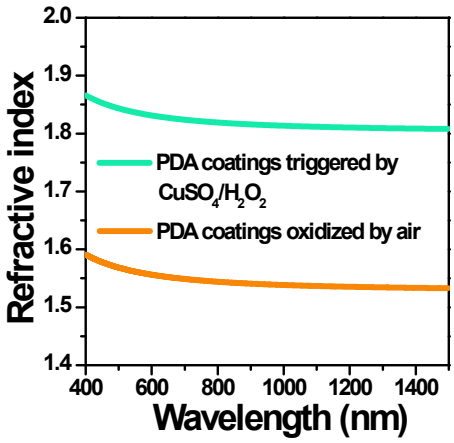

Figure S6. Refractive index of PDA coatings deposited on silicon wafers with different oxidants, as determined by ellipsometry.

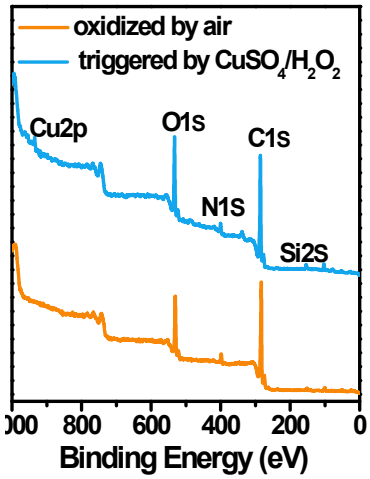

Figure S7. XPS spectra of PDA coatings with different oxidants.

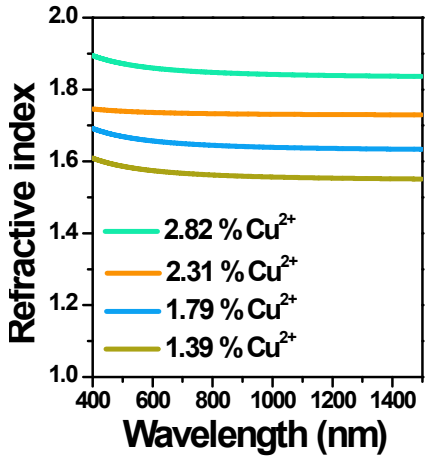

Figure S8. Refractive index of PDA coatings with different contents of copper ions, as determined by ellipsometry.

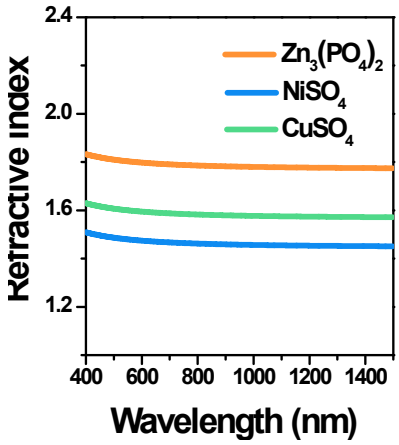

Figure S9. Refractive index of PDA coatings oxidized by different metal ions, as determined by ellipsometry.

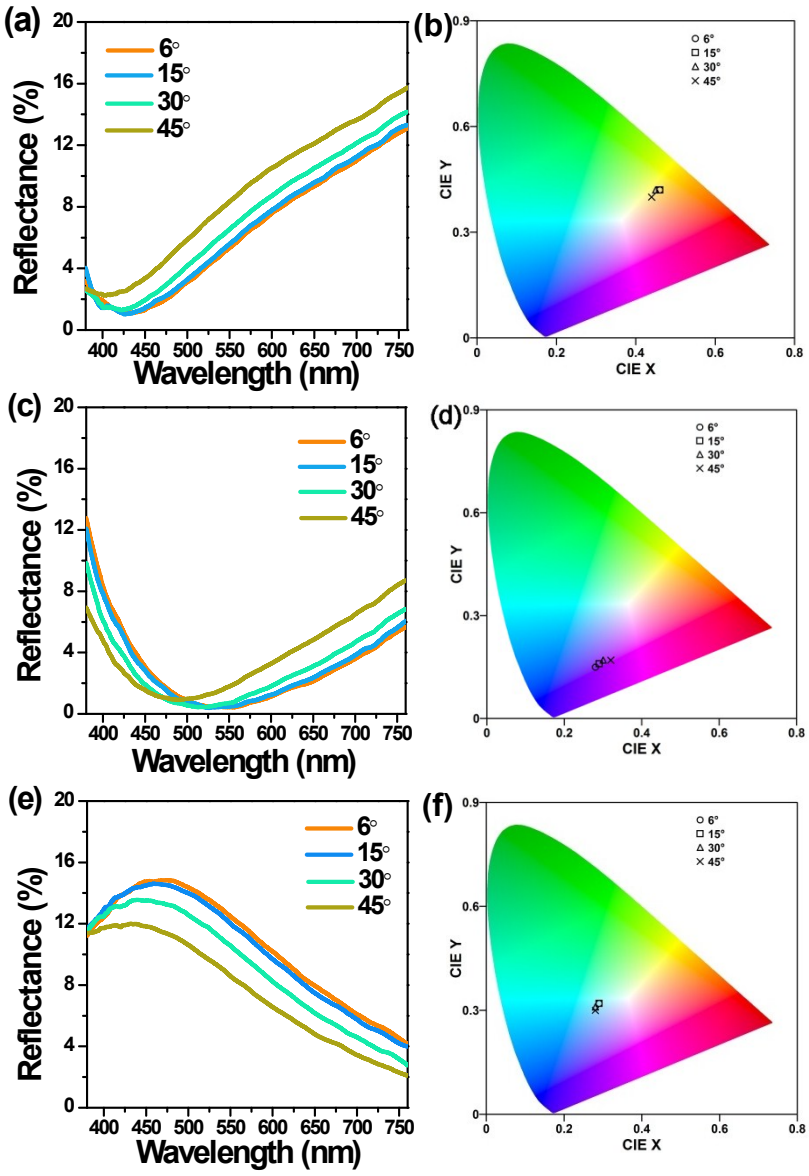

Figure S10. Measured angle-resolved reflectance spectra and according CIE 1931 chromaticity coordinates of PDA coatings on silicon wafers with different thicknesses: (a, b) 64.6 nm, (c, d) 80.0 nm and (e, f) 118.0 nm.

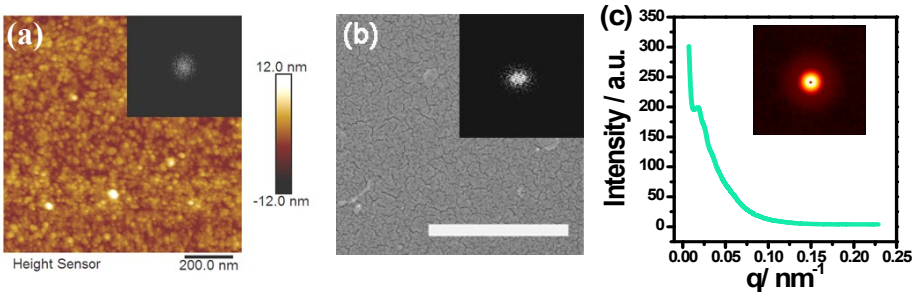

Figure S11. AFM image (a), FESEM image (b) and SAXS spectrum (c) of PDA coatings. The inset in (a), (b) and (c) is 2D-FFT spectrum from images and SAXS pattern, respectively. The scare bar is 1 μm.

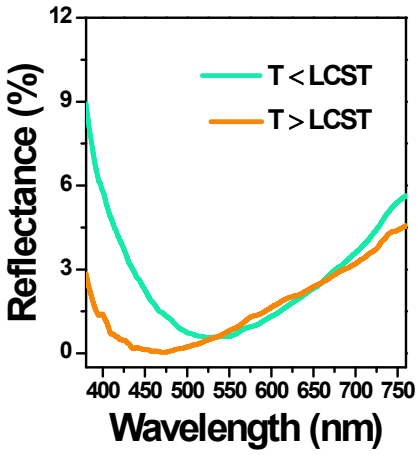

Figure S12. Reflectance spectra of PDA/PNIPAM coatings on silicon wafers exposed at different temperatures.

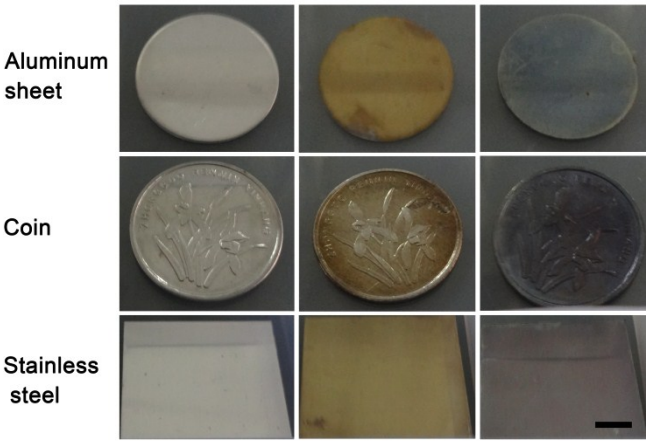

Figure S13. Digital images of PDA coatings on different substrates. The scale bar is 1 cm.