Supporting Information

Multi-mode structural-color anti-counterfeiting label based on physically unclonable amorphous photonic structures with convenient artificial intelligence authentication

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Experimental Section

Fabrication of the structural color anti-counterfeirting label with APS. The mono-disperse latex spheres with diameters of 150 nm, 180 nm, 205 nm, 225 nm and 254 nm were synthesized by the facile emulsion polymerization approach.[1] The silica micro-spheres with sizes of 5 – 10 μm were purchased from Fuhong nanomaterials corporation. The substrates treated with water-proof black ink or black substrates were directly used. The black substrates were then coated with dispersions of silica nanoparticles (average diameter, ~ 30 nm) and acrylic modified polyurethane resin in water (silica content, 10 wt%, acrylic modified polyurethane/silica, 0.8:1, wt/wt) by a drop coating method and was dried at ambient conditions, forming the intermediate layer with water-induced transparency. Next, aqueous latex dispersions (latex content, ~ 10 wt%, without or with optimized 25 wt% silica microspheres, silica/latex) with varying particle diameters was spray-coated onto the dry intermediate layer, producing the encrypted amorphous photonic structures (APS) anti-counterfeiting label.

Optical and Structural Characterization. The photos and videos of the anti-counterfeiting label were taken by a digital camera (Canon EOS 80D) under diffusive light illumination. The optically microscopic images of the samples doped with silica microsphere were taken by a motorized optical microscope **(**Leica DM6M). The structure**s** of the intermediate and APS patterned layers were characterized by field emission SEM (Hitachi S-4800). The time- or angle-resolved reflectance spectra of the APS patterned layer and the time-resolved transmission spectra of the intermediate layer of the anti-counterfeiting label were measured by UV-vis optic fiber spectrometer (Ocean Optics Inc., HR 4000) via a 600 μm broadband optical fiber (Ocean Optics Inc., QP600-1-UV/vis). Xenon lamp was used as light source with wavelengths from 250 to 800 nm (DH2000-BAL, Ocean Optics Inc.). A standard diffusive white board (ws-1, Ocean Optics Inc.) was used as the reference.

Deep learning and validation methodology. The deep learning and validation was carried out according to the literature.[2] To generate a large enough training set from theoretical images for 18 layer ResNet,[3] a data augmentation procedure to the original synthetic images is applied. The deep learning networks used here were based on PyTorch.[4] The input images were resized to 256 × 256 using pixel area relation for training. The learning process takes ~ 2 h. For validation, the consumers take a picture of a security label by a portable microscope and sent it to the AI. AI can then automatically recall the accurately corresponding relationship and output the indexing name with a detailed match score. Based on the scores, we can easily differentiate the fake labels from the genuine ones.

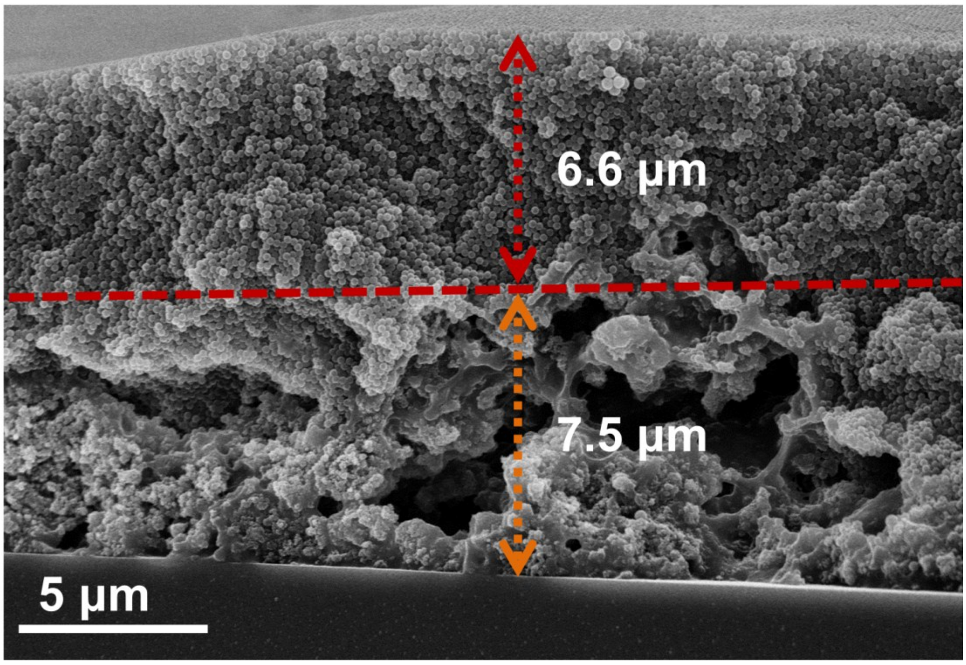

Figure S1. Cross-sectional SEM image of the anti-counterfeiting label, which consisted of the APS layers and the intermediate layers.

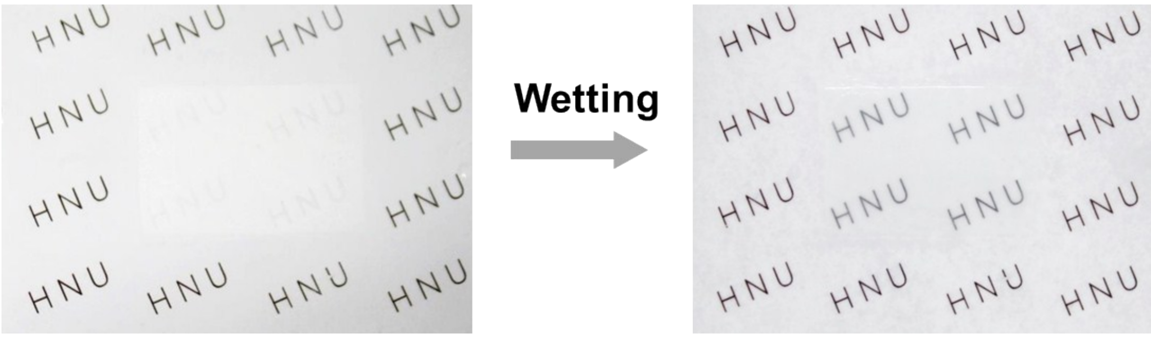

Figure S2. Photographs of the intermediate layer at the dry and wet conditions. Wetting the layer with water led to its transitions from white to transparency.

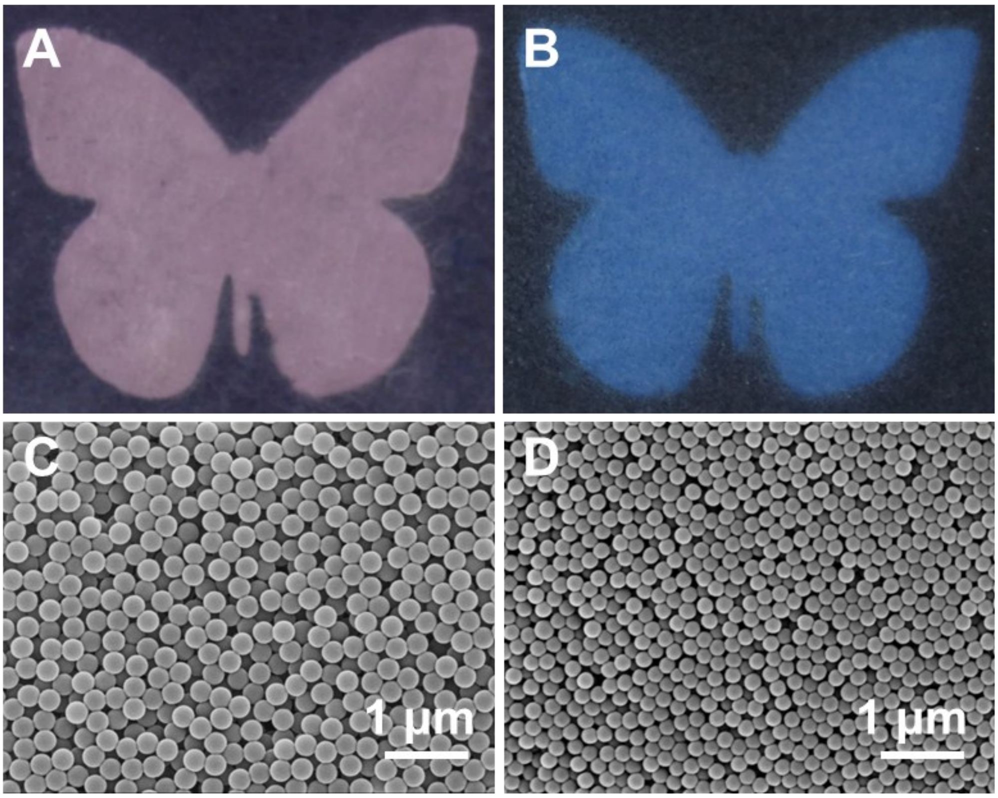

Figure S3. The photographs of the emerged red **(A)** and blue **(B)** butterfly patterns assembled with 180 nm and 254 nm latex spheres. **(C), (D)** The SEM images of the photonic structures of the butterfly pattern regions, which showed the random arrangements of the latex particles.

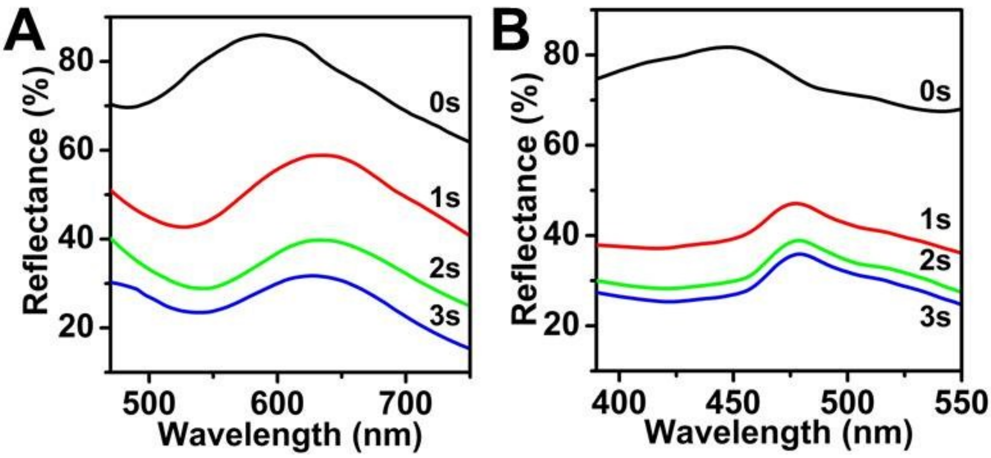

Figure S4. Time-resolved reflectance spectra of the patterned structural color layers after being immersed in water with red structural color **(A)** and blue structural color **(B)**.

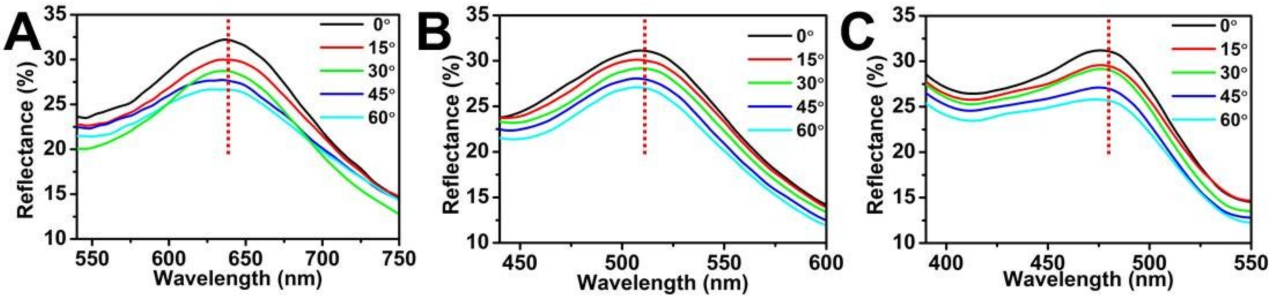

Figure S5. Reflectance spectra of the emerged red **(A)**, green **(B)** and blue **(C)** structural colors at varying detecting angles relative to the normal to the film surface. The unvarying reflectance peaks indicated the non-iridescent nature of these structural colors.

Figure S6. Diverse pattern design of the structural color anti-counterfeiting label by the facile spray-coating method.

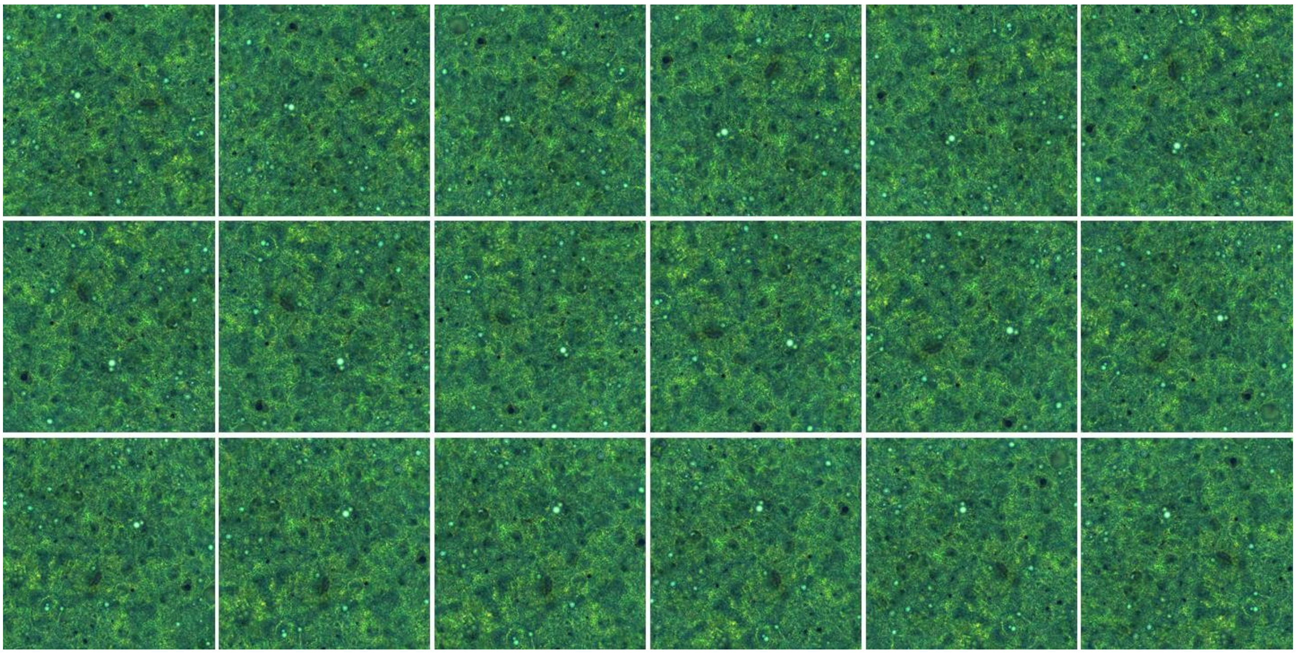

Figure S7. Selected 18 images (corresponding to A1) captured for deep learning.

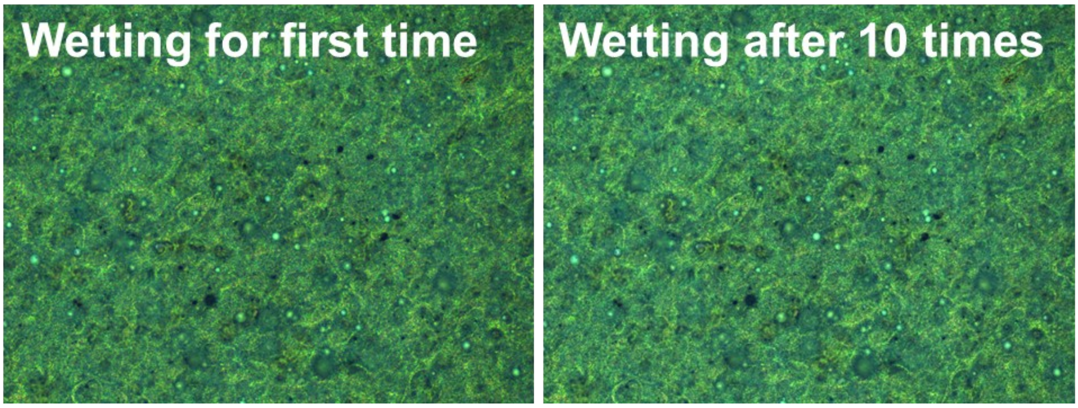

Figure S8. The optical microscope images after repeated wetting of the anti-counterfeiting labels, which showed that the random arrangements of the silica microspheres kept stable.

References

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