

Engineering designer materials with birdinspired structural colors using nanoparticlebased supraballs

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Effect of monodisperse binary nanoparticle mixture (220nm-diameter melanin and 220nm-diameter silica; melanin, blue spheres; silica, yellow spheres) composition and mixing state on the supraball color reflectance. -- (A) Visualizations of the cross-section of binary mixture supraballs with varying levels of particle mixing in the increasing order from top to bottom and varying relative proportion of silica in the increasing order from left to right. (B) Corresponding structural colors, represented as RGB color panels, of the binary mixture supraballs. Credit: *Science Advances*, doi: 10.1126/sciadv.adf2859

Materials scientists are often bioinspired, and in a new study, birdinspired by structural colors exhibited by avian species to form noniridescent nanoparticle assemblies. Such nanoparticle mixtures varying in particle chemistry and size can affect the color produced to identify structure-color relationships and create designer materials with tailored color.

In a new report on *Science Advances*, Christian M. Heil, and a research team at several international, multidisciplinary research institutes in the U.S., Belgium, and Germany, showed how to reconstruct the assembled structures via <u>small-angle scattering measurements</u>.

The research team successfully and quantitatively predicted the experimentally observed colors in mixtures with strongly absorbed <u>nanoparticles</u> to demonstrate the influence of a single layer of segregated nanoparticles and produce a color of interest. The versatile computational approaches integrated in this work were useful to engineer synthetic materials with desired colors suited for paints, cosmetics, and food coloring applications.



Synthetic color fabrication and characterization.

The color fabrication of <u>synthetic materials</u> is inspired by <u>diverse arrays</u> <u>of color</u> in nature. They can arise from the spatial organization of nanostructured materials that are resistant to color degradation. Materials with consistent, periodic nanostructures can form iridescent colors, while those with short-range ordering produce non-iridescent colors.

Materials scientists can mimic natural non-iridescent structural colors by self-assembling polymeric nanoparticles via amorphous assemblies of inorganic nanoparticles. They can vary the colors of nanoparticle assemblies by regulating their <u>structure and optical properties</u> to provide increased structural diversity. Scientists can adjust the nanoparticle size ratio between the components and the composition by generating a myriad of diverse structures and structural colors.

Characterizing designer materials

Heil et al. used small-angle scattering and electron microscopy to obtain structural information of the <u>designer materials</u>. They further integrated small-angle neutron and X-ray scattering to provide well-suited methods to examine the nanoscale and <u>bulk structural information</u>. For optical modeling of complex nano-assemblies, they used the <u>finite-difference time-domain</u> (FDTD) method.

The small-angle-scattering experiments generated a scattering experiment profile, which they interpreted with computational reverseengineering analysis for scattering experiments (CREASE) to generate nanoparticle assemblies.

The researchers combined CREASE-FDTD methods to understand binary nanoparticle assemblies forming <u>supraballs</u> to produce a wide



spectrum of colors. The outcomes characterized the <u>optical properties</u> of nanoparticle assemblies and showed the capacity to design colors on demand for wide-ranging applications in paints, cosmetics and food coloring.



Applying the CREASE method to reconstruct the binary nanoparticle mixture assembly structure from SANS profiles. (A) Schematic describing the CREASE method operation. (B to D) SANS plot of I as a function of q for NCM (left top; gray dots) and MCM (left bottom; yellow dots) condition of binary mixture supraballs overlaid with the CREASE output structures' scattering profile for NCM and MCM condition for (B) 1:4, (C) 1:1, and (D) 4:1 melanin:silica compositions. (B) plots the NCM and MCM from CREASE as orange and green, respectively, with an χ^2 scattering error of 2.35, (C) colors the NCM and MCM from CREASE as black and red, respectively, with an χ^2 scattering error of 2.01. (B) to (D) show, on the right,



transmission electron micrograph of the cross section of a representative binary mixture supraball (right top; melanin, lighter spheres; silica, darker spheres; scale bars, 500 nm) and Visual Molecular Dynamics (VMD) visualization of the central portion (3 μ m by 3 μ m by 10 μ m) of reconstructed 3D binary nanoparticle mixture assembly with yellow spheres representing silica chemistry and blue spheres representing melanin (right bottom). (C) Scattering profile and CREASE results originally from (50). (E) Scanning electron micrographs of a representative 1:4 binary mixture supraball and supraball surface (left top; scale bars, 1 μ m and 100 nm, respectively) and lognormal size distribution of the melanin nanoparticles used to form the surface segregated shell after CREASE reconstruction (left bottom). VMD visualization of the central portion (3 μ m by 3 μ m by 10 μ m) of reconstructed 3D binary nanoparticle mixture assembly with the added melanin shell layer (right). Credit: *Science Advances*, doi: 10.1126/sciadv.adf2859

Structural color diversity with binary nanoparticle mixtures

The <u>materials scientists</u> examined the composition and phase morphology of binary mixtures of absorbing and non-absorbing species of similar size to provide a method to regulate the prediction of structural colors upon which the relative sensitivity of structural color relied. This work produced a rich diversity of colors to show the need to know the relative composition of two nanoparticle types in a binary mixture alongside their phase segregation and the extent of mixing.

To monitor this process, Heil et al. used an experimental and computational approach, which provided information about the internal morphologies of supraballs to predict each reflectance spectra and their resulting color.





Optical modeling and color analysis comparison between experimental supraballs and FDTD calculations on the CREASE output structures. (A) Reflectance spectra (solid curve, experimental; dashed curve, computed), RGB color panel, CIE 1976 chromaticity values, and CIE 1931 chromaticity coordinates' comparisons for the 1:4 melanin:silica binary mixture supraball system. The quantitative difference between FDTD and experimental colors is given by a color difference (ΔE) value that is ~0.9 times the average JND value. The black box in the inset of the optical micrograph of corresponding supraball represents the size of the area (3 µm by 3 µm) probed during optical measurements using



microspectrophotometer. (B) Similar to (A) but for 1:1 melanin:silica binary mixture supraball system with a ΔE value that is ~1.9 times the average JND value. (C) Similar to (A) but for 4:1 melanin:silica binary mixture supraball system with a ΔE value that is ~1.4 times the average JND value. The lighter colored envelopes coming from the experimental and computational lines in the reflectance plots indicate the standard deviation (SD). The SD of the computational approach is from FDTD simulations of three independently generated CREASE structures, and the SD of the experimental measurements is from ~15 independent measurements of different supraballs. Credit: *Science Advances*, doi: 10.1126/sciadv.adf2859

CREASE and small-angle neutron scattering data

The team conducted <u>small-angle neutron scattering experiments</u> for three different mixtures of nanoparticles and produced suspensions of micron-scale supraballs. They obtained morphological information of the interior of supraballs by isolating the scattering contribution of individual components within the structure to collect total scattering from the nanoparticles of interest.

Using computational reverse-engineering analysis for the scattering experiments, the team analyzed scattering results from multicomponent structures to obtain 3D structural reconstruction and optical simulations of the products.

The outcomes of the structural analysis based on computational reverseengineering analysis for scattering experiments (CREASE), provided the coordinates of all nanoparticles within supraballs. This information was useful to calculate the scattering of light by employing the finitedifference-time domain method (FDTD).

The combined CREASE-FDTD method represented a close structural



match to experimental systems with optical modeling comparisons. Based on the remarkable agreement between these two combined methods, the team modeled optical properties underlying a complex binary mixture of nanoparticles to understand the impact <u>of size</u> <u>dispersity</u> and surface segregation on structural color.



Optical modeling and color analysis comparison between FDTD calculations on the CREASE output structures with and without the melanin shell. (A to C)



Reflectance spectra (solid curve, with melanin shell; dashed curve, without melanin shell), structure visualizations, CIE 1931 chromaticity coordinates' comparisons, and RGB color panel comparisons for the (A) 1:4, (B) 1:1, and (C) 4:1 melanin:silica binary mixture supraball systems. The lighter colored envelopes coming from with and without shell computed reflectance profiles in the reflectance plots indicate the SD. The SD is from FDTD simulations of three independently generated CREASE structures for each case. Credit: *Science Advances*, doi: 10.1126/sciadv.adf2859

Outlook

In this way, Christian M. Heil and colleagues combined two key methods; computational reverse-engineering analysis for scattering experiment (CREASE) and the finite-difference-time domain method (FDTD) to provide a new platform to model structural colors of nanoparticle-based supra-assemblies. The bird-inspired <u>structural colors</u> formed non-iridescent nanoparticle assemblies, while those with shortrange ordering produce non-iridescent colors.

The team used two primary methods to determine the outcomes as a function of size, dispersity, phase morphology and strongly absorbing optical properties of the nanoparticles. This combined method produced reconstructed 3D structures of nanoparticle assemblies with scattering profiles that were a close match to those obtained via small-angle neutron scattering measurements. The combined method is well-suited for multiscale modeling to study optical properties of much larger assemblies of supraballs, such as packed films and pigment dispersions.

This method can design programmable colors for practical applications in coatings, paints and cosmetics to help materials researchers better understand and design complex materials for applications across the electromagnetic spectra, while predicting material properties such as



thermal, electrical conductivity and mechanical properties, which depend on varying structural compositions of the designed materials.

More information: Christian M. Heil et al, Mechanism of structural colors in binary mixtures of nanoparticle-based supraballs, *Science Advances* (2023). DOI: 10.1126/sciadv.adf2859

Vukusic P. et al, Photonic structures in biology. *Nature* (2023), <u>DOI:</u> <u>10.1038/nature0194</u>

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