White nanolight source for optical nanoimaging

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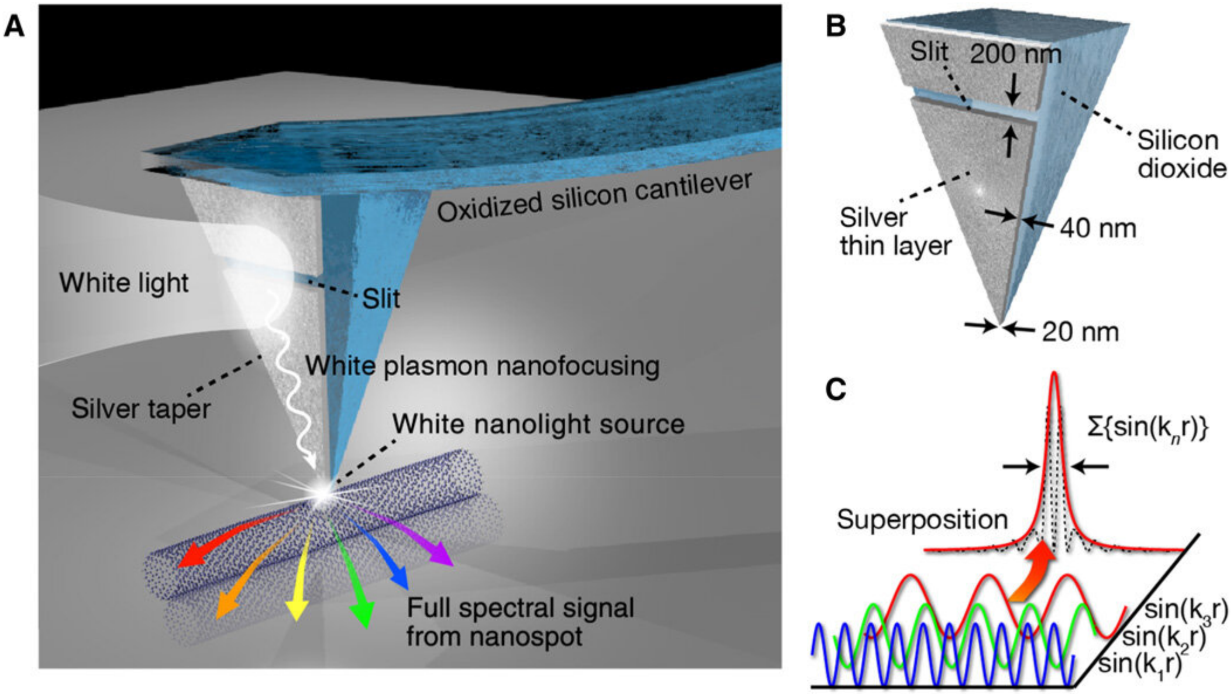
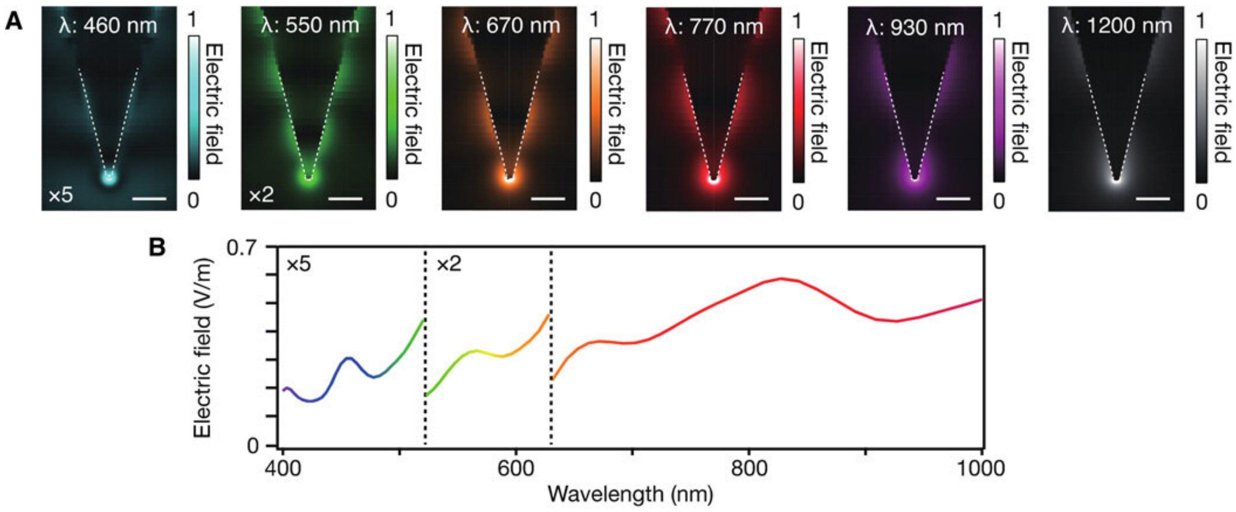


Plasmon nanofocusing of white light for full spectral nanoanalysis. (A) Schematic of plasmon nanofocusing for white light and spectral bandgap nanoanalysis. (B) Schematic of tapered metallic structure used for simulation. (C) Superposition of waves with different wave vectors. Credit: Science Advances, doi: 10.1126/sciadv.aba4179

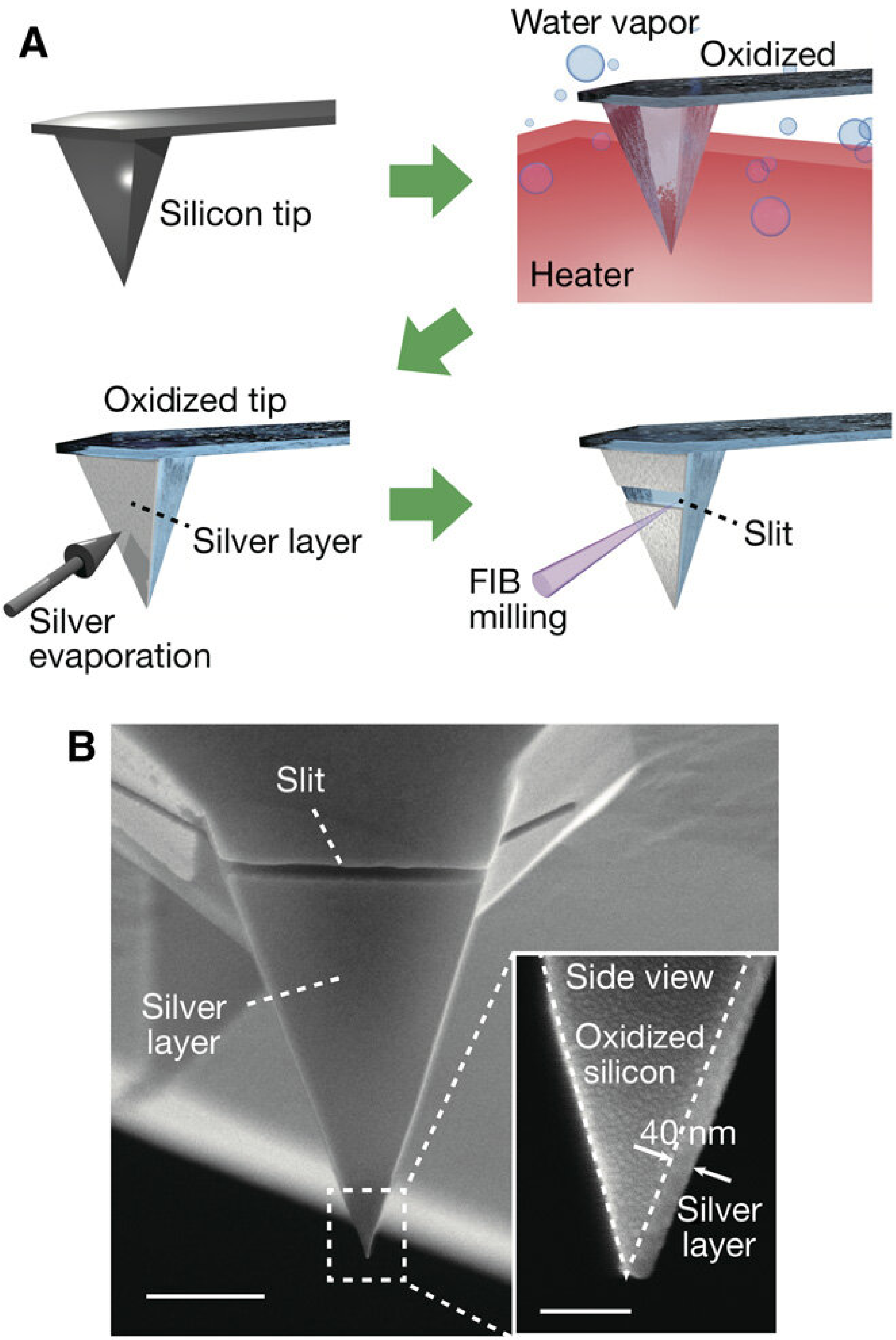
Nanolight sources based on resonant excitons of plasmons near a sharp metallic nanostructure have attracted great interest in optical nanoimaging. However, the resonant phenomenon only works for one type of wavelength that resonates with plasmons. Compared to plasmonic resonance, the alternative plasmon nanofocusing method can generate a source of nanolight by propagating and compressing plasmons on a tapered metallic nanostructure, independent of wavelength, due to its reliance on propagation. In a new report on *Science Advances*, Takayuki Umakoshi and a research team in applied physics and chemistry in Japan generated a white nanolight source spanning across the entire visible light range through plasmon nanofocusing. Using the process, they demonstrated spectral bandgap nanoimaging of carbon nanotubes (CNTs). The experimental demonstration of the source of white nanolight will enable diverse research fields to progress toward next-generation, nanophotonic technologies.

The co-existence of multiple wavelengths of light in a confined nanometric volume can constitute an interesting optical effect. The unique nanolight is therefore a promising platform for diverse research fields by providing opportunities to probe a sample across a range of wavelengths, or induce light-light interactions between different wavelengths at the nanoscale. Optical antennas have played an important role in recent decades to confine light at the nanoscale through localized plasmon resonances in metallic nanostructures, leading to unprecedented research in nanolight, including light-field enhancement. Since plasmon resonance is a resonant phenomenon, it cannot facilitate broadband nanolight generation, therefore, as a result, plasmon nanofocusing has gained wider attention as an alternative to generate sources of nanolight. During the process, a nanoscale light source can be engineered by propagating and superfocusing surface plasmon polaritons (SPPs) at the apex of a metallic, tapered superstructure. The work led to enormous enhancement of the light field at the nanoscale, at the apex and resulted in background-free illumination. Scientists have explored the resulting broadband property for four-wave mixing with a high nonlinear conversion efficiency. The plasmon-nanofocused broadband light source is a powerful tool across diverse research fields.



Broadband property of plasmon nanofocusing evaluated by FDTD simulations. (A) Electric field distribution maps in the vicinity of the apex of the tapered silver structure produced by FDTD simulations. Scale bars, 100 nm. The plasmon coupler slit, where white light was illuminated, is not shown, as it is out of the frame. (B) Simulated near-field spectrum detected 6 nm below the apex. Credit: Science Advances, doi: 10.1126/sciadv.aba4179

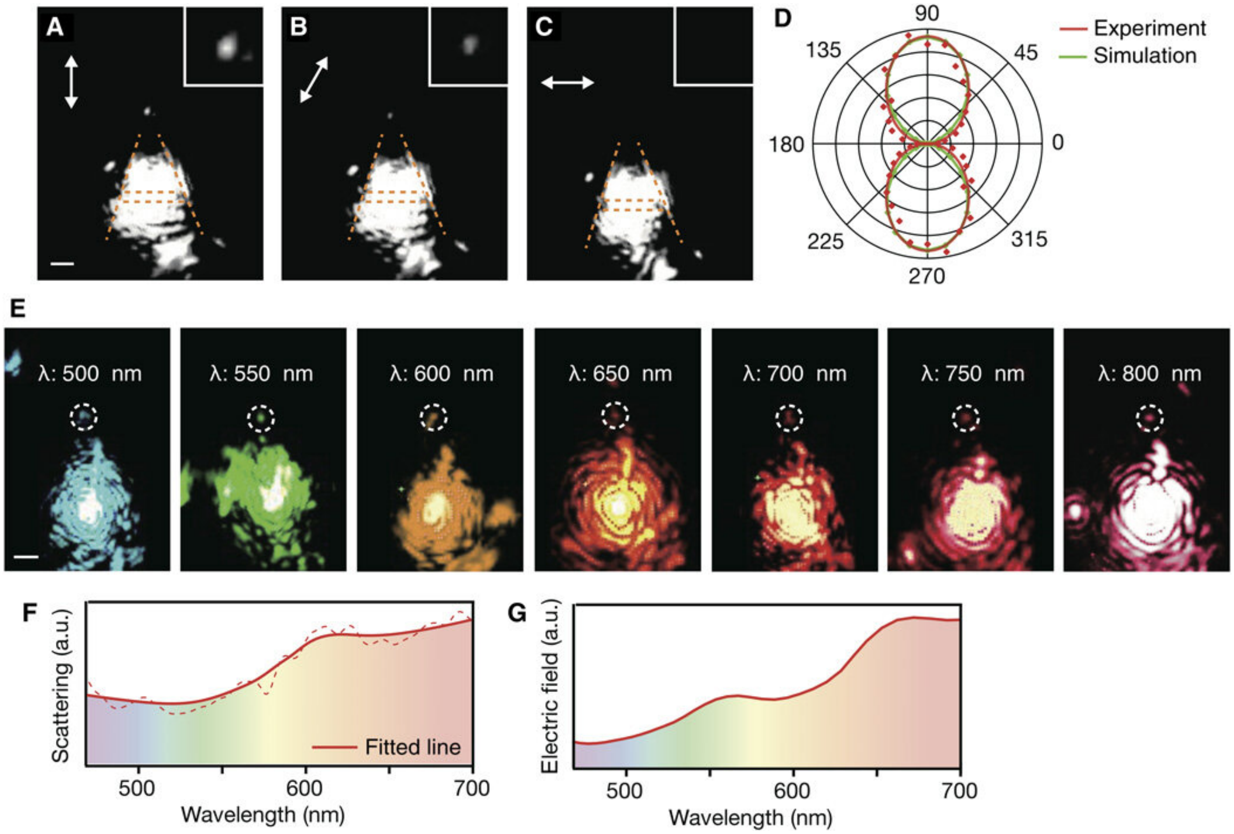
In this work, Umakoshi et al. introduced a white nanolight source spanning across the entire visible wavelength range—generated via plasmon nanofocusing. They showed broadband energy bandgap optical imaging of carbon nanotubes using the white nanolight source. Although plasmon nanofocusing can be excited in a broad wavelength range, researchers have only used it in the near-infrared range due to limitations of materials constituting the tapering structure. They had used gold as a material to form conical tapered structures and lower ohmic losses, but such experiments remained in the near-infrared range and not in the visible or ultraviolet range. Umakoshi et al. had also recently developed an efficient fabrication method to form tapered metallic structures based on thermal evaporation, where the construct included a commercially available silicon cantilever with a pyramidal tip. Using a surface of the pyramid as a base, they obtained a two-dimensional metallic taper and created an extremely smooth metallic coating applicable to a range of metal types, including silver. Using the silver taper, the team obtained highly efficient plasmon nanofocusing with 100 percent reproducibility at 642 nm and conducted white plasmon nanofocusing across a broad range of visible wavelengths.


A
Water vapor
Oxidized
Silicon tip
Heater
Oxidized tip
Silver layer
Silver evaporation
Slit
FIB milling
B
Slit
Silver layer
Side view
Oxidized silicon
40 nm
Silver layer

Fabrication of a tapered silver structure on a cantilever tip. (A) Schematic of fabrication process of the tapered silver structure on a cantilever tip. (B) Scanning electron microscopy image of the fabricated tapered silver structure on the cantilever tip. The inset shows a side view of the silver layer. Scale bars, 2 μm (inset, 200 nm). Credit: Science Advances, doi: 10.1126/sciadv.aba4179

Designing and engineering a tapered metallic structure for broadband plasmon nanofocusing

Umakoshi et al. developed a tapered metallic structure to maintain a broadband white nanolight source on an oxidized silicon pyramidal tip with a thin silver layer coated on a surface of the pyramid. Using a single slit of 200 nanometer (nm) in silver they coupled light in the visible range, and calculated the electric field distributions in the vicinity of the apex at multiple excitation wavelengths using the finite-difference-time domain (FDTD) method. The team observed strong electric fields confined at the apex tip at excitation wavelengths ranging from 460 nm to 1200 nm. The work showed how a 200-nm-wide slit generated a broadband nanolight source spanning across the entire visible region to even reach the near-infrared region. During the fabrication process, the scientists used a commercially available silicon cantilever tip with a pyramidal shape. They oxidized the silicon cantilever and developed a smooth silver coating of 1 nm surface roughness to reduce energy loss during SPP (surface plasmon polariton) propagation.

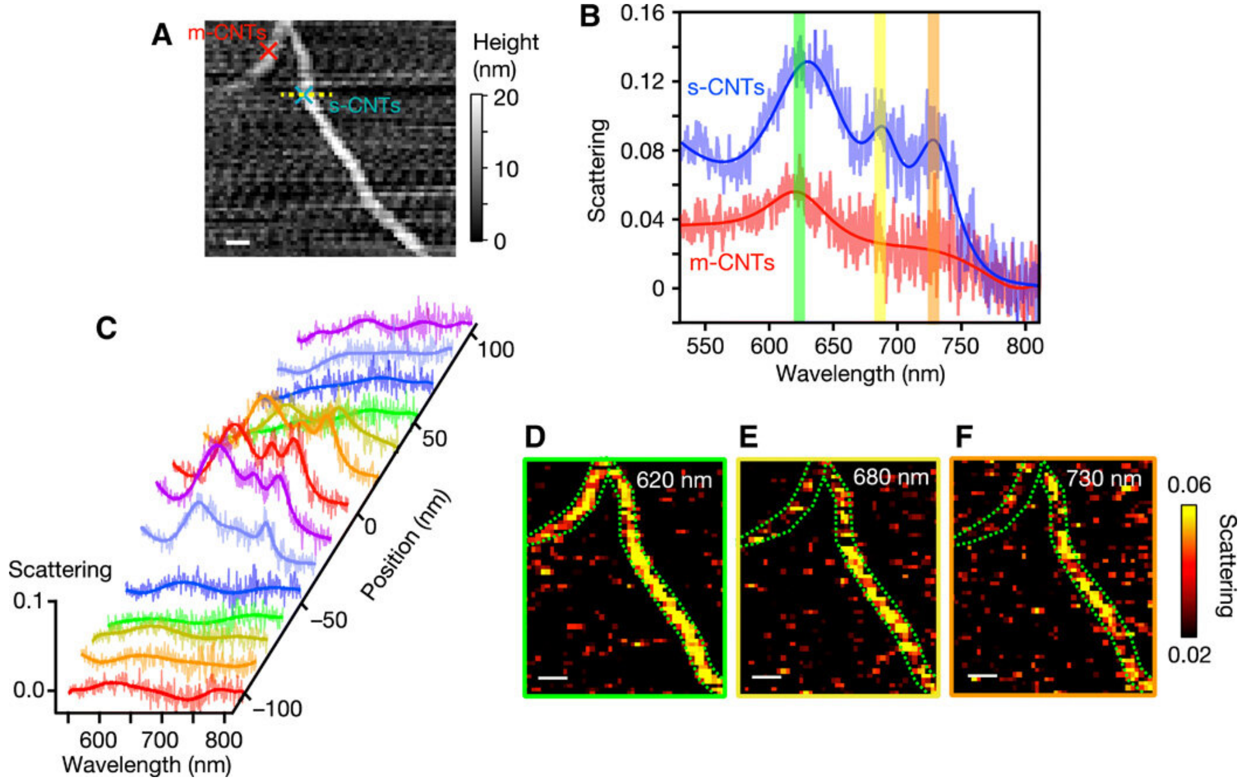


Optical observation of a white nanolight source generated through plasmon nanofocusing. (A) Optical image of a tapered silver structure under illumination by supercontinuum laser at its slit. The locations of the boundaries of the tip as well as the slit are indicated by dashed lines. The inset shows a zoomed image of the apex. Incident polarization was normal to the slit as indicated by the arrow. (B and C) Optical images of the same tapered silver structure with supercontinuum laser illumination at different incident polarizations, as indicated by the arrows. (D) Polar graph of the light spot intensity at the apex with respect to the incident polarization; 0° and 90° correspond to parallel and perpendicular polarizations, respectively. (E) Optical images of the tapered silver structure illuminated with a supercontinuum laser, observed through a series of band-pass filters indicated by their central wavelengths. (F) Scattering spectrum of the optical spot at the apex of the tapered silver structure. a.u., arbitrary units. (G) Simulated near-field spectrum calculated at the tip apex. Scale bars, 2 μm (A and E). Credit: Science Advances, doi: 10.1126/sciadv.aba4179

Generating a white light source via plasmon nanofocusing and conducting spectral bandgap imaging

To understand the process of confined white light production through the tapered structure based on plasmon nanofocusing, the team illuminated the slit structure with a coherent supercontinuum laser that spanned across a wide range of wavelengths. When the incident polarization was perpendicular to the slit, they noted the best coupling in the setup in agreement with simulations. As the wavelength shortened, the scattering efficiency increased. Therefore, the team experimentally observed a higher intensity in the shorter wavelength range.

They used the plasmon-nanofocused white light source to perform spectral nanoanalysis of CNTs (carbon nanotubes). The white nanolight source localized at the tip of the apex interacted with CNT bundles containing multiple bandgaps during the experiment. The scattering signal increased during the experiment to indicate photons with the same energy that corresponded to the bandgaps of the CNTs. Umakoshi et al. then combined the approach with Raman spectroscopy to examine chirality of the CNT sample.



Optical nanoimaging of CNTs using the white nanolight source. (A) An AFM image of CNT bundles. The structures observed on the left and the right parts of the image are the metallic (m-CNTs) and semiconducting (s-CNTs) CNTs, respectively, as identified during the sample preparation process. Scale bar, 100 nm. (B) Near-field spectra of s-CNTs and m-CNTs, obtained from the locations indicated by the blue and red crosses, respectively, in (A). (C) Near-field spectra obtained pixel by pixel along the dotted line in (A). (D to F) Bandgap images constructed at 620, 680, and 730 nm, respectively. Scale bars, 100 nm. Credit: Science Advances, doi: 10.1126/sciadv.aba4179

The plasmon-focussed white light source in this work is a fundamental and effective state of light for bandgap nanoimaging. This work will pave the way for a variety of possible applications, including probing biomolecules to understand their absorption properties at nanoscale spatial resolution. A mid-infrared broadband nanolight source will also be productive across materials science and molecular biology. This technique can also boost the analytical capability of surface-enhanced Raman spectroscopy to investigate molecular vibrations.

In this way, Takayuki Umakoshi and colleagues generated a white nanolight source at the apex of a tapered silver structure using plasmon nanofocusing to perform nanoanalysis of carbon nanotubes. The team designed and engineered a tapered structure that induced plasmon nanofocusing across a broad wavelength range. The spectral bandgap technique will have wide-ranging applications at the nanoscale across materials science and biological research. The demonstrated work is only a single example, with diverse applications possible based on a powerful and fundamental nanoscale optical tool with excellent wavelength flexibility.

More information: Takayuki Umakoshi et al. White nanolight source for optical nanoimaging, *Science Advances* (2020). DOI: 10.1126/sciadv.aba4179

Satoshi Kawata et al. Plasmonics for near-field nano-imaging and superlensing, *Nature Photonics* (2009). DOI: 10.1038/nphoton.2009.111

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