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Transdimensional Photonics

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In all areas of science ranging from mathematics and physics to data analysis and computation, moving from one dimensionality to the other reveals new phenomena and brings remarkable performance changes. Historic examples from surface science and the very recent area of optical metamaterials[1−5] and metasurfaces[6,7] have shown that transitioning from bulk, three-dimensional (3D) structures to two-dimensional (2D) counterparts uncovered truly new physics unattainable with 3D systems and enabled novel applications. However, the *transdimensional* regime that by definition relates to in-between dimensionalities remains somewhat overlooked.

In the highly dynamic fields of nanophotonics and plasmonics[8,9]—or metal-based nanoscale optics—extensive research has focused on utilizing either conventional materials in the form of thin films or nanoparticles with bulk material properties, or 2D structures including metal−dielectric interfaces and novel 2D materials.[10] Yet, the material optical properties' evolution in the transitional—transdimensional regime between 3D and 2D—is still underexplored. In such *transdimensional materials* (TDMs) that have a thickness of only a few atomic layers, the material's optical and electronic properties are expected to show unprecedented tailorability including unusually strong dependences on structural parameters such as thickness (number of atomic monolayers), composition (stoichiometry, doping), strain, and surface termination compared to their 3D counterparts and conventional thin films (that are usually described by the bulk material properties). Similar to truly 2D materials such as graphene,[11,12] transdimensional materials are also expected to show extreme sensitivity to external optical and electrical stimuli.[13] With the recent discovery of 2D and quasi-2D materials[14] as well as the development of novel plasmonic materials that can be grown as ultrathin layers (down to a few atomic layers)[15−17] one can now probe the exciting properties of TDMs as they evolve from a single atomic layer to a larger number *N* when the properties become similar to those of the bulk material.

Plasmonic TDMs, or ultrathin metallic materials, that offer uniquely strong light confinement via highly localized *surface plasmon* (SP)[9] excitations are of particular interest, as they are expected to show higher transparency, unusual tailorability, dynamic tunability, and novel quantum phenomena compared to conventional plasmonic films. More interestingly, strongly confined SPs in plasmonic TDMs could enable normally forbidden atomic transitions[18] since they have wavevectors with exceptionally large momentum. While some theoretical predictions on tunability, anomalous dispersion, and strong light confinement in ultrathin plasmonic films have been made,[13,19] much remains unclear in the evolution of the materials properties including their linear and nonlinear optical response as well as quantum and nonlocal effects.

Thus far, the experimental progress in plasmonics utilizing ultrathin materials has largely been impeded by the challenges in producing atomically thin films maintaining their metallic properties. With the recent experimental demonstrations of ultrathin noble metals[20] and plasmonic transition metal nitrides,[21] the rich materials "playground" has now been extended from noble metals to emerging tailorable plasmonic materials such as transition metal nitrides/carbides and their quasi-2D counterparts, MXenes, semimetals, 2D materials, and their van der Waals heterostructures as well as low-loss transparent conducting oxides, giving us access to excellent platforms for probing TDMs.

One recent example of investigating the optical properties of TDMs, ultrathin films down to 2 nm (around 10 atomic layers) with a persistent metallic character, has been demonstrated using TiN.[21] Atomically smooth, epitaxial thin films down to 1 nm are enabled by the film growth of TiN on lattice-matched substrates, such as MgO, via DC reactive magnetron sputtering. Despite the decrease in the metallicity with reducing thickness, a carrier concentration on the order of 10^{22} cm^{-3} is retained even in the 2 nm film, indicating a strong plasmonic character. Additionally, first-principles density functional calculations (DFT) reveal that as the film thickness is decreased, strong confinement shifts the interband transitions, and hence the optical properties, providing a unique approach to tune the optical response by controlling the film thickness.[22]

Another avenue to tailor the optical properties of TDMs includes strain engineering. Below a critical thickness, an epitaxial thin film will retain strain induced by the substrate. It has been theoretically demonstrated that on varying the in-plane lattice parameter of an ultrathin film, the optical response can be tuned.[22] This can be achieved experimentally by growing strained ultrathin films on lattice-mismatched substrates. In conjunction with the thickness dependence of TDMs, strain engineering offers a novel way to tailor the optical response of plasmonic materials.

Dynamic electrical control of optical properties is yet another exciting avenue TDMs enable.[13] So far electrical modulation in noble metals has remained elusive due to the associated difficulties in growing metals down to only a few atomic layers. Advances in producing ultrathin transition metal nitrides that are only a few atomic layers in thickness have enabled an ideal test bench to probe the limits of electrical tuning. Preliminary studies have revealed that the free electron density can be modulated over 7% in 1-nm-thick TiN films

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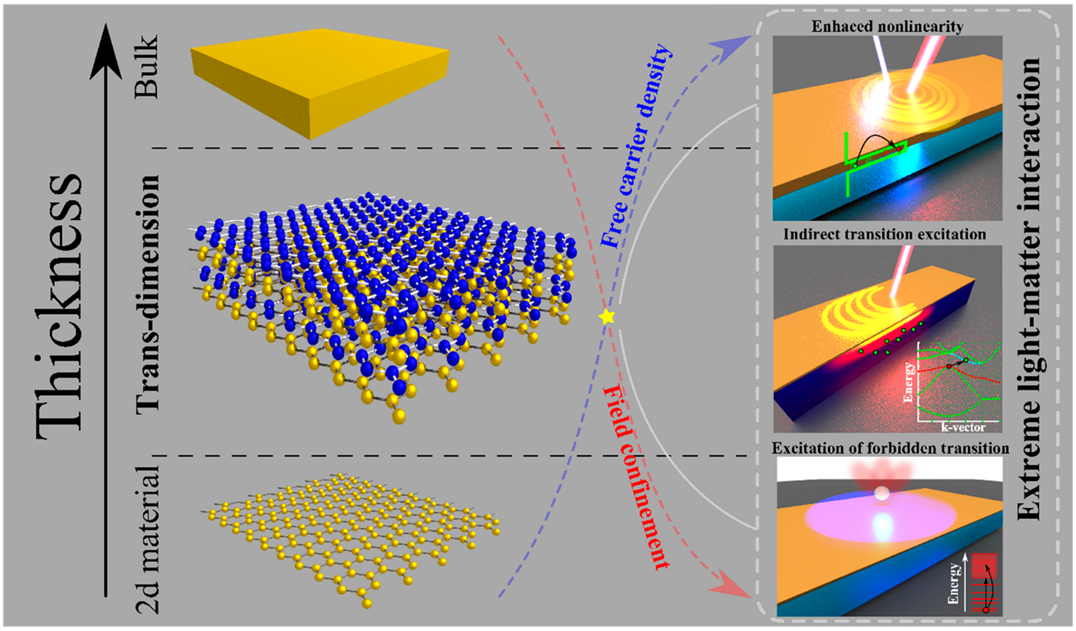

Figure 1. Schematics.

using existing solid-state gating approaches,[23] offering promising ways toward the realization of dynamic plasmonic and nanophotonic components.

Ultratight light confinement and extreme tailorability and tunability in TDMs are expected to open up a new chapter in extreme light−matter interactions and advance the fields of plasmonics and optical metasurfaces. For example, the sensitivity of the TDMs' optical response to external electrical and optical stimuli as well as ultrahigh localization of light could enable the realization of novel *space−time* optical metamaterials/metasurfaces with unmatched functionalities where the properties are dynamically modulated in both space and time. The exciting outcomes of transdimensional photonics might also include demonstrations of exotic effects including unprecedented quantum confinement, strong indirect and forbidden transitions in solids and atoms, and novel photonic designs for dynamic parity−time symmetry breaking. Discovering new, unexpected phenomena in transdimensional photonic materials will no doubt open up a new chapter in light−matter interactions.

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Notes

Views expressed in this editorial are those of the authors and not necessarily the views of the ACS.

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