Research Summary: Engineering and Spectroscopy of Photon Momentum
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(References in bold included in package)

Electromagnetic wavefront-shaping technologies exploit *control* of photon momentum to perform, e.g. beam-steering and light-focusing functions. Reciprocally, *measurements* of photon momentum provide substantial insight nanomaterial optical properties. Research in my group synthesizes these topics and is divided into two main themes: 1) controlling photon momentum with active metasurfaces and 2) developing and using momentum-resolved spectroscopies for fundamental investigations of nanomaterial optical properties.

Theme 1: Engineering Photon Momentum
Introduction and Overview of Pre-Tenure Accomplishments
The ability to engineer optical properties at subwavelength dimensions has led to metamaterials that provide unprecedented control of electromagnetic waves. Metamaterials are typically constructed from metal-based plasmonic elements. Despite my expertise in the area of plasmonics (as evidenced by a 3500+ cited 1st-author 2010 Nature Materials review article), my own research focuses instead on semiconductor-based metamaterials. My Ph.D. research on semiconductor optical antennas (Physical Review Letters 2007, Nature Materials 2009, Nature Photonics 2009) produced some of the earliest work demonstrating metamaterial phenomena deriving from non-metallic constituents. This early work particularly influenced subsequent research on dielectric and semiconductor metasurfaces. When I joined UCSB in 2012, most metasurfaces exploited *static* effects derived from nanopatterning. My group's early research focused instead on *active* metasurfaces for dynamically controlling external infrared beams. A fundamental challenge is achieving large tunability in deeply subwavelength elements. My group addressed this challenge with investigations of large-magnitude optical tunability achieved through free-carrier refraction (ACS Photonics 2015, Nano Letters 2015, Advanced Optical Materials 2016) and thermo-optic effects (Nano Letters 2017, Nature Communications 2017).

Progress Since Tenure (Awarded in Summer 2017)
My group's research on active metasurfaces has progressed nicely since I was awarded tenure. Our experimental demonstration of an electrically tunable semiconductor heterojunction metasurface element (**ACS Photonics 2019)** represents the culmination of a 6+ year effort to conceive, design, and implement subwavelength metasurface constituents capable of electrically-tuning reflection phase over a full 2π radians. These results demonstrate the promise of our approach, but also reveal myriad challenges that must be overcome to achieve our ultimate goal of a fully programmable metasurface. Complementary work on spatially-uniform thermo-optic tuning demonstrates new, easier opportunities to construct e.g., tunable metasurface lenses (Physical Review Applied 2018) and filters (Nanophotonics 2018). We are simultaneously pursuing research into active infrared metasurfaces that exploit the metal-insulator phase transition in Vanadium Oxide (VO_2). Typical research in this area, including some of our own work (ACS Photonics Feb. 2018), focuses on *switchable* behavior as the material toggles between metallic and insulating states. In contrast, our most recent work exploits the percolative thin-film VO_2 metal-insulator transition to achieve electrically-controlled *continuous tuning* of infrared reflectivity (**ACS Photonics 2018**).

My group's expertise in active metasurfaces has led to an exciting new research direction: light-emitting metasurfaces. Traditional metasurfaces operate on an incident light beam and provide precise control over transmitted or reflected waveforms. Adapting metasurface concepts for incoherent light emission is significantly more complex due to the lack of a phase-defining incident beam. We recently demonstrated the first-ever example of directed light emission from a phased-array metasurface (**Nature Photonics 2020**). Our ability to create asymmetric, unidirectional emission patterns is bolstered by a more than 100-fold increase in the photoluminescence external quantum efficiency. Moreover, we developed accompanying theoretical and analytical models for predicting and explaining the complex and counter-intuitive behavior of light-emitting metasurfaces. Our subsequent demonstration of focused light emission (**Nature Communications 2021**) exemplifies the usefulness and validity of these models, and highlights the potential for exploiting metasurface functionality in future light-emitting devices.

Theme 2: Spectroscopy of Photon Momentum

Introduction and Overview of Pre-Tenure Accomplishments

Complementing efforts to *control* photon momentum in active metasurface devices, we *measure* photon momentum to gain new insight into nanomaterial properties. These efforts build off my postdoctoral work pioneering the use of momentum-resolved photoluminescence (mPL) measurements for quantifying optical anisotropies in nanomaterials (Nature Nanotechnology 2013). This technique is now commonly used by many other research groups across the world. At UCSB, my group has significantly expanded the repertoire of momentum-resolved optical measurements. For instance, by adding momentum-resolved excitation (mPLE) we demonstrated the ability to also resolve the orientations of absorbing (mPLE) dipoles (Physical Review B 2016). We subsequently developed a new "model-blind" momentum-resolved reflectometry technique for measuring optical constants (Optics Express 2016). In complementary work, we demonstrated how control of photon-momentum through beam engineering, can be used to select and enhance specific multipolar light-matter interactions (Physical Review B 2015). Collectively, these investigations demonstrate the power of momentum-resolved techniques, an integral component of my group's ongoing research.

Progress Since Tenure (Awarded in Summer 2017)

Since tenure, my group's investigations have focused on hybrid organic/inorganic perovskites (HOIPs), an important and widely-studied class of solution-processable optoelectronic semiconductor. Our initial studies focused on optical anisotropies in 2D Ruddlesden-Popper HOIPs, which comprise alternating layers of atomically-thin inorganic semiconductor quantum wells and insulating organic spacer molecules. We recognized that this layered structure, in essence, is a naturally occurring metamaterial. Interpreting our momentum-resolved measurements within the context of metamaterial effective medium theory, we showed that the large optical anisotropies derive almost entirely from classical electromagnetic field inhomogeneities (**ACS Nano 2019**). In doing so, we resolved apparent disagreements between experiments and DFT calculations. In the process of making these measurements we made an exciting, unexpected, and unique discovery—2D HOIPs exhibit extraordinarily bright magnetic dipole (MD) light emission (**Science Advances 2020**). Semiconductors, and other extended materials, are uniformly treated within the electric dipole approximation—optical properties are assumed to arise solely from the interaction of electric fields with atomic-scale electric dipoles. Our discovery of the only known extended material (i.e. not an atom or point defect) to exhibit optical frequency MD light emission challenges this assumption, suggests atomic-scale optical MDs may be more prevalent than previously thought, and clarifies the origins of a commonly observed 2D HOIP sideband emission feature. In subsequent studies, we used temperature and morphology-resolved effects to explain the origins of MD light emission—even-parity self-trapped excitons (**ACS Nano 2020**). Moving forward, we recently submitted a manuscript showing that these 2D HOIPs also possess the only known non-unity optical frequency magnetic permeability. As such, Theme 2 research has come full circle. Initial efforts exploiting metamaterial concepts to better understand atomic-scale optical properties now inform efforts to realize metamaterial phenomena at atomic and molecular length scales.