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Exploring Mie Resonances, Anapole States, and Anapole-Exciton Polaritons in Nanopatterned TMD Materials Using STEM EELS

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To reach next-generation photonics applications we need to improve our control over the interactions between light and matter at the smallest possible length scales. Strong light-matter interactions occur when the Rabi frequency, which is the rate of coherent energy exchange between an electromagnetic mode confined in an optical cavity and a dipolar excitation within matter, exceeds the hybrid systems decay rates [1,2]. This strong coupling results in Rabi splitting and the genesis of two new coupled modes called polaritons. Polaritons exhibit inseparable light and matter characteristics that can significantly modify the systems properties, for example the chemical reactivity [3], phase transitions [4], and electronic transport [5]. These discoveries are enabling breakthroughs in quantum and nonlinear optics and material science.

Strongly coupled polaritons have historically been studied at low temperatures using optical microcavities coupled to quantum emitters such as atoms or quantum wells. More recently, polariton research has been extended to hybrid nanoscale systems, such as those composed of metal nanoparticles and transition metal dichalcogenide (TMD) materials. These single nanoparticle platforms enable the creation of plasmon-exciton polaritons (plexcitons) at room temperture via the interaction between excitons in the TMD and localized surface plasmons in the nanoparticle [6]. Optical based characterization methods have made significant advances in understanding plexciton physics. However, because of the optical diffraction-limited spatial resolution, the optical methods lack the ability to directly map these polaritons at their characteristic nanometer length scale. Recently, we have demonstrated for the first time that electron energy loss spectroscopy (EELS) in a scanning transmission electron microscope (STEM) can measure strongly coupled polaritons and map nanoscale variations in plexcitons behavior [7]. STEM EELS was later used to study polaritons in other hybrid systems, such as nanoparticle surface plasmons coupled to quantrum dots [8] and phonons in hBN [9]. These results demonstrate that STEM EELS is a powerful technique for expanding our knowledge of strong light-matter interactions beyond what is accessible using current optical spectroscopy.

Typically achieving strong coupling relies on bright and dark cavity modes. However, recently optically dark scattering states have shown the possibility to also coupled to dipolar excitations. For example, there is growing interest in optical anapole states, which are non-radiating current configurations with vanishing scattering [10,11]. These states are created by the superposition of electric and toroidal multipoles and have radiation patterns that lead to destructive interference in the far-field and a

fascinating radiation-less behavior. Strong coupling between these anapole states and excitons has emerged as an alluring route towards enriching traditional nanophotonics approaches [12,13].

In this work, we explore Mie resonances, optical anapole states and the strong coupling between anapoles and excitons in tungsten disulfide (WS_2) nanodiscs by utilizing experimental and theoretical STEM EELS. To this end, we synthesized high refractive index WS_2 nanodiscs by transferring a mechanically exfoliated WS_2 flake onto a 50nm thick SiN membrane and performing a combination of e-beam lithography and dry etching. We utilize a JEOL Mono NEO ARM 200F instrument to characterize the structure, composition, and nano-optical behavior of the WS_2 nanodiscs as a function of disc diameter. High angle annular dark field (HAADF), secondary electron (SE), and energy dispersive X-ray spectroscopy (EDS) STEM was used to confirm the size, morphology, elemental distribution, and single crystalline nature of the WS_2 nanodiscs (Figure 1). Monochromated STEM EELS with an energy resolution down to 19 meV was used to probe the low-energy-loss response of the nanodiscs (Figure 2). EELS data from the edge of the largest disc exhibits extremely rich low-loss signatures composed of multiple sharp peaks and valleys. As the disc size is reduced, these signals shift to higher energies, and when they shift above the ~2 eV WS_2 A exciton they dampen and blur.

EELS numerical simulations performed using COMSOL Multiphysics software were used to predict and interpret the experimental signals. These calculations confirm that the experiments are measuring a series of multipolar Mie resonances and that the lowest energy large dip in the EELS data corresponds to the first anapole state, which is created by the interference between the electric and toroidal dipoles. In addition, the larger discs host higher order anapole states that also manifest as dips in the EEL spectra. By varying the nanodisc diameter, the Mie modes and anapole states can be tuned to overlap the WS_2 A exciton, leading to anapole-exciton self-hybridization in the strong coupling regime. EELS spectrum imaging was used to spatially map these signals inside and outside the nanodiscs, revealing the spatial behavior of the Mie modes, anapole states, and strongly coupled polaritons. Our results can constitute an avenue for enhancing light-matter interactions using dark scattering states, such as the anapole state, which can now be accessed by electron microscopy [14].

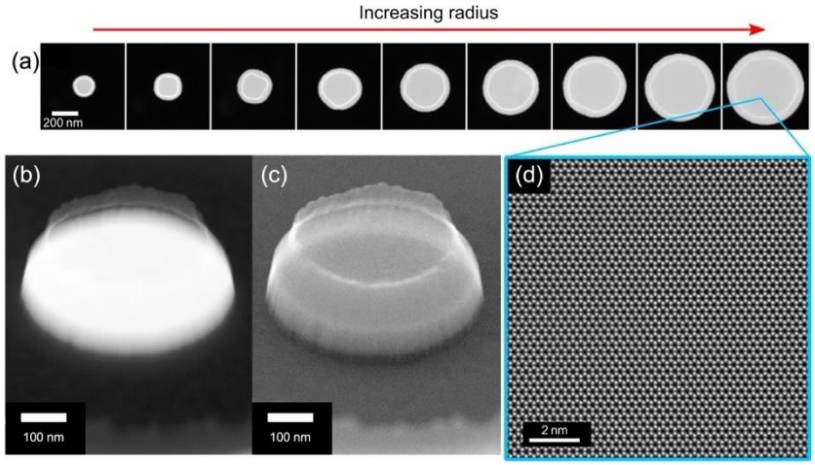

Figure 1. (a) Plan view HAADF STEM images of nine WS_2 nanodiscs with diameters ranging from around 100 to 600 nm. (b,c) 53 degree tilted HAADF STEM and SE STEM images, respectively, of a nanonodisc revealing the morphology of nanodisc and residual resist material. (d) Atomic resolution HAADF STEM from the center of a nanodisc.

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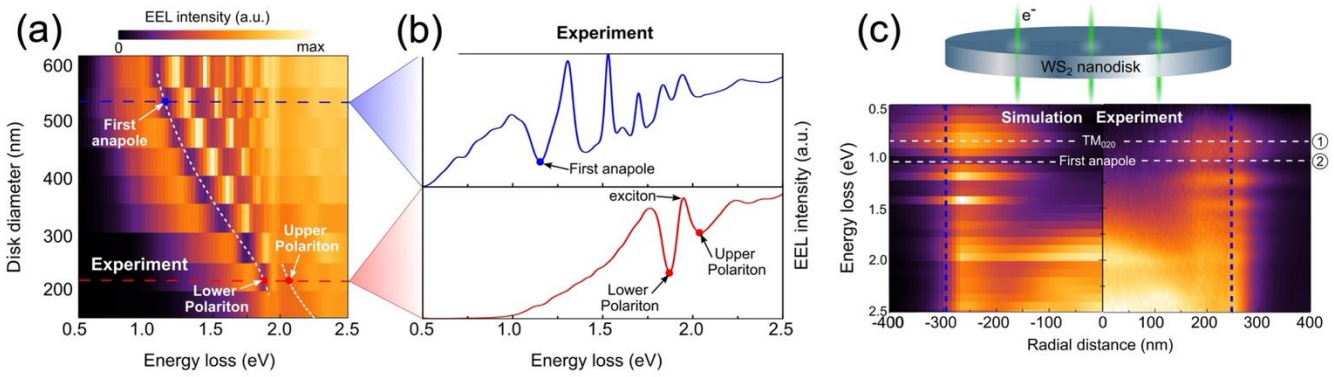

Figure 2. (a) Experimental EEL spectra as a function of disc diameter with two of the spectra displayed in (b). (c) Experimental and simulated EEL spectra as a function of radial position inside and outside the disc with 0 nm radial distance being the disc center. The edge of the disc in the experiment and simulation are marked by the blue dashed lines. The dips in the spectra corresponding to the first anapole are indicated in each figure.

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