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Visualizing Strong Light-matter Interactions Using Fast Electrons

Andrew Yankovich[1], Battulga Munkhbat[1], Denis Baranov[1], Jorge Cuadra[1], Erik Olsén[1], Hugo Lourenço-Martins[2], Luiz Tizei[2], Mathieu Kociak[2], Timur Shegai[1] and Eva Olsson[1]

[1]Chalmers University of Technology, Gothenburg, Vastra Gotaland, Sweden, [2]CNRS, Orsay, Ile-de-France, France

Enhancing our control over the interactions between light and matter at the smallest possible length scales is crucial for realizing next-generation photonics-based applications. In the strong coupling regime, light and matter can couple to create new quasiparticles called polaritons. Polaritons exhibit inseparable light-matter properties that modify the system's properties and have already enabled remarkable breakthroughs in quantum and nonlinear optics and material science. The strength of the interaction depends on the magnitudes of the system's decay rate and the Rabi frequency, which is the rate of coherent energy exchange between the optical component and the quantum emitter. Spontaneous photon emission occurs in the weak coupling regime when the Rabi frequency is slower than the decay rates. Strong coupling occurs if the Rabi frequency exceeds the decay rates, resulting in Rabi splitting and the genesis of two new coupled polaritons. Historically, polaritons have been studied at low temperatures using large optical microcavity devices coupled to quantum emitters, such as atoms or quantum wells. However, recently, plasmon-exciton polaritons (plexcitons) have been realized in hybrid systems composed of transition metal dichalcogenide (TMD) materials and metal nanoparticles, which provide a platform for the interaction of excitons and localized surface plasmons (LSPs), respectively. These single nanoparticle hybrid systems extend polaritonic concepts to room temperature and nanoscale systems [1].

Despite the enormous progress in understanding TMD-based plexcitons using optical-based methods, experimental evidence of plexciton formation has remained indirect and mapping their nanometer-scale characteristics has remained an open challenge because of the optical diffraction-limited spatial resolution. The unprecedented spatial resolution and impressive energy resolution provided by modern electron energy loss spectroscopy (EELS) in a scanning transmission electron microscope (STEM) have led to various discoveries about the basic nature of LSPs and excitons. However, STEM EELS has been underutilized to investigate plexciton physics, even though the possibility has been recently theoretically predicted [2].

In this work STEM EELS experiments were performed on two instruments at 80 and 60 keV respectively: 1) A FEI Titan 80-300 microscope equipped with a Schottky field emission gun, Wien type monochromator, probe aberration corrector, and Gatan Imaging Filter (GIF) Tridiem 866 spectrometer. The monochromator was operated in accelerating mode, which allowed for significantly improved energy resolution compared to normal monochromator use [3]. 2) A JEOL MonoNEOARM 200F microscope equipped with a Schottky field emission gun, double Wien type monochromator, probe and image aberration correctors, and GIF Continuum HR spectrometer. In order to resolve the polariton splitting in EELS, specific techniques were required to ensure high energy resolution and improved signal to noise ratio [4-5].

We demonstrate that plexcitons generated by a hybrid system composed of a single Ag truncated nanopyramid (TNP) and a few-layer WS_2 flake (Figure 1a) can be spectroscopically mapped with sub-nanometer spatial resolution and < 40 meV energy resolution using monochromated STEM EELS. The hybridization occurs between a single dipole LSP mode of the Ag TNP and a single A-exciton state of the WS_2, producing polariton splitting in EELS up to 130 meV (Figure 1b). The polariton EELS observations

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correlate well with optical dark-field spectroscopy data and numerical optical and EELS simulations. Experimental EELS anti-crossing measurements (Figure 1b) using the absorption-dominated extinction signal provide stronger evidence for plexciton hybridization than the typically used optical DF scattering. The anticrossing data, in combination with the analysis of the extracted coupling strength using the coupled mode theory, show that this system is at the onset of the strong coupling regime. Spatially resolved EELS maps (Figure 2) of the polaritons reveal that they adopt a plasmon-like charge and field distribution. Additionally, the mapping reveals the existence of unexpected nanoscale variations in the deep-subwavelength nature of the plexcitons generated by this system. Our results demonstrate that STEM EELS provides a platform for expanding our knowledge of strongly coupled polaritons beyond what is accessible with current optical spectroscopy. These findings pioneer new possibilities for in-depth studies of polariton-related phenomena with sub-nanometer spatial resolution in hybrid material systems. [5-6]

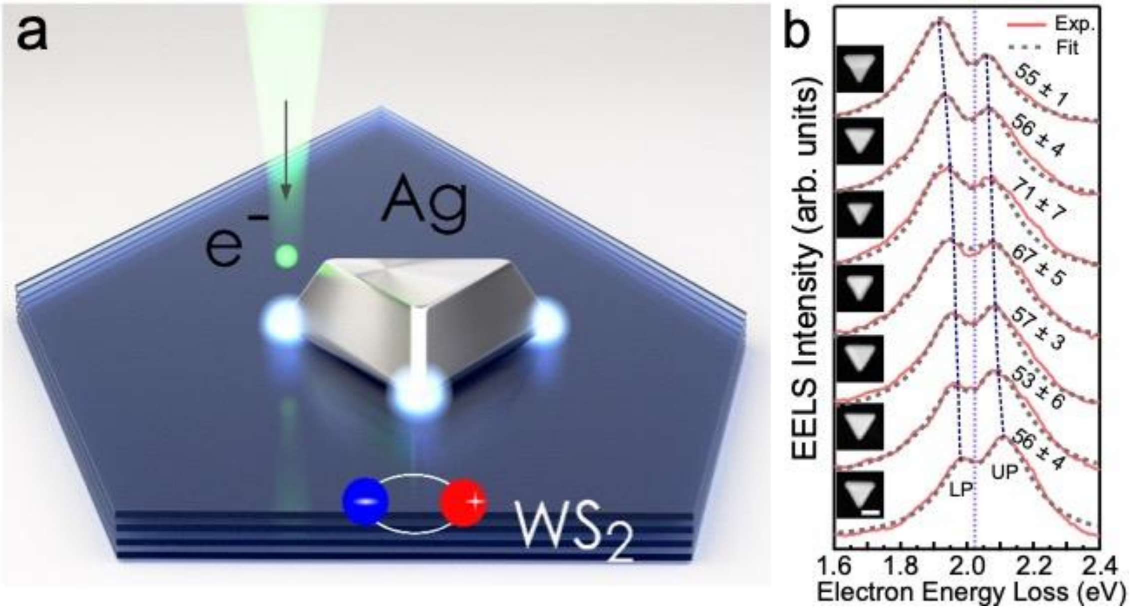

Figure 1. (a) Schematic diagram of a strongly coupled plexciton system composed of a Ag TNP and few-layer WS2 being excited by an electron beam. (b) The red experimental EEL spectra were acquired from seven different TNPs on a 6 layer WS2 flake with various degrees of detuning. The dashed black lines show the shifts of the lower polaritons (LP) and upper polaritons (UP). The vertical dashed line shows the energy of the WS2 A-exciton. The HAADF STEM images of each TNP are shown in the inset images. The scale bar is 50 nm. The grey dashed lines are the fits of the experimental EEL spectra using the coupled mode theory and the extracted plasmon-exciton coupling strength (meV) is reported beside each spectrum.

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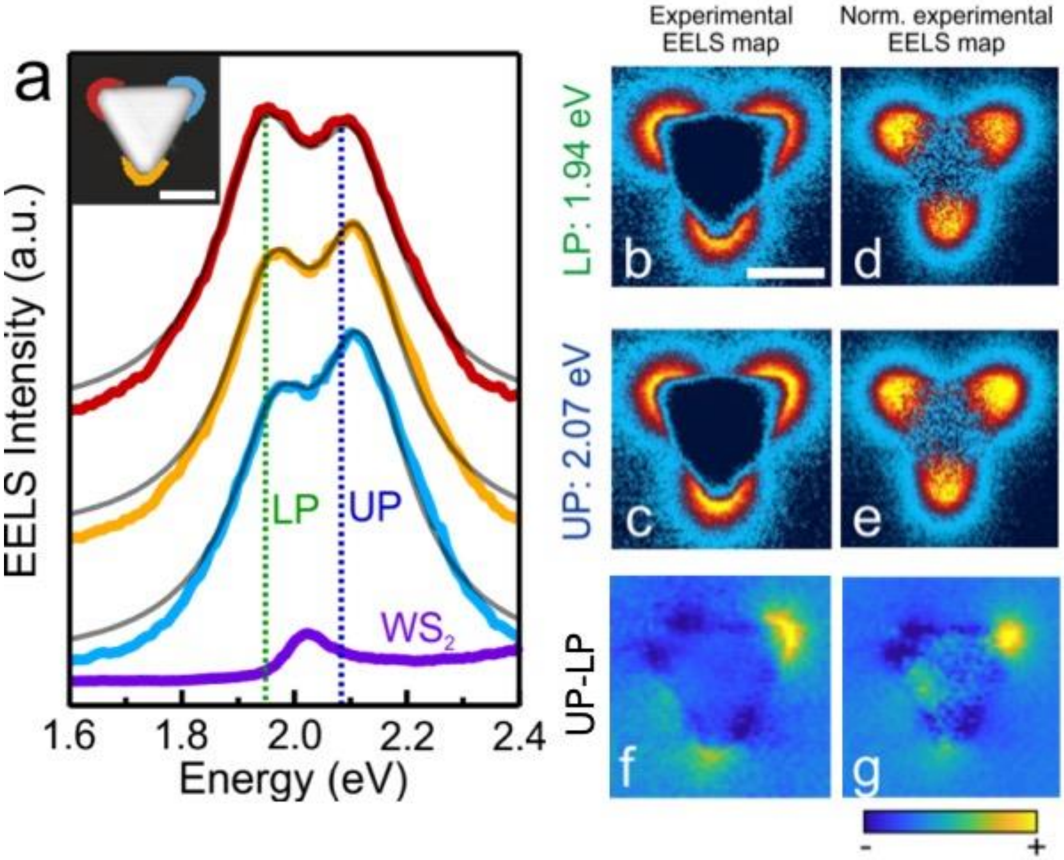

Figure 2. (a) EEL spectra extracted from a spectrum image from each corner of the coupled TNP – 6 layers WS2 system that is shown in the inset HAADF STEM image. The LP and UP peaks are marked by the vertical dotted lines at 1.94 eV and 2.07 eV, respectively. The grey lines are the fits of the experimental EEL spectra using the coupled mode theory. The bottom purple EEL spectrum is from the uncoupled WS2 flake far away from the TNP. (b) and (c) are EELS maps extracted from a spectrum image that has not been normalized to the zero-loss peak height at the LP and UP energies, respectively. (d) and (e) are EELS maps extracted from the spectrum image that has been normalized to the zero-loss peak height at the LP and UP energies, respectively. (f) and (g) are maps of the difference in intensity between the UP and LP maps (UP-LP) extracted from the not-normalized and normalized spectrum images, respectively. The scale bars in (a) and (b) are 50 nm.

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