Probing exciton-polaritons with fast electron beams

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Polaritons are compositional light-matter quasiparticles that have enabled remarkable breakthroughs in quantum and nonlinear optics, as well as in material science. Recently, plasmon-exciton polaritons (plexcitons) have been realized in hybrid material systems composed of transition metal dichalcogenide (TMDC) materials and metal nanoparticles, expanding polaritonic concepts to room temperature and nanoscale systems that also benefit from the exotic properties of TMDC materials. Despite the enormous progress in understanding TMDC-based plexcitons using optical-based methods, experimental evidence of plexcitons formation has remained indirect and mapping their nanometer-scale characteristics has remained an open challenge. Here, we demonstrate that plexcitons generated by a hybrid system composed of an individual silver nanoparticle and a few-layer WS2 flake can be spectroscopically mapped with nanometer spatial resolution using electron energy loss spectroscopy in a scanning transmission electron microscope. Experimental anticrossing measurements using the absorption dominated extinction signal provide the ultimate evidence for plexciton hybridization in the strong coupling regime. Spatially resolved EELS maps reveal the existence of unexpected nanoscale variations in the deep-subwavelength nature of plexcitons generated by this system. These findings pioneer new possibilities for in-depth studies of the local atomic structure dependence of polariton-related phenomena in TMDC hybrid material systems with nanometer spatial resolution