Exploring electronic coupling of optical and phonon excitations at the nanoscale

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A new domain for electron energy-loss spectroscopy (EELS) is now available with the recent development of monochromators in scanning transmission electron microscopy (STEM) [1]. This has allowed to study the nature of low energy (< 1 eV) excitations in materials with an unprecedented combination of spectral and spatial resolution. Two recent examples are the detection of vibrations of different carbon isotopes in amino acids [2] and the direct atomic-scale observation of how a single atomic impurity can influence the phonon response of a material [3].

Another area in which EELS can now contribute is in the understanding of mutual coupling between quasiparticles such as phonons, plasmons, or excitons. For instance, it has been recently shown that it is possible to detect, and spatially map, a strong phonon-plasmon coupling between a silver nanowire and a thin substrate of hexagonal boron nitride [4].

Similarly, recent work by Yankovich and colleagues [5] presented direct evidence of plexcitons (plasmon-exciton polaritons) in a silver nano-triangle supported on a few-layer sheet of WS₂. However, the study reported that the plexcitons had anomalies (different intensity strengths) that could not be directly explained by the experiments. Shape irregularities of the nano-triangle as well as a moiré potential, arising from the mismatch on the interface between Ag and WS₂, were suggested as possible culprits.

Here, we will present a methodology of how the spatial dependence of an exciton-plasmon and phonon-plasmon coupling can be easily determined from EELS data guided by theoretical calculations. We will also show our experimental efforts to locally study electronic states associated with moiré potentials at a 2D/3D interface between few-layer MoS₂ and an Au nanostructure [6], as shown in Figure 1. Finally, we will discuss the future experimental conditions required to study electronic excitations such as those arising from superconducting and topological materials [7, 8].
Figure 1. Averaged EEL spectrum of a Au nanostructure on top of 3-layer MoS$_2$ acquired at room temperature with an energy resolution of 20 meV, using a Nion monochromated aberration-corrected (MAC)-STEM operated at 60 kV. The inset shows a Z-contrast image of the system studied and the electron probe location where the spectrum was acquired.

References
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