Probing Low-energy Hyperbolic Polaritons In Van Der Waals Crystals With An Electron Microscope

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Only recently, specially designed instrumentation for spatially-resolved electron energy-loss spectroscopy (EELS) has been developed to substantially improve spectral resolution and operating spectral range [1]. This progress has dramatically broadened application potential of EELS in probing low-loss vibrational excitations. Pioneering experiments demonstrated capability of probing vibrational signal in organic samples [2], ionic crystals [3], and also van der Waals materials [4].

In our work [4], we theoretically and experimentally studied the very low-loss EELS of multilayer hexagonal boron nitride (h-BN), a representative van der Waals structure. The weak coupling between individual atomic layer results in extreme optical anisotropy, which gives rise to hyperbolic phonon polaritons (h-PhPs): coupled excitations of optical phonons and light with hyperbolic dispersion in the range of 90 - 200 meV [5]. H-PhPs might be a key to many emerging photonic technologies relying on nanoscale light confinement and manipulation [5,6]. Thus, efficient design and utilization of h-BN structures require spectroscopic studies with adequate spatial resolution, which can be provided by EELS utilizing electrons as localized electromagnetic probes.

To that end, we performed spatially-resolved EELS on a simple h-BN flake structure with an optimized STEM-EELS tool [7], which revealed the peak energy dependence on the h-BN thickness and on the proximity of the electron beam to the h-BN edge (Fig. 1A). Such behavior is a consequence of the polaritonic nature of the induced excitations. Indeed, with help of the classical dielectric response theory for EELS, applied to anisotropic slabs and edges, we demonstrate that the electron energy loss in h-BN is dominated by h-PhP excitation and not directly by bulk phonons as in preliminary interpretations [1]. This finding describes and quantitatively matches experimental observations (see an example of h-BN loss spectra in Fig. 1B). We thus suggest that EELS can be a technique complementary to scanning near-field optical microscopy [6] for characterization of low-energy phonon polaritons.

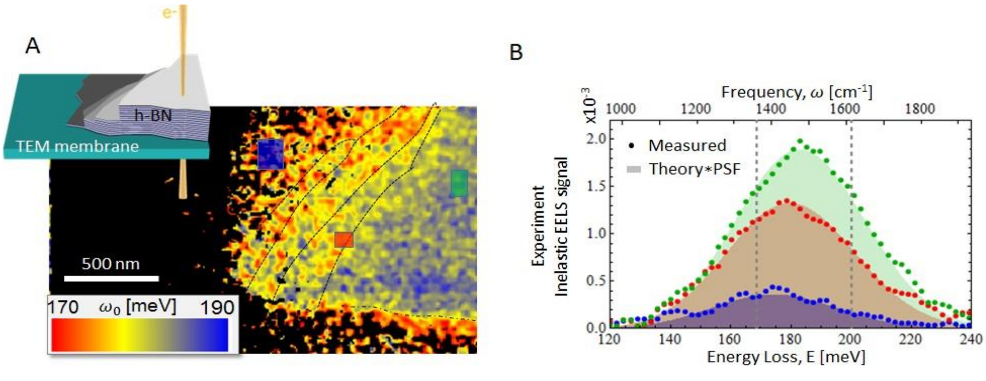

Figure: A) Peak position EELS map of a hBN flake (sample geometry is displayed in the inset showing the variation of thickness from a few monolayers to 30 nm). B) Experimental EELS spectra from different areas as marked on the map in A) are in excellent agreement with theoretical calculations obtained for the corresponding hBN thicknesses.

References

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