Probing low-energy hyperbolic polaritons in van der Waals crystals with an electron microscope

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Van der Waals materials (vdW) are a wide class of 2D systems in which individual atomic layers are only weakly bound by van der Waals interaction [1]. Much of the vdW materials functionality results from the large anisotropy in the bonding strength of atoms in the direction parallel to atomic layers and across them, and is often intimately connected with the corresponding phonons. The investigation of phononic excitation in vdW materials thus requires low mid-IR energies with nanoscale spatial resolution. Electron energy loss spectroscopy performed in scanning transmission electron microscopy (STEM-EELS) is a versatile technique capable of performing spectroscopy at the nanoscale [2]. However, conventional STEM-EELS has limited capabilities for mid-IR spectroscopy primarily due to poor monochromaticity of the primary electron beam and limited resolution of the detection system, which mask low energy excitations below 200 meV with the zero loss peak (ZLP) originating from the elastic electron scattering.

Here, we reduced the ZLP width of a conventional STEM-EELS system down to 50 meV which allowed us to probe low-energy phononic excitations (down to 100 meV) in vdW materials. Particularly, we performed a nanoscale mapping of electron energy loss (EEL) in hBN flakes, a representative vdW material. Surprisingly, we observe a variation in EEL peak position as a function of flake thickness and the edge prximity, which can not be explained by the bulk phonon excitations. Instead, our developed theory reveiles that the energy loss is dominated by the excitation of phonon polaritons, which exhibit a hyperbolic dispersion [3] owing to the layered crystal structure of hBN. The propagating nature of these hyperbolic polaritons explains the observed phenomena and suggests that STEM-EELS is highly suitable for the investigation of optical/polaritonic properties of vdW materials, with our work laying the foundation for such investigations. With ongoing developments in the monochromator and electron gun designs, we expect further improvement of STEM-EELS spectral resolution and the applicable energy ranges in near future, allowing for correlative studies of polaritons and structural properties in vdW materials.

References

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Figures

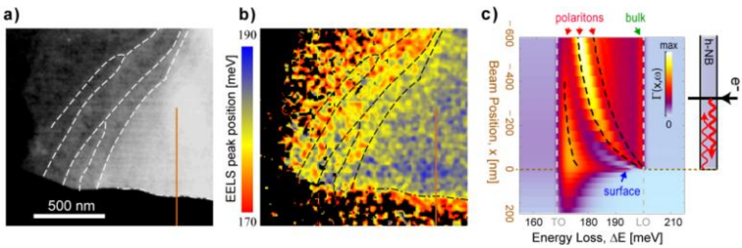

Figure 1: (a) HAADF image of hBN flake. (b) EELS peak position map. (c) Calculated EELS probability profile (along the brown line in panels a and b) reveiling the dominant role of hyperbolic polaritons.