Spatiotemporal Imaging of 2D Polariton Wavepacket Dynamics Using Free Electrons

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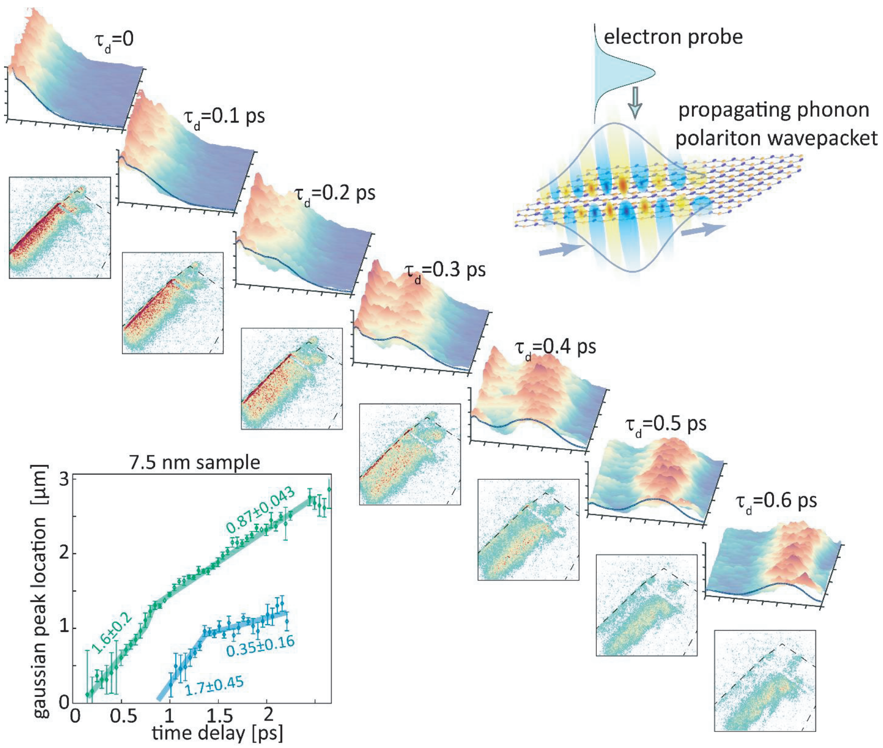
Abstract— Coherent optical excitations in van der Waals materials (2D polaritons) span a large range of optical phenomena which arise from their extraordinary dispersion relations [1]. Among the different polaritons, it is particularly interesting to examine polaritons in hyperbolic materials (hyperbolic phonon-polaritons, hPhP) which show rich physical behavior, ranging from negative refraction and subdiffraction imaging. The phononic resonance creates a dispersion relation that contains multiple branches, which were shown to be tunable by the 2D material geometry, thickness, and surrounding environment [2], reaching relatively low losses in room temperature [3].

Among the various experimental techniques that are used in the field of 2D polaritons, scanning nearfield optical microscopy (SNOM) and its variants dominates the direct nearfield imaging of 2D polaritons. Specifically, recent advances in time-resolved SNOM allowed addressing the polariton's dynamical properties as group velocity from the changes of interference of scattered polaritons with different time delays [4]. However, this interferometric technique cannot image the wavepacket dynamics, as it excludes the phase difference between different wavelengths. Different imaging techniques such as electron energy loss spectroscopy (EELS), where the 2D



polariton dispersion was recently extracted [5], obtains only time-independent information on the polaritonic modes.

Here we present how an ultrafast transmission electron microscope (UTEM), driven by femtosecond mid-infrared pulses, can probe the spatiotemporal dynamics of 2D wavepackets.

We utilize the strong light-matter interactions between free electrons and the hPhPs to measure the wavepacket's trajectory inside the sample, revealing unforeseen physical behaviors: multi-branch wavepacket splitting and wavepacket acceleration and deceleration. These phenomena are especially surprising since conventional wavepackets propagate in a fixed group velocity. We show that it is the unique dispersion relations of the hyperbolic PhPs that facilitates these phenomena, while recording group velocities between c/45 to c/850. These results examplify the wide physical spatiotemporal dynamical phenomena that can be probed through a non-destructive measurement using the unique combination of femtosecond temporal and nanometer spatial resolution of the UTEM.

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