Moiré excitons confined by twisted hBN substrates

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Abstract: We demonstrate a new approach to confining excitons in a M oSe$_2$ monolayer via the electrostatic potential from a twisted hBN substrate, which offers more flexibility in controlling exciton properties in a moiré superlattice.

1. Main Text

When two atomically thin van der Waals (vdW) layers are vertically stacked together, the atomic alignment between the layers exhibits periodical variations, leading to a new type of in-plane superlattices known as the moiré superlattices. New exciton resonances emerge in such a superlattice [1]. If the electron and hole mostly reside in different (the same) monolayers, they are referred to as interlayer (intralayer) excitons. Both intra- and interlayer excitons experience lateral quantum confinement from the moiré supercell. The supercell sizes are tunable by varying the twist angle between the two layers. Properties of moiré excitons, including optical selection rules, resonant energies, lifetimes, and diffusion, vary systematically with the twist angle. Thus, the moiré superlattices offer great opportunities for controlling excitonic properties.

There are some limitations with the existing semiconductor moiré superlattices. When the supercell sizes are reduced to enhance lateral quantum confinement, a shift between the conduction band minimal and valence band maximal occurs, introducing an indirect transition element near the K-valleys where excitons reside in the momentum space. This shift leads to a significantly longer exciton lifetime [2], which may reduce the quantum efficiency in light-emitting processes. Secondly, the conduction and valence bands exhibit complex and different energy landscapes within a moiré supercell. Thus, optically excited electrons and holes may not reside in the same location within the supercell. Furthermore, the depth of the moiré potential is not tunable.

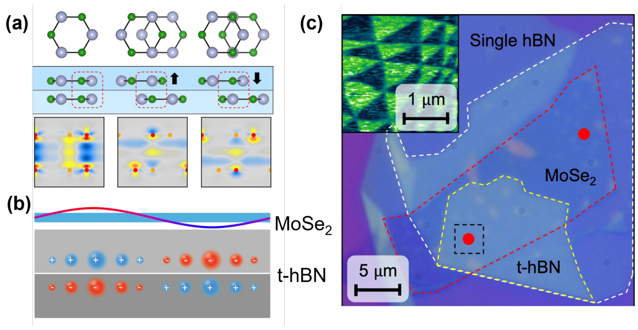

Fig. 1. (a) Three different atomic alignments at the interface of a t-hBN bilayer and calculated charge redistribution. (b) Electrostatic potential on the surface of t-hBN substrate modulates properties of an adjacent MoSe$_2$ monolayer. (c) Optical image of a stacked structure where a MoSe$_2$ monolayer overlaps with either a single hBN layer or a twisted hBN bilayer. Inset is a KPFM image of a t-hBN substrate.

Hexagonal boron nitride (hBN) as a wide-gap insulator has played an essential role in van der Waals (vdW) materials and heterostructures. In the vast majority of the studies, hBN layers act as passive layers such as atomically smooth substrates, capping layers, or ultrathin tunnel barriers, drastically improving charge carrier mobility or reducing inhomogeneous broadening of optical resonances. More recently, twisted hBN bilayers have been found to exhibit ferroelectricity in both transport and scanning probe measurements. When the B and N atoms are aligned vertically, the structural inversion symmetry is broken, and the charge redistribution can be explicitly calculated as shown in Fig. 1a. It has been proposed that the electrostatic potential on the surface of a twisted hBN (t-hBN) bilayer or a multilayer can be used to impose a universal moiré potential on an adjacent functional layer as illustrated in Fig. 1b.

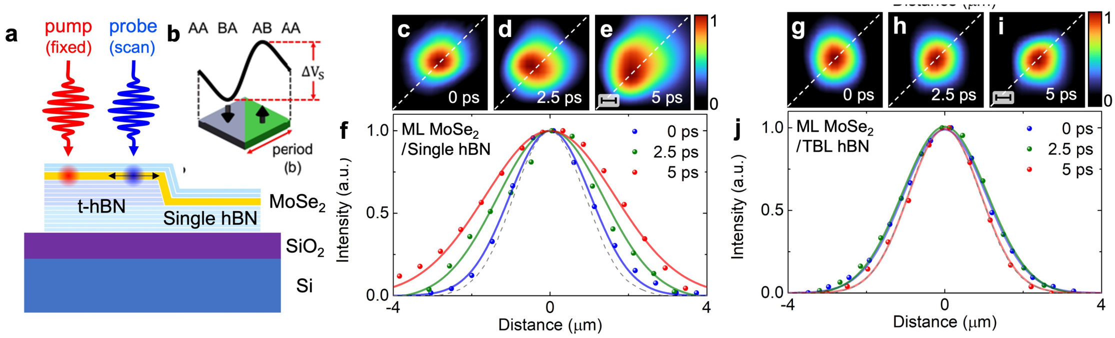

Fig. 2. (a) Illustration of the pump-probe experiment and the layered structure. (b) definition of the moiré potential depth. Spatial images of exciton diffusion in a region with (c-e) a single hBN substrate and (g-i) the t-hBN substrate at the pump-probe delay of 0, 2.5, and 5 ps. (f, j) line cuts taken along the white dashed line shown in c-e and g-i.

Here, we demonstrate that the electrostatic potential from a twisted hBN bilayer can confine excitons in an adjacent $MoSe_2$ monolayer [3]. The layered structure illustrated in Fig. 2a corresponds to the sample shown in Fig. 1c. We compare exciton diffusion in two regions of the sample with either a single hBN layer or a t-hBN bilayer with supercells of $\sim$ 700 nm as marked by two red dots. The moiré supercells formed in the t-hBN bilayer region are visualized by frequency modulation Kelvin probe force microscopy (KPFM) measurements as shown in the inset of Fig. 1c. The moiré potential depth varies with the supercell size and top hBN layer thickness. For the t-hBN region where optical measurements are taken, the potential depth ΔV_s defined in Fig. 2b exceeds $\sim$ 150 meV.

The spatial images of exciton diffusion taken from the $MoSe_2$/single hBN region are taken with spatially-resolved pump-probe technique as illustrated in Fig. 2a. The wavelength of the laser pulses are tuned to be resonant with the A exciton in the $MoSe_2$ monolayer. A series of images are taken with increasing delay between the two pulses in Fig. 2c-e while the line cuts (white dashed lines) from each image are shown in Fig. 2f. With increasing pump and probe delay, exciton diffusion beyond the excitation laser spot (black dashed curve) is clearly observed in the location where there is only a single layer of hBN as the substrate. In contrast, no exciton diffusion is observable from the $MoSe_2$/t-hBN region in spatial images (Fig. 2g-j) and line cuts in Fig. 2j. In many previous studies of TMD monolayers encapsulated by hBN, exciton diffusion was observed. These experiments demonstrate that electrostatic potential from the t-hBN substrate functions differently from encapsulation layers and effectively impedes exciton diffusion in the $MoSe_2$ monolayer. In our current experiments, the supercell size of the t-hBN bilayer is much larger than the exciton Borh radius. Thus, moiré potential imposes weak lateral confinement. Future experiments based on near-field optical techniques with higher spatial resolutions would be necessary to elucidate the localization mechanism.

References

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