Locally probing interlayer states in van der Waals materials

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We study van der Waals (vdW) systems using low-energy electron microscopy (LEEM) and spectroscopy. Specifically, reflection vs. energy, i.e. R(E) spectra, with 0<E<50 eV, give us direct information on the local layer structure with ~2 nm resolution. For graphene and hBN an (unoccupied) interlayer state (or rather: 2D-band) is added with each additional layer. These interlayer states hybridize in a tight-binding manner, such that the number of states scales with the number of layers (See Fig. 1). If an incoming electron is resonant with a state, it will be transmitted, leading to a minimum in R(E). Hence, the local R(E) curve gives direct information on the number of layers and their stacking [1-3]. We take this concept one step further by studying the 2D-dispersion relations of these unoccupied bands (Fig. 1). For that, we have developed 'angle-resolved reflected-electron spectroscopy' (ARRES) [2,3]. Interestingly, whereas at resonance reflection minima are expected, maxima are anticipated in transmission. To test this, we created eV-TEM, i.e. transmission EM at very low energies (0-100 eV) [1]. Indeed, the transmission vs. energy T(E) curves for freestanding graphene show maxima at the interlayer state energies. Moreover, the combination of T(E) and R(E) allows us to study inelastic path lengths for electrons of these energies [1].
Summarizing, we are able to measure the unoccupied band structure of vdW materials (above E_{vac}) directly. Since such data are obtained on the nanoscale, our experiments allow for extremely accurate sample (quality) characterization, a necessity when studying the intriguing physics of vdW systems such as twisted bilayer graphene [4].

References

[1] D. Geelen et al. PRL 123, 086802 (2019).
[2] J. Jobst et al., Nat. Comm. 6, 8926 (2015)
[3] J. Jobst et al., Nat. Comm. 7, 13621 (2016)
[4] Lisi et al., Nature Phys. 17, 189 (2021)

Figure

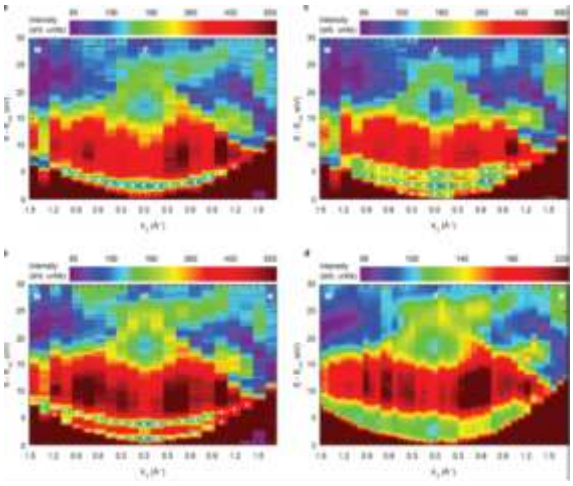

Figure 1: ARRES maps of multilayer graphene taken on 10x10 nm^2 areas. The number of inter-layer states, showing parabolic dispersion, increases with thickness. a) 2 layers, b) 3 layers, c) 4 layers, d) bulk (graphite). The spectra can roughly be understood using a simple tight binding model. [2]