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Local probing of 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 $\alpha E < 50$ 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].

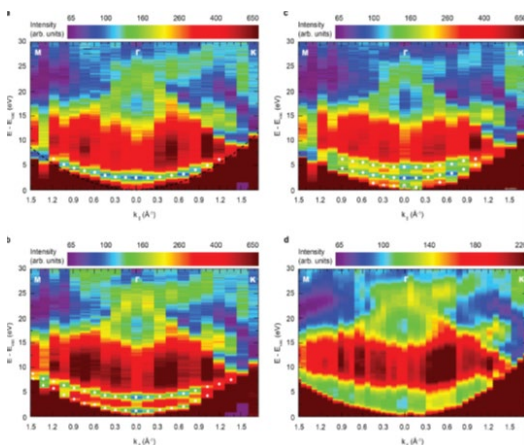


Figure 1: ARRES maps of multilayer graphene taken on 10×10 nm² areas. The number of inter-layer states, showing parabolic dispersion, increases with thickness. a) 2 layers, b) 3 layers, c) 4 layers, d) bulk (graphite). The spectra can roughly be understood using a simple tight binding model. [2]

- [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)