PbTe Nanocrystal Arrays on Graphene and the Structural Influence of Capping Ligands - Supplementary Information

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S1. Residual Organic Contamination

Figure S1a demonstrates the low levels of organic contamination deposited on the graphene support, with the figure showing the edge of a PbTe monolayer close-packed array in the top left of the image and clean graphene identifiable as the uniform grey area in the rest of the image field. Areas of residual organic contamination are imaged as darker grey regions, often with a dark border. It can be seen that a large fraction of the sample is pristine graphene, with contamination limited to small areas of thin mono- to few-layers of amorphous material. We have found that depositing nanocrystals on to pre-cleaned graphene was a crucial preparation step, as it permitted the unambiguous resolution of the nanocrystal edge structure. Extended imaging of nanocrystals can result in the migration of metal atoms across the sample, as can be seen in Figure S1b. On the graphene adjacent to some nanocrystals denser and thicker amorphous carbon residue could be seen, as shown in Figure S1c and d. The location of these denser patches of contamination next to the nanocrystals, and the prevalence of darker spots of metals atoms within them, suggests that they are comprised of excess ligand that was not rinsed away.

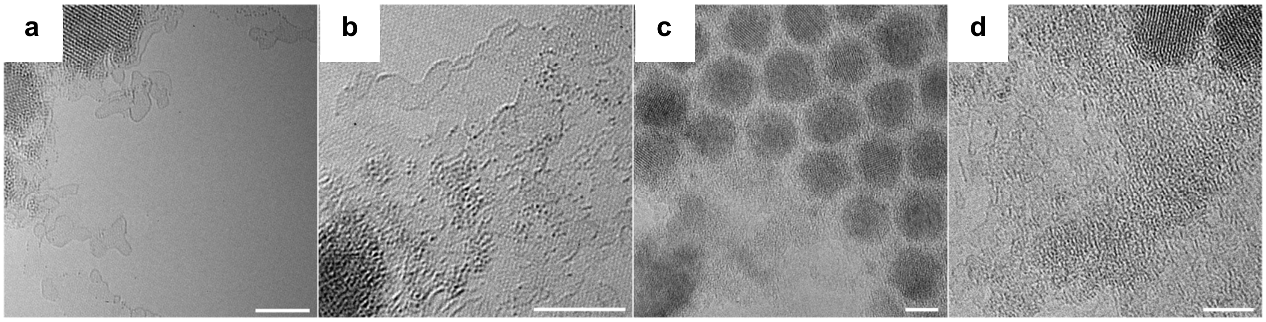

Figure S1. (a) TEM image of the boundary of the nanocrystal superlattice. The uniform light grey area is graphene and the nanocrystals are located in the top left. The marginally darker grey areas located on the graphene are regions of thin amorphous carbon residue. (b) The area near to a nanocrystal after extended imaging. The dark contrast dots are metal atoms. (c) The edge of a nanocrystal array, showing thick and dense amorphous residue on the graphene. (d) High magnification image showing the denser residue. All scale bars are 5 nm.

S2. Few-Layer PbTe Superlattice

Closer examination of the few-layer stacked regions confirms the stacking sequence as hexagonal close packing (ABA), rather than cubic packing (ABC). Figure S2a shows an AC-TEM image of a region confirmed from the intensity profile analysis in Figure 2i as tri-layer. It is possible to discern circular nanocrystals with varying contrast, some of which contain lattice information. These regions are annotated in Figure S2b, with the discrepancy in contrast between different nanocrystals attributed to their occupancy of different layers. The stronger contrast seen in nanocrystals highlighted by yellow circles arises from their location closer to the optimal focal plane and the additional effect of the aligned third layer, supporting HCP stacking. Analysis of the moiré pattern in the white dotted region in Figure S2b further confirms the presence of a third layer in an ABA stacking sequence. Figure S2c shows this analysis of the moiré pattern, the existence of which confirms that the third layer is aligned with the first, consistent with an hcp stacking sequence.

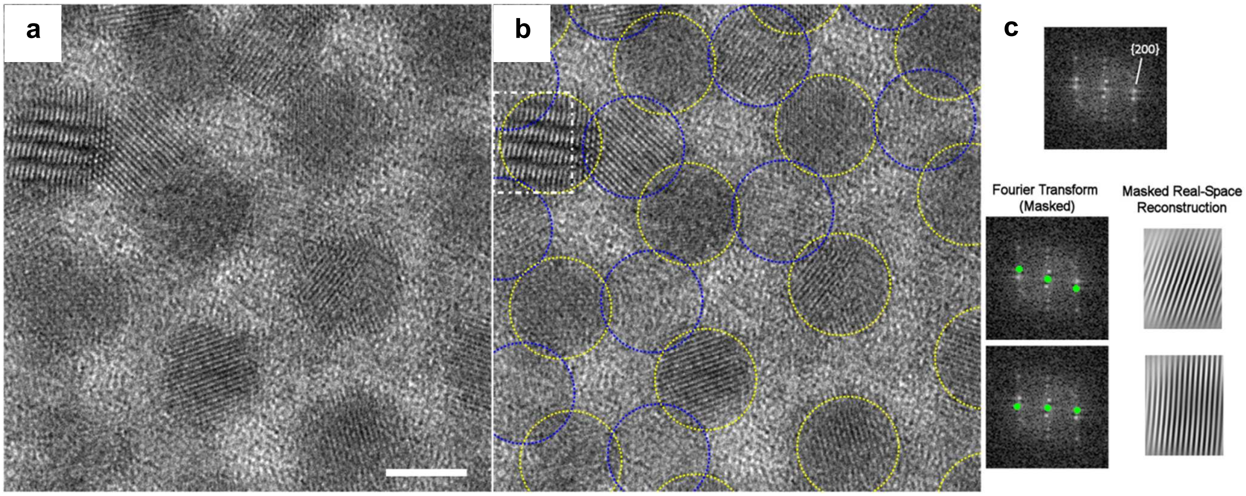

Figure S2. (a) High magnification image of a tri-layer region of the superlattice. (b) Annotated version of the image from panel (a) illustrating the AB hcp stacking. Yellow circles denote nanocrystals belonging to one layer and blue circles show nanocrystals of fainter contrast that belong to a second layer. (c) Power spectrum calculated from the image within the white dashed region in panel (b). The reflection corresponding to the {200} planes is indicated. Taking positive masks (green) of the two {200} spots it is possible to generate real-space reconstructions that demonstrate the corresponding lattice directions which constitute the moiré pattern. Scale bar is 5 nm.