## Strain-Originated Flat Band-Induced Integer Charging and the associated Phase Transitions in Double-Aligned Monolayer Graphene

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Van der Waals crystals can slide, rotate and reconstruct relative to each other adjusting themselves to minimize the total energy arising from the van der Waals interactions, elastic energies and Coulomb forces. Therefore, in complex heterostructures with multiple aligned interfaces – multiple competing structural configurations, phase transitions between them should be observable by varying the balance between different energy contributions. Here we demonstrate, for the first time, that in a system of monolayer graphene, double aligned to two hexagonal boron nitride (hBN) crystals, one of which is a monolayer hBN<sup>1</sup> – a structural phase transition (whose change of crystalline symmetry is visualized by STM) could be triggered by electrical charging of graphene, or by magnetic field or temperature. Most importantly, such structural phase transition results in a novel electronic phase transition, manifesting as a completely novel quantum phenomenon: precise integer doping of graphene, which demonstrates doping with quantized integer number of electrons (or holes) per moiré plaquette. This anomalous integer doping is attributed to the charge-trapping in localized, strain-induced pseudo-Landau levels resulting from moire reconstruction enhanced by monolayer hBN(which corresponds to the formation of the flat bands in the kspace). The ability to engineer structural and electronic phase transitions opens up a new frontier in designing novel electronic devices, controlling the formation of strain-induced flat band and exploring strain-Coulomb correlations in Van der Waals heterostructures.

## References

[1] Z. Wang et al, Composite super-moiré lattices in double-aligned graphene heterostructures. Science Advances 5, eaay8897



**Figure 1: (A)** Schematic of the mhBN/Gr/hBN transport device. Inset: The FFT (Fast Fourier transformation) of the AFM image confirms the double alignment (**B**) Longitudinal resistance  $R_{xx}$  of the states before (blue) and after (red) the transition. The neutrality point, labelled "NP," shifts from p-doping to n-doping upon transition. The top axis marks doping levels corresponding to integer multiples of electrons or holes per moiré unit cell. (**c**) The doping levels of other same-structured devices with different periods of **moiré** plaquette  $\lambda_m$  before (blue dots) and after (red dots) transition.

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