Quantum Phases of Dipolar Interlayer Excitons in Tri- and Quad-Layered Atomically Thin Heterostructures

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Dipolar excitons in multilayered heterostructures provide a promising platform for studying strongly correlated quantum phases [1, 2, 4, 3, 5, 6, 7, 8]. Beyond purely planar configurations, multilayer arrangements introduce a new layer degree of freedom and highlight the anisotropic nature of dipolar interactions.

Motivated by recent experimental advances, we investigate dipolar excitons in trilayer and quadrilayer heterostructures using large-scale quantum Monte Carlo simulations. We establish the low-temperature phase diagram as a function of key physical parameters and make concrete predictions for experimentally relevant observables.

In trilayer systems, we identify the emergence of a quantum quadrupolar exciton liquid, driven by rapid hole tunneling between the two outer layers. Consistent with experimental findings, our simulations reveal a quadratic redshift in exciton energies under an applied polarizing electric field. Remarkably, at high exciton densities and mass, we observe a new, staggered crystal phase, stabilized by the attractive interaction of antiparallel dipoles.

For quadrilayer heterostructures, where dipoles in the top and bottom layers are oriented antiparallel, we demonstrate the formation of a trimer bound state, consisting of two dipoles in the lower layers and one in the upper layer. At sufficiently low temperatures, these trimers condense, giving rise to a trimer superfluid. Increasing exciton density induces a quantum phase transition where in addition unbound dipoles condense.

Our results provide theoretical insights into the complex phase diagrams of multilayered dipolar excitons with anisotropic extended interactions, offer an interpretation of existing experimental data, and guide future investigations in this field.

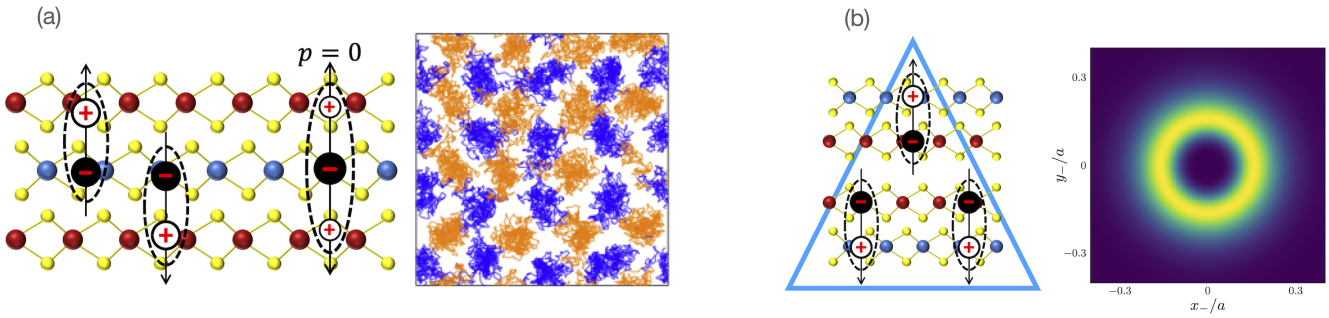

Figure 1: (a) Quantum quadrupolar versus staggered dipole configuration and Monte Carlo snapshots of dipolar world lines arranged in a dipolar staggered lattice (b) Trimer bound state configuration and spatial wave function probability density.

References

[1] Colin Hubert et al. "Attractive dipolar coupling between stacked exciton fluids". In: *Physical Review X* 9.2 (2019), p. 021026.

[2] Yevgeny Slobodkin et al. "Quantum Phase Transitions of Trilayer Excitons in Atomically Thin Heterostructures". In: *Physical Review Letters* 125.25 (Dec. 2020). ISSN: 1079-7114.

[3] D. J. Choksy et al. "Attractive and repulsive dipolar interaction in bilayers of indirect excitons". In: *Physical Review B* 103.4 (Jan. 2021). ISSN: 2469-9969.

[4] Michal Zimmerman, Ronen Rapaport, and Snir Gazit. "Collective interlayer pairing and pair superfluidity in vertically stacked layers of dipolar excitons". In: *Proceedings of the National Academy of Sciences* 119.30 (July 2022). ISSN: 1091-6490.

[5] Tian Li et al. "Emergent ferroelectricity in moiré heterostructures". In: *Nature Materials* 22.10 (2023), pp. 1102–1108.

[6] Zhen Lian et al. "Quadrupolar excitons and hybridized interlayer Mott insulator in a trilayer moiré superlattice". In: *Nature Communications* 14.1 (2023), p. 4604.

[7] Wei Zhang et al. "Observation of Topological Photonic States in an Array of Coupled Resonators". In: *Physical Review Letters* 131.18 (2023), p. 186901.

[8] Cheng-Cheng Liu, Wanxiang Feng, and Yugui Yao. "Spin-orbit coupling induced topological phase transition in silicene". In: *Physical Review B* 110.20 (2024), p. L201402.

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