# Engineering spin configurations in graphene architectures

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Abstract

Graphene nanostructures with custom-designed shapes can exhibit unconventional paramagnetism due to open-shell states in their conjugated π-electron lattice [1]. A key challenge in utilizing nanographene as quantum registers, e.g. collections of qubits, is the localization of highly entangled spins with tunable interactions.
In this work, we present an on-surface synthesis strategy to engineer highly tunable spin states in nanographenes by combining structural design with heteroatom doping. Beginning with an aza-triangulene unit [2], we construct more complex architectures (Fig. 1) [3, 4] that showcase a broad spectrum of spin properties, ranging from high-spin states to topological polyradicalism. The incorporation of a nitrogen heteroatom adds one additional electron to the π-conjugated lattice, disrupting the half-filled scenario predicted by Lieb's theorem [5] and introducing a new parameter for tuning spin states.
We demonstrate that molecular orbital filling plays a critical role in understanding the spin properties of these aza-nanographene platforms. To explore this, we combine low-temperature scanning probe microscopy with quantum chemistry simulations, revealing the nature of the unpaired electrons and their interactions within each nanostructure. Our findings identify promising candidates for a three-qubit register with long coherence times [1], which may be operable via ESR-STM.
We also have worked out strategies to tune spin and charge states by working of substrates of different electronegativity and magnetic configuration, such as $GdAu_2$ and MgO. In this presentation I will also review several results from our group and collaborators that allow us to tune the charge states of the nanographenes and, hence, their spin configuration.

References

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Figures

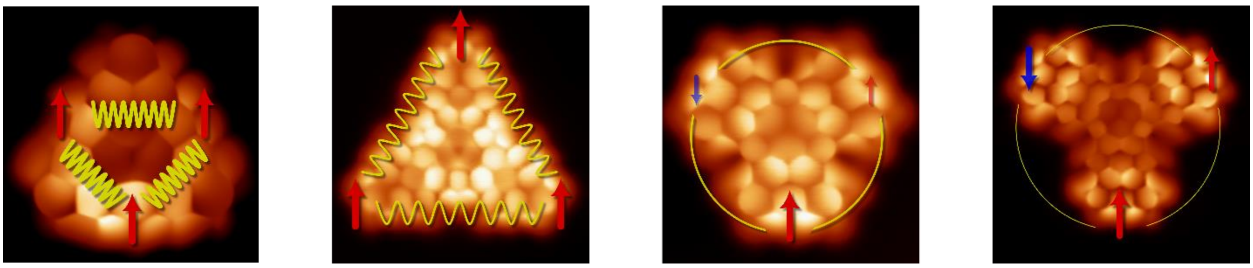

**Figure 1:** Bond-resolved STM images of four aza-nanographene platforms hosting unpaired spins of different nature and interactions.