Control of the local magnetic states in graphene with voltage and gating

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ABSTRACT SUBMISSION

Magnetism of graphene can be created by atomic defects, either hydrogen adsorption or single-carbon vacancy formation, owing to the unpaired π electrons around the defects. Based on rigorous first principles calculations, we explore the possibility of voltage manipulation of two such types of π magnetism in graphene via a scanning tunneling microscope tip. For the hydrogen, the magnetic moment can be switched on and off with voltage-induced doping, whereas, for the carbon vacancy, the spin splitting of the π bands persists, almost independent of the extent of doping, due to the coupling between the π and the σ bonds. Furthermore, the local atomic structures near the vacancy can be reversibly manipulated by a coordination mechanism between an intermediate tip-defect distance and a moderate tip voltage, consequently leading to the reversal of spin polarization of the π bands. Voltage control of the local magnetic states may open a new avenue for potential applications in spintronics.

INTRODUCTION

Voltage control of magnetism at the atomic scale is one of the grand challenges in spintronics and has the potential to bring about revolutionary new spintronic devices down to subnanometer scale[1]. It is known that graphene can become magnetic by hydrogen adsorption[2], or vacancy formation [3], which introduces defect states in the π band through its unpaired p_z orbitals. These defects states are localized in space and show preference to one sublattice, according to Lieb's theorem, and can offer a unique candidate for atomic-scale magnetic operation and qubits.

MODEL

The SIESTA/TRANSSIESTA code was employed for the spin polarized calculations. All the calculations were carried out with a low electronic temperature of 50 K. Physical quantities like density of states, transmission, current, and spin density were extracted by using SISL.

SAMPLE SECTION

Figure 1 shows the transport setup for one H atom adsorbate on graphene. The total magnetic moment is 1 and 0 μ_B at 0.0 and 0.4 V with $g = 0$ gating, respectively. This means the spin states of such system can be switched on and off with voltage-induced doping. Moreover, the calculated current is around 0.016 μA at 0.4 V, which implies the mechanism here is governed by the tip-induced field or potential shift with little current effect. The tip acts as a local electrostatic gate, akin to local doping or gating and tunes the energy of the localized defect state.

Figure 2 shows the transport properties of graphene with single-carbon vacancy. At zero bias, three magnetic states, HS, LS, and the non-magnetic state (NS), can be realized as the Au tip-CV distance. Interestingly, a spin transition from the LS to HS states occurs at a bias of −0.4 V, corresponding to the atomic geometry changing from a nonplanar structure to a nearly flat structure. Moreover, the charge-induced forces on the carbon atoms close to the vacancy suggest that it remains an out-of-plane structure at 0.4 V and favors a planar structure at −0.4 V, as shown in Fig. 4(c).

CONCLUSION

In conclusion, we have performed first-principles DFT- non-equilibrium Green's function calculations to study the manipulation of the

localized π magnetism at two types of defects in graphene by using a STM tip. It is found that the magnetism of the π states strongly depend on both the tip voltage and the tip-defect distance. Our findings support the prospect of the local control of graphene magnetism on the atomic scale. We believe that it is versatile and could find potential applications in spintronics and magnetic sensing.

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REFERENCES

[1] W. Han, R. K. Kawakami, M. Gmitra, and J. Fabian, *Graphene spintronics*, Nat. Nanotechnol. **9**, 794 (2014).

[2] H. González-Herrero *et al.*, *Atomic-scale control of graphene magnetism by using hydrogen atoms*, Science **352**, 437 (2016).

[3] Y. Zhang *et al.*, *Scanning Tunneling Microscopy of the π Magnetism of a Single Carbon Vacancy in Graphene*, Rev. Lett. **117**, 166801 (2016).

[4] F. Gao *et al.*, *Control of the local magnetic states in graphene with voltage and gating*, Phys. Rev. B **103**, L241402 (2021).

[5] Y. Zhang *et al.*, *Characterization and Manipulation of Intervalley Scattering Induced by an Individual Monovacancy in Graphene*, Phys. Rev. Lett. **129,** 096402 (2022).

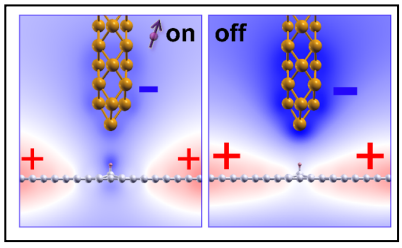

Fig. **1**. Hydrogen adsorbate on graphene. Electrostatic potential profile at 0.0 and 0.4 V with g = 0 gating.

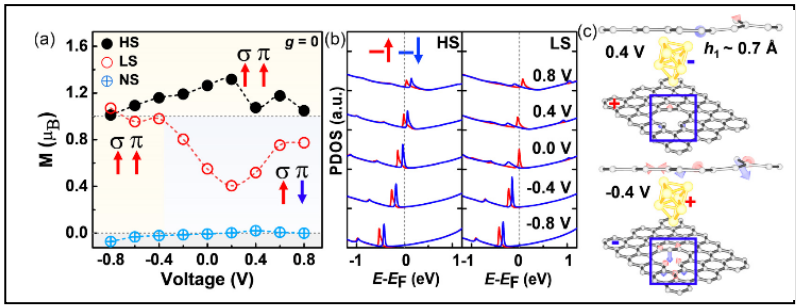

Fig. 2. Carbon vacancy. (a) The magnetic moment as a function of applied voltage for the three different vacancy spin states and conformations, HS, LS, and NS, respectively. (b) The density of states projected on the p_z orbitals. The red and blue lines represent the spin-up and spin-down states. (c) Charge-induced force in the LS state at ±0.4 V. The absolute force on the C atom is 0.24 nN at −0.4 V.