Edge Magnetism and Stability of Edge States in Graphene Nanoribbons

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Abstract— We critically discuss the stability of edge states and edge magnetism in zigzag edge graphene nanoribbons (ZGNRs). We point out that magnetic edge states might not exist in real systems and show that there are at least three very natural mechanisms -edge reconstruction, edge passivation, and edge closure- which dramatically reduce the effect of edge states in ZGNRs or even totally eliminate them. Even if systems with magnetic edge states could be made, the intrinsic magnetism would not be stable at room temperature. Charge doping and the presence of edge defects further destabilize the intrinsic magnetism of such systems.

Since the first successful isolation of mono- and multilayers of graphene [1], this material has raised tremendous attention within the scientific community. This is mainly due to the fact that graphene is an almost perfect two-dimensional system with unique electronic properties that might ultimately lead to a new generation of nanoelectronic devices [2,3].

Graphene nanoribbons (GNRs) are one-dimensional stripes "cut" from graphene, which are a few nanometers in width and quasi-infinite in the perpendicular direction. Initially GNRs were primarily discussed in the theoretical literature [4,5], but meanwhile GNRs can be fabricated and studied in the laboratory [6-9]. GNRs with zigzag edges (ZGNRs) are of particular interest, because they are forming edge states [4,5]. The latter are localized electronic states that decay exponentially toward the center of the ribbon [2]. The decay lengths are in the range of a few nanometers [10]. Such edge states do not only exist in perfect ZGNRs but in any graphene system that has zigzag edge segments [11].The localized nature of the edge states in ZGNRs gives rise to flats bands (i.e., parts of bands with little dispersion) in the electronic band structure and thus to a pronounced peak in the electronic density of states (DOS) right at the Fermi level (E_F). This peak has been measured in the local DOS of monoatomic zigzag step edges of graphite by scanning tunneling microscopy [10,12,13]. In those measurements the peak in the DOS did not appear right at E_F but very close to it. However, no such measurements for a monolayer of graphene or GNRs are known up to date.

According to numerous theoretical studies based on density functional theory or on the mean-field Hubbard model [4,14], it is believed that the peak in the DOS at E_F gives rise to a magnetic instability, where the edge states become spin polarized. In the magnetic ground state a band gap at E_F is opened, and the atoms are ferromagnetically ordered along one edge and antiferromagnetically ordered between opposite edges. Potential applications of edge magnetism for spintronics are widely discussed. But up to now, the edge magnetism in ZGNRswas never directly observed in local probe microscopy. Only an indirect observation has been reported quite recently [15].

In this article we critically discuss the present state of research on graphene edge states and edge magnetism. We point out that magnetic edge states might not exist in real systems. And even if they do, the intrinsic magnetism would be so weak, that realistic applications in spintronic devices will not be feasible. In order to use a physical effect in a practical device, the effect must be (i) a strong intrinsic effect and (ii) it must be stable at room temperature. We show that graphene edge magnetism does not fulfill these two criteria: (i) the effect is easily destroyed by a multitude of mechanisms and (ii) it is not stable at room temperature, even in perfect samples (see Figure below).

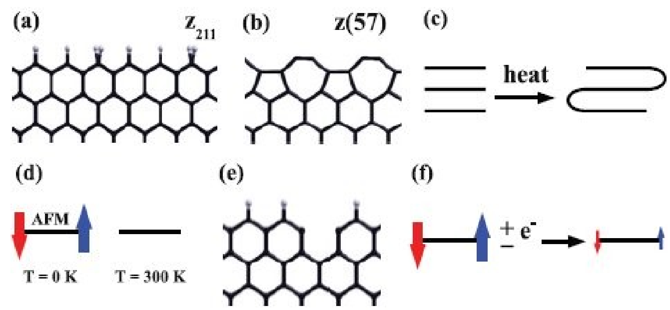

Figure: Graphical outline of this article. (Top) Three mechanisms that strongly affect the edge states in ZGNRs: (a) the atomic structure of the z211 passivated edge, (b) the atomic structure of the zz(57) reconstructed edge, and (c) edge closure. (Bottom) The edge magnetism in (d) ideal ZGNRs is not stable at room temperature. The low-temperature magnetism is destabilized (or even totally disappears) whenever the system has (e) edge defects or (f) is charge doped.

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