To U or not to U – that is the GNR question

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Abstract

The tight-binding (TB) description of bulk graphene uses only first nearest-neighbour hopping (*i.e.,* a simple tight-binding (STB) model) to calculate its electronic properties [1]. Unlike the bulk, the TB minimal model description of graphene nanoribbons (GNRs) requires extended hopping terms, up to third nearest-neighbour for armchair [2], and with mean-field Hubbard-U interaction for zigzag GNRs [3-6]. These terms are necessary in the TB model to reproduce the semi-conductor properties of GNRs, as seen in both density functional theory (DFT) simulations [7], as well as experiment [8]. In addition, the mean-field Hubbard-U is also required for ZGNRs to produce the DFT-predicted magnetism [7].

For mixed-edge GNRs (which are experimentally more realistic), a generalised TB (GTB) model has been proposed, which includes up to third nearest-neighbour hopping with Hubbard-U and uses a single parameter set to accurately reproduce the low-energy band structure and band gaps obtained from local spin-density functional theory GNR results [9]. The GTB model has an advantage in that it is computationally efficient against DFT, exact diagonalisation, CI and other methods for calculating reduced symmetry systems with large unit cells that are more relevant to experiment (*i.e.,* with realistic defects and patterning).

In this presentation, we provide several examples of why 'to U' is an essential question. Specifically, we demonstrate the importance of the GTB model for the study of magnetism in GNRs. With the computational efficiency of the GTB model, we have been able to understand the effect of random edge-vacancies, as well as random edge-disorder. In the systematic edge-vacancy study of Huang *et al.*, it was shown that at ~33% edge-vacancy concentration, ZGNRs become non-magnetic [10]. This is, in fact, contrary to our random edge-vacancy results, which are beyond the scope of DFT, and show that the spin-polarisation does not disappear irrespective of the random edge-vacancy concentration (Fig. 1). For random edge-vacancy and edge-disordered ZGNRs, we have also investigated the interplay between Anderson localisation and the Hubbard-U within the GTB model applied to coherent transport studies. Contrary to other research, which uses the STB description to model disordered ZGNRs [11], we demonstrate that Hubbard-U effects remain important and that these effects cannot be ignored. We have also been able to demonstrate spintronics and spin-filtering by applying the GTB model to patterned ZGNRs both with and without strain, thereby using the U in GNR device design (Fig. 2) [12].

'To U' is therefore essential, particularly in the recent wake of experimental evidence, which shows signatures of magnetism in scanning tunneling spectroscopy results of chiral GNRs [12]. We would like to introduce the formalism behind this computational efficient model, and provide evidence to convince more theoreticians to take up the U when studying graphene on the nanoscale.

References

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Figures

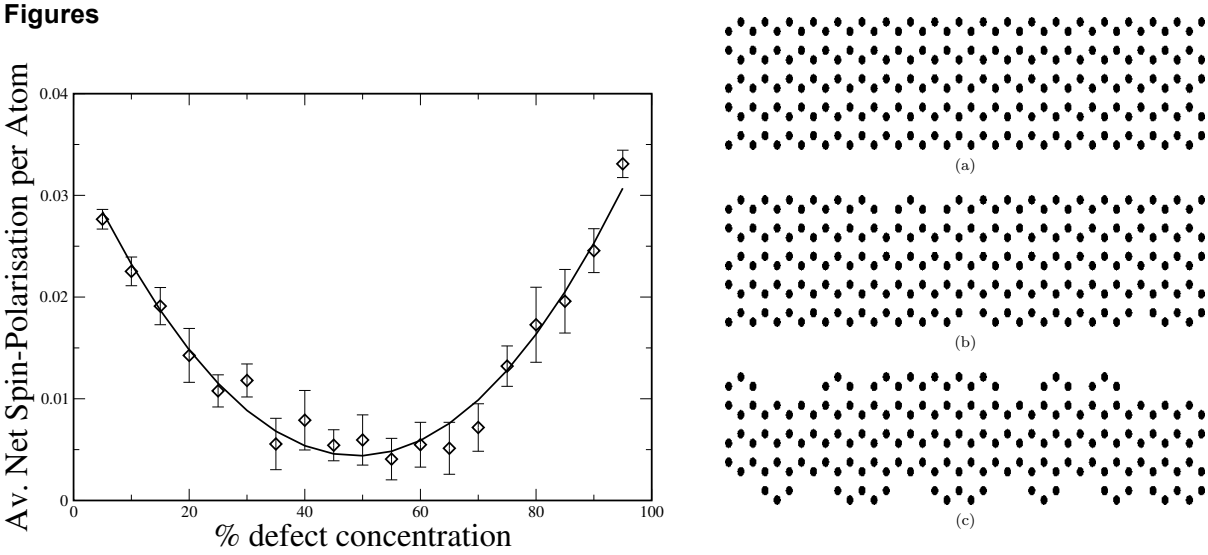

Fig. 1 Average net spin-polarisation per atom as a function of the % defect concentration (ensemble averaged) for a 5-ZGNR ribbon of length 49.2 Å and width 9.24 Å shown on the right with (a) zero, (b) 5% and (c) 41.3% random edge-vacancy defect concentrations calculated using the GTB model. In comparison, an ideal 5-ZGNR has an average net spin-polarisation per atom of 0.033. *N.b.,* Huang *et al*.'s systematic defected system has a total loss of spin-polarisation at 33.3% edge-vacancy defects [10].

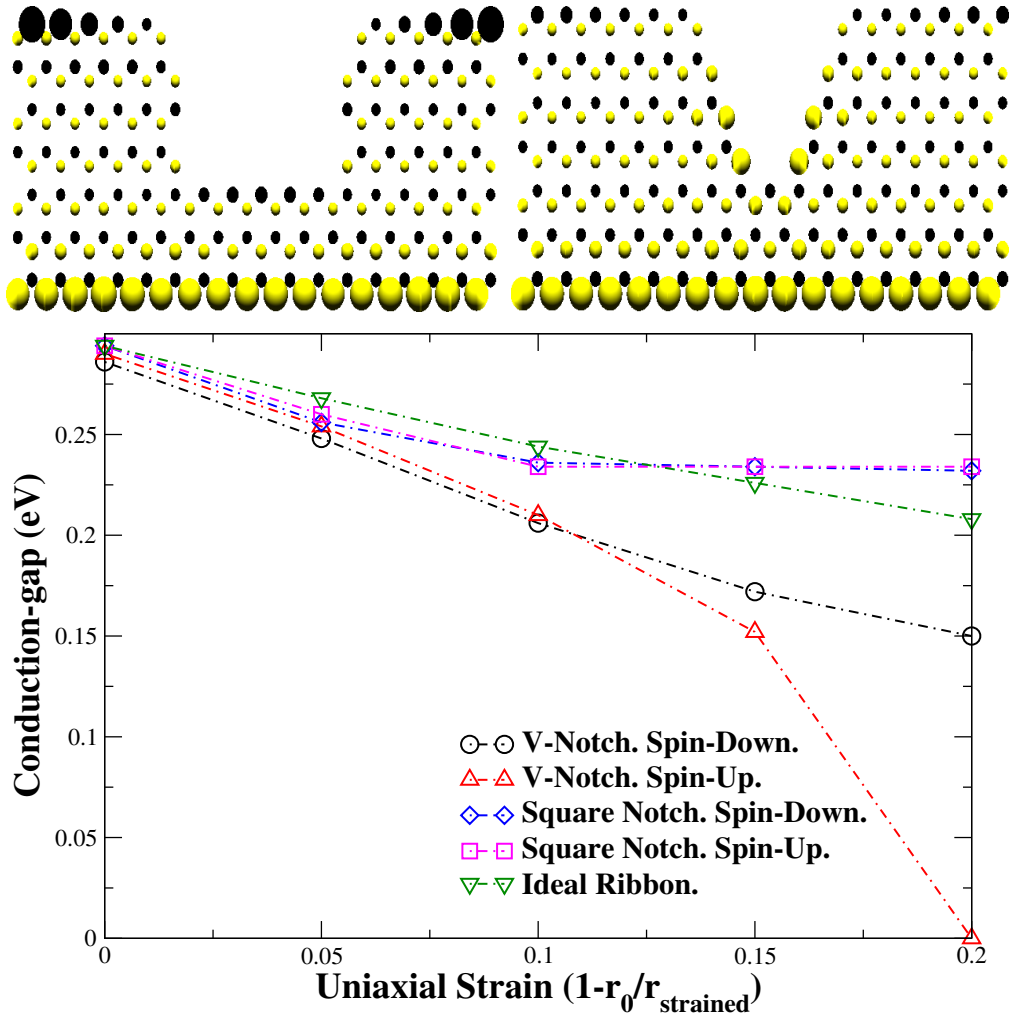

Fig. 2 Effect of uniaxial strain in the *y*-direction on the spin-dependent conduction-gap for ideal, square (left top) and V-shaped notch (right top) devices calculated using the GTB model within the coherent transport formalism. In the V-shaped notch system there occurs a spin-dependent result, which at 20% strain leads to spin-filtering [11].