Ferromagnetism in Graphene due to Heteroatom Doping: Insights from Experiment and Electronic-structure Calculations

Piotr Blonski[a]
Jiri Tucek,[a] Zdenek Sofer,[b] Petr Simek,[b] Martin Petr,[a] Martin Pumera,[c] Michal Otyepka,[a] and Radek Zboril*[a]

a) Regional Centre of Advanced Technologies and Materials, Department of Physical Chemistry, Faculty of Science, Palacky University, 17. listopadu 1192/12, 771 46 Olomouc, Czech Republic.
b) Department of Inorganic Chemistry, University of Chemistry and Technology Prague, Technicka 5, 166 28 Prague 6, Czech Republic.
c) Division of Chemistry & Biological Chemistry, School of Physical and Mathematical Sciences, Nanyang Technological University, 637371 Singapore.

piotr.blonski@upol.cz

Heteroatom doping of graphene by light elements opens a doorway to tailor electronic properties of this material, and, hence, its physical properties, meeting thus the requirements of a given application. Recently, it has been proved[1] that doping of graphene lattice with non-carbon atoms offers a promising approach for imprinting the magnetic order into graphene. Graphene doped with 4.2 at% of sulfur shows strong ferromagnetic properties below ≈62 K with a saturation magnetization reaching ≈5.5 emu g^{-1}, which is among the highest values reported so far for any graphene-based system or even carbon-based nanomaterials. Electronic-structure calculations indicated that in a narrow concentration window (4-6 at%) magnetic interactions mediated by the π-electron system emerges between the substitution-generated paramagnetic centers leading to a ferromagnetic order. The theory showed that the gamma-thiothiapyrone motif of sulfur in the graphene lattice, as magnetically active configurations, is responsible for observed magnetic properties at low temperatures. Furthermore, the theory predicted that the observed magnetic behavior is solely due to a substitution mechanism, as the addition of sulfur atoms on-top of the graphene sheet cannot promote a self-sustainable magnetism. The possibility of imprinting magnetic order in graphene by another *n*-type dopant, with a complex interplay between the concentration and configuration of the dopant atoms in the host lattice, will be also discussed. The heteroatom *n*-type doping of graphene offers viable highly conductive material with a strong potential for spintronic applications.

References

[1] J. Tucek, P. Blonski, Z. Sofer, P. Simek, M. Petr, M. Pumera, M. Otyepka, and R. Zboril, Adv. Mater. 28 (2016) 5045