Possibility of application of two-domain model for graphenic materials with high electrical conductivity

Oleksiy V. Khavryuchenko[1], Maria Augustyniak-Jablokow[2], Roman Strzelczyk[2], Gilles H. Peslherbe[3]

[1] Research and Development Department, TMM LLC, Volodymyrska str. 49a, Kyiv, Ukraine
oleksiy.khavryuchenko@tmm.ua
[2] Institute of Molecular Physics Polish Academy of Sciences, Mariana Smoluchowskiego Str. 17 60-179 Poznań, Poland
[3] Centre for Research in Molecular Modeling and Department of Chemistry and Biochemistry, Concordia University, Sherbrooke Street West 7141, Montréal, QC, Canada

Abstract

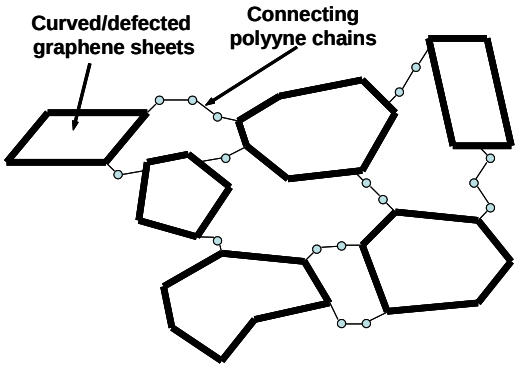
Origin of magnetism in carbon materials is often ascribed to dangling bonds on zigzag edge of graphene planes. However, for amorphous carbon experimental evidences are scarce due to two reasons: first, there is no definite model of amorphous carbon, second, carbon tends to react with *3d*-metals, while even traces of paramagnetic metal ions can corrupt picture from magnetism of carbon as it is. Is it possible that their magnetism and high electric conductivity is due to the same mechanisms as in graphenic materials with sp^2 hybrydization?

Here we report the first step in a complex approach of solving this problem with synthetic, spectroscopic and computational methods. The aim of the work on this stage was to synthesize ultra-pure carbon, prove its purity and amorphicity, characterize its magnetic properties by EPR spectroscopy and magnetic measurements, and propose a zeroth-order computational model of spin centers able to describe the magnetic properties. Very well known carbon material produced by carbonization of phenol-formaldehyde (PhF) resin was chosen due to large number of works, allowing various methods of material characterization, simple synthesis and relatively easy purification of the precursors. The resulting material showed no traces of paramagnetic metals, and its microstructure was characterized by TEM. Molecular model for the magnetic and EPR data interpretation was constructed basing on recently developed model of amorphous carbon produced from PhF resin [1]. These results were compared with EPR data on reduced graphene.

Neither magnetism of the edge-located paramagnetic defects [2], nor antiferromagnetic ordering observed for magnetic moment associated with carbon vacancies in graphene [3], can explain the temperature dependence of EPR signal. More sophisticated model comprising both graphene-derived parts and connecting carbene atoms was used, and DFT calculations were applied to simulate its thermal-dependent properties. Combined experimental and computational demonstrated that untypical temperature dependence of EPR signal intensity can be ascribed to interaction between localized and delocalized (conducting) electrons in the bulk, with ratio between them being related to conformation of carbene-type atoms connecting graphene sheets.

References

[1] V. D. Khavryuchenko, O. V. Khavryuchenko, J. Phys. Chem. C, **117** (2013) 7628.
[2] M. Augustyniak-Jablokow, M. Maćkowiak, K. Tadyszack, R. Strzelczyk, Acta Phys. Polonica C, **127** (2015) 537.
[3] M. A. Augustyniak-Jabłokow, K. Tadyszak, M. Maćkowiak, Y.V. Yablokov, physica status solidi (RRL)-Rapid Research Letters, **5** (2011) 271.



Schematic representation of two-domain model of amorphous carbon

Acknowledgments

Computational resources were provided by the Centre for Research in Molecular Modeling (CERMM) and Calcul Québec.