Spin Doublet Point Defects in Graphenes: Predictions for ESR and NMR Spectral Parameters

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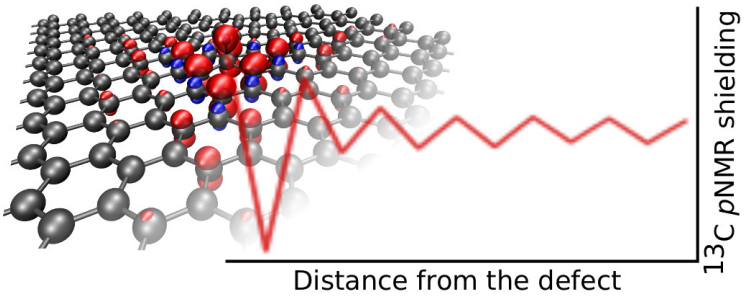
An adatom on graphene surface may carry a magnetic moment causing spin-half paramagnetism. This theoretically predicted phenomenon has recently also been experimentally verified [1]. The measurements of defect-induced magnetism are mainly based on magnetometric techniques where artefacts such as environmental magnetic impurities, are hard to rule out. Spectroscopic methods such as electron spin resonance (ESR) and paramagnetic nuclear magnetic resonance (pNMR) are conventionally used in the development of magnetic materials, $e.g.$, to study paramagnetic centers.

The present density-functional theory study demonstrates with calculations of the ESR g-tensor and the hyperfine coupling tensors, as well as the pNMR shielding tensor, that these spectroscopies can be used to identify the paramagnetic centers in graphenes [2]. The studied defects are hydrogen and fluorine adatoms on sp^2-hybridized graphene, as well as hydrogen and fluorine vacancies in the sp^3-hybridized graphane and fluorographene, respectively. The directly measurable ESR and pNMR parameters give insight to the electronic and atomic structures of these defects and may contribute to understanding carbon-based magnetism via the characterization of the defect centers.

We show that missing hydrogen and fluorine atoms in the functionalized graphane and fluorographene, respectively, constitute sp^2-defect centers, in which the magnetic resonance parameters are greatly enhanced. Slowly decaying adatom-induced magnetic resonance parameters with the distance from the sp^3-defect, are found in pure graphene.



[1] R. R. Nair, M. Sepioni, I.-L. Tsai, O. Lehtinen, J. Keinonen, A. V. Krasheninnikov, T. Thomson, A. K. Geim, I. V. Grigorieva, I. V., Nature Physics **8**, 199 (2012).

[2] J. Vähäkangas, Jiří Mareš, P. Lantto and J. Vaara, Journal of Chemical Theory and Computation **8**, 199 (2012).