# Abstract

The magnetic tetrapyrrole molecules (such as porphyrins and phthalocyanines) with an active transition metal atom in their centre are currently intensively studied as prosperous potential elements of devices for high-density information storage and processing, It has been recently proved that by means of external factors one could induce two stable fully controllable molecular states. Therefore, hybrid systems consisting of such magnetic molecules and suitable carriers from family of two-dimensional materials are often considered as promising highly scalable spintronic systems that could in the near future lead to novel industrial applications.

The main goal of this dissertation is to provide understanding of physical mechanisms that determine the stability of such hybrid systems and shed light on their electronic and magnetic properties, on one hand, and deliver reliable quantitative predictions that could facilitate the design of devices, on the other hand. To reach this goal, the methodology based on the first-principles theory is necessary. The computations in the frameworks of the density functional theory (DFT) have been employed for issues of the hybrid systems' stability and morphology, and the multi-reference method going beyond the standard DFT has been used to study the magnetism in the hybrid systems. Two hybrid systems have been chosen as the case studies: iron phthalocyanine (FePc) on graphene, FePc/Graphene, and FePc on titanium carbide ($Ti_2C$) MXene magnetic 2D system, FePc/$Ti_2C$.

The FePc/Graphene hybrid system has been studied in the first part of the thesis. In these studies the adsorption of the FePc to pristine and defected graphene layer is investigated. We consider typical defects in graphene, such as Stone-Wales (SW) defect, substitutional impurities, of boron, nitrogen, and sulphur, and also complex defects involving SW defect and B, N, and S impurities in its interior. We determine first the stability and geometry of these defects in graphene, and then we study whether these defects can facilitate the adsorption of FePc, and finally the defect induced modifications of the electronic and magnetic properties of the hybrid systems.

The DFT studies of the FePc/Graphene hybrid systems have been carried out within two types of boundary conditions: (i) superlattice geometry with periodic boundary conditions, and (ii) cluster geometry, where the carbon dangling bonds at the edges of the graphene layer are saturated with hydrogen atoms. In the case of superlattice and cluster geometries, the numerical packages *Quantum Espresso* and *ORCA* have been employed, respectively. We observe fairly good agreement between geometries of the studied systems as obtained with two kinds of boundary conditions. This allows us to use the results obtained with the DFT calculation involving the cluster boundary conditions as a starting point for studies of excited states employing multireference methods, which is a prerequisite of the correct description of the magnetic states of the FePc/graphene hybrid system.

For the multireference calculations, the graphene layer has been mimicked by pyrene molecule consisting of four carbon hexagons. This concept makes it possible for the first time to go beyond the one-determinant approximation in studies of such complex system as the FePc/Graphene hybrid structures, even with very restricted computer resources available. As it turns out, in systems with B and N impurities, only multi-reference method leads to reliable results.

In the second part of the thesis, we perform the spin polarised DFT studies of FePc/$Ti_2C$ hybrid system. The most relevant issue in this part is the interaction between magnetic atoms: Ti from MXene substrate and Fe from phthalocyanine. Four various magnetic configurations of FePc/$Ti_2C$ have been considered. The significant ferromagnetic interaction between the iron atom and the upper titanium layer plays important role in the reorientation of the iron atom magnetic moment. We also analyse a model of the system in which the FePc molecule is in a quintet state (the ground state of an isolated molecule is a triplet). To get a better understanding of the physics of the FePc/$Ti_2C$ hybrid system, we also study the hybrid systems with single iron atom and non-magnetic $H_2$ phthalocyanine on the $Ti_2C$ layer, Fe/$Ti_2C$ and $H_2$Pc/$Ti_2C$, respectively, which nicely explains the role of the Pc ligand in the FePc/$Ti_2C$ hybrid system.