Spin-orbit coupling caused spin splitting in doped graphene like layered materials

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We provide a general effective Hamiltonian, deduced from the theory of invariants, which describes relativistic band structure in pristine and decorated graphene-like systems for the most relevant region of low energy around the Dirac $K$-point. The effective Hamiltonian embraces systems such as decorated graphene (also silicene, germanene, and stanene), h-BN, and MX$_2$ (e.g., MoS$_2$). Together with relativistic ab initio calculations in the framework of the density functional theory (DFT) that provide the material constants, the developed scheme constitutes the general theory of spin splitting caused by the spin-orbit coupling (SOC) in layered materials. In this report, we provide exemplary results for graphene decorated with Ge, Sn, and Pb atoms (where the symmetry is lowered from the original $D_{6h}$ symmetry to $C_{3v}$) and with Ca, and Sr (where the symmetry is lowered to $C_{6v}$).

Graphene and other two-dimensional systems have emerged recently as very promising candidates not only for charge electronics but also for spintronics. However, prerequisites for spintronic devices based on these systems are (i) deep quantitative understanding of the spin-orbit caused spin splitting in the electronic structure, and (ii) possibility to tune spin splitting by external factors, such as decoration of layers with various atoms.

The effective Hamiltonian for the band structure around the Dirac $K$-point (of the $D_{6h}$ symmetry) has the form $H_{\text{eff}} = H_0 + H'$, where $H_0$ is usual low energy k.p Hamiltonian [1], and $H'$ describes the effects of the perturbation (decoration) coupled to the S-O interaction, and was established by us on the basis of the theory of invariants [2]. It has the form of 4x4 matrix, $H' = \begin{pmatrix} A & 0 \\ 0 & B \end{pmatrix}$, where $A = \begin{pmatrix} \Delta + \delta & 0 \\ 0 & \Delta - \delta \end{pmatrix}$ and $B = \begin{pmatrix} \Delta^* + \delta^* & 0 \\ 0 & \Delta^* - \delta^* \end{pmatrix}$, and contains four material constants, $\Delta$, $\delta$, $\Delta^*$, $\delta^*$, which can be determined from ab initio calculations. We perform DFT calculations taking relativistic effects non-perturbatively and employing relativistic PAW pseudopotentials as implemented in the VASP code. The full relaxation of atomic positions in the supercell is performed and provides us the energetically favorable position of the dopants in the graphene backbone. Extremely high degree of accuracy is enforced to be able resolve the electronic structure on the scale of $\mu$eV. For the case of Ca decorated graphene, the parameters of the effective Hamiltonian $\Delta$, $\delta$, $\Delta^*$, and $\delta^*$ are computed to be equal [in eV] to -2.4076, 0.0006, -2.4084, 0.0002, respectively. The excellent agreement between the eigenvalues of the effective Hamiltonian and the electronic dispersions from DFT computations for the graphene decorated with Ca atoms (in the hollow position) at 13% concentration is illustrated in the figure below.

We discuss also physical mechanisms leading to the constants in the effective Hamiltonian.