

metry. Moreover, with increasing concentration of the H-vacancies ($N > 8$) we found that the number of localized states can be smaller than the number of defects. For the *AA*-distribution the size of the gap is found to gradually decrease from 3.75 to 2.72 eV with increasing number of defects from $N = 2$ to $N = 18$. Additionally, with a growing number of defects a significant distortion of the planarity of graphane, such as buckling of the lattice inherent for the *AA*-distribution, was observed.

STABLE FERROMAGNETISM

A prediction of the ordering of the *AA*-distributed localized states on the graphane surface is controversial. According to results in [24] for semihydrogenated graphene, i.e. graphene hydrogenated from one side (the so called graphone), the non-hydrogenated side of graphane is possessing the localized states and the spins all of them are ferromagnetically ordered. We believe that this behavior is highly debatable because it is known for the carbon-like materials, such as diamond and graphitic structures, that the total magnetization is suppressed by increasing the vacancy concentrations, and particularly for graphitic structures this occurs more rapidly [10]. In contrast, authors of Ref. [20] did not consider the ferromagnetic ordering of the states in defected graphane at all because they found that for the vacancy defects located on the neighbors ($d = 2a_{C-C}$), the spins are paired, indicating a nonmagnetic state. When the distance exceeds $2a_{C-C}$ for which pairing of the spins can not occur, the interaction between the localized states vanish, thereby favoring the antiferromagnetic ordering of spins. However, we believe that pairing of the spins of the localized states located on the neighboring carbon atoms is unlikely, considering the significant distance between the vacancies (~ 2.55 Å). Moreover, pairing may occur only when the spins are antiferromagnetically ordered (that for the states localized on the same sublattice is against the Lieb's theorem [5]) and it implies the formation of the bond, which again seems unlikely because of the large distance between the localized states (the typical *C*–*C* bond length in organic compounds is ~ 1.4 Å against ~ 2.55 Å for the states separated by $d = 2a_{C-C}$ in graphane). Therefore, the question of ordering of spins of the localized states in their *AA* distribution is unclear as yet which we set out to investigate here.

We found that for the *AA*-distribution of the localized states formed by the H-vacancies, the ferromagnetic ordering of their spins is possible when the vacancies are placed on the neighboring carbon atoms (see $E_{(\frac{8}{2};\frac{1}{2})}^0$ for the *AA*-distribution in Table I), thereby generating a state characterized by a large spin number (see an example for graphene in Ref. [23]). Just as in [20] it was noticed that increasing the distance between the vacancies leads to vanishing of π - π interaction between local-

ized states resulting in the antiferromagnetic ordering of spins. Therefore, we investigating here the stability of the ferromagnetic ordering of the spins of the localized states placed on the neighboring carbon atoms ($d = 2a_{C-C}$) and formation of domains of defects depending on the concentration of the defects in domain and number of domains.

We found that with increasing concentration of defects (N) the stability of the state with a large spin number, i.e., the difference in total energy between the state with a large spin number and the singlet states, can increase. Therefore, for two parallel lines of defects $E_{(\frac{4}{2};\frac{1}{2})}^0 = -3.32$ eV for $N = 4$ (Fig. (a) for the spin density distribution of $N = 4$), $E_{(\frac{6}{2};\frac{1}{2})}^0 = -5.03$ eV for $N = 6$ and $E_{(\frac{8}{2};\frac{1}{2})}^0 = -6.56$ eV for $N = 8$. However, for $N = 8$ the states characterized by the lower spin number are close in energy to the state with the larger spin number ($E_{(\frac{8}{2};\frac{6}{2})}^0 = -0.035$ eV, $E_{(\frac{8}{2};\frac{4}{2})}^0 = -0.065$ eV and $E_{(\frac{8}{2};\frac{2}{2})}^0 = -0.12$ eV) thereby destabilizing it and limiting the number of defects having ferromagnetically ordered spins. A further increase of the defect concentration ($N > 8$) leads to significant suppression of the energy difference between the ferromagnetic and singlet states. Thus, $E_{(\frac{10}{2};\frac{1}{2})}^0 = -0.25$ eV for $N = 10$, $E_{(\frac{12}{2};\frac{1}{2})}^0 = -0.17$ eV for $N = 12$ and $E_{(\frac{14}{2};\frac{1}{2})}^0 = -0.14$ eV for $N = 14$. Therefore, just as for graphene [6–9], there is a critical value of defect concentration above which the ferromagnetic ordering of the spins of the localized states occupying the same sublattice no longer exists. Another destabilizing factor for ferromagnetism in graphane containing many H-vacancies in the *AA*-distribution is the lattice relaxation leading to the buckling of the graphane structure ($E_{(\frac{4}{2};\frac{1}{2})}^0 = -1.49$ eV for $N = 4$ and $E_{(\frac{8}{2};\frac{1}{2})}^0 = 1.29$ eV for $N = 8$). There is also a significant decrease in stability when the defects are divided in groups, because the state characterized by antiferromagnetic ordering of spins between the groups is close in energy to that of the state with ferromagnetic ordering of all spins (see the spin density distribution for $N = 4$ in Fig. (b)). Finally, since graphane is a wide gap semiconductor and possess no localized states at the edges which could interact (π – π interaction) with states formed by the H-vacancy defects, the increasing size of the graphane flakes leading to suppression of the graphane gap should not drastically alter the interaction of the localized states and their ordering, which was the case in graphene [8].

To summarize, for the *AA*-distribution of the H-vacancies the size of the band gap can be tuned by the level of hydrogenation – the gap slowly decreases with increasing number of defects, while for the *AB*-distribution the size of the gap fluctuates in the range of 1.2 – 3.7 eV depending on the level of the broken sublattice symmetry. Moreover, formation of the H-vacancy defects redistributed over one side of the graphane plane