Optimize the probability of single atom manipulation in scanning transmission electron microscope

Su, C.[1], Li, J.[2], Idrobo, J.[3], Susi, T.[4], Kotakoski, J.[4], Wang, Z.[5], Dong, M.[6], Yan, Q.[7] and Su, G.[7]

[1] MIT, United States, [2] MIT, United States, [3] Oak Ridge National Lab, United States, [4] University of Vienna, Austria, [5] Arhuus Univeristy, Denmark, [6] Arhuus University, Denmark, [7] University of Chinese Academy of Sciences, China

Aberration-corrected scanning transmission electron microscopy (STEM) provides a powerful way of identifying dopant atoms with atomic resolution imaging and electron energy-loss spectroscopy (EELS) [1,2]. Single Si [3,4], N [4,5] and B [6,7] dopant atoms in graphene have already been imaged and their chemical bonding distinguished from the EELS near-edge fine structure. Substitutional P atoms has also been observed in STEM and identified by EELS, while in the meantime, it demonstrates a greater variety of dynamic processes.

In this work, we discusses the four distinct dynamical processes of dopant atom in graphene and demonstrated the possibility of manipulating a single phosphorus dopant in the graphene lattice using STEM. We have further explained the mechanisms of these dynamics using first-principles calculations, revealing that most processes involve the interaction of the electron beam with a C atom neighboring the dopant. Some other dynamic processes will be explained by climbing-image nudged elastic band method.

In addition to providing physical insight, we have proposed an architecture for optimizing the probability of manipulating dopant atoms. Here, we introduce two important concepts: *dynamic space* and *manipulation decision tree*. Dynamic space is a space partitioned into different areas to represent dynamic processes. It is a convenient way of calculating the scattering cross section of these processes, which lays the foundation of calculating the probability of each branch in the decision tree. This architecture offers strategies for a more precise manipulation of dopants in graphene.

References

1. Suenaga, K. & Koshino, M. Atom-by-atom spectroscopy at graphene edge. Nature 468, 1088 - 1090 (2010).

2. Krivanek, O. L. et al. Atom-by-atom structural and chemical analysis by annular dark-field electron microscopy. Nature 464, 571 - 574 (2010).

3. Zhou, W. et al. Direct determination of the chemical bonding of individual impurities in graphene. Phys. Rev. Lett. 109, 206803 (2012).

4. Ramasse, Q. M. et al. Probing the bonding and electronic structure of single atom dopants in graphene with electron energy loss spectroscopy. Nano Lett. 13, 4989 - 4995 (2013).

5. Nicholls, R. J. et al. Probing the bonding in nitrogen-doped graphene using electron energy loss spectroscopy. ACS Nano 7, 7145 - 7150 (2013).

6. Bangert, U. et al. Ion implantation of graphene-toward IC compatible technologies. Nano Lett. 13, 4902 - 4907 (2013).

7. Kepaptsoglou, D. et al. Electronic Structure Modification of Ion Implanted Graphene: The Spectroscopic Signatures of p- and n-Type Doping. ACS Nano 9, 11398 - 11407 (2015).