Machine-Learned Structure Search for Silicene on Ag(111)

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Silicene, a silicon analog of graphene, has been intensively studied in the last decade because it is a promising candidate for the quantum spin Hall system and is more compatible with silicon-based nanotechnologies than graphene. Although freestanding silicene has an alternately buckled honeycomb lattice [1], scanning tunneling microscopy (STM) has revealed that silicene on a substrate exhibits a variety of reconstructions. For example, silicene on the Ag(111) surface shows e.g., 4×4, $\sqrt{13} \times \sqrt{13}$, $2\sqrt{3} \times 2\sqrt{3}$ periodicities with respective to the Ag surface, which are further classified into several phases with distinct buckling patterns. Although the geometries of representative phases have been identified, there remain phases which are difficult to clarify experimentally. A typical example is the T phase, which is observed as round dots with a $\sqrt{13} \times \sqrt{13}$ periodicity in STM images. However, detailed analyses have revealed that some of the dots are elongated and orientated randomly [3], whose origin has never been clearly explained based on the structural models proposed so far. Theoretically, it is difficult to apply expensive density functional theory (DFT) calculations to such less ordered systems because of the complexity inherent to the buckling structure. To overcome the difficulty, we here adopt global optimization with first-principles energy expressions (GOFEE) algorithm [4] to search for the stable structures of silicene on Ag(111). GOFEE outperforms conventional methods based on DFT and evolutionary algorithms (EA) by relaxing the EA-generated structures using the inexpensive surrogate model of Gaussian process regression. Our results demonstrate that GOFEE can not only reproduce the representative stable structures but also generate metastable ones. In particular, the structures with the $\sqrt{13} \times \sqrt{13}$ periodicity (Figure 1) include phases with one (d), two (e), and three (f, g) protrusions, whose energies are close to each other. This indicates that the less ordered nature of the T phase can be attributed to the mixture of these phases.

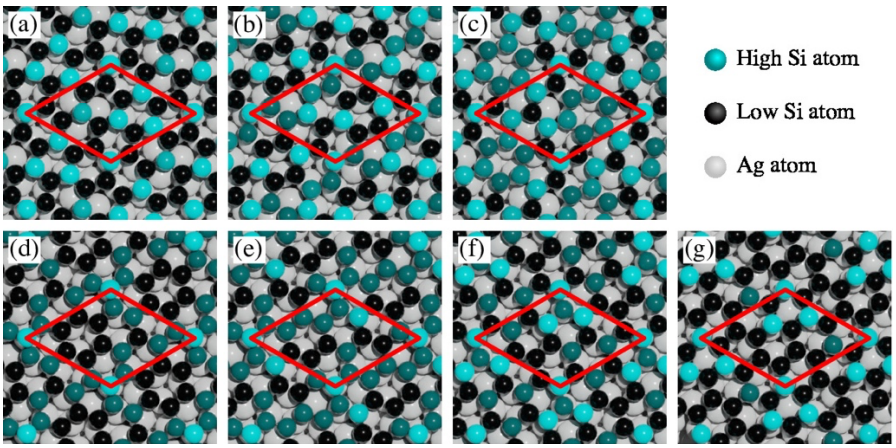


Fig. 1. $\sqrt{13} \times \sqrt{13}$ phases of silicene on the Ag(111) surface obtained by the GOFEE search. The rhombus denotes the unit cell with the $\sqrt{13} \times \sqrt{13}$ periodicity relative to the Ag(111) surface.

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[4] M. K. Bisbo and B. Hammer, Phys. Rev. Lett., **124**, 086102 (2020).