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Electronic, structural, and vibrational properties of monolayer materials and their heterostructures

単層物質とその複合系の物性研究

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Outline

This thesis presents first-principles theoretical works on monolayer materials and their heterostructures within the framework of density functional theory (DFT). Atomically thin two-dimensional materials such as graphene and a hexagonal boron nitride (h-BN) monolayer have attracted much interest because of their interesting properties and possible application as nanoscale devices. Recently the heterostructures composed of these monolayer materials have been realized in experiments. Therefore, the current situation is approaching the arbitrary fabrication of artificial new materials. The theoretical results presented in this thesis will be a basis of the future researches on monolayer heterostructures.

We study the structural properties of three-dimensionally stacked superlattices composed of graphene and h-BN. We discuss the possibility of lattice matching in graphene/h-BN monolayer and graphene/h-BN bilayer superlattices because there is 1.6 % difference in their lattice constants of h-BN. We adopt twisted graphene/h-BN superlattices to simulate the interlayer interactions in lattice-mismatched superlattices. We find that lattice-matched superlattices are more stable than a lattice-mismatched superlattice when they have a specific kind of interface stacking geometries between graphene and h-BN. The atomic configuration of this interface stacking geometry of lattice-matched superlattices has the short boron-carbon distance, indicating a strong interaction between the carbon and the boron atoms.

We also study the electronic structure of the graphene/h-BN superlattices. The electronic structure of the lattice-matched graphene/h-BN monolayer superlattices is metallic due to the band dispersion along the direction perpendicular to the layers, which is caused by the inter-graphene interaction mediated by a h-BN monolayer. On the other hand, the dominant parts of the electronic properties of the lattice-matched graphene/h-BN bilayer superlattices are almost determined by only sublattice symmetry breaking. The graphene/h-BN bilayer superlattices exhibit semiconducting or graphene-like two-dimensional electronic properties depending on their stacking sequences. We further discuss the electronic properties of lattice-mismatched graphene/h-BN monolayer superlattices. Lattice-mismatched h-BN monolayers should still mediate inter-graphene interaction although the dispersion along the direction

perpendicular to the layers is not large. We also find that the dispersion depends on the moire periodicity of the superlattice.

Next we study finite layer thin films of graphene and h-BN. We report that the interlayer interaction energies are inversely proportional to the number of layers. The infinite-layer limit of the interaction energies is consistent with the energies of the graphene/h-BN superlattices. We again discuss the possibility of lattice matching by using a method similar to the superlattice case. Thin films with four or more layers can also have stable lattice-matched stacking geometries. We find that the maximum value of the band gaps of lattice-matched thin films having the same layer number but different stacking sequences reduces with respect to the number of the layers. The band gaps of these thin films can be tuned by using an external electric field. We also propose six-layer thin films with h-BN bilayers, which have 99 meV band gap or graphene-like linear dispersion depending on their stacking sequences, similar to graphene/h-BN bilayer superlattices.

Finally, we study another important kind of monolayer materials, self-assembled molecular monolayers. We investigate the vibrational properties of a self-assembled monolayer of adamantane molecules on a Au(111) surface. Such a self-assembled monolayer is also of importance for the size reduction of nanoscale devices. We reveal the considerable effect of the gold substrate on the infrared (IR) spectrum of adamantane molecules by using the first-principles IR spectrum simulation methods based on DFT. One of three gas-phase IR peaks is significantly redshifted due to adamantane–substrate interactions. We also find that another gas-phase IR peak loses its IR intensity due to adamantane–adamantane interactions. Our theoretical IR spectrum well agrees with experimental spectra. We expect that the analysis method used in this thesis can be applied to other molecule/substrate systems.