2O03 The application of accelerated quantum chemical molecular dynamics program in rare earth system

○羅　一[1], 草谷友規[1], 伊藤優基[1], 遠藤　明[1], 久保百司[1], 今村　詮[2], 宮本　明[1,3]
[1]東北大学大学院工学研究科 (〒980-8579 仙台市青葉区荒巻字青葉 07)
[2]広島国際学院大学 (〒739-0321 広島市安芸区中野 6-20-1)
[3]東北大学未来科学技術研究センター (〒980-8579 仙台市青葉区荒巻字青葉 04)

[Introduction]

Dramatic changes were reported in the optical properties of lanthanum and yttrium hydride films with changing hydrogen content, e.g., films that switch from a shiny mirror to a yellow transparent window in a time of order seconds [1]. The electronic structure of lanthanide hydrides (LaH_x) with switchable optical properties, which is poorly understood, has attracted theoretical interest [2]. To challenge this topic, quantum theory is considered to be a powerful tool. However, it is pointed out that standard local density approximation (LDA) calculations do not predict a metal-insulator transition at all [3]. Recently, we have extended the application of accelerated quantum chemical molecular dynamics program to rare earth system, and challenged the theoretical aspects on LaH_x ($2< x < 3$) system. In this symposium, the interaction between lanthanum and hydrogen and the dynamics behavior of hydrogen in lanthanum metal and LaH_2 will be discussed.

[Method]

The molecular dynamics (MD) simulations were performed by “Colors” code [4], developed by our group, which is based on tight-binding theory. The velocity form of the Verlet algorithm is used with a time step of 0.1×10^{-15}s to integrate the equations of motion. For comparison, density functional theory method (DFT) was also applied with the consideration of relativistic effective core potentials.

[Results]

The dynamics behaviors of hydrogen in lanthanum metal and LaH_2 bulk were investigated, respectively. To realize high accuracy, all the parameters for “Colors” were determined on the basis of DFT calculations. “Colors” results can be compared to available experimental result and that obtained by DFT calculation results in the electron density distribution and atomic energy of single atom, the potential curve of pair atoms, binding energy, charge population as well as the cell size. The dynamics simulation results show that hydrogen in lanthanum metal prefers the central site of tetrahedron. The octahedral hydrogen in lanthanum dihydrides prefers interacting with tetrahedral hydrogen. Under higher temperature, the hydrogen in lanthanum metal carries more negative charge, which shows that more electron transfer from La to H. Fermi energy level indicates the metallic property of LaH_2, which is agreement with the experimental fact that LaH_2 is a conductor. Moreover, our results also indicate that the presence of one octahedral hydrogen dose not change the metallic property of LaH_2. At 500 K, the octahedral hydrogen is further from tetrahedral hydrogen and get more negative charge compared to that at other selected temperature. The quite different orbital interaction between H and La was observed. That is, H-1s interacts with La-6s orbital in lanthanum metal, however, La-5d orbital in LaH_2 bulk.

[References]

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