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First-principles Analysis of Carbon Diffusion in Ferromagnetic bcc-Fe

Minoru Ikeda, Takahiro Yamasaki and Chioko Kaneta

Nano-Electronics Research Center, Fujitsu Laboratories Ltd.,
10-1 Morinosato- Wakamiya, Atsugi, Kanagawa 243-0197, Japan

The acetylene (C_2H_2) molecular gas is deposited on the surface of Fe catalysis and grapheme sheets are formed experimentally. The actual state of C_2H_2 molecule on ferromagnetic (FM) bcc-Fe (110) surface and the formation process of a carbon network on Fe surface is reported previously [1]. There arises an open question whether a carbon atom can penetrate into Fe catalysis or not during the growth process. According to Jiang and Carter [2], a carbon atom hardly invades FM bcc-Fe substrate using 4x4x4 super cell geometry (128 Fe atoms). We further increase the super cell geometry from 2x2x2 (16 Fe atoms) until 7x7x7 (686 Fe atoms) systematically and investigate solution enthalpy of C into FM-bcc-Fe. In this paper, we first discuss C diffusion in FM bcc-Fe and possibility of C incorporation into FM bcc-Fe

Throughout this paper, we employ the projector augmented plane wave method (PAW) as the first-principles calculations as implemented into PAW-VASP codes [3-5]. The generalized gradient approximation and the spin polarization are also included to treat ground state properties of Fe. The Brillouin zone integrations are summed up to 4 k points. The barrier height energy (BHE) of C diffusion in FM-bcc Fe is estimated using the nudged elastic band (NEB) method [6].

We first investigated a diffusion of C atom in FM bcc-Fe. The initial position of C is located at the interstitial octahedral site. Then we elevated the temperature up to 1200K. In Fig. 1(a), trajectories of atoms in the cell are shown for 15 ps. While Fe atoms are confined near bcc sites (red lines), whereas the interstitial C (green line) moved in one lattice constant interstitially. The most stable site of interstitial C in a 6x6x6 super cell is shown in Fig. 1(b). The BHE of C diffusion in FM bcc-Fe is also estimated using NEB calculations. In Fig.2(a) and (b), the configurations of C along the NEB path and the BHEs for 4 different super cell sizes are shown. Our estimated BHE is 0.84 eV for the 6x6x6 super cell, which is close to the experimental value 0.87 eV. Our obtained results indicate that C atoms in FM bcc-Fe diffuse fast.

We define the solution enthalpy ΔHs as $\Delta Hs=E(Fe_nC_1)-E(Fe_n)-E(C_1)$ [2], where $E(Fe_nC_1)$ and $E(Fe_n)$ are cohesive energies of the corresponding super cell. And $E(C_1)$ is cohesive energy of graphite. In Fig.3, we plot ΔHs as a function of C density in the super cell. It should be noted that the negative value of ΔHs is exothermic, whereas the positive value is endothermic. Therefore below 0.05% C in FM bcc-Fe, FM bcc-Fe system with interstitial C is energetically favorable. Thus interstitial C atoms are incorporated into the FM bcc-Fe at the initial stage of grapheme formation process. And above critical concentration (0.05%) of C never penetrates into Fe catalysis. Using larger super cell we can obtain these results, being different ones of ref.[2]

References

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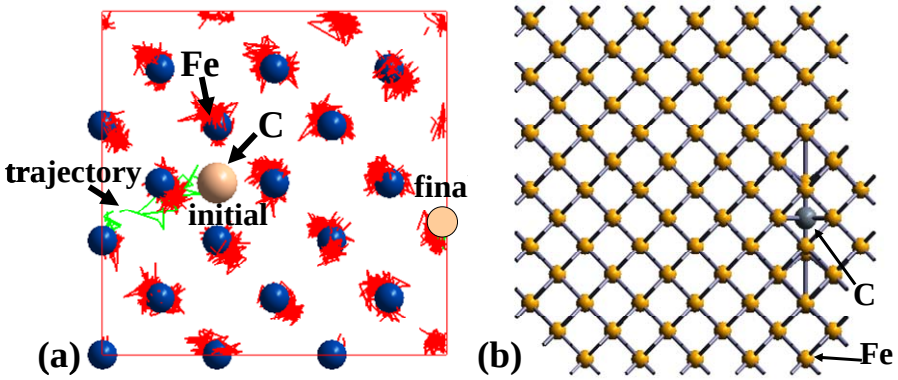

FIG. 1. The trajectories of atoms during 15 ps simulated annealing for 3x3x3 super cell are shown in (a). The stable position of interstitial C in 6x6x6 super cell is shown in (b).

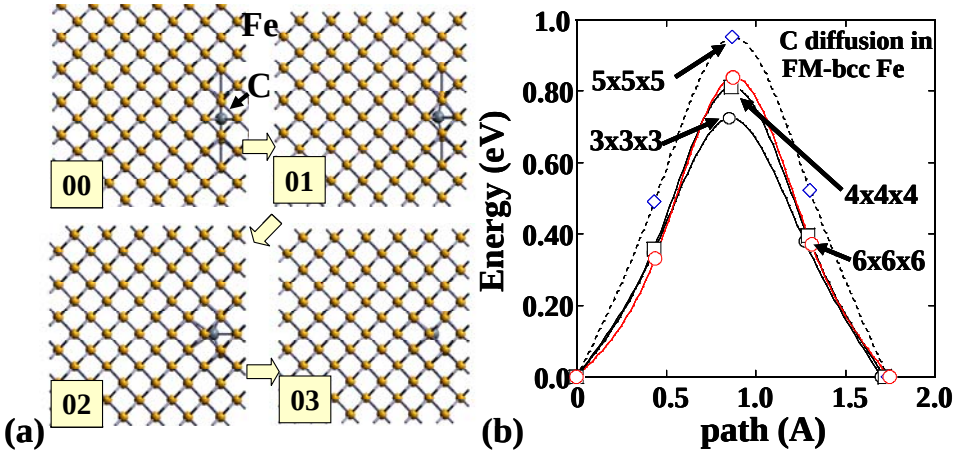

FIG. 2. The configurations of C along the NEB path are shown in (a). The BHEs for 4 different super cell sizes are illustrated in (b).

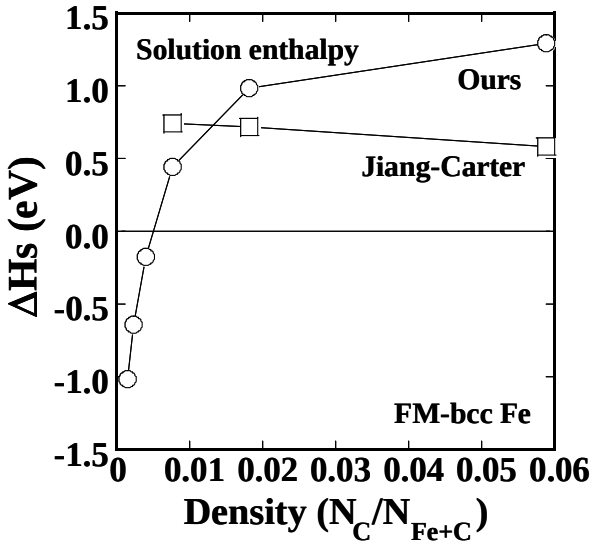

FIG. 3. The solution enthalpy of carbon in FM bcc-Fe is plotted as a function of density of C by increasing the size of a super cell. The Jian-Carter's results also also plotted.