

Determining Surface Phase Diagrams including Anharmonic Effects

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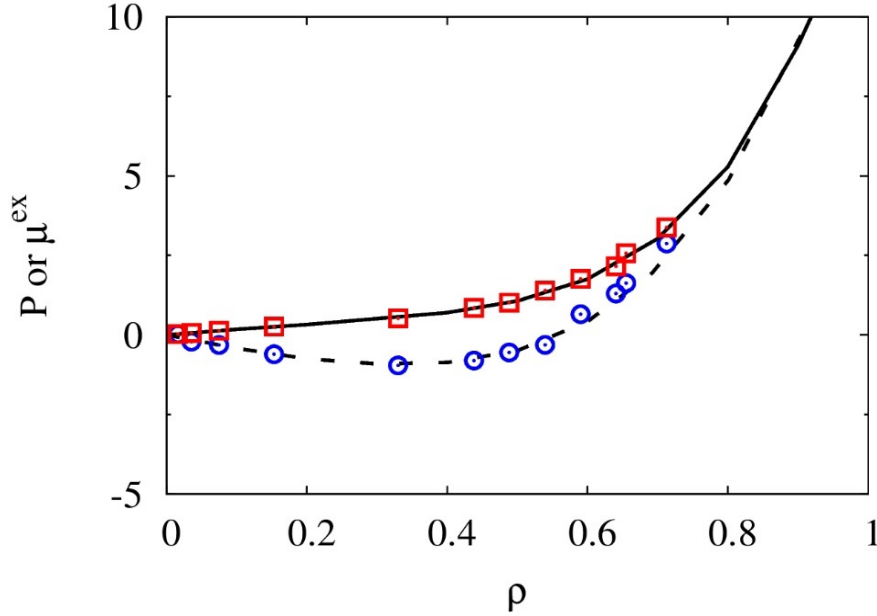


FIG. S1. Equation of state of the Lennard-Jones fluid; isotherm at $T = 2.0$. The solid line is the equation of state of Johnson et al. [Molecular Physics 78, 591 (1993)] and the squares are the results from our grand-canonical simulations. The dotted line is the excess chemical potential as calculated from the equation of state of Johnson et al [Molecular Physics 78, 591 (1993)] and the circles are the results of the simulations. All variables are in reduced units. The translation of reduced units to real units for Lennard-Jones argon are the same in Ref [D. Frenkel and B. Smit, Understanding molecular simulation: From algorithms to applications (Elsevier, 2002).]

(200, -2.4)	(200, -2.3)	(200, -2.2)	...	(200, -1.0)	(200, -0.9)
(250, -2.4)	(250, -2.3)	(250, -2.2)	...	(250, -1.0)	(250, -0.9)
(300, -2.4)	(300, -2.3)	(300, -2.2)	...	(300, -1.0)	(300, -0.9)
⋮	⋮	⋮	⋮	⋮	⋮
(550, -2.4)	(550, -2.3)	(550, -2.2)	...	(550, -1.0)	(550, -0.9)
(600, -2.4)	(600, -2.3)	(600, -2.2)	...	(600, -1.0)	(600, -0.9)
(650, -2.4)	(650, -2.3)	(650, -2.2)	...	(650, -1.0)	(650, -0.9)

Table. S1 The set of (μ, T) states used for REGCMC simulations of the LJ system. In each bracket, the first value represents temperature in unit of Kelvin (K), while the second value indicates the chemical potential in unit of electron Volts (eV). There are 10 temperatures ranging from 200 to 650 K, with an interval of 50 K, and 16 chemical potentials ranging from -2.4 to -0.9 eV, with an interval of 0.1 eV.

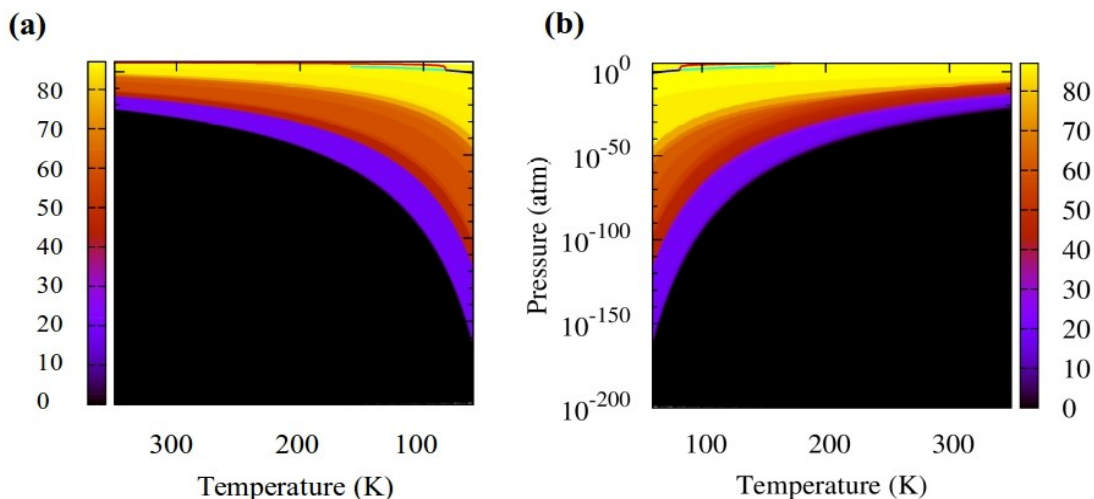


FIG. S2. Phase diagrams of a LJ gas-phase (particles B) in contact with a frozen fcc(111) LJ frozen surface calculated by aiAT (a) and MBAR (b) at low temperatures conditions ranging from 60 to 350 K.

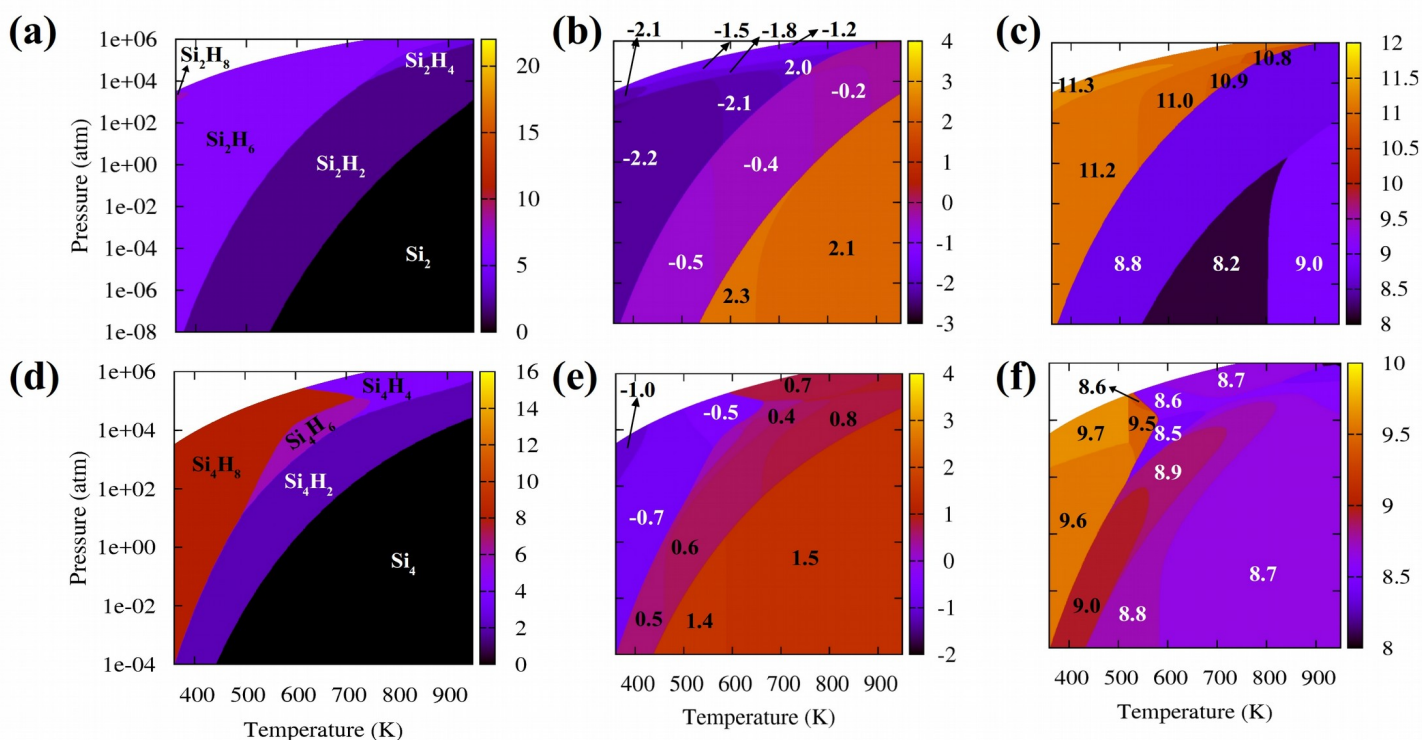


Fig. S3 Phase diagrams of chemisorbed Si_2H_N (a), vertical electronic affinity (VEA) (b) and vertical ionization potential (VIP) (c) of Si_2H_N at reactive gas phase, respectively. Phase diagrams of chemisorbed Si_4H_N (a), vertical electronic affinity (VEA) (b) and vertical ionization potential (VIP) (c) of Si_4H_N at reactive gas phase, respectively. The scale of panels (a) and (d) is the number of chemically adsorbed H atoms. VEA and VIP in panels (b), (c), (e) and (f) are in eV.

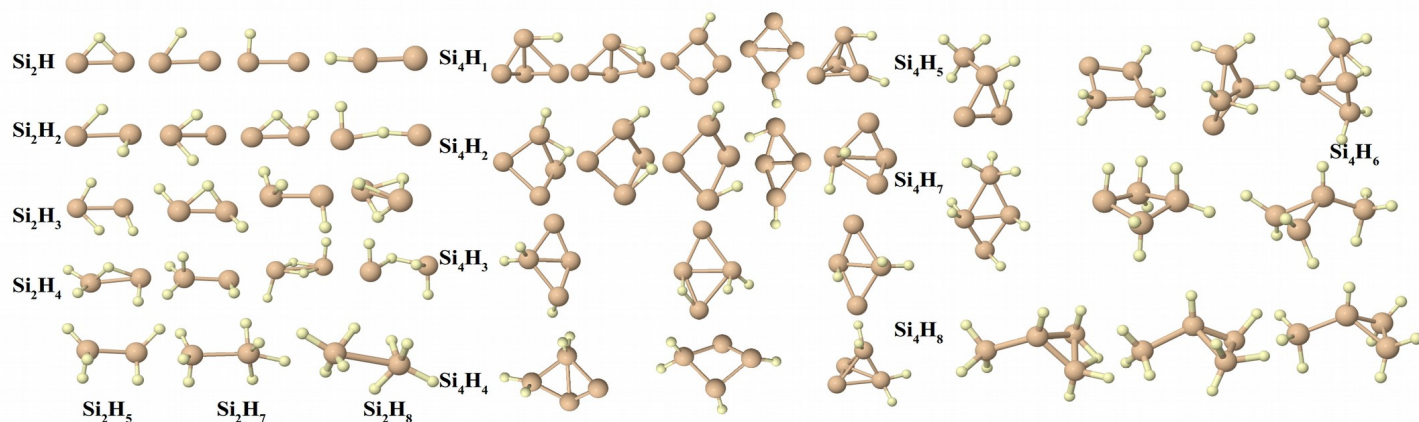


FIG. S4. Structures of isomers of each $\text{Si}_{\{2,4\}}$ with H_N size, respectively.

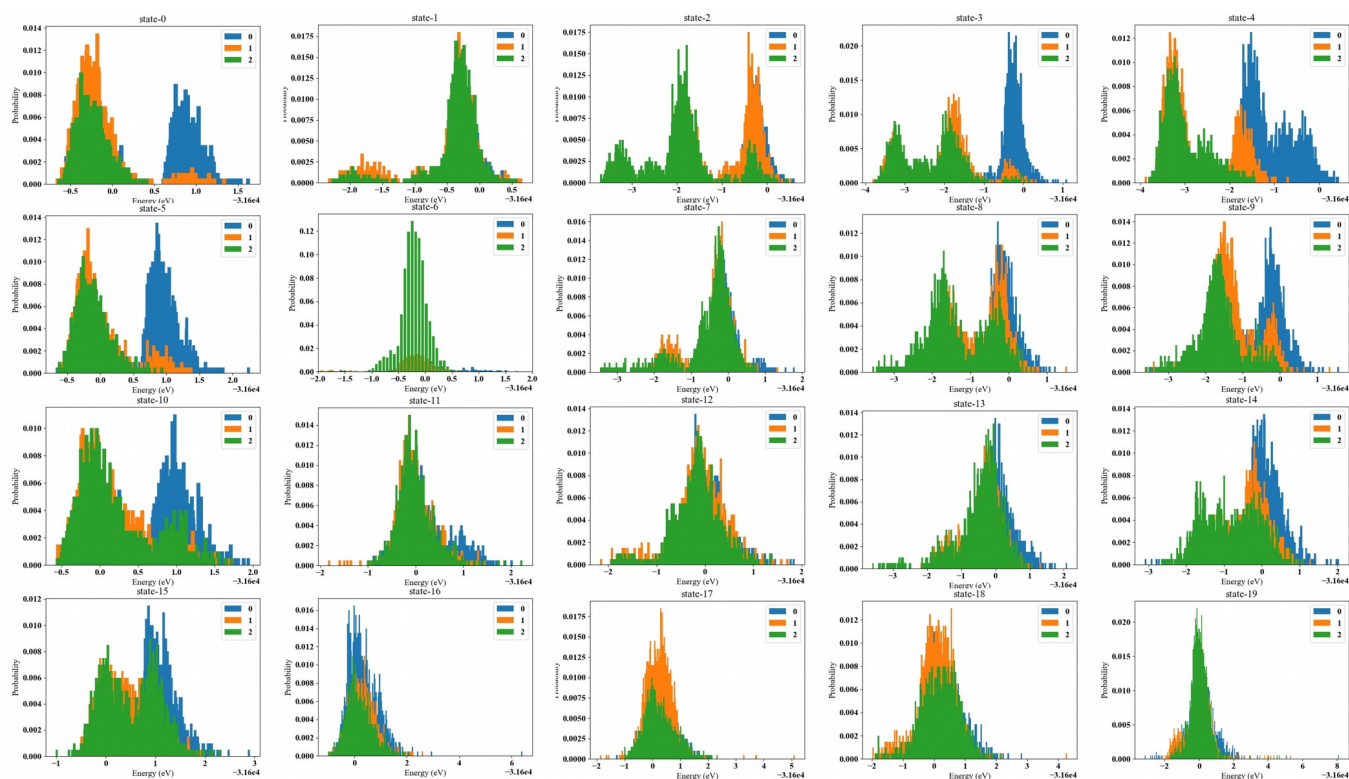


FIG. S5. Distribution of *adsorption energy* ($E_{\text{Si}_4\text{H}_N-N}/2E_{\text{H}_2}$) of each thermodynamical state in the REGCMD simulation for Si_4 . Each trajectory is divided into three same interval (2000 REGCMD steps). The distribution is calculated at every interval for each trajectory. The blue, orange and green bars indicate the energy distribution of first, second and third interval of each trajectory, respectively.

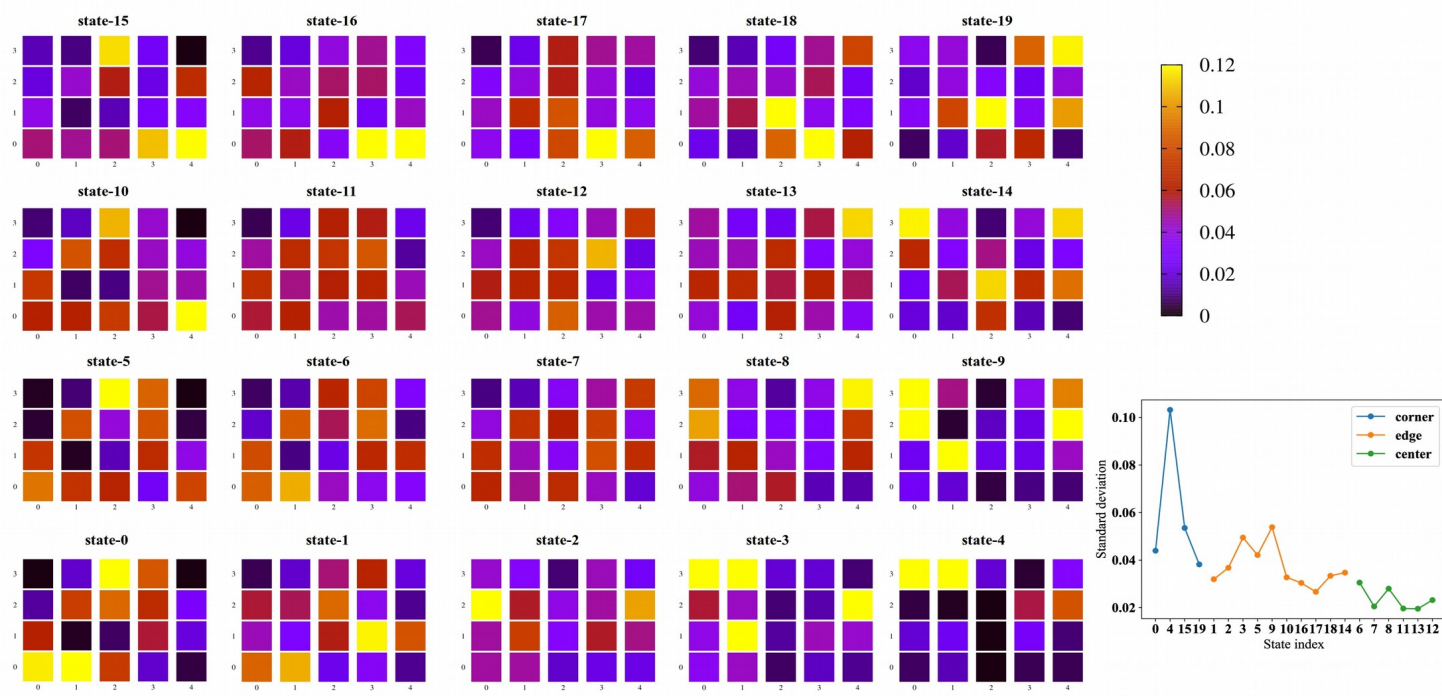


FIG. S6. The diffusion probability of each thermodynamical state in each configuration (replica) in the REGCMD simulation for Si_4 ; Inset on the bottom right: the standard deviation of each state.