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First principle investigation of phase transition and thermodynamic properties of SiC

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ABSTRACT

First principle calculations are employed to investigate the phase transition and thermodynamic properties of cubic silicon carbide. The Perdew–Burke–Ernzerhof (PBE) and the Perdew–Burke–Ernzerh of for solids (PBEsol) generalized gradient approximation (GGA) are used as the exchange–correlation potential respectively to reproduce the phase transition from zinc-blende (ZB) structure to rock-salt (RS) structure of silicon carbide. Then, they are compared with previous results and experiments which are calculated with different pseudopotentials. Both of the two pseudopotentials show reasonable results on equilibrium properties of SiC. However, Further analysis show that the calculated lattice constants, bulk modulus and elastic constants with PBEsol are indeed better than the calculations with PBE, while the PBEsol pseudopotentials has a lower evaluation than the PBE and other pseudopotentials on calculating the phase transition pressure of cubic silicon carbide. Moreover, the thermodynamic properties are studied with the quasi-harmonic Debye model. The isothermal bulk modulus and the adiabatic bulk modulus, the specific heat at constant volume and pressure, the Debye temperature and the entropy under different pressures and temperatures have been successfully obtained and discussed in detail.

1. Introduction

Silicon carbide (SiC) is a kind of promising engineering material due to its excellent chemical stability and physical properties, such as low density, high stiffness, high hardness and high thermal conductivity. It is widely used in the friction members and optoelectronic devices, such as gas turbines, heat exchanger and ceramic fans [1,2]. Sorted by different orders of twin molecular layers, there are many kinds of crystal structures of SiC. Among these, the most common is the cubic-crystal structure.

Some experiments have been carried out to better understand the physical properties of silicon carbide under high pressure and temperatures. X-ray diffraction measurements have shown that 3C-SiC undergoes a phase transition into rock-salt structure at 100 GPa or higher accompanied by a volume reduction of 20.3%, while the 6H polytype of SiC remained stable to 95 GPa [3]. Later, the shock compression of 6H polytype SiC experiments have shown that 6H SiC transformed to a sixfold-coordinated phase, most probably rock-salt structure at 105 GPa [4]. Phonon dispersion curves for some polytype SiC have been constructed from first-order Raman scattering data [5]. Also, lots of theoretical studies on SiC have been carried out. First principle is a promising theory in calculating materials properties which is based on density functional theory. The first principle calculations start from the basic laws of physics (the Schrodinger equations), then obtain the corresponding energy eigenvalues and get the related physical parameters of materials. Those physical parameters can be used to explain or predict the physical properties of materials. Many first-principle investigations on pressure-induced phase transition of SiC polytypes have been done [6–14]. The calculated pressures of transition were mostly around 60 GPa with local-density approximation (LDA) [11,12] or Perdew–Wang (PW) generalized gradient approximation (GGA) together with the Troullier–Martins pseudopotentials [13]. The DFT–LDA calculations usually tended to underestimate critical pressure for structure phase transition [8]. A revised Perdew–Burke–Ernzerhof generalized gradient approximation that improved equilibrium properties of densely packed solids and their surface was introduced [15]. The effects of the same exchange and correlation functional on transition metals were different [16]. Moreover, the elastic properties under pressure were also studied to find the relationship between elastic constants and pressures [17–19]. Extensive works on electronic structure [20–23], dielectric properties [24,25] and magnetic properties [26,27] have been accomplished. However, few investigations on the thermodynamics properties of SiC under high

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pressure and temperature have been done. As far as we know, some thermal properties of cubic silicon carbide have been calculated in the early years [28,29]. Recently, some thermodynamics properties and bulk modulus under pressure and temperature have been done [30]. However, the information about Debye temperature seems to be scarce and many other properties are not discussed in detail. On the whole, we would like to reproduce some elastic properties and the phase transition with the PBE and PBEsol exchange correlation potential and make a more comprehensive comparison to previous work with different pseudopotentials. Furthermore, we try to give more thermodynamic information to better understand the properties of SiC under high pressures and temperatures and discuss these properties in detail.

2. Theoretical method

The calculations in this work are performed using VASP code [31] which is based on density functional theory. The pseudopotential presented by Troullier–Martins [32] is employed to describe the interactions between electrons and core ions. The Perdew–Burke–Ernzerh (PBE) generalized gradient approximation and the revised Perdew–Burke–Ernzerh for solids (PBEsol) generalized gradient approximations [15] are used as the exchange correlation potential respectively. The typical pseudo atomic calculations are performed for Si $3s^23p^2$ and C $2s^22p^2$. In order to make sure that the ground state energy is converged within a specified tolerance, the optimize tests on k-points and cut-off energy are accomplished first. According to the test results, a well converged cutoff energy of 600 eV and the Brillouin-zone k-points with $9 \times 9 \times 9$ Monkhorst–Pack scheme [33] are employed for all calculations. The tolerance of the total energy is 10^{-6} eV/atom. The maximum atomic force is within 0.03 eV/Angstrom. All the stress components on optimized cell are smaller than 0.05 GPa.

3. Results and discussion

3.1. The crystal parameters and elastic constants

The total energy E and the corresponding primitive volume V are calculated firstly for both ZB structure and RS structure of cubic SiC, as it can be seen in Fig. 1. Fig. 1(a) and (b) shows that

Table 1
The lattice constants, bulk modulus (GPa) and its pressure derivation, elastic constants (GPa) of the ZB and RS structures of SiC at $P = 0$ GPa, $T = 0$ K.

	Ours (PBE)	Ours (PBEsol)	Others [8,11,12,34,36]	Experiments [35–38]
ZB structure				
a	4.374	4.361	4.30; 4.315; 4.34; 4.36; 4.37	4.36
B_0	219.7	224.2	200; 212; 223; 223.6; 227.1	225
B_0'	3.70	3.70	3.7; 3.77; 3.8; 3.79; 7.3	3.57
C_{11}	392.5	390.0	420; 415.1	390
C_{12}	135.9	137.6	126; 131.9	142
C_{44}	249.1	246.3	287; 265.4	256
RS structure				
a	4.06	4.04	4.04; 3.97	–
B_0	249.0	259.3	252.3; 266.6	–
B_0'	4.08	4.06	4.26; 4.64	–
C_{11}	411.5	416	484.8	–
C_{12}	184.5	182	174.7	–
C_{44}	320.1	313	383.4	–

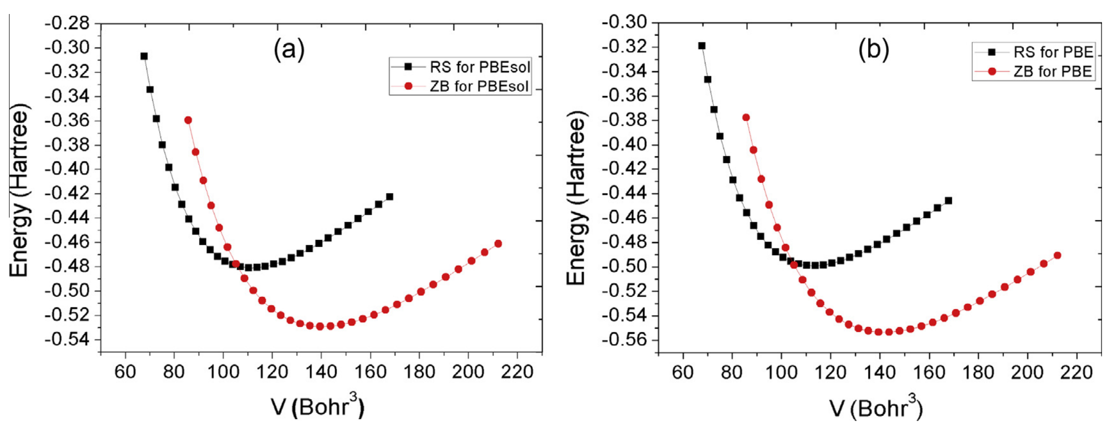

Fig. 1. (a) The calculated energy of ZB and RS structure as a function of volume with PBEsol (b) the calculated energy of ZB and RS structure as a function of volume with PBE.

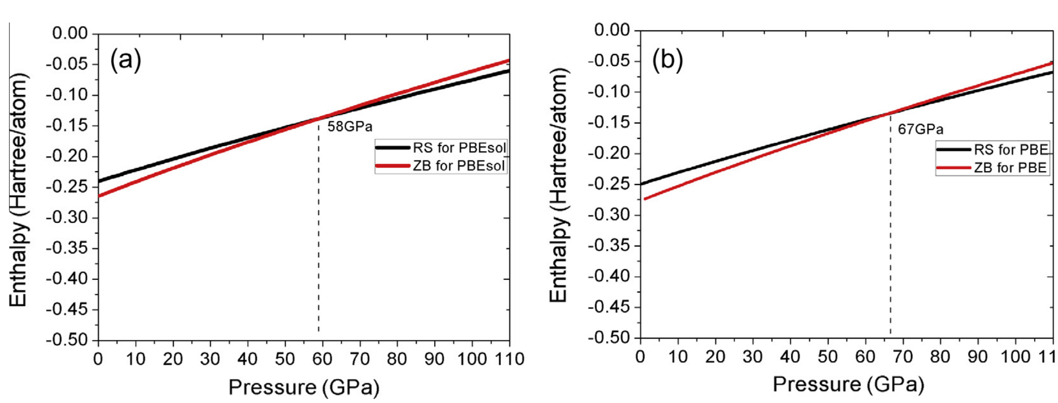

Fig. 2. The calculated enthalpy of ZB and RS structure as a function of pressure with (a) PBEsol and (b) PBE.