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An atomistic study of phase transition in cubic diamond Si single crystal subjected to static compression

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ABSTRACT

It is been widely experimentally reported that Si under static compression (typically in a Diamond Anvil Setup-DAC) undergoes different phase transitions. Even though numerous interatomic potentials are used for numerical studies of Si under different loading conditions, the efficacy of different available interatomic potentials in determining the phase transition behavior in a simulation environment similar to that of DAC has not been probed in literature which this manuscript addresses. Hydrostatic compression of Silicon using seven different interatomic potentials demonstrates that Tersoff(T0) performed better as compared to other potentials with regards to demonstration of phase transition. Using this Tersoff(T0) interatomic potential, molecular dynamics simulation of cubic diamond single crystal silicon has been carried out along different directions under uniaxial stress condition to determine anisotropy of the samples, if any. β-tin phase could be observed for the [001] direction loading whereas Imma along with β-tin phase could be observed for [011] and [111] direction loading. Amorphization is also observed for [011] direction. The results obtained in the study are based on rigorous X-ray diffraction analysis. No strain rate effects could be observed for the uniaxial loading conditions.

1. Introduction

Silicon is one of the most important elements used in modern nanodevices. It has been reported through experimental observations that Si undergoes phase transition under application of pressure [1–6]. The experimentally reported phases of Si on application of high pressure in compression are shown in the supplementary file (supplementary Fig. 1). It should also be noted that on decompression Si does not return back to its same phase but produces other different phases as shown in the supplementary file (supplementary Fig. 2). Most of these experimental investigations use a Diamond Anvil Cell (DAC) for application of pressure which typically applies a hydrostatic type loading situation on the sample. The Si specimen generally chosen for investigation is either amorphous or polycrystalline cubic diamond form. It has been reported that both of these forms of Si undergo phase transition, albeit at different pressures [7,8].

There has also been numerous numerical literature on molecular dynamic simulations of Si in which phase transition was demonstrated under different types of compression using different types of interatomic potentials [1,9–11,7,12,13]. Some of these literature [13,7,9–12] considers different variations of the Tersoff potential (hereby referred to as T0 [14], T1 [15], T2 [16], T3 [17] and T4(Erhart-Albe) [18] respectively). Other literature considers Modified Stillinger Weber potential(SW) [19,20] for their simulations. Interestingly none of these studies validate their used potential in its ability to correctly predict phase transformation behavior in Si. It should be noted that while developing a interatomic potential, many different considerations are tested but their ability to perform phase transition at initial ambient temperature and pressure conditions under application of load (as observed in experimental conditions) is not investigated.

The static compression loading mechanisms applied in the above literature also differs significantly. Some literature focus on nano-indentation [10,7,12,9] and some on abrasion at nanoscale [11]; in both of these the resulting stress state is quite different compared to uniaxial or hydrostatic stress type compressive loading condition carried out in this manuscript. There are also studies which considers uniaxially compressed silicon nano-spheres [13], but since it is a nanosphere the boundary conditions and the resulting stress conditions are quite different from either uniaxial or hydrostatic type loading situation considered in this study. The literature [1] even though is on uniaxial compression, focus on the amorphous form of Si and not on the cubic diamond crystal phase, as is considered in this manuscript. It should be noted that literature pertaining to static compression has only been considered in this manuscript. There are numerous studies on dynamic compression such as shock compression reporting phase transformations, but these studies have not been reviewed in this literature review

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since it is beyond the scope of this study.

The mechanism for identification of phase transformation in a sample on application of static compression also varies from study to study and none of these are confirmatory in nature. Some literature [7] considers changes in the Gibbs free energy to demonstrate phase transformation. Even though change in Gibbs energy denotes phase transformation in a confirmatory manner at T = 0 K where the entropy of the system can be neglected. However, at ambient temperature and pressure conditions (situation in which typical experimental investigations are carried out) the Gibbs free energy is not the total potential energy since the entropy of the system cannot be neglected. Therefore, under ambient temperature and pressure conditions it is not possible to determine phase transition using Gibbs energy. Moreover, since in Si there are numerous phases which have been reported and using Gibbs energy calculations it is not possible to determine which phase (or if a combination of phases) are present in the material. Based on these studies, determination of which phase has been made from purely coincidental evidence from experimental investigations reporting phase transitions at those pressure ranges, albeit under different loading conditions. For the case of Radial distribution function (RDF) (used in literatures [1,9,11]) and Angular distribution functions (ADF) phase change may be observed based on changes in the second and third coordination shell pattern but identification of the specific phase cannot be done with accuracy. Moreover it should also be realized that if the phase exists in small amounts then its effect cannot be identified using these methods. Coordination number (CN) has also been used in numerous literature [10,12,11,9] to demonstrate phase transformation and identify the phase. However, even though from a theoretical viewpoint CN is ideal for determination of a specific phase but its implementation in Lammps [21] requires specification of a cut-off radius. Ideally speaking the cutoff radius should be the distance where the first coordination shell in the RDF (after the peak) touches the x-axis. One should note that as a sample is loaded, this coordination shell shifts and thereby unless one changes the cutoff radius during simulation (based on amount of strain given to the sample), the results are bound to give higher coordination shell numbers which may not be entirely realistic to demonstrate a phase transformation. In this regard it should be noted that this method of CN may be acceptable if the load is applied locally (such as in the case of a nanoindentation) and phase transformation is expected to happen in small regions. However, it should be noted that this method fails if there is a significant shift in the first coordination shell peak distance and along with load the second coordination shell peak left shifts to the region of the initial first coordination shell peak. In the literatures on Si [10,12,11,9], the cutoff distance chosen by researchers ranges from 2.6 to 3.5 Å. Given the above mentioned methods for phase identification, the best way to identify change in crystalline phase is through X-ray diffraction (XRD) methods. As a matter of fact, if there are multiple phases in the sample, XRD will show peaks corresponding to different phases. XRD has been done in this manuscript for identification of the phases as cubic diamond single crystal is statically compressed hydrostatically as well as uniaxially along different directions.

It should be mentioned that there also exist DFT (Density Functional Theory) studies which focus on pressure induced phase transformation [22,8] (in which energy calculations demonstrate existence of high pressure phases of cubic diamond Si) but it should be understood that DFT studies typically show the most stable phase of Si under a specified pressure and 0 K temperature. In DFT simulations, there are issue also with regards to the use of basis functions; whether plane wave hypothesis is being utilized or it is a linear combination of all atomic orbitals and various other such considerations based on which the results may vary significantly. It should be mentioned that there is no study in open literature comparing different DFT methods for studying phase transition in Si.

The primary objective of this study is to determine which interatomic potential (out of the available ones and being commonly utilized) works best to predict phase transformation in Si under hydrostatic compression as reported from experimental observations. Apart from that another objective of the study is to determine the orientational dependence of cubic diamond crystal of Si under uniaxial compressive pressure loading (typically observed in static compression experiments involving universal testing machines and dynamic compression experiments involving Kolsky-bars).

In this regard, it should be noted that there has been no work in published literature which investigates the topic of orientational dependence of Si under isentropic/static pressure loads (not involving any significant rise in temperature). Under shock loads, involving both rise in temperature and pressure, there are studies which focuses on the issue of orientational dependence [23,24]. It should be noted that compression as a result of shock loading is significantly different compared to the uniaxial stress type compressive loading condition considered in this literature. Uniaxial strain conditions are representative of shock loading situations in high velocity impact experiments. Uniaxial stress type loading has been considered in this study apart from hydrostatic loading. Differences in between different types of compressive loading scenarios have been described in details in literature [25].

2. Simulation methodology

Cubic diamond single crystal silicon with periodic boundary conditions in all orthogonal directions is created and equilibrated using isothermal-isobaric, NPT ensemble integration scheme (for 80 ps with time step of 1 fs using Nose-Hoover Thermostat and barostat algorithm) at ambient temperature and pressure conditions. The observed density of the final equilibrated Si using different interatomic potentials are 2.32 ± 0.02 gm/cc in comparison to that of recorded experimental density of 2.329 gm/cc. Utilizing suitable interatomic potentials these samples were subjected to hydrostatic and uniaxial (along different directions such as [0 0 1] representing high symmetry direction, [0 1 1] and [1 1 1] representing low symmetry directions) compressive loading. Non-equilibrium molecular dynamics(NEMD) simulation have been performed on the samples to investigate the phase transition of single crystal cubic diamond silicon. For details of samples with regards to orientation, size and number of atoms used in simulation, please see Table 1.

Simulation of the hydrostatic compression corresponds to experimental Diamond Anvil Cell compression. Hydrostatic pressure was ramped up by 1 GPa pressure in 20 ps followed by relaxation for 2 ps using NPT ensemble. The sample was loaded till the final pressure

Table 1
Necessary details of the initial configurations of samples.

Sample No.	Orientations			Size(Å)			No. of atoms
	X	Y	Z	X	Y	Z	
1.	⟨1 0 0⟩	⟨0 1 0⟩	⟨0 0 1⟩	271.869	271.869	2718.69	10,000,000
2	⟨$\bar{1}$ 0 0⟩	⟨0 $\bar{1}$ 1⟩	⟨0 1 1⟩	276.988	276.988	2769.88	10,575,360
3	⟨2 1 1⟩	⟨0 $\bar{1}$ 1⟩	⟨1 1 1⟩	268.627	259.089	2902.43	10,053,120