Computational Materials Science 128 (2017) 121–126

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

Simulations of the inelastic response of silicon to shock compression

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ARTICLE INFO

Article history:
Received 16 June 2016
Received in revised form 27 October 2016
Accepted 6 November 2016
Available online 18 December 2016

Keywords:
Silicon
Shock-compression
Molecular dynamics
Phase-change
Lagrangian elastic code

ABSTRACT

Recent experiments employing nanosecond white-light X-ray diffraction have demonstrated a complex response of pure, single crystal silicon to shock compression on ultra-fast timescales. We present here details of a Lagrangian code which tracks both longitudinal and transverse strains, and successfully reproduces the experimental response by incorporating a model of the shock-induced, yet kinetically inhibited, phase transition. This model is also shown to reproduce results of classical molecular dynamics simulations of shock compressed silicon.

1. Introduction

The response of matter to rapid shock compression has been a field of study for well over a century. One material that has been a subject of particular interest for several decades is single crystal silicon [1–4]. Given that this element can be manufactured in an almost perfect, defect-free form, it might first appear to be an ideal test-bed for studying the fundamental physics of shock compression. However, in many ways the opposite has seemed to be true, in that despite many attempts, a full understanding of how such perfect single crystals react at the lattice level to rapid uniaxial loading has remained surprisingly elusive, with apparently differing results and interpretations being put forward between gas gun experiments [5,6] and those performed on a shorter timescale employing laser-plasma-based drivers [7].

However, recent work employing nanosecond white-light Laue diffraction to diagnose laser-driven shocks in single crystal silicon shocked along the [100] axis has re-confirmed that a complex elastic response, first observed by Loveridge-Smith et al. [7], indeed occurs [8]. This work showed that when silicon is shock-compressed to stresses in the regime of a few 10's of GPa on nanosecond timescales, a leading double elastic-wave structure can form in compression, which, upon breakout from a free surface, can also result in a state of elastic tension.

In the work of Ref. [8], it was shown that the observed experimental results were consistent with simulations based on a simple Lagrangian code, which incorporated in an empirical manner a pressure dependent, but kinetically inhibited (delayed) phase transition, with the complex elastic behaviour being a result of the relatively large volume collapse associated with the change in phase. However, within the work of [8], for the sake of brevity no details of the code, and the assumptions that underpin it, were presented. Within the paper presented here we rectify this situation, giving a full description of the code, and a description of how states of strain within the shocked sample are determined, allowing us to predict time-dependent X-ray diffraction patterns that can be compared directly with experimental results.

Before describing the code in detail, we briefly recap the main features of the particular experiment it is designed to model. Within this experiment, 30 μm thick samples of [001] silicon, over-coated with a 15 μm layer of parylene-N ablator, were shock compressed by irradiation with a 5 ns square pulse of 351 nm light at an irradiance of 4×10^{14} W cm^{-2}. At a time of 5 ns after the onset of this drive pulse, a laser-plasma-generated, quasi-white-light beam of X-rays (3–10 keV) were diffracted from the rear undriven surface of the target, with the X-rays being collimated such that they impinged on a central region of 0.8 mm diameter directly opposite the 5 mm diameter drive spot. The timing of the X-rays was such that they were diffracted from as-yet unshocked material, as well as the elastic compression waves that moved toward the rear surface of the target, and also the regions of tension that formed upon shock breakout. Further and more complete details of the experimental set-up can be found in Ref. [8].

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http://dx.doi.org/10.1016/j.commatsci.2016.11.006
0927-0256/

In order to model the sample response we utilise and adapt a simple two-step algorithm to solve the elasticity equations within a Lagrangian framework put forward by Horie [9]. As we show below, this model allows us to keep track of the time and space-dependent elastic strains within the sample, and subsequently from them predict X-ray diffraction patterns. This approach has recently proven to be successful in modelling femtosecond diffraction patterns recorded from copper as it is shock compressed on picosecond timescales [10,11].

The paper is laid out in the following manner. Firstly the relevant phase transition in silicon is introduced, followed by the formalism for the elastic code, including the equations that govern the phase transition. We discuss the necessary prerequisites for the code's function, and the extent of fitting required. Finally, we show a comparison between the new code and molecular dynamics simulations, before making concluding remarks.

2. Theory

2.1. Cubic diamond → β-Sn transition in silicon

We start by summarising the physics of the relevant phase transition in silicon; the first order transition from the ambient phase, which has a cubic diamond (cd) structure, to the higher pressure β-Sn structure, which occurs at 13 GPa on the hydrostat [1]. The crystal structures for these two phases are shown in Fig. 1. It should be noted that although we choose to refer to β-Sn in this paper, a similar analysis would apply to the closely related Imma phase, which is found to become stable between 15–32 GPa.

Several features of this transition lead to complexity in its modelling. Firstly, silicon exhibits a large volume collapse of 21% between cd and β-Sn. Molecular dynamics simulations suggest that, for compression of single crystals along [001], this proceeds via a significant contraction along the compression direction, and an accompanying transverse expansion. Moreover, previous studies suggest a significant enthalpy barrier, and thus kinetic effects must be considered [12,8].

In the following sections we will develop the theory of the Lagrangian elasticity (LE) code to allow for meaningful simulation of such a system.

2.2. Lagrangian elasticity code formalism

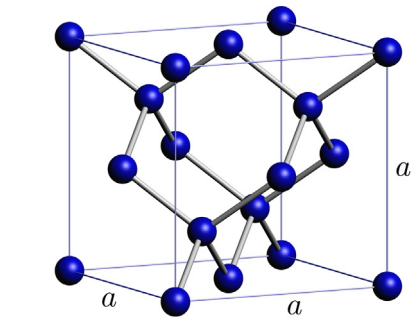
The approach described here is based on the two step integration of the elasticity equations described by Horie [9,10]. This method allows for solution of the 1d Lagrangian wave equations of Taylor [13]

$$\rho_0\left(\frac{\partial u}{\partial t}\right)+\left(\frac{\partial \sigma_n}{\partial z}\right)=0, \tag{1}$$

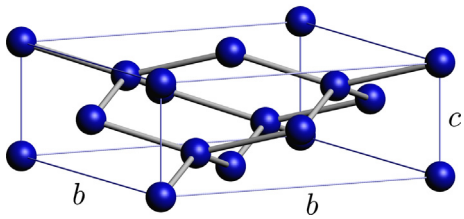
$$\left(\frac{\partial u}{\partial z}\right)+e^{-\epsilon_n}\left(\frac{\partial \epsilon_n}{\partial t}\right)=0, \tag{2}$$

where ρ_0 is the initial density of the material in a Lagrangian element, u is its position, σ_n the normal stress, and ϵ_n the normal true strain. Note the use of true, rather than engineering strain, leads to an additional factor of $e^{-\epsilon_n}$ not present in Taylor's description.

By utilising the equations of elasticity we can extend our model, tracking both longitudinal and transverse properties, while retaining a 1d integration scheme. We modify Taylor's treatment by considering a relation based on the full, strain dependent, compliance tensor, $\epsilon_i = S_{ij}(\bar{\epsilon})\sigma_j$. In order to allow for the strain dependence of the compliance tensor, we define the components in terms of small changes in stress and strain, which in the case of tetragonal symmetry gives



(a) Cubic diamond (cd)



(b) Beta (β)

Fig. 1. The two Si phases of interest. (a) The cubic diamond phase: a face-centered cubic lattice with a basis of $[(0,0,0),(\frac{1}{4},\frac{1}{4},\frac{1}{4})]$. (b) The β-Sn phase: a tetragonal lattice, with the same basis as cd. We use values of $a = 5.431$ Å, $b = 6.897$ Å and $c = 2.548$ Å, found by minimising the energy of the unit cell in MD, while holding the cell at zero external pressure.

$$\begin{pmatrix}\Delta\epsilon_t\\ \Delta\epsilon_t\\ \Delta\epsilon_n\\ 0\\ 0\\ 0\end{pmatrix}=\begin{pmatrix}S_{11} & S_{12} & S_{13} & & & S_{16}\\ S_{12} & S_{11} & S_{13} & & & -S_{16}\\ S_{13} & S_{13} & S_{33} & & & \\ & & & S_{44} & & \\ & & & & S_{44} & \\ S_{16} & -S_{16} & & & & S_{66}\end{pmatrix}\begin{pmatrix}\Delta\sigma_t\\ \Delta\sigma_t\\ \Delta\sigma_n\\ 0\\ 0\\ 0\end{pmatrix}. \tag{3}$$

where we have employed Voigt notation, and have defined $\epsilon_3 = \epsilon_n$ and $\epsilon_1 = \epsilon_2 = \epsilon_t$ (with similar definitions for the stress tensor), where subscripts n and t denote directions normal to and transverse to the compression direction respectively. We assume here that the stress and strain tensors are diagonalised, and that the two transverse stresses, and thus strains are equal. This leads to stress/strain relations of the form

$$\Delta\epsilon_n = S_{33}\Delta\sigma_n + 2S_{13}\Delta\sigma_t, \tag{4a}$$

$$\Delta\epsilon_t = S_{13}\Delta\sigma_n + S_{1112}\Delta\sigma_t, \tag{4b}$$

where we have relabeled $(S_{11}+S_{12}) = S_{1112}$ to make clear that the relations require only three independent elastic constants. These equations are analogous to Eq. (4) in Ref. [10][1], where only two elastic constants are needed to describe a cubic system.

2.3. Phase transition

The formalism outlined above allows for integration of stress in a purely elastic solid. Previous work has used an additional model for plastic relaxation due to dislocation motion, however, in this

[1] It should be noted that the original paper [10] contains a typographical error in these equations, and that they should read, in the original nomenclature:

$$\sigma_n = \left(K+\frac{4\mu'}{3}\right)\epsilon_n^e + \left(2K-\frac{4\mu'}{3}\right)\epsilon_t^e,$$
$$\sigma_t = \left(K-\frac{2\mu'}{3}\right)\epsilon_n^e + \left(2K+\frac{2\mu'}{3}\right)\epsilon_t^e.$$