# Phase field approach for stress- and temperature-induced phase transformations that satisfies lattice instability conditions. Part I. General theory

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## ABSTRACT

Recently, results of molecular dynamics (MD) simulations were obtained for the crystal lattice instability conditions for the phase transformations (PTs) between semiconducting Si I and metallic Si II under action of all six components of the stress tensor (Levitas et al. (2017a, b)). These conditions are linear in terms of stresses normal to the cubic faces of Si I and are independent of the shear stresses. In the current paper, we (a) formulated the requirements for the thermodynamic potential and transformation deformation gradient tensors and (b) developed a phase field approach (PFA) for the stress-induced martensitic PTs for large strains while allowing for interfacial stresses, which are consistent with the obtained instability conditions. The general system of equations for coupled PFA and nonlinear elasticity is presented. Crystal lattice instability criteria are derived within a PFA, and it is proven that they are independent of the prescribed stress measure. In order to reproduce the lattice instability conditions obtained with MD: (a) one has to use the fifth degree polynomial interpolation functions of the order parameter for all material parameters; (b) each component of the transformation strain tensor should have a different interpolation functions; and (c) the interpolation functions for tensors of the elastic moduli of all ranks should have zero second derivatives for the parent and product phases, so that terms with elastic moduli, which are nonlinear in stresses, do not contribute to the lattice instability conditions. Specific interpolation and double-well functions have been derived for all parts of the Helmholtz free energy and for two models for the transformation deformation gradient. For these models, explicit expressions for the Ginzburg-Landau equations and lattice instability conditions are derived. Material parameters have been calibrated using results of MD simulations. In Part II of this paper, the developed model is further refined and studied, and applied for the finite element simulations of the nanostructure evolution in Si under triaxial loading.

## 1. Introduction

The PFA is broadly used for modeling martensitic PTs (Artemev et al. (2001); Chen (2002); Jin et al. (2001); Levitas and Preston (2002a,b); Levitas et al. (2003); Mamivand et al. (2014); Mamivand et al. (2013); Paranjape et al. (2016); Rogovoy and Stolbova (2016); Wang and Khachaturyan (2006); Zhu et al. (2017)) and reconstructive PTs (Denoual et al. (2010); Salje (1990)). Here, we will

consider the PT between the parent phase $P_0$ and the product phase $P_1$, without including multiple symmetry-related martensitic variants for brevity. This PT is parameterized by the order parameter $\eta$, with $\eta = 0$ for the phase $P_0$ and $\eta = 1$ for the phase $P_1$. The Helmholtz free energy consists of both the local part and gradient energy $0.5\beta|\nabla\eta|^2$, the latter penalizes the interface energy. Some theories Artemev et al. (2001); Chen (2002); Jin et al. (2001); Wang and Khachaturyan (2006)) did not specify the physical meaning of the order parameter, but others utilized some components of the strain tensor as the order parameters (Barsch and Krumhansl (1984); Falk (1983); Finel et al. (2010); Jacobs (1992); Vedantam and Abeyaratne (2005)).

The key points in formulating the local thermodynamic potential is to interpolate all material properties along the transformation paths and to introduce proper energetic barriers between phases. The only requirements imposed in Artemev et al. (2001); Barsch and Krumhansl (1984); Chen (2002); Falk (1983); Finel et al. (2010); Jacobs (1992); Jin et al. (2001); Vedantam and Abeyaratne (2005); Wang and Khachaturyan (2006) for the local energy are that it has as many local minima as there are phases (including martensitic variants) that one considers and that the energy is invariant with respect to an exchange of any symmetry-related martensitic variants, which is sufficient for qualitatively reproducing a complex multivariant martensitic structure. As a consequence, the order parameter for the product phase $P_1$ was not fixed to 1 but depended on stresses and temperature. This did not allow for precisely reproducing the known material properties of the product phase. In Levitas and Preston (2002a, b); Levitas et al. (2003) additional important requirements have been formulated, which will be discussed below and which results in constraint Eqs. (12) and (16) for an interpolation function $\varphi_m(\eta)$ for any material property $M$. Also, the PT criteria for direct and reverse PTs should follow from the crystal lattice instability criteria. The PFAs that satisfy these requirements have been developed within the fourth degree potential $(2 - 3 - 4)$ in terms of the order parameter and the six degree potential $(2 - 4 - 6)$ in terms of the even degrees of $\eta$. These theories reproduce desired stress-strain curves. The order parameter in Levitas and Preston (2002a, b); Levitas et al. (2003) was related to the transformation strain tensor rather than to the total strain. In fact, we did not find a way to impose these constraints for the theories based on the total strain-related order parameters, that is why they will not be considered any further. These requirements are important to ensure that the thermodynamically equilibrium material properties of both parent and product phases are reproduced in the PFA, which was not the case in the previous theories. As it is shown in Levitas and Preston (2002a), the transformation strain and stress hysteresis in Artemev et al. (2001); Barsch and Krumhansl (1984); Chen (2002); Falk (1983); Finel et al. (2010); Jacobs (1992); Jin et al. (2001); Vedantam and Abeyaratne (2005); Wang and Khachaturyan (2006) strongly depend on temperature, and stress-strain curves do not reproduce the main features observed in experiments for shape memory alloys, steels, and some ceramics.

The PFA developed in Levitas and Preston (2002a, b); Levitas et al. (2003) was applied for modeling microstructure evolution during multivariant martensitic PTs (Cho et al. (2012); Idesman et al. (2008); Levitas and Lee (2007); Levitas et al. (2010)). It was also generalized for large strain formulation in Levitas (2013a); Levitas et al. (2009) with corresponding finite element simulations in Levin et al. (2013). This approach was based on the interpolation of the transformation deformation gradient between parent and product phases.

It was found in Tuma and Stupkiewicz (2016); Tuma et al. (2016) that such an interpolation does not allow for properly separating the volumetric part of the transformation strain and change in shape. In particular, for twinning it produces volume-conserving shear after complete transformation only, while there is a volume change during the transformation. Interpolation of the logarithmic transformation strain which keeps the volume conserved was suggested in Tuma and Stupkiewicz (2016); Tuma et al. (2016). This was done within the theory which uses volume fraction of phases as order parameters, which is suitable for the microscale models (as in Idesman et al. (2005); Levitas et al. (2004)) but not for the nanoscale ones. However, it was shown in Basak and Levitas (2017) that interpolation for logarithmic transformation strain produces artificial elastic interfacial stress for a variant-variant interface which are more than two times larger than for the interpolation used in Levitas (2013a); Levitas et al. (2009).

Interfacial stresses for martensitic PTs were introduced in Levitas (2013b,c, 2014a); Levitas and Javanbakht (2010) for small strain formulation. These theories were generalized for large strains in Levitas (2014b) for isotropic interface energy and stresses and in Levitas and Warren (2016) for an anisotropic case. However, the lattice instability conditions in Levitas (2013a) were not yet extended for the model with interfacial stresses, which will be done in the current study.

In the previous theories the constraint on the interpolation functions were limited to the values of the functions and their first derivatives at $\eta = 0$ and 1, see Eqs. (12) and (16). The lattice instability conditions operate with the second derivatives of the interpolation functions (Levitas (2013a); Levitas and Preston (2002a,b); Levitas et al. (2003)); however, since they were not known from experiment or atomistic simulations, no limitations were imposed. At the same time, lattice instability conditions are crucial for understanding barrierless nucleation during martensitic and reconstructive PTs for relatively low temperatures, when thermal fluctuations play a minor part (Olson and Cohen (1972, 1986); Olson and Roytburd (1995)). They are especially important for high pressure PTs for which transformation pressure is much higher than the phase equilibrium pressure. For example, the phase equilibrium pressure for the PT from hexagonal to superhard cubic BN at room temperature is even negative (i.e., cubic BN is stable at atmospheric pressure), see Solozhenko (1995); however, highly disordered hexagonal BN does not transform up to at least 52 GPa, i.e., lattice instability pressure is even higher (Ji et al. (2012)).

Recently, lattice instability conditions for the PTs between semiconducting Si I and metallic Si II under action of all six components of the stress tensor were found with the help of MD simulations (Levitas et al. (2017a, b)). These conditions are linear in terms of stresses normal to the cubic faces of Si I, $\sigma_i$, and are independent of the shear stresses. Thus, the instability conditions can be presented in a 3D stress space $\sigma_i$ as two planes (Figs. 4 and 5), one for direct and another for reverse PTs. It is important that these planes are not parallel and consequently not consistent with the instability conditions within $2 - 3 - 4$ and $2 - 4 - 6$ thermodynamic potentials. In addition, after the intersection of these planes they coincide for part of the stress space (Fig. 5). Thus, new conditions have to be formulated for the interpolation functions in order to make phase field equations consistent with the lattice instability conditions obtained with MD simulations and new PFA should be developed that satisfies these conditions.