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Ab initio simulation of a tensile test in MoSi₂ and WSi₂

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ABSTRACT

The tensile test in transition metal disilicides with C11_b structure is simulated by *ab initio* electronic structure calculations using full potential linearized augmented plane wave method (FLAPW). Full relaxation of both external and internal parameters is performed. The theoretical tensile strength of MoSi₂ and WSi₂ for [001] loading is determined and compared with those of other materials.

INTRODUCTION

Transition metal (TM) silicides are considered as a very promising basis for a new generation of high-temperature structural materials that can significantly improve the thermal efficiency of energy conversion systems and advanced engines. The reason is that at high temperature they combine the ductility and thermal conductivity of metals with high strength and corrosion resistance of ceramics. Intrinsic oxidation resistance is due to the formation of silicon oxide films at surfaces and high creep strength is related to low diffusion coefficients. The melting temperature is much higher than that of Ni-based superalloys or Ni and Ti-based aluminides and is comparable to that of silicon-based ceramics. The largest impediment is low ductility and/or toughness at ambient temperatures.

The purpose of this paper is to investigate, from first principles, the electronic structure and ground state of TM-disilicides, MoSi₂ and WSi₂, with C11_b structure, to simulate a tensile test for ideal crystal without defects, including full relaxation of both external and internal parameters, and to determine thus the theoretical tensile strength for the [001] loading.

TENSILE TEST SIMULATION

The tensile strength of materials is usually limited by presence of internal defects, mostly dislocations. In a defect-free crystal, the tensile strength is several order of magnitude higher and is comparable with elastic moduli. Most of the calculations of theoretical (ideal) strength is based on empirical potentials with the parameters adjusted to experimental data. However, most of these experimental data correspond to the equilibrium ground state. Therefore, the

semiempirical approaches adapted to the equilibrium state may not be valid for materials loaded close to their theoretical strength limits.

In the first-principles (*ab initio*) electronic structure calculations, we start from the fundamental quantum theory. The only input is atomic numbers of the constituent atoms and, usually, some structural information. This approach is reliable even for highly non-equilibrium states.

To simulate the tensile test, we first calculate the total energy of the material in the ground state. Then, in the second step, we apply some elongation of the crystal along the loading axis (in the [001] direction in the present case; the loading axis is denoted as 3) by a fixed amount ε_3 that is equivalent to the application of a tensile stress σ_3 . Subsequently, we fully relax both the stresses σ_1 and σ_2 in the directions perpendicular to the axis 3 as well as the internal degrees of freedom. In this way, we find the contractions ε_1 and ε_2 which correspond to zero tensile stresses σ_1 and σ_2 and the new values of internal parameters. The $C11_b$ structure is tetragonal and keeps its tetragonal symmetry during the tensile test along the [001] axis. Therefore, $\sigma_1 = \sigma_2$ and we minimize the total energy as a function of lattice parameter a and internal parameter Δ (defined e.g. in [1, 2], see also Fig. 2).

The tensile stress σ_3 is then given by

$$\sigma_3 = \frac{2}{c_0} \frac{\partial E}{\partial \varepsilon_3} \frac{1}{a^2},$$

where E is the total energy per basis (i.e. one TM atom and two Si atoms) and c_0 is the lattice parameter in the direction of loading in the ground state. The inflexion point in the total energy dependence yields the maximum of the tensile stress; if some other instability does not occur before reaching the inflexion point, it also corresponds to the theoretical tensile strength.

DETAILS OF THE CALCULATIONS

In order to obtain reliable *ab initio* total energies of materials during tensile test simulations, the methods using a shape approximation of the crystal potential (for example spheroidization, as in the LMTO-ASA method or in standard KKR and APW approaches) are not adequate [3, 4]. Instead, full-potential treatments must be employed. In this study we utilized the full-potential linearized augmented plane waves (FLAPW) code described in detail in [5]. The electronic structure calculations were performed self-consistently within the local density approximation (LDA).

When simulating the tensile test, crystal lattices are severely distorted and some atoms may move very close together. Therefore, the muffin-tin radii must be sufficiently small to guarantee non-overlapping of the muffin-tin spheres at every stage of the test. We use the muffin-tin radii equal to 2.3 a.u. for transition metal atoms and 2.1 a.u. for silicon. These are kept constant in all calculations presented here. The product of muffin-tin radius and the maximum reciprocal space vector, $R_{MT}k_{max}$, is equal to 10, the maximum l value for the waves inside the atomic sphere, l_{max} , and the largest reciprocal vector \mathbf{G} in the Fourier expansion of the charge, G_{max} , are set to 12 and 15, respectively, and the number of \mathbf{k} -points in the first Brillouin zone is equal to 2000.

RESULTS AND DISCUSSION

First we determined the ground state properties for MoSi₂ and WSi₂. The values of lattice parameters, a and c , internal parameter Δ , and ratio of calculated ground-state volume over the experimental one, V/V_{exp} (Ref. [2]), are summarized and compared with other calculations and experiments in Table 1. Our results are in very good agreement with both experimental and previous theoretical data.

	parameters of MoSi ₂			
	a	c	Δ	V/V_{exp} [2]
FLAPW-this work	6.004	14.689	0.0021	0.974
exp. Ref. [2]	6.051	14.836	0.0019	1.000
exp. Ref. [6]	6.059	14.830	0.0020	1.002
exp. Ref. [7]	6.047	14.855	–	0.999
FLAPW Ref. [1]	6.089	14.897	0.0022	1.017
LMTO-FP Ref. [8]	6.021	14.740	–	0.984
LMTO-ASA Ref. [9]	6.026	14.780	–	0.988
	parameters of WSi ₂			
FLAPW-this work	6.020	14.721	0.0013	0.975
exp. Ref. [2]	6.068	14.870	0.0014	1.000
exp. Ref. [7]	6.070	14.891	–	1.010
FLAPW Ref. [1]	6.104	14.866	0.0018	1.020

Table 1: Ground-state lattice parameters of MoSi₂ and WSi₂.

The theoretical tensile strengths are summarized in Table 2. They are comparable with the tensile strengths predicted for, e.g., NiAl [10] or W [11]. Unfortunately, to the best of our knowledge there are no measurements of ideal tensile strength of MoSi₂ and WSi₂ (whiskers, nanoindentation) and, therefore, we were not able to compare our results with the experimental ones.

material	MoSi ₂	WSi ₂	NiAl [10]	W [11]
structure	C11 _b	C11 _b	B2	A2
σ_{th} (GPa)	37	38	46	29
ε_3	0.18	0.18	0.21	0.12

Table 2: Theoretical tensile strengths σ_{th} of MoSi₂ and WSi₂ and the corresponding ε_3 compared with those of other materials.

In Figure 1, we display the dependences of total energy E , internal parameter Δ , tensile stress σ_3 and the lattice parameter a on ε_3 in MoSi₂. The total energy has a parabolic shape around the minimum; it becomes almost flat in the neighbourhood of the inflexion point corresponding to the maximum of tensile stress. During the deformation the value of lattice constant perpendicular to the loading axis decreases nearly monotonously. On the other hand, the internal parameter increases with increasing ε_3 except for small neighbourhood of

the ground state where it is almost constant. Let us note that the tendency of Δ to increase with ε_3 is in agreement with the interpretation of recent experimental data [12].

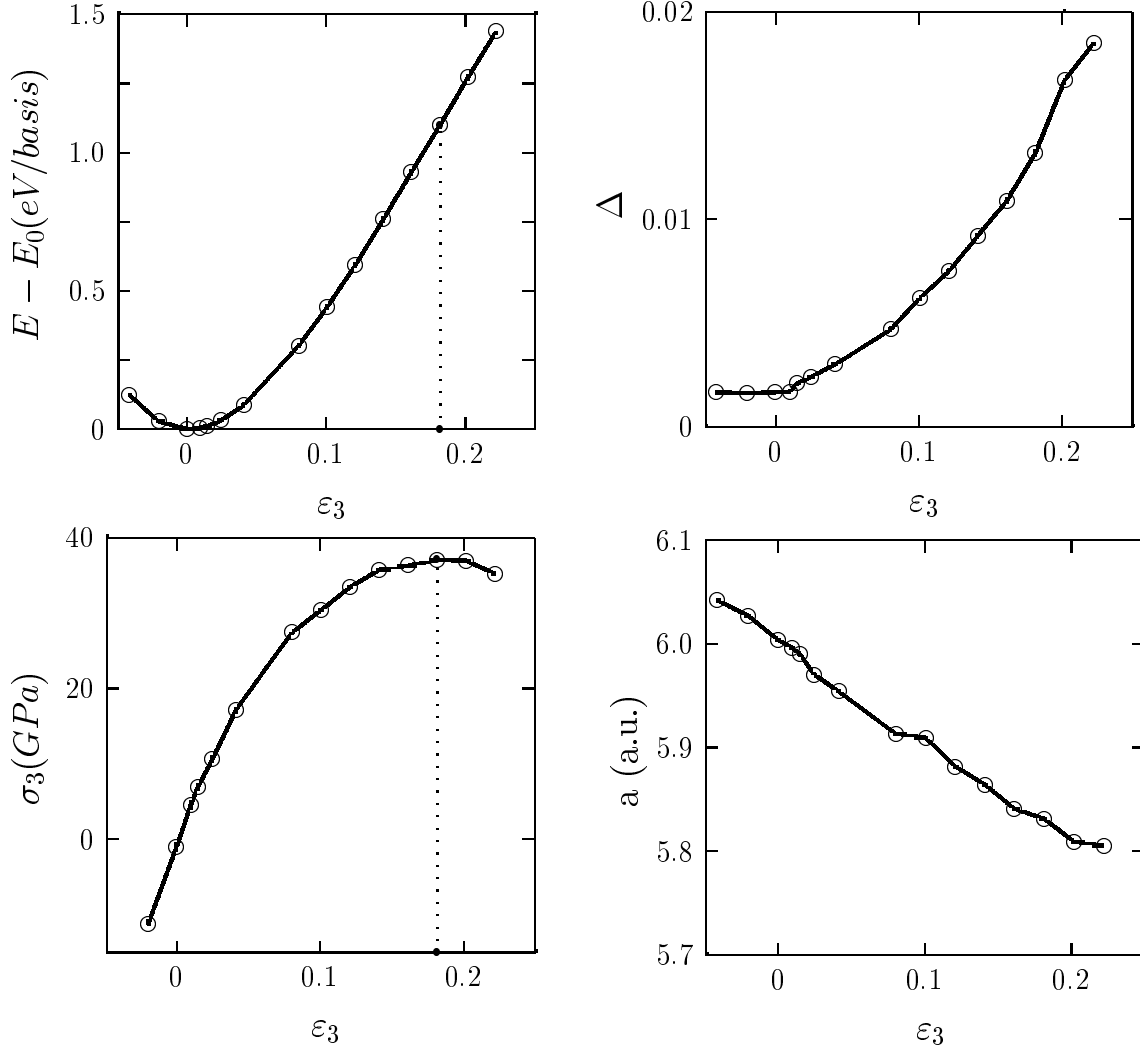


Figure 1: Variations of total energy E per basis (one Mo and two Si atoms), internal parameter Δ , tensile stress σ_3 and lattice parameter a during the simulation of the tensile test in MoSi_2 . Here E_0 is the ground-state energy and ε_3 is the strain in the [001] direction. The position of the inflexion point in the energy dependence and the maximum of the tensile stress are denoted by a dashed line.

In Figure 2 we remind the definition of the internal parameter Δ [1, 2] and show four different types of bonds between constituent atoms of the repeat cell in MoSi_2 . It was proposed by Tanaka et al. [2] that the Mo-Si bonds along the [001] direction (thin dash-dotted lines in Fig. 2) are weaker than the Mo-Si bonds in the other directions (thick dashed lines in Fig. 2) and, on the contrary, Si-Si bonds along the [001] direction (thick full lines in Fig. 2) are stronger than those in the other directions (thin dashed lines in Fig. 2). Our calculations confirm this suggestion regarding the Mo-Si bonds. In the $C11_b$ structure,

four Mo atoms in the (001) plane form a square of side a which constitutes the basis of a bipyramid completed by two Si atoms above and below the center of this square. It seems that the atoms forming these bipyramids try to keep together during the tensile test in the [001] direction. Namely, the edges of the bipyramid are the Mo-Si bonds the length of which is nearly constant during the test even when exceeding the theoretical tensile strength (full circles in the left-hand part of Fig. 2). In agreement with Ref. [2], they can be denoted as "strong" bonds. The Si-Si bonds in the [001] direction, constituting the height of the bipyramid, are elongated under tension (open triangles in Fig. 2); this is related to the decrease of the lattice parameter a (Fig. 1). However, the Si-Si bonds in the other directions are extended approximately in the same way (open circles in Fig. 2).

The behavior of the Si-Si bonds is somewhat different under compression. The length of the non-[001] Si-Si bonds is nearly constant whereas the length of the [001] Si-Si bonds changes significantly, similarly as the length of the [001] Mo-Si bonds. Therefore, we can distinguish the "strong" and "weak" Mo-Si bonds, but it is not possible to introduce the "strong" and "weak" Si-Si bonds. The situation in WSi_2 is similar.

Let us note here that the relaxation of the internal parameter Δ during the tensile test is crucial. If Δ were kept constant the [001] Si-Si and Mo-Si bonds would behave very similarly. The same would be true for non-[001] Si-Si and Mo-Si bonds.

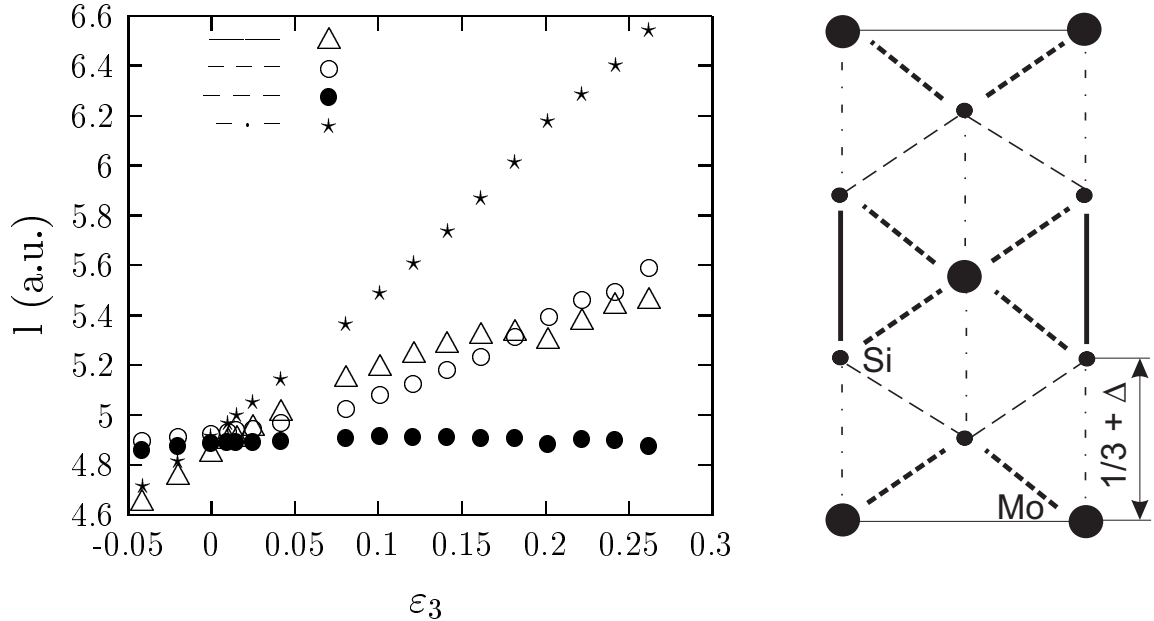


Figure 2: Variations of the length of atomic bonds during the tensile test simulation in MoSi_2 . The right-hand side of the figure displays the (110) plane in MoSi_2 with the $C11_b$ structure; the Si atoms are represented by small circles and Mo atoms by large circles. Bonds between Mo-Si and Si-Si atoms are shown as thick dashed and thin dash-dotted lines (the "strong" and "weak" Mo-Si bonds) and thick full and thin dashed lines (the [001] and the other Si-Si bonds). The internal parameter Δ is defined as deviation from the ideal value of $1/3$ [1, 2].

CONCLUSIONS

We simulated the tensile test in ideal MoSi₂ and WSi₂ loaded along the [001] axis using the first-principles full-potential electronic structure calculations and determined theoretical tensile strength of those materials. The analysis of bond lengths variation under an uniaxial stress shows that, in accordance with Ref. [2], it is possible to distinguish "strong" and "weak" TM-Si bonds but the behavior of Si-Si bonds is more complex and exhibits a tension-compression asymmetry.

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