

## Analysis of the transformation strain associated with the hexagonal-to-orthorhombic transition in $Ti_3Sn$

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*This review is dedicated to the analysis of the hexagonal-to-orthorhombic transition into  $Ti_3Sn$ . The place of martensitic transformation into broader category of displacive (diffusionless) transformations is shown from the point of view of a transformation strain associated with the transition. The transformation strain regarded to the transformation into  $Ti_3Sn$  was analyzed and the displacive shear-dominant (martensitic) origin of this transition was shown. The latter part of the work is dedicated to the comparison of the volume change and transformation temperature in  $Ti_3Sn$  with those into other materials undergoing martensitic transformation.*

**Keywords:** *martensitic transformation, transformation strain, shape memory alloys.*

### Introduction

Intermetallic compound  $Ti_3Sn$  and some  $Ti_3Sn$ -based alloys were reported to exhibit high damping capacity in the frequency range 0,10—10 Hz [1, 2] and unusual mechanical properties such as low Young's modulus and good room temperature plasticity [3—5]. Such properties in conjunction with strength and high melting temperature are of interest for materials for aerospace components operating under vibration and extremely difficult environment. According to [1, 2] the origin for high damping and unusual mechanical behaviour is a phase transformation occurring in the intermetallic compound  $Ti_3Sn$  at about 350 K which is accompanied by a peak of damping capacity and a reduction of Young's modulus.

According to [6] the room temperature  $Ti_3Sn$  has orthorhombic  $Cmcm$  structure (space group № 63, lattice parameters  $a = 0,585$  nm,  $b = 1,034$  nm,  $c = 0,475$  nm) and undergoes a phase transformation to the hexagonal  $DO_{19}$  structure (space group № 194, lattice parameters  $a = 0,5938$  nm,  $c = 0,4749$  nm) under heating. Microstructural observations by TEM have revealed self-accommodated twinned microstructure with compound (110) twins in room-temperature single-phase  $Ti_3Sn$  [2—6].

For the phase transition into  $Ti_3Sn$  the ratio between transformation temperature and melting temperature

$$\frac{T_{trans}}{T_{melt}} = \frac{343 \text{ K}}{1943 \text{ K}} = 0,18$$

is low enough to suggest the diffusionless nature of the transition. A

formation of specific twinned self-accommodated microstructure [3—6] points to its martensitic origin. Most of them undergoes diffusion-displacive transition and several intermetallic compound doesn't undergo any transformation. Ni<sub>3</sub>Sn and Fe<sub>3</sub>Mg were reported to exhibit martensitic transformation, but these two

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compounds have ordered cubic DO<sub>3</sub> or L1<sub>2</sub> high-temperature phase which transforms into hexagonal phase [7]. However the Ti<sub>3</sub>Sn compound crystallizes into hexagonal close-packed DO<sub>19</sub> superlattice from the melt. It is unusual for the case of martensitic transformations because the high-temperature phase is mostly cubic. Into our previous work [6] it is shown that hexagonal lattice transforms to an orthorhombic *Cmcm* structure in Ti<sub>3</sub>Sn. Carrying out analysis of the transformation strain we aimed to show the martensitic (displacive) origin of the transition, and compare it with martensitic transformations into other alloys and intermetallic compounds.

### Classifications of diffusionless phase transitions

Buerger in 1951 have made a classification of phase transformations into solid state based on the mechanism of the transition and on the structural relations between parent and product phases. According to this classification [8] phase transformation can be divided on *reconstructive*, *displacive* and *order/disorder* transformations.

During *reconstructive* transition chemical bonds are broken and new bonds formed. In this case the space group symmetries of parent and product phases are unrelated. Reconstructive transformations are quite abrupt and have no order parameter. These transformations are of first order thermodynamic character, they occur by nucleation and growth, show thermal hysteresis; parent and product phases coexist at equilibrium.

*Displacive* transitions involve the distortion of bonds. In this case of parent and product phases show group/subgroup relations. Low-symmetry phase approaches the transition to higher symmetry-phase continuously. Order parameter exists and measures a 'distance' of the low-symmetry to the high-symmetry structure. These transitions can be of second or weak first order and are characterized by vanishing or small latent heat, volume jump and thermal hysteresis.

During *order/disorder* transitions the structural difference between phases is related to the different chemical occupation of the same crystallographic sites. These transitions have second order character and show vanishing enthalpy and volume changes.

Phase transformations which involve long range diffusion are regarded as reconstructive rather than displacive. However some transformations requiring diffusion also have displacive character;

they are called "diffusionless-displacive transformations" to emphasize their mixed characteristics; sometimes the term "nonferrous bainites" is used for them [9]. Displacive transformations involve atom's displacement within unit cell and long range diffusion is not required.

Cohen et al. in 1979 proposed a classification scheme that identifies a class of displacive transformations. According to his classification [9], martensitic transformation is a subclass of broader category of displacive transformation and is defined as a shear-dominant, lattice-distortive, diffusionless transformation occurring by nucleation and growth. This classification scheme is reproduced on fig. 1.

The first subdivision is made between "*shuffle transformations*", in which kinetic and morphology are dominating by shuffle displacements, and "*lattice-distortive transformations*" which are driven by lattice-distortive strain energy. A shuffle is a coordinated movement of atoms that alters the symmetry or

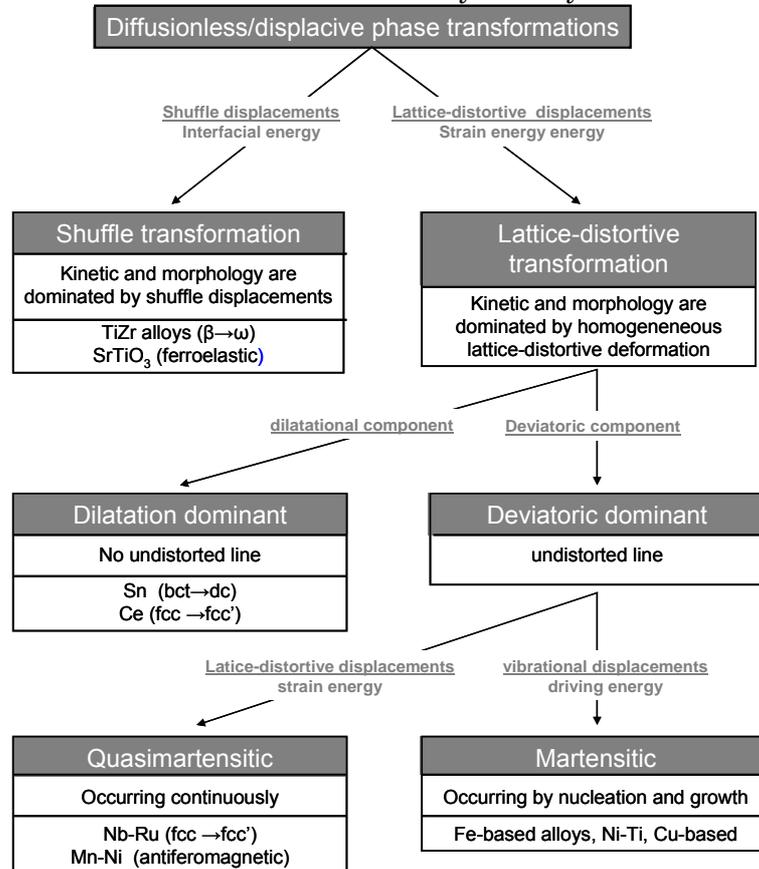


Fig. 1. Classification scheme of diffusionless/displacive phase transformation proposed by Cohen et al. [9].

structure of the crystal and rearranges the atom positions within a unit cell. In the case of lattice-distortive transformations the kinetic

and morphology are dominated by homogeneous lattice-distortive deformation; that is a homogeneous strain that transforms the lattice of parent phase into a lattice of product phase.

A further subdivision was made between “*dilatation-dominant*” and “*deviatoric-dominant*” transformations according to the relative magnitudes of the two components of the homogeneous lattice deformation, i. e., the dilatational and the deviatoric (shear) components.

The next subdivision is made between *martensitic transformation*, occurring by nucleation and growth and *quasimartensitic transformation* occurring continuously according to the relative magnitudes of the lattice-distortive displacements and lattice vibrational displacements. Martensitic transformation requires heterogeneous nucleation and passes through a two-phase mixture of parent and product phases; it is a first order diffusionless phase transformation. In the case of quasimartensitic transformations these two displacements are comparable. In this case a nucleation is not required since domains of product phase appear simultaneously throughout the material by continuous strain modulation.

The “martensitic transformations” are therefore the displacive and shear-dominant phase transformations where the lattice-distortive strain is large enough to dominate the kinetics and the morphology of the transformation. These transformations are characterized by existence of invariant line which remains unchanged during transformation.

The homogeneous lattice-distortive strain can be represented by a matrix according to

$$\mathbf{y} = S \mathbf{x},$$

where the strain  $S$  deforms the lattice vector  $\mathbf{x}$  into a lattice vector  $\mathbf{y}$ . This strain is homogeneous because it transforms straight lines into other straight lines. A spherical body of the parent phase will thus be transformed into another sphere or into an ellipsoidal body. The actual shape of the ellipsoid depends on the deformation along the three principal axes. The ellipsoid obtained after a pure shear intersects the original sphere; hence a set of vectors exist, whose lengths remain unchanged. The initial and the isotropically dilated spheres have no intersection and it is therefore not possible to find a vector whose length has not been changed by the transformation. An undistorted line can only result from a homogeneous lattice deformation if the deviatoric or shear component sufficiently exceeds the dilatational component.

Bilby and Christian [10] stated that for an undistorted line to exist after homogenous deformation, that one of the principal distortions must be greater than unity, one must be less than unity and one must be equal to unity.

Fig. 2 shows one section of the sphere which represents a unit sphere of parent phase and a section of ellipsoid which represents a

product phase. The ellipsoid has a unity principal distortion along y axis, greater than unity distortion along x axis, less than unity distortion along z axis. The invariant line of contact between sphere and ellipsoid can be found; it is A'O'B' line. The plane A'O'B' is a plane of zero distortion and represents a habit plane into crystal lattice.

Thus during martensitic transformation the transformation strain exists and displays the volume change that accompanies the transition. This differs from martensitic transformation from shuffle transformation. Shear-dominant nature of martensitic transformation requires the existence of an invariant line which leads to the necessary condition that one of the principal strains must be greater than unity, one must be less than unity and one must be equal to unity. This differs from martensitic transformation from dilatational transformation. In the latter case all principal

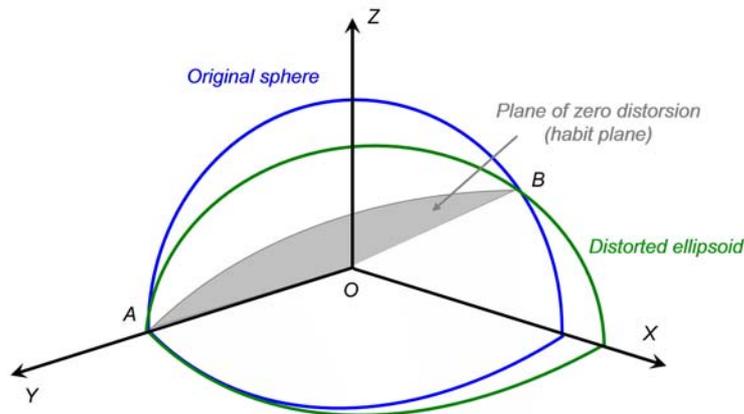


Fig. 2. Bain distortion with one strain of zero, one greater than zero and one less than zero [10].

strains higher or smaller than unity. Quasimartensitic and martensitic transitions can be distinguished by the magnitude of volume change, latent heat or thermal hysteresis.

### Transformation into $Ti_3Sn$

As reported in [6],  $Ti_3Sn$  high temperature hexagonal  $DO_{19}$  ( $P6_3/mmc$ , space group #194) phase transforms to orthorhombic  $Cmcm$  (space group #63) phase. These structures obey group-subgroup relations thus transition from  $P6_3/mmc$  to  $Cmcm$  structure can be displacive. Below we analyze the lattice transformation strain in order to show the existence of an invariant line and to show its shear-dominant origin.

The defined orthorhombic lattice of  $Ti_3Sn$  can be viewed as a distorted hexagonal phase with a relation between lattice parameters:  $a_{orto} \approx a_{hex}$ ,  $b_{orto} \approx \sqrt{3}a_{hex}$ ,  $c_{orto} \approx c_{hex}$ . Based on the dimensions of orthorhombic and hexagonal structures, a crystallographic relation between these phases can be suggested. A schematic of the crystal structures of the orthorhombic and

hexagonal phases and their lattice correspondences are shown in fig. 3, *a*. Therefore the orientation relationships between orthorhombic and hexagonal phases become  $(110)_{\text{ortho}} \parallel (100)_{\text{hex}}$ ,  $\langle 001 \rangle_{\text{ortho}} \parallel \langle 001 \rangle_{\text{hex}}$ . The transition from hexagonal to orthorhombic phase is accompanied by a shortening of 1,5% along *x* axis and an elongation of 0,5% along *y* axis (into orthorhombic basis) leading to the total transformation strain about

$$\det U = \det \begin{pmatrix} \frac{a_{\text{ortho}}}{a_{\text{hex}}} & 0 & 0 \\ 0 & \frac{b_{\text{ortho}}}{\sqrt{3}a_{\text{hex}}} & 0 \\ 0 & 0 & \frac{c_{\text{ortho}}}{c_{\text{hex}}} \end{pmatrix} = \det \begin{pmatrix} 0,985 & 0 & 0 \\ 0 & 1,005 & 0 \\ 0 & 0 & 1 \end{pmatrix} = 0,990$$

which corresponds to reduction of crystal lattice volume of about 1%.

Principal strains of the pure lattice dilatation can be deduced as a difference between calculated transformation strains and unit matrix and equal to  $e_1 = -0,015$ ,  $e_2 = 0,005$ ,  $e_3 = 0$ . This evidences that the strain ellipsoid of orthorhombic phase is contracted along *x* axis, dilated along *y* axis and not

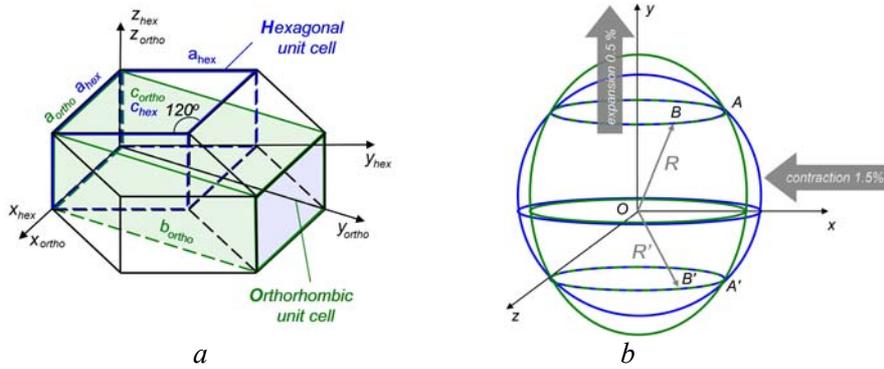


Fig. 3. Lattice correspondence between orthorhombic and hexagonal phases (*a*) [6]; a sphere of the hexagonal phase and dilated ellipsoid of orthorhombic phase (*b*). Undistorted invariant plane can be found as an intersection between the ellipsoid and the sphere (AB and A'B' circles).

altered along *z* direction compared to the strain sphere of the hexagonal phase. fig. 3, *b* shows a sphere representing the hexagonal phase and an ellipsoid of orthorhombic phase which is contracted on  $e_1 = -0,015$  (1,5%) along *x* axis, dilated on  $e_2 = 0,005$  (0,5%) along *y* axis and not altered along *z* axes.

Therefore, an undistorted invariant plane can be found as an intersection between dilated ellipsoid of orthorhombic phase and initial sphere of hexagonal phase (AB and A'B' circles). The presence of an invariant plane indicates that this orthorhombic-to-hexagonal transition is lattice-distortive and shear-dominant, and thus martensitic transformation according to the definition given by Cohen et al [9]. The transformation strain matrix correspondent to

hexagonal-to-orthorhombic transition in  $\text{Ti}_3\text{Sn}$  has  $\lambda_2 = 1$  which points to good geometrical compatibility [11—13] between parent and product phases and might explain small thermal hysteresis measured into previous works [14].

The volume change and transformation strain associated with the hexagonal-to-orthorhombic transition into  $\text{Ti}_3\text{Sn}$  are deducted and this transition can be compared with those into other materials.

Table 1 shows volume changes, transformation and melting temperatures for some alloys and intermetallic compounds undergoing martensitic transformation. Crystal structures of parent phases of the SMAs are either of ordered cubic superlattice (B2) or disordered cubic (fcc) lattice, as seen in the table 2. Product phase have low-symmetry orthorhombic or monoclinic lattice.

It is seen from table 1 and fig. 4 that alloys with lower  $M_s$  have negative volume change while into materials with higher transformation temperature parent B2 phase is denser than martensite. Shape memory materials can be distinguished on low- (if  $M_s < 400$  K) and high-temperature (if  $M_s > 400$  K) ones [22].  $\text{Ti}_3\text{Sn}$  having  $M_s = 343$  K belongs to low-temperature materials. As it seen from fig. 1 the volume change associated with the transition in  $\text{Ti}_3\text{Sn}$  is negative and quite big compared to other low-temperature SMA.

$M_s/T_m$  ratio is a measure of interplay between diffusion processes and martensitic transformation. Into high-temperature SMA diffusion processes

**T a b l e 1. Volume changes, transformation and melting temperatures for some alloys undergoing martensitic transformation**

Composition, % (at.)	$V_P^*$ , $10^{-3}$ nm	$V_M$ , $10^{-3}$ nm	$\Delta V/V_M$ , %	$M_s$ , K	$T_m$ , K [15]	$M_s/T_m$
Ti—49,8Ni	13,7035	13,6573	-0,34	330 [16]	1313	0,24
AuCd	18,3303	18,2177	-0,62	325 [17]	629	0,52
Ag—45Cd	18,1982	17,9047	-1,64	199 [18]	740	0,27
Ti49,5Ni40,5Cu10	13,9091	13,9119	0,02	400 [17]		
Ti—4Mo	17,3389	17,4777	0,79	780 [19]	3146	0,25
Ti—20Nb	17,7254	17,6805	-0,25	493 [20]	2846	0,17
Ti—24Nb	17,7358	17,7249	-0,06	338 [20]	2766	0,12
Zr—49,9Cu	17,5311	17,6094	0,45	413 [21]	1461	0,28
$\text{Ti}_3\text{Sn}$	18,1269	17,9577	-0,94	343 [1]	1943	0,18

\*  $V_P$ ,  $V_M$  — volume per atom for parent and martensitic phase;

$$\Delta V = \frac{V_M - V_P}{V_M} \times 100\% \quad \text{— volume change during martensitic transformation.}$$

**T a b l e 2. Crystal structures of parent and martensitic phases of the alloys listed into table 1 [15]**

Composition, % (at.)	Parent phase		Martensitic phase	
	Space group	Lattice parameters, nm	Space group	Lattice parameters, nm
Ti—49,8Ni	B2	$a = 0,3015$	B19'	$a = 0,2889,$ $b = 0,4120,$ $c = 0,4622,$ $\beta = 96,8^\circ$
AuCd	B2	$a = 0,3322$	B19	$a = 0,4766, b =$ $= 0,3151, c =$ $0,4859$
Ag—45Cd	Cubic	$a = 0,3314$	Orthorhombic	$a = 0,30968,$ $b = 0,48651,$ $c = 0,47536$
49,5Ti— 40,5Ni— 10Cu	B2	$a = 0,303$ [16]	B19	$a = 0,2881,$ $b = 0,4279,$ $c = 0,4514$ [16]
Ti—4Mo	$\beta$	$a = 0,3261$ [19]	$\alpha''$	$a = 0,3012,$ $b = 0,4983,$ $c = 0,4658$ [19]
Ti—20Nb	$\beta$	$a =$ $0,328505$ [20]	$\alpha''$	$a = 0,31257,$ $b = 0,48704,$ $c = 0,46456$ [20]
Ti—24Nb	$\beta$	$a =$ $0,328569$ [20]	$\alpha''$	$a = 0,31785,$ $b = 0,4812,$ $c = 0,46355$ [20]
Zr—49,9Cu	B2	$a = 0,3273$ [22]	B19'	$a = 0,3237,$ $b = 0,4138,$ $c = 0,5449,$ $\beta = 105,19^\circ$ [22]
Ti <sub>3</sub> Sn	DO <sub>19</sub>	$a =$ $0,5938,$ $c = 0,4749$ [6]	Orthorhombic	$a = 0,585,$ $b = 1,034, c =$ $0,475$ [6]

of internal stress relaxation inhibit the shape recovery if  $M_s/T_m$  is high [23, 24]. In materials with low-temperature martensitic transformation the interplay between diffusion processes of point defects rearrangement might lead to twinning pseudoelasticity [25]. Twinning pseudoelasticity i. e. strain recovery below transformation temperature was observed into AuCd, In—Tl, some Cu-based alloys and was not found into Ni—Ti [25]. For Ti<sub>3</sub>Sn this ratio is 0,18 (table 1) meaning that diffusion processes are not expected to deteriorate mechanical effects associated with martensitic transformation. Although twinning pseudoelasticity is not expected

for  $Ti_3Sn$  because of low  $M_s/T_m$ , the strain recovery below transformation temperature was observed in the non-stoichiometric  $Ti_{75.5}Sn_{24.5}$  [5].

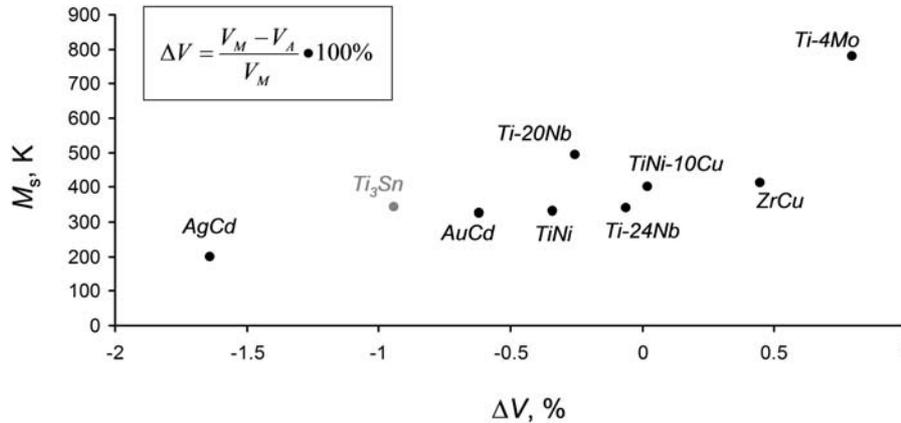


Fig. 4. Transformation temperature  $M_s$  versus volume change accompanied martensitic transformation according to table 1.

### Summary

The transformation strain matrix associated with orthorhombic to hexagonal in  $Ti_3Sn$  contains eigenvalues  $\lambda_1 < 0$ ,  $\lambda_2 = 1$ ,  $\lambda_3 > 0$  and corresponds to the volume change of about 1% ( $\det U = 0,99$ ). These imply that the transformation is lattice-distortive and shear dominant, i. e. martensitic. The condition  $\lambda_2 = 1$  points to good geometrical compatibility between parent and product. The volume change accompanied the transition in  $Ti_3Sn$  is negative and quite big compared to other low-temperature SMA.

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### **Аналіз деформації перетворення, пов'язаної з перетворенням з гексагональної в орторомбічну фазу в $Ti_3Sn$**

О. М. Иванова

*Огляд присвячений аналізу перетворення із гексагональної в орторомбічну фазу в  $Ti_3Sn$ . Показано місце мартенситного перетворення в ширшій категорії бездифузійних перетворень з точки зору деформації перетворення. Деформація перетворення в  $Ti_3Sn$  проаналізована і зсувний (мартенситний) характер перетворення проілюстровано. Порівняно зміни об'єму та температури перетворення в  $Ti_3Sn$  з такими в інших матеріалах, що зазнають мартенситного перетворення.*

**Ключові слова:** мартенситне перетворення, деформація перетворення, сплави з ефектом пам'яті форми.

### **Анализ деформации превращения, связанной с превращением из гексагональной в орторомбическую фазу в $Ti_3Sn$**

О. М. Иванова

*Обзор посвящен анализу превращения из гексагональной в орторомбическую фазу в  $Ti_3Sn$ . Показано место мартенситного превращения в более широкой категории бездиффузионных превращений с точки зрения деформации превращения. Деформация превращения в  $Ti_3Sn$  проанализирована и сдвиговый (мартенситный) характер превращения показан. Проведено сравнение изменения объема и температуры превращения в  $Ti_3Sn$  с этими характеристиками в других материалах, претерпевающих мартенситное превращение.*

**Ключевые слова:** мартенситное превращение, деформация превращения, сплавы с эффектом памяти формы.