

The Interaction of Point Defects with the Martensitic Transformation: A Prototype of Exotic Multiscale Phenomena

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Abstract

The martensitic transformation has so far been studied without considering its interaction with point defects. In this article, we shall show that such interaction, which stems from a universal symmetry property of point defects, can create a rich spectrum of exotic multiscale phenomena in martensitic materials. These phenomena include unique short-range diffusion at the atomic or nano level, remarkable domain-pattern memory at the mesoscopic level, and peculiar rubber-like behavior and aging-induced two-way shape memory at the macroscopic level. Exotic multiscale phenomena may also be found in a wide range of transforming materials, such as ferroelastic, ferroelectric, and ferromagnetic materials. These novel effects may provide new opportunities for these important materials.

Keywords: atomic diffusion, intermetallic alloys, phase transformation, shape memory.

Introduction

Point defects, such as vacancies, solute atoms, impurities, or antisite defects in ordered compounds, are ubiquitous in crystalline solids and are important in controlling many physical and mechanical properties of materials. Their critical role in *diffusional* transformation has long been recognized and well understood; in sharp contrast, their role in *martensitic/diffusionless* transformation is largely unknown and has been neglected for a long time. Traditionally, it has been considered that all features of martensite can be understood once the lattice correspondence between the parent phase and the martensite is es-

tablished because the martensitic transformation involves only a diffusionless lattice distortion. In fact, as martensitic alloys always contain point defects, they can produce many unexpected interesting effects through their interaction with the martensitic transformation and during the subsequent aging/diffusion process.

In this article, we shall give a concise review of recent advances in the understanding of the important role of point defects in martensitic transformations and associated exotic multiscale phenomena. We shall show that all these interesting effects stem from an important and general

symmetry property of point defects.^{1,2} Due to the generality of this symmetry property, we expect that novel multiscale phenomena may also be found in a wide range of materials.

Historical Background

Before explaining the central idea of the symmetry property of point-defect configurations, we first give a brief historical account of the field closely related to the present topic, because the symmetry concept of point defects actually developed out of this background.

Since 1932,³ it has been known that a number of martensitic alloys (including Au-Cd,^{3,4} Au-Cu-Zn,⁵ Cu-Zn-Al,^{6,7} Cu-Al-Ni,⁸ In-Tl,^{9,10} Ti-Pd,¹¹ Ti-Pd-Ni,¹² and Ni-Ti-Hf⁽¹³⁾) show an unusual kind of deformation behavior in the martensitic state. These alloys, after aging for some time in the martensitic state, can be deformed like a soft and pseudoelastic rubber with a recoverable strain as large as a few percent. An example is shown in Figure 1a.¹⁴ The fresh, or unaged, martensite shows a plastic-deformation behavior ($t = 0$ s curve in Figure 1a), as expected from a normal, irreversible twinning process in the low-symmetry martensite. However, after aging for some time, the deformation behavior changes into a strange pseudoelastic or rubber-like behavior, together with an increase in twinning stress. This rubber-like behavior involves an unexpected reversible twinning process (the original twin pattern is restored after stress is removed) in the martensitic state without involving any phase transformation or structure change. Therefore, it differs from superelasticity, which occurs by means of the stress-induced martensitic transformation from the high-temperature parent phase and its subsequent reverse transformation (for more details, see the introductory article by Otsuka and Kakeshita in this issue). Also, owing to the structure invariance in the twinning process, the possibility of a "pseudotwinning" process (which can also give rise to a pseudoelastic behavior through a structure-changing process) was also excluded. The rubber-like behavior is completely unexpected from the viewpoint of the common notion that a twinning process (without structure change) is not reversible and always yields plastic deformation; thus, this phenomenon had remained unexplained for over 60 years.

Accompanying the martensite aging is the development of martensite stabilization, an increase of the reverse-transformation temperature with aging time (an example is shown in Figure 1b).¹⁵ This effect had also remained controversial for a long time.

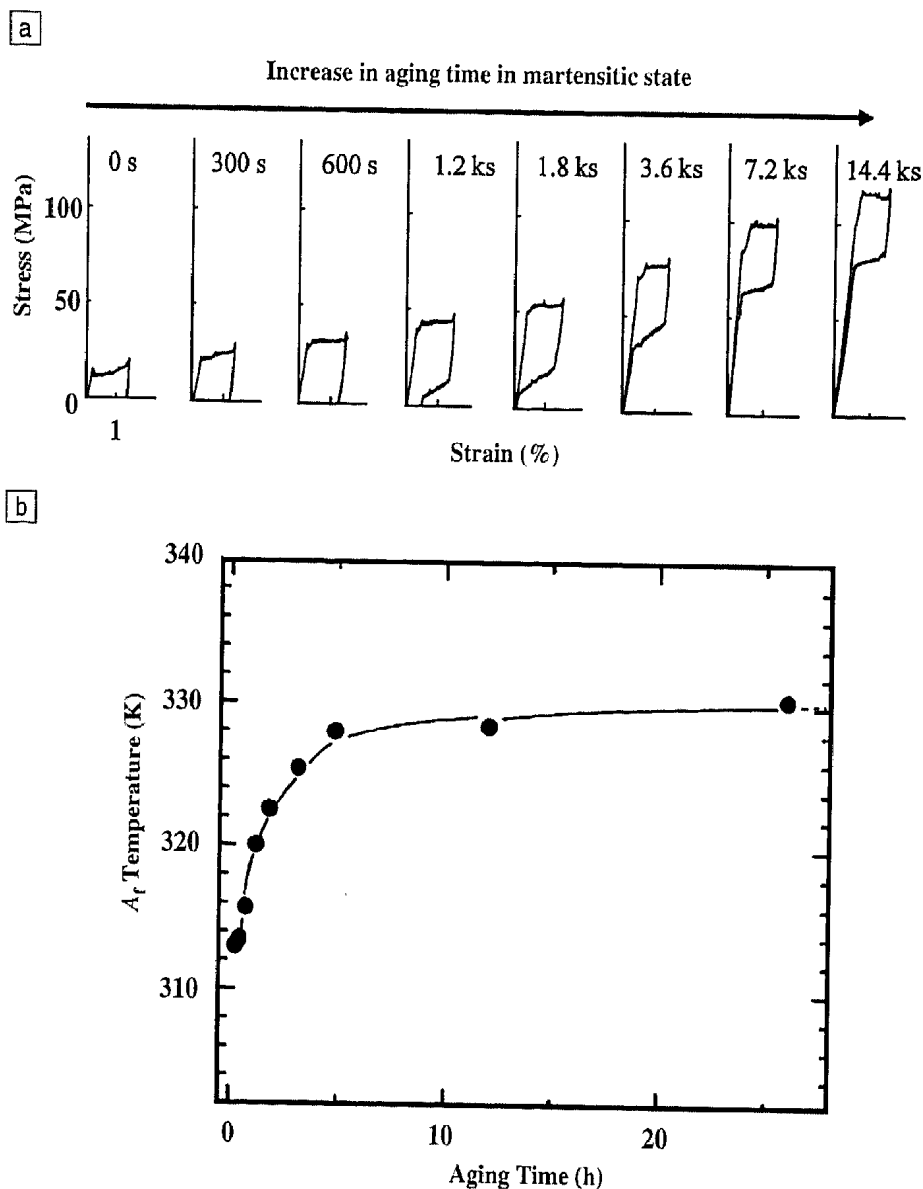


Figure 1. Changes in physical properties of a Au-49.5at.%Cd alloy during aging in the martensitic state: (a) from soft plastic behavior (unaged martensite) to rubber-like behavior (aged martensite); (b) the martensite stabilization effect, corresponding to the increase in the A_f temperature (reverse-transformation-finishing temperature) with aging time.

Since it is the aging in the martensitic state that causes these significant effects, the key to understanding these phenomena is to understand what happens during martensite aging. The central question concerning martensite aging is why fresh martensite is not stable and why it needs to change.

Great efforts have been made over the past 50 years to find an answer to these curious aging phenomena. However, the explanation was seriously complicated by the simultaneous decomposition of many metastable martensites (mainly Cu-based alloys) during aging. These metastable martensites have an inborn tendency to decompose into their equilibrium products.

As long as diffusion is possible, some decomposition is unavoidable. Because the time-dependence of this inevitable process coincides with the martensite aging process, the martensite aging effect was frequently mistaken as being more or less related to such decomposition. However, the decomposition is obviously an extrinsic or secondary effect because equilibrium martensite like AuCd also exhibits a strong aging effect without decomposition.⁴ For equilibrium martensite, there is no reason for its average structure to change during aging. Indeed, an x-ray diffraction experiment¹⁶ has indicated that there is no change in the average martensite structure in equilibrium martensite. Therefore, the

biggest challenge in explaining the martensite aging effect is to understand why significant changes in properties can be realized even without a change in structure.

Many workers¹⁷⁻²² have contributed to the understanding of this challenging problem. Ahlers and his co-workers,¹⁷⁻¹⁹ Tadaki et al.,²⁰ Marukawa and Tsuchiya,²¹ and Suzuki et al.²² proposed different microscopic models for martensite aging effects. They all realized that there must be some kind of atomic rearrangement or diffusion during aging that is responsible for the aging effects. However, there was no consensus on what kind of atomic rearrangement/diffusion produced the observed aging effects. Their models explain part of the experimental findings, but they do not explain why a significant change in properties can occur even without a detectable change in the martensite structure during aging, as found in Au-Cd^{14,16} and Au-Cu-Zn alloy.²³ Interested readers may refer to References 24-26 for a more detailed review of different aging models.

Behind a long-standing puzzle there is always something completely new to be discovered, and something very simple in many cases. Recently, a general but simple idea was proposed.^{1,2} It has only two necessary ingredients: symmetry and point defects (which have the possibility of rearrangement/diffusion). In the following, we first introduce this general idea and then use it to explain these long-standing puzzles. We will show that these puzzling phenomena are no more than a portion of a wide spectrum of exotic multiscale phenomena caused by the interaction of point defects with crystal-symmetry changes (the martensitic transformation).

The Universal Symmetry Property of Point-Defect Configuration at the Atomic/Nano Level: A Symmetry of Probability

Even from a purely geometrical argument, it is natural to consider that the short-range-order (SRO) distribution of point defects has a simple symmetry property. For simplicity, we consider two-dimensional structures, as schematically shown in Figure 2, but the argument is applicable to any three-dimensional structure because only symmetry is of relevance. Let the system contain atoms A and B (here, B is regarded as a point defect; it can also be a vacancy or an antisite atom in an intermetallic compound). Now consider a given defect B in a high-symmetry (square) lattice (Figure 2a). Due to the fourfold crystal symmetry, the four nearest-neighbor sites 1, 2, 3, and 4 are equivalent to defect B. Thus, it is natural to expect that when in

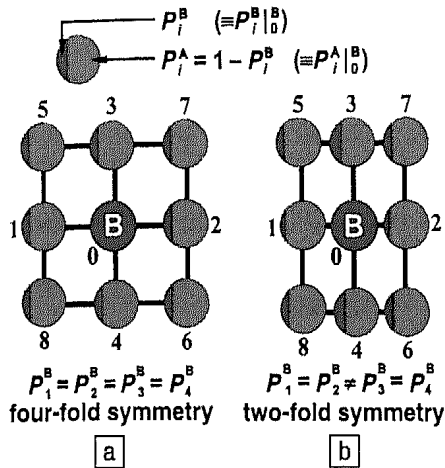


Figure 2. The symmetry-conforming short-range order (SC-SRO) property of point defects in crystals. (a) Fourfold crystal symmetry; (b) twofold crystal symmetry. $P_i^A|_0^B$ is the conditional probability of an A atom occupying site i ($i = 1, 2, 3, \dots, 8$) if there is a defect B at site 0; $P_i^B|_0^B$ is the conditional probability of a defect B occupying site i if there is a defect B at site 0. The relative values of these probabilities are indicated by the green and red areas, respectively.

equilibrium, the probability P of finding another defect B around the defect at site 0 has the same fourfold (square) symmetry as the crystal symmetry, that is, $P_1^B = P_2^B = P_3^B = P_4^B$. On the other hand, if the crystal is of lower symmetry (rectangular) (Figure 2b), the environment around B becomes less symmetrical, and it is natural to expect $P_1^B = P_2^B \neq P_3^B = P_4^B$ when in equilibrium, following the twofold (rectangular) crystal symmetry. Apparently, this symmetry consideration also applies to sites farther from B, but the effect becomes weaker owing to the weakening of the influence of B on farther sites. This means that this is an SRO effect. Therefore, Figure 2 suggests that there exists a universal symmetry property for the SRO distribution of point defects at the atomic/nano level. That is, when in equilibrium, the probability of finding another point defect around a given point defect (this conditional probability uniquely defines the SRO parameter) has the same symmetry as the crystal symmetry. This is the *symmetry-conforming SRO* (SC-SRO) principle of point defects, and can be expressed as follows:

$$\text{SRO symmetry} \Rightarrow \text{crystal symmetry.}$$

Note that here SRO symmetry is the symmetry of SRO probabilities (conditional

probabilities), not the local symmetry of atom species.

Important Consequences of the Point-Defect Symmetry Property in Displacive/Martensitic Transformations

When the parent phase (Figure 3a) transforms into martensite, all of the probabilities must remain unchanged despite the crystal-symmetry change, since the transformation is diffusionless (Figure 3b). That is, $P_1^B = P_2^B = P_3^B = P_4^B$. However, this high-symmetry SRO configuration is no longer a stable configuration for the low-symmetry martensite structure, according to the SC-SRO principle. Therefore, if SRO symmetry of point defects is taken into account, martensite immediately after the transformation is in a metastable state. During martensite aging, if diffusion is allowed (i.e., the temperature is not too low), such a metastable SRO configuration gradually

changes into a stable one that conforms to the martensite symmetry, as shown in Figure 3c. Because the equilibrium martensite structure should be maintained (for stable martensite), this process must proceed by atomic rearrangement or relaxation *within the same sublattice*. This is the only way that martensite can lower its free energy without altering the average structure (equilibrium phase). It is a kind of short-range ordering within the same sublattice. This SRO stems from a requirement that the symmetry of the SRO parameters of point defects should attempt to conform to the crystal symmetry.

One can see clearly from this that there is a time lag between crystal-symmetry change and SRO-symmetry change; the former can change abruptly, while the latter must take some time to change (by diffusion) and to follow the crystal symmetry. The time lag also occurs during the reverse martensitic transformation

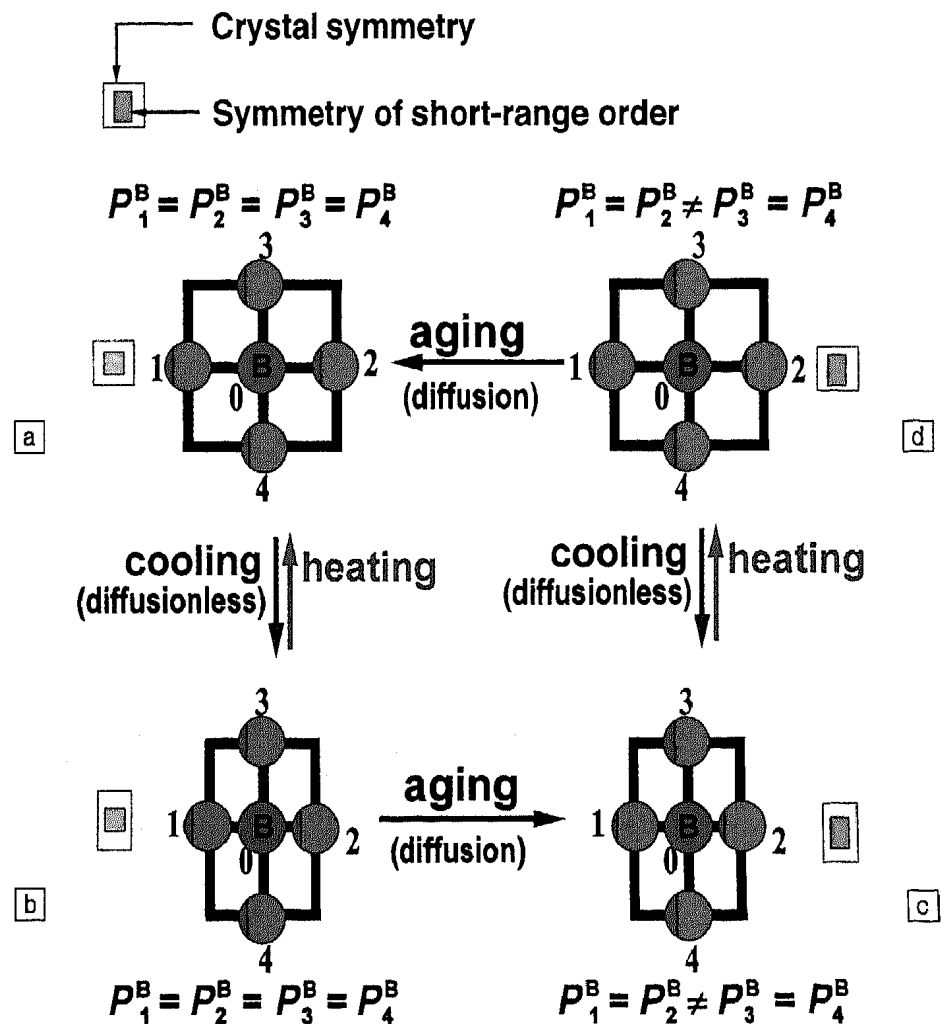


Figure 3. (a)–(d) Time lag between the change in crystal symmetry and the change in SRO symmetry. Take note of the two symmetry symbols (crystal symmetry and SRO symmetry) in order to understand the mechanism in Figure 4.

and during parent-phase aging (from Figures 3c, to 3d, to 3a). Immediately after the reverse transformation from Figure 3c to Figure 3d, the low SRO symmetry of martensite does not follow the crystal symmetry of the parent phase, thus this state (Figure 3d) is unstable and changes into Figure 3a after aging in the parent state. In the next section, we show that this time lag can generate many interesting multi-scale phenomena.

Exotic Multiscale Phenomena Associated with the Interaction of Point-Defect SRO Symmetry and the Martensitic Transformation

It is known that fresh martensite (without aging) behaves normally and exhibits one-way shape memory with respect to temperature and plastic twinning deformation with respect to stress. These prop-

erties can be understood simply by considering the diffusionless nature of the transformation and the normal twinning process (see the introductory article by Otsuka and Kakeshita in this issue), but the aging phenomena cannot be explained. On the other hand, we have already shown that point-defect symmetry in martensitic crystals can produce important consequences and may be the origin of the aging phenomena. Then, naturally, comes the question of whether both the normal behavior of the fresh martensite and the abnormal aging effects (rubber-like behavior and stabilization of aged martensite) can be explained when point defects are considered. In the following, we shall show that the answer is yes. In Figure 4, the central illustrations show schematically the change in SRO symmetry, crystal symmetry, domain pattern, and macro-

scopic shape during heating/cooling and stress-loading/unloading. The photographs and micrographs on the left show experimentally observed mesoscopic domain patterns and macroscopic shape change for Au-Cd martensite without aging, while those on the right show Au-Cd martensite after aging. Also see Figure 3 for the atomic/nano-level illustration of SRO symmetry and crystal symmetry, which are crucial to understand the following.

Normal Behavior of Unaged Martensite in the Presence of Point Defects

When the equilibrium parent phase (Figure 4a) transforms into martensite (Figure 4b), a characteristic martensite domain/variant pattern is formed. However, the SRO symmetry is still cubic, owing to the diffusionless nature of the transfor-

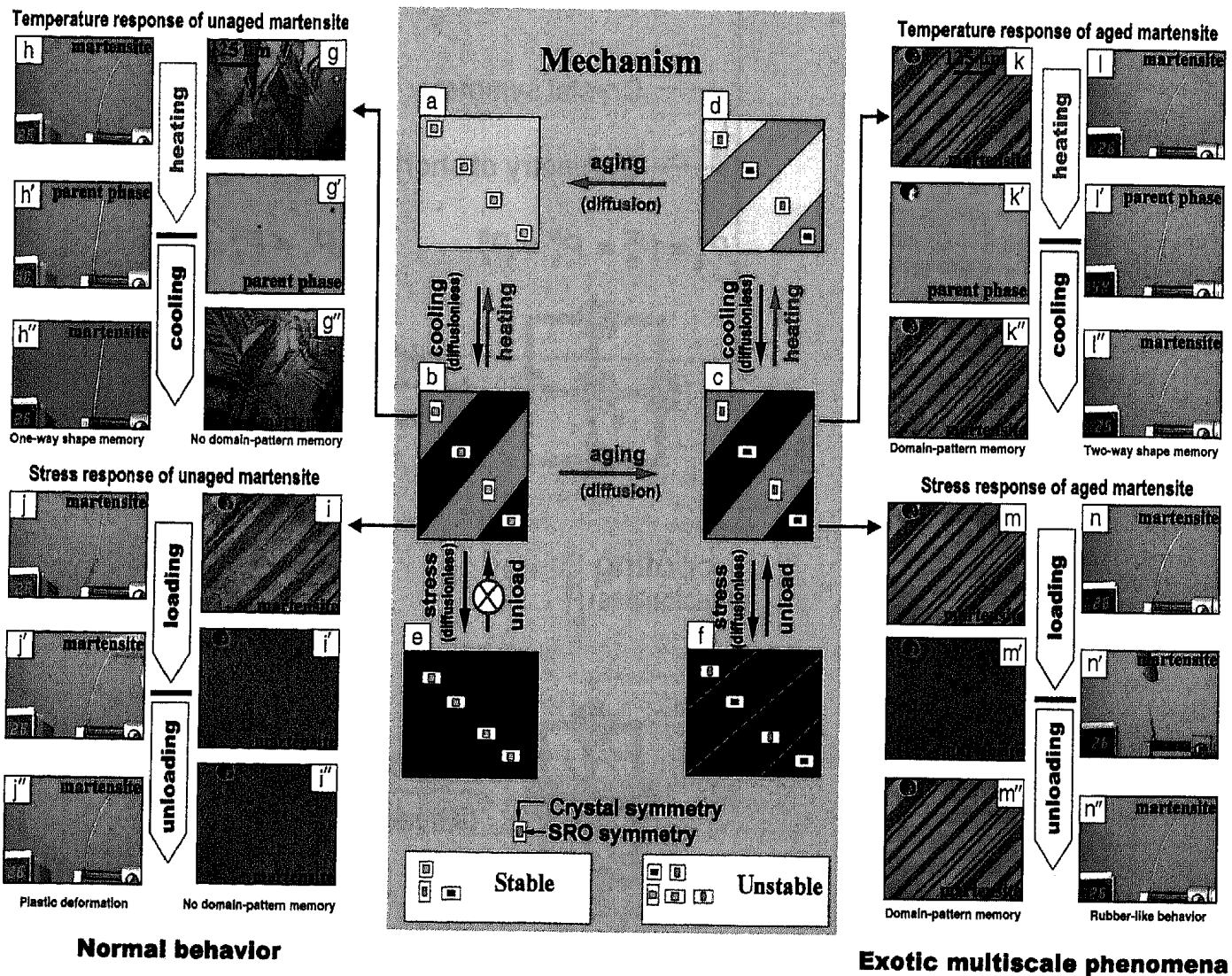


Figure 4. (a)–(f) Mechanism and experimental observation of the normal behavior of (g)–(j) unaged martensite and (k)–(n) exotic multiscale phenomena of aged martensite. All micrographs were taken at the same scale. See text for explanation. Note that (a)–(d) here correspond to (a)–(d) in Figure 3.

mation (refer to the discussion of Figure 3). Although this is an "uncomfortable" state for point defects, the discomfort is the same for different domains (light-blue and dark-blue ones) because they are related by a rigid-body rotation (which does not change the internal energy). Therefore, different domains (twins) actually have the same energy, as if the SRO of point defects did not exist. As a result, fresh martensite behaves like an ideal martensite in which no point defects exist. On the mesoscopic level, the martensite domain pattern cannot be reproduced after one transformation cycle (compare Figures 4g and 4g'). This means that the macroscopic shape of the martensite cannot be restored after one transformation cycle, as shown in Figures 4h, 4h', and 4h''. Therefore, we have only one-way shape memory (because the macroscopic shape of the parent phase can always be reproduced).

Also, owing to the energy equality of different domains, martensite shows plastic twinning deformation (Figures 4e, 4i, 4i', and 4i'', and Figures 4j, 4j', and 4j''), because there is no reason for a particular domain pattern (which corresponds to a particular macroscopic shape) to be energetically favored over other domain patterns (which correspond to other macroscopic shapes).

Exotic Multiscale Phenomena in Aged Martensite

When martensite is aged, it is possible to change the SRO symmetry through diffusion. Following the SC-SRO principle, the low-symmetry SRO configurations in different martensite domains are oriented differently (Figure 4c), following their corresponding crystallographic orientation. When the equilibrium martensite (Figure 4c) transforms into the cubic parent phase (Figure 4d) by reverse transformation, the crystal symmetry becomes cubic everywhere. However, due to the diffusionless nature of the transformation, the low SRO symmetry of the martensite is inherited by the cubic parent phase. Consequently, there exists a difference in SRO orientation in the parent phase between what previously were different martensite domains. This orientation difference in SRO will produce an "SRO domain" pattern in the cubic phase (Figure 4d) that is identical to the martensite domain pattern shown in Figure 4c. This contrast has been observed by TEM,² which is a structure-sensitive imaging technique, but it cannot be observed with optical microscopy because there is no surface relief involved.

Now, when we let this "nonequilibrium" parent phase (Figure 4d) cool down and transform into martensite again, it is

apparent that it will adopt the previous martensite domain pattern (Figure 4c) so as to make the SRO orientation conform to the martensite crystallographic orientation everywhere, as a consequence of the SC-SRO principle. This is a kind of "domain-pattern memory," or "mesoscopic memory," induced by martensite aging and has been found experimentally² (Figures 4k, 4k', and 4k''). The macroscopic manifestation of this microstructure memory is the memory of the previous martensite shape² (Figures 4l, 4l', and 4l''). Therefore, we can show that aged martensite exhibits domain-pattern memory at the mesoscopic level and the associated two-way shape memory at the macroscopic level. The aging-induced two-way shape memory is different from another kind of two-way shape-memory effect produced by a complicated thermomechanical "training" process,²⁷ where the formation of favorably oriented dislocations is responsible for the recovery to the martensite shape, and no aging treatment is required. The present mechanism for two-way shape memory associated with martensite aging can also account for the similar two-way shape memory associated with the aging of stress-induced martensite.^{28,29}

When aged martensite (Figure 4c) is deformed, domain reversal (i.e., detwinning) occurs (Figure 4f) as a result of the accommodation of the strain. Because this twinning process is also diffusionless, the SRO orientation shown in Figure 4c is inherited by the new domain, as shown in Figure 4f. Such a configuration, however, is not the stable one for the new domain. Therefore, a driving force that tries to restore the original domain (Figure 4c) is engendered. This gives rise to another kind of domain-pattern memory due to stress (Figures 4m, 4m', and 4m''). When the external stress is released, this restoring force reverts the new domain (Figure 4f) to the original one (Figure 4c) by de-twinning, and macroscopically the sample exhibits a pseudo-elastic behavior (Figures 4n, 4n', and 4n''). This is the origin of the rubber-like behavior, which had resisted explanation for over 60 years.

Therefore, the atomic/nano-level SRO symmetry and its interaction with the martensitic transformation create many exotic multiscale phenomena, ranging from temperature-driven domain-pattern memory to two-way shape memory, stress-driven domain pattern memory, and rubber-like behavior. We can also see how simple it is to explain the long-standing puzzle of rubber-like behavior and to predict all of these new effects, if we use the SC-SRO principle.

Stabilization Effect

The stabilization effect refers to an increase in the reverse-transformation temperature with increasing aging time, as shown in Figure 1b.¹⁵ When the SRO symmetry of point defects is considered, this effect can be easily understood as well.

When the aged martensite (Figure 4c) is heated up and transforms back (diffusionlessly) into the parent, the stable SRO configuration for the martensite is inherited by the parent (Figure 4d). From the SC-SRO principle, it is obvious that Figure 4d is not the stable configuration for the parent. From a thermodynamics point of view, this corresponds to an increased reverse-transformation temperature. This is the origin of martensite stabilization.

Parallel to the development of the microscopic explanation for the origin of the martensite aging effect, in recent years notable phenomenological models have been proposed by Bhattacharya et al.³⁰ and Ohta.³¹ These models assume that there exists a so-called time-dependent variable coupled to the transformation strain/shuffle, and they are able to reproduce the martensite aging effects in a semi-quantitative way. Transformation strain refers to a uniform displacement of atoms in the unit cell upon transformation; a shuffle is a displacement of an atom or atoms within the unit cell. Now we can understand that the microscopic nature of this time-dependent variable should be the time-dependent SRO symmetry of point defects.

The Effect of Point-Defect Concentration: Evidence for the Central Role of Point Defects in the Aging Effect

So far, we have given a simple and unified explanation for all of the known effects of martensite aging and some interesting new effects, based on the interaction of the SRO symmetry of point defects with the crystal symmetry. If all of these effects are due to point defects, one will naturally expect that the size of these effects should strongly depend on point-defect concentration. Now we show that this is really the case.

As point-defect concentration changes with alloy composition, we can expect a strong composition-dependence of the aging effect. Figure 5 shows that this is true. One may refer to Reference 32 for a closer correlation of the type and the concentration of point defects with the magnitude and rate of the aging effect. Therefore, there is no doubt that all of the aging effects are due to the SRO rearrangement of point defects.

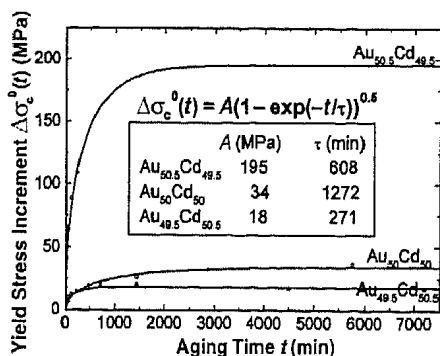


Figure 5. Composition-dependence (or point-defect-concentration-dependence) of the martensite aging effect, characterized by the increment of critical twinning stress (yield stress of Figure 1a). Experimental data are fitted with a Chipman function, where A represents the magnitude of the martensite aging effect and τ represents the aging rate (relaxation time). (From Reference 32.)

Because point defects are a crucial factor in martensite aging effects, a quantitative evaluation of point-defect concentration becomes important. It is noted that most shape-memory alloys are ordered B2 compounds; thus, a modeling of point-defect formation in B2 compounds is indispensable. Recently, a unified model has been proposed^{33,34} and is applicable to different types of B2 compounds.

Conclusion and Outlook

Point defects have a simple and universal symmetry property, that is, the short-range-order symmetry of point defects tends to follow the crystal symmetry. This important property is the origin of atom/defect rearrangement after diffusionless transformation (symmetry change) and is also the origin of all of the exotic multi-scale phenomena introduced in this article. Although the examples discussed are confined to martensitic alloys, it is expected that similar effects, together with some novel electrical and magnetic effects, may

be found in ferroelectric and ferromagnetic materials because they also undergo abrupt symmetry change. Furthermore, Zener relaxation³⁵ and directional anisotropy in cubic³⁶ or amorphous materials³⁷ induced by stress annealing or magnetic annealing may be also due to the same cause, although the way of changing crystal symmetry is different (i.e., by elastic distortion or magnetostriction). The symmetry-conforming short-range order principle may open a new research area with promising opportunities.

Acknowledgments

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