Precursor Phenomena of Martensitic Phase Transformation

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The unique aspect of martensitic transformation is pointed out to lie in coexistence of apparently contradictory features of 'non-locality' and 'discreteness' during the ordering process. As a remarkable manifestation of this unique feature, the precursor phenomenon called 'ghost lattice effect' observed in X-ray diffraction experiments is discussed. Based on thermodynamical treatment within Ginzburg-Landau scheme, it is shown that soliton-like fluctuations of the order parameter in highly non-linear medium, called embryonic fluctuation, is the origin of this precursor. It is suggested that this key word seems to bring the unique feature of martensitic transformation into clearer understanding.

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I. Introduction

It is generally accepted that the martensitic phase transformation of metals is characterized by the following transformation properties.

(i) It is a first order transition: A phase transition is classified to be first order when there is discrete and finite jump of the order parameter at the transition point. In fact, the product phase of the martensitic transformation exhibits discrete change from the parent phase both in the lattice parameters as well as in the internal atomic positions.

(ii) It is a non-diffusional transition: The structural change from the parent phase to the product phase is attained by the *displacement* of the constituent atoms by a fraction of the lattice constant rather than the *replacement* of atom species through a diffusion process.

There seems, however, to be another important feature to make the martensitic transformation distinguished from other first order, displacive phase transitions which are commonly observed in various ionic materials: As is well known, the first order phase transition ordinarily proceeds through nucleation-growth process. That is, once an embryo of the product phase is nucleated beyond a critical size, it continues to grow until it covers the whole system. The growth velocity is critically dependent on the 'quench depth', or the degree of metastability. Hence, by controlling the quench depth experimentally in the vicinity of the transition point, we can control the growth velocity by orders of magnitude⁽¹⁾.

On the other hand, in the case of martensitic transformation, it seems to proceed through somewhat different process: As the specimen is quenched below M_s -temperature macroscopic part of the specimen transforms almost

instantaneously, whose growth velocity can not be controlled by the quench depth.

In order to clarify the difference between these processes, we may use the key word 'local' vs 'non-local' event. The ordinary first order transition proceeds as a *local* event like 'domino game': In domino game, a standing piece of domino chips does not know that the others have fallen, until the neighbouring piece actually touches to push it down. Similarly, in the case of ordinary first order transition a part of the untransformed region stays metastable until the phase boundary reaches the position to influence a short range force *locally*. In contrast, in the second order transition, the local informations of stability is exchanged between the part of the specimen separated by a macroscopic distance through *non-local* fluctuations such as soft phonons, which enables the system to transform uniformly and instantaneously.

In this context, the martensitic transformation bears both of the apparently contradictory features of 'discreteness' (first order) and 'non-locality' (second order).

The above uniqueness is manifested in the so-called precursor effect of the transformation. Precursor phenomena mean that the system prepares for the phase transition before it actually takes place, which has definite similarity to the well known critical phenomena at second order phase transition. It is therefore of importance to elucidate the properties of precursor phenomena in order to bring the unique feature of martensitic transformation into clearer understanding.

In this paper, we first establish the order parameters of martensitic transformations exhibited by various bcc-based alloys⁽²⁾. This section serves to make our standpoint clear to discuss the martensitic transformation on thermodynamical basis. In Section III, we describe the particular precursor phenomenon called ghost lattice effect observed by diffraction experiments and its thermodynamical interpretation. The last section is devoted to discussions on this precursor phenomena in connec-

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tion with the uniqueness of martensitic transformation as discussed above.

II. Order Parameters of Displacive Phase Transitions

In the case of displacive phase transition, the symmetry change of the crystal is associated with the displacement field of the atoms from the stable positions in the high symmetry phase. In general the displacements should be expressed by some linear combination of the normal coordinates of the phonons within the high symmetry phase because the normal coordinates form the complete set of the variable associated with the freedom of motion of the constituent atoms. Practically, only very limited number of the normal modes belonging to so-called 'soft phonon' branches are enough to describe the displacements. Thus the order parameter of the displacive transition is expressed by the amplitude of these 'frozen' soft modes.

1. bcc-based metals

To be specific, let us consider the case of bcc-based alloys. A number of metals with conduction electron-atom ratio $e/a \sim 1.5$ tend to stabilize bcc structure at high temperatures because geometrical properties of bcc Brillouin zone favors stabilization of electronic band energy known as Hume-Rothery rule. As the temperature is lowered bcc phase become unstable and the system tends to take close-packed structures.

This intrinsic instability of bcc lattice manifests itself in the anomalously low-lying dispersion surface of a particular phonon branch, that is, the TA branch within the (111)-zone of the bcc reciprocal lattice. Experimentally, this anomaly is observed as the diffuse 'sheet' of TDS (thermal diffuse scattering) perpendicular to the [111]-directions⁽²⁾. In particular, we focus our attention to the two symmetric directions within the (111)-zone; the $[1\bar{1}0]$ - and $[2\bar{1}\bar{1}]$ -directions. (See Fig. 1.)

As is shown in Fig. 2, the dispersion curves along these directions have very low energies throughout the Brillouin zone⁽³⁾. Sometimes, the dispersion curve gives upward curvature or even dips⁽⁴⁾. Therefore we infer that the displacement of the atoms in the martensite phases is expressed by the normal coordinates of the phonon modes on the symmetric directions, $[\zeta, \bar{\zeta}, 0]$ and $[2\xi, \bar{\zeta}, \bar{\zeta}]$ belonging to this characteristic low-lying TA dispersion surface.

Based on this consideration, we express generally the displacement of the atoms in martensite phase from original bcc positions as follows:

$$u_j^v(r_i) = \sum_{k=1}^m [\xi_k e_k^v(q_0^{(k)}, \text{TA}) e^{iq_0^{(k)} \cdot r_i} + \text{c.c.}] + \varepsilon_1^{(k)} \cdot r_{ij} + \varepsilon_2^{(k)} \cdot r_{ij}, \quad v=x, y, z. \quad (1)$$

Here, $u_j^v(r_i)$ is the displacement of the j 'th atom in the i 'th cubic unit cell sitting at r_{ij} in bcc phase, $e_k^v(q_0^{(k)}, \text{TA})$

Fig. 1 Reciprocal space of the bcc lattice. The shaded plane gives the [111]-zone, in which strong TDS is observed. The wave vectors of the frequently observed soft modes are at $\blacktriangle$: $1/3[110]$, $\blacksquare$: $1/2[110]$, $\circ$: $1/3[211]$.

is a component of the normal coordinate of the relevant frozen modes, the index k specifies one of the equivalent q_0 's (such as $[\zeta, \bar{\zeta}, 0]$, $[0, \zeta, \bar{\zeta}]$, etc.), and ξ_k is the amplitude of the mode which plays the role of the 'order parameter' of the martensitic transformation.

The second term $\varepsilon_1^{(k)}$ is the strain tensor to describe the uniform deformation of the lattice to attain a close-packed structure (Bain strain);

$$\varepsilon_1^{(1)} = \begin{pmatrix} \varepsilon & 0 & 0 \\ 0 & \varepsilon & 0 \\ 0 & 0 & -2\varepsilon \end{pmatrix}, \quad (2)$$

and the third term is the uniform shear perpendicular to $q_0^{(k)}$;

$$\varepsilon_2^{(1)} = \begin{pmatrix} \varepsilon' & 0 & 0 \\ 0 & -\varepsilon' & 0 \\ 0 & 0 & 0 \end{pmatrix}. \quad (3)$$

Any of the martensite structures of bcc-based alloys should therefore be classified by the set of $\{q_0, m, \varepsilon, \varepsilon'\}$. Hitherto reported various kinds of martensite structures are classified on this basis in Table 1.

Several comments would be necessary in connection with the table.

Fig. 2 Examples of phonon dispersions which display low-lying TA branches. (a) AuCuZn_2 (after Ref. (3)). (b) NiAl (after Ref. (4)).

(i) Single q vs. multiple- q structure

Because of the high symmetry (O_h) of the original bcc structure, there are several equivalent q -vectors which form the 'star' in the first Brillouin zone. Since the energy for these modes with the equivalent q 's should be equal, any linear combination formed by these modes is energetically equally stable (within the lowest order quadratic energy term). Therefore, the atomic displacement in the martensite phase is not necessarily be expressed by a mode with single- q value, such as $[\zeta, \bar{\zeta}, 0]$, but may also be given by a linear combination of a set of the equivalent modes, such as $\{[0, \zeta, \bar{\zeta}], [\bar{\zeta}, 0, \zeta], \dots\}$. The choice of the set of equivalent q 's, $\{q\}$, directly determines the overall lattice symmetry of the resultant martensite phase.

In the case of single- q mode structure with $q = 1/3[110]$, for instance, the lattice symmetry of the resultant martensite should be orthorhombic. On the other hand, the triple- q mode structure which is given by the linear combination of the three modes with a set of q 's; $\{1/3[011], 1/3[101], 1/3[110]\}$ results in a rhombohedral lattice with the threefold axis along the $[111]$ -

direction.

We could consider the extreme case when all of the equivalent modes condenses simultaneously, in which case the lattice symmetry of the martensite structure should be cubic. The multiplicity of the q -vector of individual martensite material should be determined by the properties of higher order terms than quadratic in the free energy expansion of the system.

(ii) 2H and 9R structure

Of 'single- q ' structures, the majority of martensite structures belong to either 2H or 9R. These structures are derivable from the displacement patterns of the TA phonons with $q = 1/2[110]$ and with $q = 1/3[110]$ respectively. In the case of the $1/2[110]$ mode, the derivation of the 2H structure is straightforward, while in the case of the $1/3[110]$ -mode we need additional uniform deformation (ϵ') to derive the exact 9R martensite structure.

It is noticed that, as is typically seen in Fig. 2(a), the dispersion curve does not show well defined dips at the specific values or $q = 1/2[110]$ nor at $1/3[110]$. The physical origin of the choice of these two particular q -values as the 'frozen' modes is not clear.

(iii) 7R-structure

In the case of the 7R structure, realized in NiAl alloys the dip in the phonon dispersion around $q_0 \sim 1/7$, is conspicuous (Fig. 2(b)). On the other hand, in order to construct the exact $\bar{5}2$ structure, the higher harmonics such as $q = 2q_0$ ($2/7[110]$) and $3q_0$ ($3/7[110]$) are necessarily taken into account⁽⁶⁾⁽⁷⁾.

(iv) ω -phase

The triple- q structure includes the well-known ω -phase. Usually the ω -structure is discussed in terms of the instability of a single longitudinal mode with $q = 2/3[111]$. Our standpoint is that it is physically more reasonable to associate the ω -structure with the $3-q$ state of the low-lying transverse mode with $q = 1/3[211]$.

(v) ζ -AuCd

ζ -AuCd was previously reported to be derivable from the normal mode with $2/3[110]$ LA. In 1989, the author predicted that this could more likely be expressed by $3q$ structure with $q_0 = 1/3[110]$ TA⁽¹¹⁾. Recently, Otsuka *et al.* carried out single domain structure analysis of AuCd and confirmed the prediction⁽¹¹⁾.

(vi) γ -CuZn

The ' γ '-structure (γ -CuZn etc.) is described by the instability of the same mode as the ω -phase ($q_0 = 1/3[211]$). The difference resides in the multiplicity. The γ -phase belongs to an interesting case where *all* of the q 's condenses simultaneously. The lattice symmetry in the martensite phase is still cubic⁽¹³⁾, the symmetry change at the transformation being $O_h \rightarrow T_d$.

2. Ionic materials

If the definition of the martensitic transformation is as discussed in the previous section, there is no reason that only metallic systems should show the martensitic transformation. In fact, there are a few ionic materials which exhibit similar characteristics of phase transformation as bcc alloys, including pseudoelasticity and shape

memory effect⁽¹⁵⁾⁻⁽¹⁷⁾.

For later convenience, we have listed examples of ionic crystals which are considered to belong to this class in the separate column of Table 1. In particular, $\text{Pb}_3(\text{PO}_4)_2$ will be discussed in connection with precursor phenomena in the next section.

III. Precursor Phenomena

In the preceding section, we have defined the order parameters for martensitic transformations of bcc based alloys as the amplitudes of particular TA phonon modes. Once the order parameters, ξ_k 's, are defined one can develop a phenomenological theory of phase transition based on Landau scheme. That is, the free energy is formally expanded with respect to the order parameters as

$$F = \frac{\alpha}{2} (T - T_0) \sum_k \xi_k^2 + \frac{b}{4} \sum_k \xi_k^4 + \frac{d}{2} \sum_k \xi_k^6 + \frac{1}{2} \sum_v c_v e_v^2 + (\text{coupling terms between } \xi_k \text{'s and } e_v \text{'s}), \quad (4)$$

where e_v 's are the bulk strain components and c_v 's are the corresponding elastic constants. The stable state is then given by the stability condition: $\partial F / \partial \xi_k = \partial F / \partial e_v = 0$. Usually the phase transformation scheme is classified into two classes as follows:

- (i) $b > 0$, ($d > 0$): 2nd order
- (ii) $b < 0$, ($d > 0$): 1st order.

In case (i), the higher temperature phase becomes unstable at $T < T_c$ while in case (ii), both phases remain in the local minima of the free energy and the transition takes place when the lower temperature phase ($\xi \neq 0$) becomes the absolute minimum state. There are experimental evidences that the martensitic transformation

belongs to case (ii). One of the clear evidences is that the order parameter shows finite, discrete jump at the transition point.

At the same time, however, various experimental results called 'precursor' or 'premonitory' phenomena have been reported, which have some resemblance to the critical phenomena at 2nd order transition. Thus, the martensitic transformation bears both symptoms of essentially contradictory features of first order and second order transition. In this context, the precursor phenomena seem to provide the key to understand the unique features of the martensitic transformation. In the following, we discuss in particular, the remarkable precursor effect called 'ghost lattice' behavior observed in X-ray diffraction experiments.

In bcc-based metals it was commonly observed that even at $T > T_c$, weak superstructure spots characterizing martensite phase are already present in the diffraction pattern. Salamon *et al.*⁽¹⁸⁾ reported that in TiNi alloy the superstructure spots were slightly shifted from the commensurate reciprocal lattice points of bcc lattice. Shapiro *et al.*⁽¹⁹⁾ also reported similar incommensurability in TiNi(Fe). Moreover, from precise measurements around various reciprocal lattice points, they found that the incommensurability, δq , has peculiar symmetry properties. That is, the δq -value changes from one Brillouin zone to another, and does not possess the inversion symmetry around the Brillouin zone center.

Salamon first pointed out that this anomalous incommensurability could be understood if one assumes that the satellites are at the commensurate positions of the rhombohedral lattice which is expected to be stabilized below T_c ⁽²⁰⁾. Remember all fundamental Bragg spots are consistently indexed by original cubic lattice whereas the Bragg spots corresponding to the rhombohedral

Table 1 A list of the structural characteristics of bcc-based martensities. The structures are specified by the properties of the condensing phonon modes. q_0 : wave vector, m : multiplicity, ε' : uniform strain. A few ionic materials having ferroelastic phases are also listed, which may be characterized by similar martensitic transformation.

Parent (type)	Martensite (type)	Lattice sym.	Phonon mode		Slip ε'	Examples
			q_0	m		
bcc	3R 1	cubic	$1/3[110]$	$1q$	2/3	NiAl ⁽⁵⁾
	2H $1\bar{1}$	hexagonal	$1/2[110]$	"	0	Zr, Li, Mg, Na, CuAlNi, CuSn, AgAl, AuCd
	7R $5\bar{2}$	monoclinic	$1/7[110]$	"	(1/8)/7	NiAl ⁽⁶⁾⁽⁷⁾
			$(2/7)[110]$			
	9R $(2\bar{1})_3$	orthorhombic	$1/3[110]$	"	(1/6)/3	Li, CuAl, CuSn, CuZn, CuPd, AgCd, AuCuZn ₂ , AuCd
		tetragonal	$1/4[110]$	$2q$		VRu ⁽⁸⁾
	ζ ω	rhombohedral "	$1/3[110]$ $1/3[211]$	$3q$ $3q$	0 0	AgZn ⁽⁹⁾ , AuCd ⁽¹⁰⁾⁽¹¹⁾ , TiNi ⁽¹²⁾ Zr, NbZr, TiMo
	γ	cubic	$1/3[211]$	$12q$	0	CuZn ⁽¹³⁾ , AlZn
rutile-type	CaCl ₂ -type	orthorhombic	$[\varepsilon\varepsilon 0]B_{1g}$ $\varepsilon \rightarrow 0$	$1q$		MnF ₂ ⁽¹⁴⁾
scheelite-type	ferugsonite-type	monoclinic	$[\varepsilon\varepsilon 0]B_{1g}$ $\varepsilon \rightarrow 0$	$1q$		LaNbO ₄ ⁽¹⁵⁾⁽¹⁶⁾ , NdNbO ₄ ⁽¹⁵⁾⁽¹⁶⁾
(rhombo)		monoclinic	$1/2[111]$	$1q$		Pb ₃ (PO ₄) ₂ ⁽¹⁷⁾

lattice are completely absent. (Fig. 3). These features precludes the possibility to interpret the whole diffraction pattern either by ordinary incommensurate structure or by the two-phase mixture. In order to emphasize the puzzling situation, Salamon *et al.* called this peculiar reciprocal lattice a 'ghost lattice'⁽²⁰⁾ (cubic Braggs accompanied by rhombohedral satellites).

The atomic structure in the real space corresponding to this puzzling diffraction pattern has been elucidated by a series of works by Yamada and coworkers⁽²¹⁾⁻⁽²⁴⁾. As the key words to describe the structural characteristic, they used the words 'MLR' (Modulated Lattice Relaxation) model or, in more general treatment, 'embryonic fluctuation' model.

The basic feature of the model is envisaged as follows: at $T \geq T_c$, there are random distribution of semi-macroscopic regions of martensites *coherently* embedded on the bcc matrix. Notice this is not just an incoherent inclusion of the martensite particles (embryos) in the mother matrix, but rather a soliton-like fluctuation excited in highly non-linear medium. In this context, they called this entity an embryonic fluctuation⁽²⁴⁾.

The validity of this picture was given on the basis Ginzburg-Landau scheme, in which the order parameter is allowed to be spatially varying; $\xi = \xi(r)$. They specifically studied a 1D-model system whose free energy functional is expressed by,

$$F = \int \left\{ \lambda_1 \left(\frac{\partial \xi}{\partial x} \right)^2 + \lambda_2 \left(\frac{\partial e}{\partial x} \right)^2 + f(\xi, e) \right\} dx. \quad (5)$$

Here, the first and the second terms are the so-called

gradient energy and the third term is the local free energy density given by

$$f(\xi, e) = \frac{\alpha(T - T_0)}{2} \xi^2 + \frac{b}{4} \xi^4 + \frac{d}{6} \xi^6 + \frac{c}{2} e^2 + \gamma e \xi^2. \quad (6)$$

The free energy density $f(\xi, e)$ is essentially of the same form as given in eq. (4) except that the coupling term between the strain and the order parameter is explicitly given by the last term. Setting a boundary condition that at a fixed position ($x=0$) the order parameter is 'locked' in the value to be taken in the martensite phase ($\xi(x=0) = \xi_0$), the spatial distributions of $\xi(x)$ and $e(x)$ were calculated so as to minimize the total free energy (5). As is shown in Fig. 4, $\xi(x)$ shows a soliton-like behavior: persistence of ξ -value at $\xi = \xi_0$ around $x=0$, which is followed by kink-type rapid decrease to $\xi=0$. On the other hand, $e(x)$ shows smoother decrease. The region with $\xi = \xi_0$ is nothing but the 'embryo' of the martensite phase. Using the obtained $\xi(x)$ and $e(x)$, the diffraction pattern from this 1-D system was calculated, which turned out to show clearly the 'ghost lattice' behavior.

The functional form of the free energy (4) is quite general. This implies that in fact any system undergoing a first order phase transition where the order parameter strongly couples to macroscopic strains may show the same precursor phenomena irrespective of material (metals or non-metals), crystal symmetry (cubic or non-cubic), etc. It is noticeable that recently an ionic crystal $\text{Pb}_3(\text{PO}_4)_2$ was reported to exhibit the same ghost lattice effect⁽²⁵⁾. This material undergoes a ferroelastic transition from trigonal phase to monoclinic phase at $T_c = 180^\circ\text{C}$. A detailed symmetry argument on this material has shown that, in spite of apparent difference between bcc alloys and this material, they share the common expression of the free energy functional given in eq. (4)⁽²⁶⁾.

Fig. 3 Relation between the observed satellite positions (Ref. (12)) and the rhombohedral ghost lattice. The arrows indicate the directions of the shift from the commensurate cubic positions. The satellites are consistently shifted towards the rhombohedral commensurate positions whereas the Bragg peaks remain at the cubic rel. points.

Fig. 4 The calculated spatial variation of $\xi(x)$ and $e(x)$ at $\tau (= T - T_c / T) = 0.0003$. $\xi(x)$ shows kink-like behavior while $e(x)$ decreases smoothly. The corresponding structure around $x=0$ is just the embryo of the martensite.

IV. Conclusions and Discussions

It has been shown that the extraordinary precursor phenomena in diffraction experiments called 'ghost lattice' effect is essentially due to excitation of embryonic fluctuations. That is, even above T_c , there are random (spatially and orientationally) distribution of micro-regions of martensite phase which are coherently embedded on the matrix of the parent phase.

As the temperature is decreased towards T_c , the density of embryonic fluctuations will be increased, which gives rise to the interaction between neighboring embryos to reorient the crystallographic orientation. Eventually, cooperative orientational ordering of embryos should take place at T_c to form the uniform martensite phase. This transformation scheme seems to explain the coexistence of apparently contradictory features of 'non-locality' and 'discreteness' during the ordering process, which was pointed out in Section I as the unique feature of martensitic transformation.

One important point is still left unanswered: In the treatment developed by Fuchizaki and Yamada⁽²⁴⁾, they simply assumed that the order parameter is locally 'locked' in the fixed value of the martensite structure without giving the physical origin for locking. We could consider two basically different origins of the locking. A rather extrinsic origin is provided by the local lattice imperfections such as impurities, vacancies, etc. In this case, the density of embryos should be dependent on the purity of the individual samples.

The other origin which is more intrinsic is provided by thermal excitation. It should be noted, however, that in the ordinary first order transition, once a nucleus of the ordered phase beyond a critical size is thermally created, the phase boundary continues to proceed until the total system is completely covered by the ordered phase, which precludes the possibility of equilibrium distribution of embryos with finite size as postulated in embryonic fluctuation model. However, in the case of martensitic transformation, there is strong coupling between the order parameter and bulk strain. In such the case, the domain growth process may be prevented to proceed further at some point of the process. Recently, Onuki *et al.*⁽²⁷⁾⁽²⁸⁾ studied spinodal decomposition process taking into account of the strain energies by computer simulation. It was concluded that if the conditions; (i) the medium is elastically anisotropic and (ii) there is elastic misfit between the parent and product phases are satisfied, the phase separation process becomes extremely slowed down and practically stopped at some later stage. Since the above conditions seem to be also satisfied in the case of martensitic transformation, similar feature may be expected. It is left as the important future problem to elucidate whether the 'lock-in' mechanism is intrinsic or extrinsic.

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