Diffuse scattering as an indicator for martensitic variant selection

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Abstract

Diffuse scattering is an important precursor phenomenon prior to the martensitic transformation (MT). It is related to the correlated atomic position fluctuations prior to the MT and can provide important hints of the transformation mechanism. However, the role of this precursor phenomenon in the MT is not clear so far. Here we study the evolution of diffraction patterns prior to temperature- and stress-induced MTs and consider the evolution of atomic configurations during the whole MT process, using molecular dynamics simulations on a generic body-centered cubic–hexagonal close-packed transformation as an example. Our results show that, although the diffuse scattering changes with external fields, there exists a general relationship between the transformation pathways, the diffuse scattering streaks and the martensitic products. Two preferred transformation pathways with opposite shuffle directions lead to a single specific diffuse scattering streak prior to the MT and form one pair of anti-variants after the MT. Thus the distribution of diffuse scattering acts as an indicator of the selection of martensitic variants. In addition, we find that the applied stress can change the shear order parameter of the phase transformation, and subsequently determines the preferred transformation pathways and the distribution of diffuse scattering streaks. This work establishes a relationship between the transformation mechanism, the precursor phenomenon and the products after the MT under the influence of external fields.
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1. Introduction

The martensitic transformation (MT) is a structural, first-order and diffusionless phase transformation in which an open structure at high temperature usually transforms into a close-packed structure at low temperature. The martensitic transformation in steel represents the most economically important example of this kind of phase transformation, and its increasing applications in functional materials, such as shape memory alloys, have attracted significant attention for decades.

One of the most interesting features of martensitic transformations is the ubiquitous precursor phenomenon [1–3], a signature of the impending instability of the system towards a martensitic phase. Prior to a temperature-induced MT, several important precursor phenomena have been observed as the temperature approaches the transformation temperature [1–15]; for example, most of body-centered cubic (bcc) martensitic materials exhibit a low-lying TA$_2$ phonon branch [4–6] and elastic constant $C' = (C_{11} - C_{12})/2$ softening [7–9] when approaching the transformation temperature. A number of TEM observations have reported diffuse scattering along the $\langle 110 \rangle^*$ directions [10–15]. In TiNiFe alloys, the diffuse scattering is found to correspond to tiny "domains" [11,12], rather than the tweed structure as observed in NiAl alloys [5,15]. These precursor phenomena play a significant role in the kinetics

of martensitic transformation, and provide crucial clues to understanding the transformation mechanism. However, the relationship between these precursor phenomena and the products after the MT is still not clear.

Stress can also induce a martensitic transformation. It is known that C' softening also exists prior to a stress-induced MT [16–18], and the C' softening behavior depends on the applied stress, in contrast to the temperature-induced MT, where the softening occurs in all three equivalent directions. These results suggest that the precursor behavior prior to a stress-induced MT differs from the well-known behavior prior to a temperature-induced MT. Nevertheless, it is still not clear whether other precursor phenomena exist prior to a stress-induced MT, and how they differ from that in the temperature-induced MT. In addition, the relationship between the precursor phenomena and the transformation products under stress is also not clear. The diffuse scattering intensity distribution is rich in information on structural disorder and provides an appropriate diagnostic tool for us to study the correlated atomic position fluctuations in the precursor state and reveal its role in the MT. Thus, the purpose of this work is to use molecular dynamics (MD) simulations to investigate the diffuse scattering phenomena prior to the MTs under different external fields (both temperature and stress) and to establish the relationship between the diffuse scattering prior to the MT and the products after the MT.

Using a general bcc–hexagonal close-packed (hcp) phase transformation as example, we study here the evolution of diffraction patterns prior to temperature- and uniaxial stress-induced MTs via MD simulations. Our results show that, unlike diffuse scattering prior to a temperature-induced MT, the intensity of diffuse scattering under external stress increases only along certain specific directions, while it decreases along the others. Through analysis of the atomic evolutions and pathways of phase transformation during the stress- and temperature-induced MTs, we find that there is a general relationship between the diffuse scattering precursor phenomenon, the transformation pathways and the transformation products. Two preferred transformation pathways with opposite shuffle directions lead to a single specific diffuse scattering streak prior to the MT and form one pair of anti-variants after the MT. In addition, the applied stress can change the shear order parameter of the phase transformation, and then determines the preferred transformation pathways and the distributions of diffuse scattering. The understanding gleaned from this work enables us to establish a relationship between the transformation mechanism, the increase in diffuse scattering prior to the MT and the products after the MT under external fields.

2. Simulation and calculation methods

We used the embedded atom method potentials developed by Mendelev and Ackland [19] as the input for MD simulations. This potential reproduces properties such as the cohesive energy, elastic constants, and especially the MT between bcc and hcp very well. Although the potential was developed for pure Zr, we do not expect that such a semi-empirical potential could reproduce all of the phase transformation properties of Zr exactly (e.g. the bcc to hcp transition temperature reproduced by this potential under periodic boundary conditions (192.4 K) is significantly lower than the experimental value of 1135 K [20]). The reasons for the focus on a general bcc to hcp phase transformation are: (i) it is a martensitic transformation with coupling of shuffle (primary-order parameter) and shear (secondary-order parameter), which occurs in most martensitic alloys [2]; and (ii) the transformation follows the Burgers mechanism [21]. Such a well-known phase transformation allows us to detect how the interaction between the transformation mechanism and the external fields (e.g. temperature and applied stress) affects the diffuse scattering prior to the MT and the products after the MT.

In our calculations we adopted the isothermal–isobaric ensemble, in which a Nosé–Hoover thermostat was used to control the temperature and a Parrinello–Rahman barostat was used to control the pressure. The initial MD cell (with a bcc structure) has dimensions of $50a \times 50a \times 50a$, where a is the lattice constant of the bcc structure at 0 K. To avoid the existence of free surfaces, periodic boundary conditions in three dimensions were used for all the MD simulations. The MD calculations were carried out using the LAMMPS code [22] and the atomic configurations were displayed using ATOMEYE [23].

We first started the MD simulations on a temperature-induced MT. The ensemble was first relaxed at 400 K for 100 ps to make sure that the system reached the equilibrium state. The ensemble was then cooled from 400 to 150 K at a cooling rate of 0.1 K ps^{-1}. During the cooling process we monitored the changes in MD cell axis lengths as a function of temperature. The results are shown in Fig. 1a. From this figure we obtained a bcc–hcp transformation starting temperature (M_s) of 192.4 K, which is much lower than the experimental one for pure Zr (1135 K). This means that the MT reproduced by this potential is not the same as that for real Zr; we should consider the potential to be that for a model bcc to hcp transformation. However, we note that the understanding obtained from this model material should also be valid for the bcc to hcp transformation of real Zr, as the bcc to hcp transformations in the two systems follow the same mechanism (the Burgers mechanism). Although the higher transformation temperature enhances the strength of thermal fluctuations, it does not alter the transformation mechanism. In addition, previous experimental results show the existence of diffuse scattering prior to temperature-induced bcc to hcp transformation at 1273 K [24], similar to the results obtained from the present MD simulations at low temperature.

Knowing the transformation temperature of the ensemble, we then simulated the stress-induced MT. Initially, the