Statistics of microstructure formation in structural transitions studied using a random-field Potts model with dipolar-like interactions

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Received 14 January 2009
Accepted 20 April 2009
Published 14 May 2009

Abstract. We develop a simple model for the study of a first-order cubic to tetragonal structural transition in athermal conditions in systems with a certain amount of disorder. We perform numerical simulations that allow for a statistical study of the dynamics of the transition when the system is driven from the high temperature cubic phase to the low temperature degenerate tetragonal phase. Our goal is to reveal the existence of kinetic constraints that arise from the competition between the equivalent variants of the product phase and that prevent the system from reaching optimal final microstructures.

Keywords: other numerical approaches, phase transformations (theory), random/ordered microstructures (theory)
1. Introduction

The kinetics of solid–solid structural transitions [1] involving a symmetry change and occurring without long-range atomic diffusion is a challenging problem to which physicists and material scientists have devoted several decades of studies. The complexity of the phenomenon arises from several factors. (i) The first factor is the non-trivial relation between the symmetries of the parent and product phases, especially in real 3D systems. Depending on the symmetry relations between the two phases, the product phase may appear in a number of energetically equivalent variants. (ii) The second factor is the existence of long-range correlations due to the elastic nature of the problem. The effects of the nucleation of a product phase domain in a certain point may influence the kinetics of the transition far away from this point. Minimization of the elastic energy is reflected in the tendency of the different product variants to satisfy some ‘matching’ conditions among them and with the parent phase. A common basic feature is the clear tendency for the variants to grow, forming twins. These are domains of different variants that share a common interface oriented in a certain preferred direction. (iii) A third complexity factor should be considered when transitions are of first order and high energy barriers and associated long-lived metastable states exist. The word ‘athermal’ has been used to refer to those structural transitions where thermal fluctuations do not play any role, and thus evolve through states only when the metastability limits are reached. This feature, combined with the existence of disorder in the systems, leads in many cases to history dependence and avalanche dynamics. (iv) Finally, the microscopic details of the atomic displacements during the transition are also crucial. In general, within transitions which are composition invariant, one distinguishes between martensitic transformations (true diffusionless) and massive transformations (quasi-diffusionless) in which short-range diffusion (a few lattice spacings) along or across the interface is possible [2]. A characteristic difference between the two cases [3] is as regards whether...
the product phase grows by fulfilling strict orientation relations (well determined habit planes) with the parent phase or displays orientation free growth. A lot of progress has been made for the pure martensitic case [4] for which the strict matching between the parent and product phase allows one to determine some basic microstructures. But, in many cases, real materials exhibit mixed behavior, i.e. martensitic phases that grow implying a high shuffling (AuCd [5]) or heterogeneous spontaneous strain that implies imperfect microstructural self-accommodation (NiAl [6]). Another complex example is the olivine–spinel transition in fayalite [7]. This transition has been shown to be pseudo-martensitic, as a coupling of a diffusionless anion sub-lattice transition with a short-range diffusional cation reordering.

In solid–solid transitions, the final microstructures observed in the product phase are a consequence of the complex interplay of these four factors ((i)–(iv)), and thus to have a full understanding of the problem it is not enough to reduce it to the problem of optimizing a certain Hamiltonian. The definition of a ‘kinetic constraint’ [8,9] in much simpler models has been introduced to account for the fact that some low energy states cannot be obtained due to the absence of a possible path to it. Current theories are still far from being able to predict microstructures. Details like sample size and shape, defects, annealing times, quenching rates, etc, are known to change dramatically the final state of the martensite. In fact, it has been known for centuries that obtaining a ‘good’ microstructure in a metallic alloy is an ‘art’ reserved to the best smiths or metallurgists.

Although the final microstructures in structural transitions have been widely studied [10,11], there is not much quantitative experimental information concerning the growth of the microstructure in structural transitions [12,13]. This is because most observation techniques, both direct imaging and scattering techniques (x-rays, neutrons and transmission electron microscopy (TEM)), cannot be performed in situ during the transition. One must stop the transition in an intermediate state by quenching and perform a sample post-treatment for the observation. Besides, in the case of direct imaging techniques it is difficult to extract numerical data from sequences of 2D micrographs of the system surface. There have been some in situ synchrotron radiation experiments [14] that follow the growth of the peaks associated with the product structures, but the studies have mostly concentrated on understanding the precursor effects before the real transition starts. Among the theoretical efforts to understand microstructure formation one should mention many continuum models, derived from elasticity theory [15]–[21]. Despite the advance in the computing power, the complexity of the models (and especially the long-range character of the interactions) is such that it is difficult to perform a large number of simulations and a statistical analysis of microstructures. In many cases the models have been applied only to unrealistic 2D crystals and in others the number of coexisting variants has been reduced artificially or by considering a symmetry breaking external stress.

The goal of this paper is to formulate a simple lattice model for a structural transition that allows us to discuss the existence of dynamic constraints and study the influence on the final microstructure. The model will be formulated for the simple case of a cubic to tetragonal transition and will include a certain degree of elastic interaction to generate twins in the product phase. For the cubic to tetragonal transition of interest here, the twin boundaries are planes perpendicular to the (110) direction (or any of the six equivalent ones) of the cubic phase. The overall microstructures, nevertheless,
are a combination of the different twins and usually look quite random. The cubic to tetragonal transition occurs martensitically in a number of binary metallic alloys for different ranges of concentrations: among others [4] InTl, InPb, NiAl, FePt and FePd [14]; for massive transitions, examples are zirconia based solid solutions like ZrO$_2$–ErO$_{1.5}$ [22] and Zr$_{1-x}$Ce$_x$O$_2$ [23].

Although our model will be fully athermal and will not allow for long-range diffusion, we will not take into account the strong constraints associated with the lattice integrity occurring in a martensitic transition. The growth of the product phase will be orientation free. Therefore the model is more suitable for the description of massive transformations, where the disorder is known to play a crucial role [24] or imperfect martensitic transitions involving a high degree of atomic rearrangement at the interface. The final twin structures, nevertheless, will be similar to those obtained in martensitic transitions.

In a previous paper we introduced a random-field Potts model with truncated dipolar interaction for ‘athermal’ first-order phase transitions between phases with any change of symmetry [25]. The model was based on the $T = 0$ RFIM with metastable dynamics, which was originally proposed for the study of field induced athermal transitions between two ferromagnetic symmetrically equivalent phases. The proposed modifications allowed, for instance, the following to be studied: transitions from a non-degenerate phase to a phase with three different variants, the influence of the amount of disorder on the dynamics and how the parameters of the dipolar interaction term affect the final microstructure.

In the present study we are going to go one step further. We will adapt the model to the symmetries of a cubic to tetragonal structural transition. We will extend the range of the dipolar interaction in order to obtain the twin microstructures observed in such transitions. We have found that it is enough to include up to fourth-nearest neighbors. By truncating long-range interactions we accelerate our simulations, so that we can perform a systematic study of the Hamiltonian parameters and statistics over disorder configurations.

The paper is organized as follows. In section 2 we introduce the Hamiltonian of our model, and discuss the truncation of the dipolar term. We also give details about the dynamics and the simulations. In section 3.1 we present a ground-state analysis of the microstructures that optimize the energy. This will help us in choosing the interesting range of Hamiltonian parameters. In section 3.2 we present some of the microstructures obtained by dynamically driving the system from the parent to the fully saturated product phase. The results are compared with the previous ground-state configurations. In section 3.3 we discuss the shape of the hysteresis cycles and the effect of varying the Hamiltonian parameters (disorder and dipolar intensity) and the sample size. A more quantitative analysis of the microstructures is presented in sections 3.4 and 3.5, where we show the system evolution along the hysteresis cycle in real and Fourier spaces, respectively. Finally in section 4 we both summarize and conclude.

2. The model

Let us consider a simple cubic lattice of size $N = L \times L \times L$, with lattice parameter $a$ and periodic boundary conditions. At each lattice site we define a variable which can take four different values that we will call $\hat{0}$, $\hat{x}$, $\hat{y}$ and $\hat{z}$. As in our previous work [25], we have chosen to represent our variables by considering a vector $S_i^\alpha$ ($i = 1, \ldots, N$), having three components: we will indicate the four possible values as $\hat{0} = (0, 0, 0)$, $\hat{x} = (1, 0, 0)$,
\( \hat{y} = (0, 1, 0) \) and \( \hat{z} = (0, 0, 1) \). These variables can be interpreted respectively as an elementary domain in the cubic austenite phase (vector \( \hat{0} \)) or in the three possible variants of a tetragonal product phase (\( \hat{x}, \hat{y}, \text{and} \hat{z} \)). The interaction between the variants can be described by the following Hamiltonian:

\[
H = -\sum_{\langle ij \rangle} \delta(\vec{S}_i, \vec{S}_j) + \lambda \sum_{i,j=1}^{N} \frac{|(\vec{S}_i \cdot \hat{r}_{ij})(\vec{S}_j \cdot \hat{r}_{ij})|}{|\vec{r}_{ij}|^3} - H \sum_{i} (\vec{S}_i)^2 + H_{\text{dis}}. \tag{1}
\]

The first contribution in the Hamiltonian is a Potts exchange term between nearest neighbors (NN) which accounts for the bulk energy favoring the growth of single-phase domains. The second is the long-range dipolar interaction. The absolute value taken in the dipolar term guarantees the rotational invariance that is expected in the systems that we want to model. The third Hamiltonian term represents the interaction between the external driving field \( H \) and the order parameter \( M = \sum_{i=1}^{N} (\vec{S}_i)^2 \). Note that the vector \( \hat{0} \), corresponding to an elementary domain in the austenite phase, has absolute value zero and thus does not contribute to \( M \). On the other hand, all the three variants of the product phase are here represented by vectors with modulus 1. Consequently, \( M/N \) can be read as the normalized transformed fraction of the sample. Thus, the field \( H \) drives the system from the cubic phase \( m = 0 \) (when \( H = -\infty \)) to a multi-variant tetragonal phase with \( m = 1 \) (when \( H = \infty \)). It therefore plays the role of the temperature for such an ‘athermal’ phase transition.

The last term of the Hamiltonian accounts for the interaction with the quenched disorder (impurities, dislocations, vacancies, etc), which is always present in real materials. Its precise form will be discussed later on in this section.

As explained in section 1, a first version of this model considered only nearest-neighbor dipolar interactions [25]. This produced anisotropic domains of the different variants, but not the correct microstructures observed in structural transitions. In the present work we will introduce higher order dipolar interactions. As will be seen, it is enough to include the interaction between fourth-nearest neighbors (4N), at a distance \( 2a \) from the reference elementary domain (see figure 1). It will be the interplay between the interactions to NN and to 4N (which are placed along the same spatial direction) that generates the convenient microstructures, in a way similar to what happens in the ANNNI model [26].

Although we have also studied the second- and third-nearest neighbor interactions, we have found out that these terms are not crucial for the microstructure formation and do not add any new physics. Therefore, for simplicity in most of the paper they have been neglected, except where indicated otherwise. Moreover, we have allowed the coefficients \( \lambda/|\vec{r}_{ij}|^3 \) that control the decay of the dipolar interaction to have more freedom. Since we are truncating and including only two terms, there is no need to keep the coefficients as in the original model. We have considered two general parameters \( \lambda_1 \) and \( \lambda_2 \) multiplying the dipolar interaction to NN and to 4N. Thus, after these modifications, our Hamiltonian reads

\[
\mathcal{H} = -\sum_{\langle ij \rangle} \delta(\vec{S}_i, \vec{S}_j) + \lambda_1 \sum_{ij} \frac{|(\vec{S}_i \cdot \hat{r}_{ij})(\vec{S}_j \cdot \hat{r}_{ij})|}{|\vec{r}_{ij}|^3} + \lambda_2 \sum_{ij} \frac{|(\vec{S}_i \cdot \hat{r}_{ij})(\vec{S}_j \cdot \hat{r}_{ij})|}{|\vec{r}_{ij}|^3} - H \sum_{i} (\vec{S}_i)^2 + H_{\text{dis}}, \tag{2}
\]

doi:10.1088/1742-5468/2009/05/P05009
where the second sum runs over NN and the third only over the fourth-nearest neighbors. The interaction with quenched disorder has also been simplified from what was originally proposed in [25]. Here, in fact, we will not focus on the study of the influence of disorder, but we need a certain amount of it in order to favor nucleation of the product phase in all the equivalent variants. Thus we have considered

$$H_{\text{dis}} = -\sigma \sum_i \vec{g}_i \cdot \vec{S}_i,$$

(3)

where the quenched random fields $\vec{g}_i$ are independent and identically distributed Gaussian random variables, with zero mean and unitary variance. The parameter $\sigma$ models the strength of the disorder. Note that after the simplifications, the Hamiltonian parameters are only three: $\lambda_1, \lambda_2$, and $\sigma$.

For the study of athermal behavior, besides the Hamiltonian, one must choose a certain dynamics. We chose to implement the extremal update dynamics [25]: starting from a saturated system configuration, corresponding to $m = 0$ at $H = -\infty$, we increase (decrease) the field by small steps $\Delta H$. At each field step we check the contribution to the energy of each $\vec{S}_i$. When a variable is found to decrease the total energy by varying to a new state, we change it to the value that gives the most negative energy change. In this way we guarantee that the system will reach the same state for the same applied field values, independently of the $\Delta H$ value. The states reached are equivalent both macroscopically (same transformed fraction $m$) and microscopically (same values of $\vec{S}_i$ for all the $i$ values). The simulations presented below were obtained by choosing $\Delta H = 0.05$. 

Figure 1. Schematic representation of the nearest neighbors in a cubic lattice, indicating the NN (continuous line) and 4N (dashed line) positions.
Statistics of microstructure formation

Figure 2. System configurations of $\hat{y}-\hat{z}$ twins with widths $w = 1, 2, 3, 6, \infty$, plotted for a system with size $L = 12$.

3. Results

3.1. Ground-state microstructures

Starting from the experimental observations from cubic to tetragonal transitions, our goal would be the growth of the so-called twin variants. These correspond to regions of alternating $\hat{x}$ and $\hat{y}$ (e.g.) domains separated by interfaces parallel to the $z$ axis and forming an angle of $45^\circ$ with the $x$ and $y$ axes. Equivalent structures will be the $\hat{x}-\hat{z}$ twins separated by interfaces parallel to the $y$ axis and forming $45^\circ$ with the $x$ and $z$ axes, and with the $\hat{y}-\hat{z}$ twins separated by interfaces parallel to the $x$ axis and forming $45^\circ$ with the $z$ and $y$ axes.

In addition, we would be interested in controlling the width $w$ of such twins. If one truncates the dipolar term to only first-and second-nearest neighbors, it is not possible to stabilize such twin variants, except for the case with width $w = 1$, which in fact corresponds to domains displaying a chessboard structure as illustrated in figure 2 ($w = 1$). Due to lattice geometry, third-nearest-neighbor interaction cannot lead to wider structures and it is only by including $4N$ terms that we can obtain a $w = 2$ structure, which is represented in figure 2 ($w = 2$).

In order to study the range of parameters in which the different microstructures occur, we have performed a comparison of the energies of the different configurations shown in figure 2, with $w = 1, 2, 3, 6, \infty$. The analytical results presented here are general for any system size provided that the proposed configurations are compatible with periodic boundary conditions.

The values of the energy as a function of the model parameters (in the absence of disorder) of each of the configurations considered above are given by

$$
E_1 = -N + N\lambda_2, \quad E_2 = -2N + N\frac{1}{2}\lambda_1,
$$

$$
E_3 = -\frac{7}{3}N + N\frac{2}{3}\lambda_1 + N\frac{1}{3}\lambda_2,
$$

$$
E_6 = \frac{47}{288}N(-4 + \lambda_1 + \lambda_2) + \frac{97}{288}(-6 + 2\lambda_1 + 2\lambda_2),
$$

$$
E_\infty = -3N + N\lambda_1 + N\lambda_2.
$$

From such energy functions one can obtain the phase diagram that indicates the boundaries between the structures that minimize the energy. This is shown in figure 3.

As can be seen, the equations (4) allow the existence of ground-state twinned structures $w = 1, 2,$ and $\infty$, while no other phases can exist. Although the $w = 2$ structure...
is good enough for our objective of studying kinetic constraints, we have also analyzed the modifications that one should perform on the model in order to obtain ground-state twin structures with larger widths. The guess is that higher order dipolar terms would lead to the stabilization of the wider $w$ domains. To confirm this guess, let us once again consider a $d = 12$ system, and add the seventh-nearest-neighbor term (that is, the neighbors at a distance of $3a$ from the reference elementary domain). The energies of the various phases are now

$$E_\infty = -3N + N\lambda_1 + N\lambda_2 + N\lambda_3,$$

$$E_1 = -N + N\lambda_2,$$

$$E_2 = -2N + N\frac{1}{2}\lambda_1 + \frac{N}{2}\lambda_3,$$

$$E_3 = -\frac{7}{3}N + N\frac{2}{3}\lambda_1 + N\frac{1}{3}\lambda_2,$$

where $\lambda_3$ is the coefficient of the seventh-nearest-neighbor dipolar term. An example of a projection of the phase diagram is shown in figure 4, where it can be seen that the $w = 3$ phase is now stable. The borders between the phases are calculated with equations (5).

For the remainder of the paper we will concentrate on the $\lambda_3 = 0$ case, and in the region where $w = 2$. Most of the results presented will correspond to $\lambda_1 = 10$ and $\lambda_2 = 20$.

### 3.2. Kinetically obtained microstructures

In figure 5(a) we show an example of microstructures obtained by the numerical simulations of the Hamiltonian (equation (2)) using athermal dynamics. We represent only three perpendicular projections of our 3D calculation. This example thus only supplies qualitative information. In section 3.5 we will perform a Fourier analysis in order to gain some global information.

From the figure 5(a) it is clear that some local structures of the kind $w = 2$ arise, but that we are very far from the ground states computed previously (see figure 2).
Figure 4. Phase diagram in the plane $\lambda_1-\lambda_3$ for $\lambda_2 = 2$.

Figure 5. Examples of two microstructures at saturation for $\lambda_1 = 10, \lambda_2 = 20$, $\sigma = 2$ and $L = 36$. In (a), the three-variant saturation microstructure, in (b), the stressed configuration obtained by the application of a high external compression in the $z$ direction. White lines in the $yz$ planes are guides to the eye for the identification of the regions of twinned domains.

The global structure is more complex and displays mixing of the three possible twin domains. Taking into account that there are 3 twin pairs and that each of them can occur in 2 different equivalent orientations and that, given our underlying discrete lattice, there are 4 possible translations (for our $w = 2$ case), the number of different twin domains is 24.

Another interesting example is given in figure 5(b). This figure corresponds to the saturation configuration obtained by the application of a very high external compression in the $z$ direction, disfavoring the associated $\hat{z}$ variant. In this case, the formation of diagonal $w = 2$ domains in the $x-y$ plane is already very clear at first sight, confirming the importance of kinetic constraints arising due to the coexistence of the three equivalent variants. It is interesting too to analyze the microstructure of the $x-z$ and $y-z$ planes. In this case, quite elongated domains formed by vertical alternating stripes with periodicity 2 arise, as indicated in the figure.
In this section, we study the effect of the system size and of the Hamiltonian parameters on the hysteresis cycles. As will be seen in all of the figures of this section, the two branches of the loops are strongly asymmetric, due to the physical difference between the process of transformation from a one-variant phase to a three-variant phase (ascending branch) and vice versa in the descending case.

In figure 6, we show some examples of hysteresis loops obtained by varying the amount of disorder $\sigma$. The loops have a tendency to display plateaus, which are smooth when disorder increases. An additional effect of disorder is also to tilt the hysteresis loop, extending the field range of the transition region.

In figure 7, we present examples of the effect of the dipolar parameters $\lambda_1$ and $\lambda_2$. Within the region of stability of the $w = 2$ phase and when $\lambda_1 > 0$, the plateaus are always present, but are more evident when the disorder is not too high.

Finally, in figure 8 we study the effect of system size on the hysteresis loops. The loops do not show a strong size dependence, and the plateaus are clearly not a finite-size effect. In the following we will restrict our simulations to the affordable intermediate size $L = 36$.

All the hysteresis cycles that we have shown in this section display qualitatively the same behavior, consisting of a certain number (2–3) of plateaus, where the transformed fraction remains constant in a range of $H$ values. This feature, that arises in the $\lambda_1 > 0$ regime, suggests that the transformation of the 0 phase has a tendency to split into steps, the first involving the transformation of more than 50% of the lattice. In the following sections we will try to elucidate the physical reasons behind this tendency. As we will see in section 3.4, no plateaus are shown in the $w = 2$ phase when $\lambda_1 < 0$. Thus we have two dynamic regimes that may possibly be associated with different features in the phase transition.
3.4. Real space analysis of the transition dynamics

In order to investigate the nature of the intermediate steps of the hysteresis cycles we can analyze the system configurations in real space at various points of the loops, as indicated in figure 9(a), for the value $\lambda_1 = 10$ (continuous line). Moreover, in figure 9(a) we present the hysteresis cycle obtained for $\lambda_1 = -18$ (dashed line). As can be seen, while in the first case the transition is characterized by the existence of the plateaus, in the second one we observe a very fast transition constituted basically by one single avalanche. The effect of the $\lambda_1$ sign on the hysteresis cycles can be explained on the grounds of the effect of this
parameter on the transformed domain geometry. As we already analyzed in a previous work [25], negative values of \( \lambda_1 \) lead to needle-like domains, while \( \lambda_1 > 0 \) leads to disk-like domains, whose mutual interaction during the growth and percolation process is much stronger. We will come back on this point later in this section.

We also show the derivatives \( \frac{dm}{dH} \) of the two cycles in figure 9(a), which better reflect the activity of the transition and reveal the importance for \( \lambda_1 = 10 \) of the first transformation step compared to the others. These kinds of curves can be compared with the experimental data from calorimetry or acoustic emission activity.

The microstructures at each indicated point of the ascending branch for \( \lambda_1 = 10 \) are shown in figure 10. A first inspection reveals that the transition basically proceeds by nucleation events. There is some growth, but almost no coarsening since the possibilities for two domains to match are small. One can also see that re-transformation events are common. The observed step in the hysteresis loop is clearly associated with the fact that transformed domains percolate in the sample. At this point the system needs an extra increase of the driving force to proceed. The subsequent domains will grow surrounded by a complex boundary of the product phase. Therefore, the growth dynamics will be very different from the nucleation of the first domains in a cubic phase environment. In the second part, there are many re-transformation events until the final saturated

Figure 9. Hysteresis ascendant loop branch (a) and (b) field derivative of the normalized transformed fraction \( \frac{dm}{dH} \) as a function of the field, for \( L = 36, \lambda_2 = 20, \) and \( \sigma = 2, \) and for the two values \( \lambda_1 = 10 \) (continuous line) and \( \lambda_1 = -18 \) (dashed line). In (b) the \( \lambda_1 = -18 \) curve has been divided by a factor 4 in order to fit in the figure scale. The points marked with numbers 1, 2, 3, 4, 5, 6, 7, 8 correspond to snapshots in figure 10 and have \( m = 0.125, 0.25, 0.375, 0.5, 0.625, 0.75, 0.875, 1 \) respectively.

doi:10.1088/1742-5468/2009/05/P05009
Figure 10. Snapshots of real space configurations for $L = 36$, $\lambda_2 = 20$, $\sigma = 2$, and $\lambda_1 = 10$, corresponding to the field loop points indicated in figure 9. See figure 5 for the color legend.

microstructure is obtained. After a more careful study of the snapshots one can infer that in the first step of the transformation, corresponding to the snapshots from 1 to 6, the system chooses to develop oblate (disk-like) domains. For instance, one can see in snapshot number 3 that the $\hat{x}$ phase (black) tends to be distributed in extended regions in the $y$–$z$ plane, whereas it shows elongated domains perpendicular to the $x$ axis on the $x$–$y$ and $x$–$z$ planes. Such structures correspond to the structures that optimize the NN dipolar interaction term ($\lambda_1$), as we studied in a preliminary work [25]. In a second step, when the external field is comparable to the value of the parameter $\lambda_2$, the system is able to develop the $w = 2$ structures with the desired orientation, and one starts to see regions of twin domains with the 45° orientation on the different planes. Of course, they are very much influenced by the domains that have already grown. This interpretation can even be argued for by confronting the curves for different values of $\lambda_1$ and $\lambda_2$ in figure 7. For a better quantitative understanding of this effect, it is convenient to perform the analysis of the microstructures in Fourier space.

3.5. Fourier analysis

A quite natural way to analyze the system microstructure is to study the Fourier transform (FT) of the density function. In fact, as is well known, the square modulus of this quantity represents the intensity that can be obtained from scattering experiments. In our case, at saturation we have the coexistence of three variants, $\hat{x}$, $\hat{y}$, and $\hat{z}$, and thus it is useful to separately consider three density matrices $\rho_{\hat{x}}(x, y, z)$, $\rho_{\hat{y}}(x, y, z)$, and $\rho_{\hat{z}}(x, y, z)$. In analogy we can define the density matrix of the $\hat{0}$ phase $\rho_{\hat{0}}(x, y, z)$, and transform the four matrices individually. Thus each variant $\alpha$ ($\alpha = \hat{0}, \hat{x}, \hat{y}, \hat{z}$) is associated with a scattered
intensity:

\[ I_{\alpha}(K_x, K_y, K_z) \propto \left| \sum_{b,c,d=1}^{L} \rho_{\alpha}(b,c,d) e^{i(2\pi/L)(K_x b + K_y c + K_z d)} \right|^2, \]

where \( \vec{r} = x^i + y^j + z^k = ba^i + ca^j + da^k \) is the generic position vector, \( b, c \) and \( d \) are integer numbers, and \( K_x, K_y, K_z = 0, L - 1 \). The elements of these four density matrices can take the values 1 or 0, representing, respectively, the occupation or the vacancy of a lattice site for a variant of a given color. The presence of disorder can locally favor one or the other variant, but since the disorder is randomly extracted, we expect variant behaviors to be equivalent on average. We thus restrict ourselves to studying just one variant, e.g., the \( \hat{x} \) one, without loss of generality. In the simulations we have verified that the color equivalence is effectively fulfilled. In the following we will address only this case.

In figure 11 we show the position of the peaks of the squared Fourier transform of the density of the \( \hat{x} \) variant in the ideal case \( w = 2 \). We observe that Bragg peaks occur at the position \( \vec{K} = (L/4, L/4, 0) \) and in symmetric equivalent positions. In fact, if one performs the Fourier transform of the density of \( \hat{x} \) phase corresponding, for instance, to the snapshot in figure 2, one obtains only two of the eight Bragg reflections shown in figure 11. Nevertheless, for symmetry reasons one has to consider the two possible 45° orientations and also that the phase \( \hat{x} \) would be present not only in the \( \hat{x}-\hat{y} \) twin, but also in the \( \hat{x}-\hat{z} \) twin. Translations of the configuration with the same domain orientation
Figure 12. Scattered intensity $I_\mathbf{\hat{x}}$ (linear scale): Fourier transform of the $x$ particle density for a sample with $L = 36, \sigma = 2, \lambda_1 = 10,$ and $\lambda_2 = 20$. Data are averaged over 100 realizations of the disorder. The $\mathbf{K} = (0,0,0)$ peak has been suppressed in order to enhance the contrast.

give the same contribution. Therefore, we shall expect the eight Bragg reflections shown in figure 11 with equal intensity $I_\mathbf{\hat{x}}(L/4,L/4,0) = (1/6)(N^2/8)$, where the contribution $N^2/8$ is given by the intensity of the two secondary peaks that arise for each of the four ideal configurations where the variant $\mathbf{\hat{x}}$ is present, and the factor 1/6 is due to the average over all the six possible ideal configurations. The peak at the origin stands for the fraction of the system which is covered by the $\mathbf{\hat{x}}$ phase, and thus its intensity is given by $I_\mathbf{\hat{x}}(0,0,0) = (4/6)(N^2/4)$, where again the factor 4/6 is due to the average over the six possibilities. In a real experimental case, the widths of the twin domains will be much bigger than the $w = 2$ lattice spacing and, therefore, the growth of the martensitic tetragonal domains will be revealed by satellite peaks occurring close to the cubic Bragg reflections in the directions (1, 1, 0) (and equivalent ones).

In figure 12 we show the Fourier analysis of the microstructures obtained after saturating the system from the numerical simulations. The intensity is averaged over 100 realizations of disorder. We see that the peaks appear at the positions expected from the ground-state structures, but the widths of the peaks are quite large.

In analogy with the study performed in real space, we can study the evolution of the Fourier transform along the hysteresis cycles. This is presented in figure 13. As we have already noticed, the system evolution begins with a tendency to form and grow disk-like domains which correspond to peaks at $\mathbf{\hat{K}} = (L/4,0,0)$ (and equivalent). After the first plateau the intensity is transferred to the final positions at $\mathbf{\hat{K}} = (L/4,L/4,0)$ (and equivalent).
In order to gain a quantitative insight into the dynamics of the transition, we have analyzed the evolution of the scattered intensity corresponding to some significant vectors of the Fourier space. In figure 14(a), we present the evolution of the diagonal $K_x = K_y$ with $K_z = 0$. As it can be seen, the peak is slightly shifted from the ideal position $(L/4, L/4, 0)$ (and equivalent). In particular, the position is closer to the origin, meaning that the formation of some thinner structure is affecting the average. This effect is due to the presence of the disorder and of the dynamic constraints. In figure 14(b), we present the evolution of the intensity as a function of the transformed fraction $m$. We choose $m$ and not $H$ as the independent variable since, as can be seen in figures 6–8, the jumps in the hysteresis cycles are very sharp. As can be seen, the $\vec{K} = (0,0,0)$ and the $\vec{K} = (L/4, L/4, 0)$ values grow monotonically with $m$ as one could expect, and the intensity at $\vec{K} = (L/4, L/4, 0)$ and $\vec{K} = (L/2, L/2, 0)$ tends to disappear at saturation. The scattered intensity in this latter position is a measure of the disorder in the system, since in the ideal configuration the corresponding value of $I_{\vec{k}}$ is zero. In the figure we confront the $\vec{K} = (0,0,0)$ and the $\vec{K} = (L/4, L/4, 0)$ values with the related theoretical values at saturation. The value of the $\vec{K} = (0,0,0)$ peak for $m = 1$ is in perfect agreement with the expected one, while the $\vec{K} = (L/4, L/4, 0)$, even if increasing, as it should in the presence of $w = 2$ structures, keeps far from the ideal value. Furthermore, the predominance of the $w = 2$ structures arises only in the last part of the hysteresis branch, where the reduced transformed fraction goes from $m = 0.875$ to 1. These effects are due
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Figure 14. (a) Intensity $I_\mathbf{x}$ for $m = 0.125, 0.25, 0.375, 0.5, 0.675, 0.75, 0.875, 1$ for the ascendant branch on the $K_x = K_y$ diagonal, with $K_z = 0$. (b) Values of the scattered intensity as a function of the reduced transformed fraction, for $\mathbf{K} = (0, 0, 0), \mathbf{K} = (L/2, L/2, 0), \mathbf{K} = (L/4, L/4, 0)$, and $\mathbf{K} = (L/4,0,0)$. The dot-dashed and dashed line represent the theoretical values of saturation for ideal configurations of the $\mathbf{K} = (0, 0, 0)$ and the $\mathbf{K} = (L/4,L/4,0)$ peaks. The values are averaged over 100 disorder configurations, for system parameters $\sigma = 2, \rho = 0, \lambda_1 = 10, \lambda_2 = 20$, and $L = 36$.

to the presence of disorder and of the kinetic constraints associated with the presence of many competing growing domains.

4. Conclusions

We have presented a model for the statistical study of microstructures in structural phase transitions. We have focused on the cubic to tetragonal case. The model is not aimed at exactly reproducing the microstructure details but only at providing a statistical picture of the different phenomena that may occur during the transition and that will constrain the final product state.

Our model allows some dynamical aspects of the structural transition that occur due to the existence of kinetic constraints to be studied. Among others, we have shown the existence of domain re-transformation, the fact that the transition may proceed through several steps, and the existence of intermediate structures that minimize only the short-
range term of the interaction. The Fourier transform of the microstructures obtained has enabled a quantitative analysis of such phenomena after taking averages over disorder configurations. Although some of the observed phenomena may not be totally realistic due to the cut-off that we have introduced in the long-range interaction, we have shown that the model is good enough to exhibit kinetic constraints. The possibility for extending the interactions to real long-range interactions is still open and, although it will require much longer computing times, should not be excluded for a future work.

In our opinion, the results presented quite clearly justify the importance of a statistical approach to structural phase transitions. We finally propose that, besides the real imaging experiments, many more in situ scattering experiments will be desirable in order to gain understanding of the dynamics of microstructure formation.

**Acknowledgments**

The authors acknowledge fruitful discussions with A Planes. BC would also like to thank P Moretti for very helpful conversations. This work has received financial support from CICyT (Spain), project MAT2007-61200 and CIRIT (Catalonia), project 2005SGR00969 and Marie Curie RTN MULTIMAT (EU), Contract No. MRTN-CT-2004-5052226.

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doi:10.1088/1742-5468/2009/05/P05009