Nucleation Processes in Incommensurate Phases

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Nucleation Processes in Incommensurate Phases*

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The commensurate - incommensurate phase transition in crystals in driven by the nuclei called stripples. The stripple has a structure defined by the symmetries of crystal phases. It is nucleated as a result of thermal fluctuations. Since a macroscopic number of stripples is needed to complete the phase transition a collective nucleation can take place. There are two ideal mechaniscms of collective stripple nucleations: cascade and serial ones. The stripple appearences are illustrated by the results of mocelular-dynamics simulation on the model systems.

KEYWORDS: Commensurate-incommensurate phase transition, nucleation of stripples,molecular dynamics

Some dielectrics and metallic alloys [1] exhibit incommensurate modulations. Usually such a modulated phase can be transformed to a commensurate one. Close to a phase transition point the incommensurate phase can be considered as an ordered sequence of domains of a reference commensurate phase separated by discommensuration planes. The structure of discommensuration planes are similar to that of the domain walls. Carring on the phase transition from commensurate to the incommensurate one the discommensurations appear as a final state of evolution of *stripples*. A stripple is a nucleus [2-4]. It resembles a disc built up from discommensuration planes and bordered by a deperiodization line. Stripples are nucleated by thermal fluctuations in the metastable states of usually overheated crystals, and later they grow, provided they exceed a critical size, similar to the classical nuclei in the first-order phase transition. The pecularity of the incommensurate phase is that a stripple-nuclei has already a structure, which follows from the symmetry considerations of the phase transition similar to those used in deriving possible translational and orientational domains of a crystal. The number of orientational domains is equal to the ratio p_o/p, which is the order of high-symmetry to low-symmetry point groups, respectively. The number of translational domains is given by the ratio w/w_o, where w_o and w are the volumes of the unit cells of high- and low-symmetry phases. In terms of domain walls the incommensurate phase is a periodic ordered sequence of domains $\mathbf{A}_1, \mathbf{A}_2, \ldots \mathbf{A}_S$, where $S = 1, 2, \ldots p_o w/p w_o$, with all domain wall normals oriented along the modulation direction. A period of the sequence $\mathbf{A}_1, \mathbf{A}_2, \ldots \mathbf{A}_S$ corresponds to one period of the incommensurate modulation, and the wave vector of the modulated phase is equal to $k_c \pm (lS)^{-1}$, where l is the separation distance between the domain walls. From the energy point of view the domain walls lower the free energy of the incommensurate crystal, and therefore they are called discommensuration planes. The domain walls always increase this energy.

In three-dimensional crystal the energy of the stripple consists of two essential contributions: the negative energy of the discommensuration planes which form the stripple and which is proportional to its surface $\pi R^2 E_{dc}$, and the positive energy of the core of the deperiodization line proportional to the stripple periphery $2\pi R E_{cor}$, where R is the average radius of the stripple. The maximum condition of stripple energy $\Delta E = \pi R^2 E_{dc} + 2\pi R E_{cor}$ determines the critical size $R_c = -E_{cor}/E_{dc}$. Below R_c, the stripple will shrink and above R_c, it will grow.

The peculiarity of the incommensurate phase is that one stripple adds to the system only single new modulation period, what usually means a few discommensuration planes. Thus, usually a vast number of stripples must be nucleated in order to complete the phase transition.

The nucleation processes of stripples has been studied by MD computer simulation technique. Special choise of a simple displacive type of potential energy assured that the simulated system exhibited commensurate-incommensurate phase transition. We shall refer here some results which demonstrate the structure, nucleation and growth of stripples and they collective evolution.

On Fig.1 we show the nucleation of stripple at the end of commensurate $k = 1/2$ - incommensurate phase transition [5] for the system of $30 \times 30 \times 60$ unit cells. Maps Fig.1a, b, c correspond to subsequent time moments. On the first map, Fig. 1a, the incommensurate and commensurate phases coexist and the moment of stripple nucleation is seen. It grows in lateral directions as seen in maps, Fig.1b and 1c. Its deperiodization line, as a closed cirle, is visible in the $X - Y$ plane.

Stripple of more elaborate structure will appear in the phase transition from commensurate phase $k = 1/4$ to the incommensurate one. The commensurate phase $k = 1/4$ is a superstructure with a unit cell four times enlarged along the modulation direction. Four translational domains exist in this case, hence, the stripple is build from four discommensuration planes. Indeed, Fig.2 shows the growth of two stripples nucleated in the hexagonal model [6], exhibiting the phase transition along vertical direction. It is quite clear that each such nucleus consists of four domains. In Ref. [7] stud-

*IMR, Report No. 2059

ies of the stripple nucleation in the strained crystal has been performed. Nucleation of six domain stripples close to commensurate phase $k = 1/3$ has been also demonstrated by computer simulation in Ref. [8].

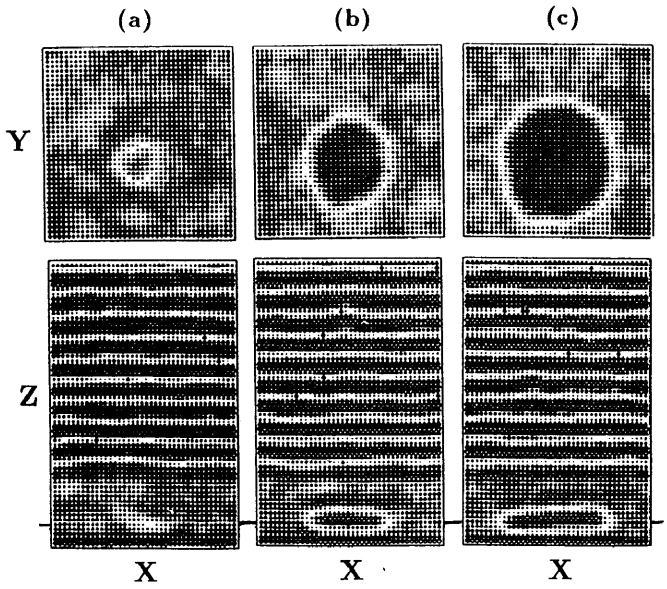

Figure 1. Maps of particle configuration showing the coexistence of the commensurate and incommensurate phases and an arising stripple. Crystallite size $30 \times 30 \times 60$ unit cells.

In the first-order phase transition one nuclei is, in principle, able to transform the whole volume of the crystal. A peculiarity of incommensurate phases is that one stripple adds to the system only single new modulation period. Consequently, it changes the modulation wave vector by a negligible quantity $\frac{1}{N}$, where N is the size of the crystal along the modulation direction. To fill the volume of the crystal with a definite density of the discommensurations, a huge number of stripples must be nucleated. The question immediately arises whether the nucleation of the vast number of stripples is correlated or not? An answer to this question was already attempted and two extreme mechanisms have been proposed: *the cascade* and *serial* ones [9,6]. The cascade mechanism takes place in the transition from the commensurate to the incommensurate phase. The cascade starts from a nucleation of a single stripple at random place. The next stripples nucleate in the vicinity of the former one. This process is repeated, and it can be considered as a frontal motion of the commensurate-incommensurate interface. This mechanism occurs when the newly created discommensurations are pinned and reluctant to shift.

In the serial mechanism, the stripples arise one after another in a random layer of the crystal, perpendicular to the modulation direction. The discommensurations produced by each stripple diffuse away to give space for the next stripples. The pinning of discommensurations might suppress this mechanism.

Summarizing, one should emphesise that the structure of the stripples can be uniquely derived from the symmetry considerations of the phases. The nucleation of stripples follows the general rules of classic nucleation theory. The collective nucleation mechanisms have been very litle studied. There is still a lot to do to understand the influence of crystal defects on the nucleation and propagation processes of discommensurations.

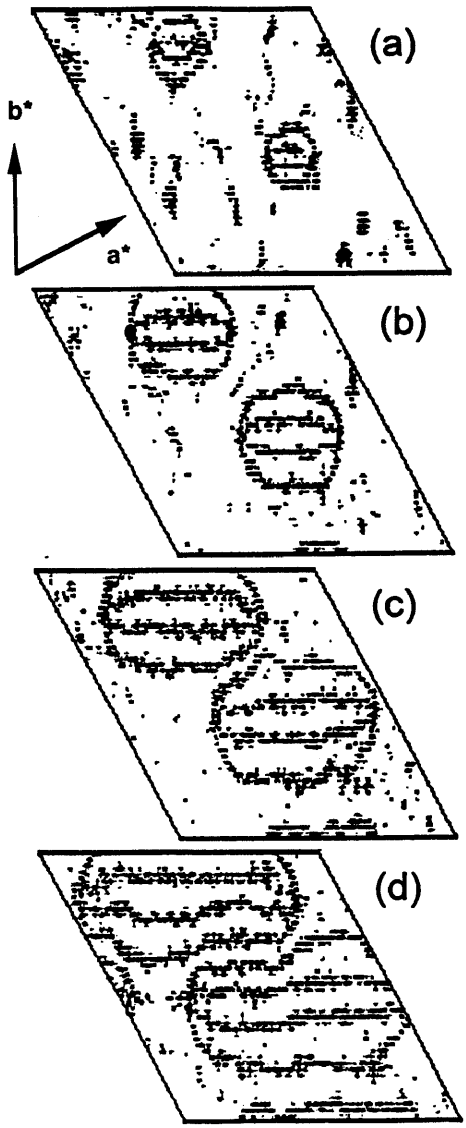

Figure 2. Maps of discommensurations in the crystallite of $88 \times 88 \times 5$ unit cells of the hexagonal model.

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