Hierarchical Phase Transformation in $Mg_{85}Y_9Zn_6$ Alloy

The formation process of long-period stacking ordered (LPSO) structures in $Mg_{85}Y_9Zn_6$ ternary alloy from melt-quenched amorphous ribbon has been examined by in-situ small- and wide-angle scattering/diffraction measurements. The 18R-LPSO structure consists of periodical stacking faults introduced every 6 atomic layers, with segregation of solute clusters of $L1_2$-Y_8Zn_6 having a $2\sqrt{3}x2\sqrt{3}$ superstructure at the stacking fault region. In-situ measurements revealed that clustering of Y and Zn occurs first, followed by spatial arrangement of clusters accompanying the introduction of stacking faults.

Mg alloys containing 3d transition metals (TM) or Al, and rare earth (RE), form a series of ordered alloys called long-period stacking ordered (LPSO) structures [1]. Due to their good performance at high temperatures, these light-weight metallic materials have attracted attention as structural materials. Mg-Y-Zn ternary alloy is a representative alloy system, where the LPSO structure is readily observed in as-cast ingots. Regarding phase transformation, how such a complex phase forms is an interesting question; it is also important to develop and design conditions for processing the material. Since the phase boundary of LPSO is reported to touch the liquidus directly, a supersaturated solid solution can not be attained by quenching. Therefore, we conducted in-situ small- and wide-angle scattering (SWAXS) measurements of melt-quenched ribbon of $Mg_{85}Y_9Zn_6$ alloy, which is reported to be a composition for single-phase 18R LPSO. Thermodynamically, it is not possible to obtain a solid solution at any temperature. Instead, the melt-spun ribbons we used in the present experiments were found to be amorphous with no macroscopic segregation/precipitation detectable by conventional SEM or laser microscope. In principle, we expect two kinds of phase transition to proceed concomitantly while heating the ribbon at a constant rate of 10 K/min. One is the formation process of LPSO structures [2], and the other is crystallization and crystal growth in the $Mg_{85}Y_9Zn_6$ ribbon [3]. In-situ SAXS clearly showed small-angle scattering of crystallites upon reaching the crystallization temperature, T_x, and another SAXS component at higher q, whose origin is solute clusters [2]. Therefore, SAXS components from crystallites and clusters in them were separately examined. **Figure 1** shows the evolution of clusters during heating of the sample. Even in the amorphous state, the cluster size suggests some association of solute atoms. Upon crystallization, the cluster size increases with temperature, eventually reaching the size of the $L1_2$-Y_8Zn_6 cluster identical to the one in the equilibrium LPSO structure. The illustrations of clusters in the figure denote the shape of the cluster corresponding to the given R_g, obtained by relaxed atomic positions calculated using VASP. This suggests that the

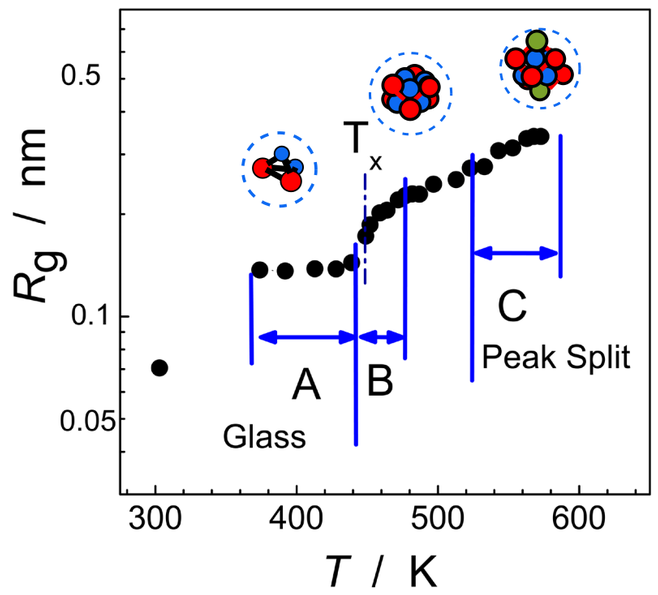

Figure 1: Growth of solute clusters during heating.

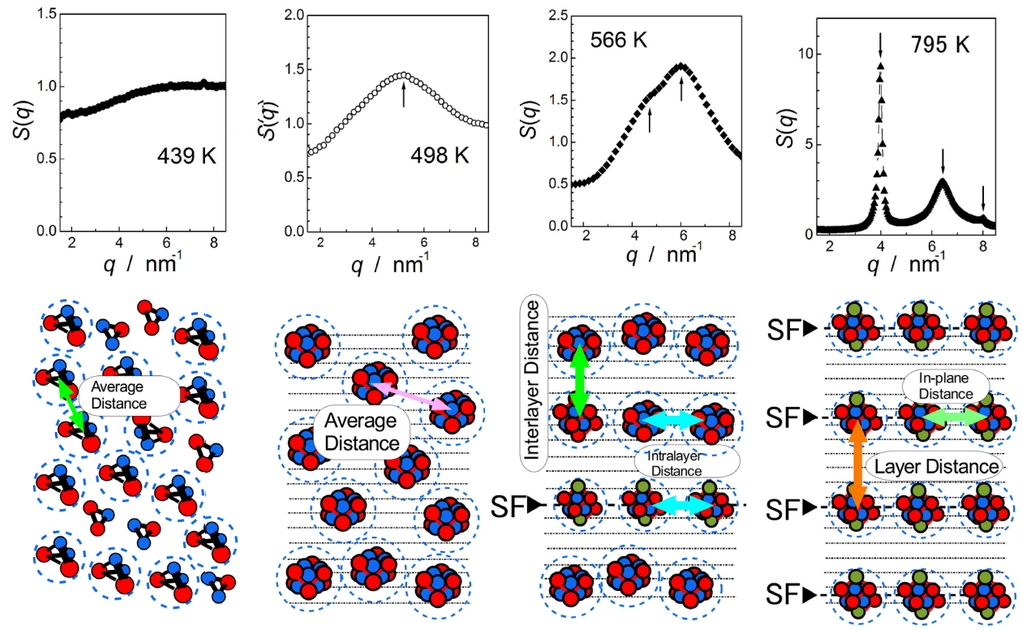

Figure 2: Structure factors representing spatial arrangements of clusters and corresponding schematic illustrations of cluster arrangements.

initial stage of formation of LPSO is not nucleation of small LPSO unit structures, but uniform nucleation and growth of small clusters that eventually form LPSO. It also rules out a thermodynamical model in which diffuse compositional modulation along the *c* axis develops to induce structural phase transition, leading to two-dimensional ordering of clusters in the later stage. Since **Fig. 1** suggests that the early stage of phase transformation is described by a cluster model, the spatial distribution of the cluster can be analyzed by using a simple form factor and structure factor model. Therefore, we used a cluster model to examine the change of spatial arrangements of clusters by calculating the structure factor. **Figure 2** shows the structure factor obtained for several temperatures, along with schematic illustrations of cluster arrangements corresponding to the structure factor. In the early stage of phase transformation, just after crystallization, the cluster size is very small, and the clusters are distributed isotropically in space as shown by a single diffuse peak of the structure factor in **Fig. 2**. As the temperature increases, the clusters grow to reach the size of the $L1_2$-Y_8Zn_6 cluster, when the introduction of stacking faults and spatial ordering of cluster arrangements, i.e., separation of the interlayer distance and in-plane distance of the clusters, are observed. To summarize, in-situ SWAXS measurements of amorphous MgYZn alloy at BL-6A revealed that the phase transformation is characterized by a hierarchical phase transition, comprising the first cluster nucleation/growth and the second cluster arrangements to form the LPSO structure.

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BEAMLINE

BL-6A

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