Deformation twinning in a Mg-Al-Gd ternary alloy containing precipitates with a long-period stacking ordered (LPSO) structure

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Abstract
A magnesium alloy containing the Mg-Al-Gd long-period stacking ordered (LPSO) platelet precipitates parallel to the basal plane was found to deform by the *c*-axis tension twinning on $\{11\bar{2}1\}$, whose passage causes the bending of the LPSO platelets, when Mg grains are oriented favourable for the extension along the *c*-axis during the plane-strain compression at room temperature. The bending of the LPSO platelets was found to be caused by the deformation twinning equivalent to the $\{11\bar{2}1\}$ twinning in the Mg matrix.

Keywords: magnesium alloys; deformation structure; twinning; transmission electron microscopy (TEM)

Recently, magnesium alloys containing Mg–TM (transition-metal)–RE (rare-earth) ternary precipitates with long-period stacking-ordered (LPSO) structures have attracted considerable attention as promising light-weight structural materials because of the simultaneous achievement of high strength (~600 MPa) and good ductility (~5 %) [1]. It is usually reported that these excellent mechanical properties are achieved only after extrusion at high temperatures above 350°C [2]. Although grain refinement of the Mg matrix in the vicinity of bent LPSO platelet precipitates as a result of recrystallization has been considered as a possible reason for the observed high strength and good ductility [3,4], the detailed mechanisms behind this have largely remained unsolved. The lack of knowledge on fundamental properties of the LPSO phase such as crystal structure, thermal stability and deformation mechanisms is largely responsible for this. Hagihara et al. [5,6] were the first who made a systematic study on the deformation mechanisms of the LPSO phase (Mg - 5 at.%Zn - 7 at.%Y) with the use of directionally-solidified (DS) ingots consisting of LPSO platelets with their platelet faces (parallel to the basal plane) being nearly parallel to the growth direction. They concluded that basal slip is by far the easiest slip system operative in most orientations while deformation bands are formed when the compression axis is almost parallel to the basal plane so that the operation of basal slip is suppressed. They further concluded from transmission electron microscopy (TEM) observations that deformation bands they observed are actually kinks formed by numerous basal dislocations accumulated perpendicularly to the basal planes in a wall, as in the classical explanation for the kink formation in hexagonal close-packed (hcp) metals by Hess and Barrett [7]. They inferred that the kink formation is responsible for the bending of LPSO platelet precipitates observed in hot-extruded Mg alloys [5]. However, the rotation angles between two crystalline regions separated by a kink boundary in their Figs. 14 and 15 of [5] are as large as 30 ~ 60° that requires a basal dislocation every few basal planes in the kink wall. On top of that, it is very difficult to see each individual dislocation in the kink wall in their Figs. 14 and 15 of [5]. These raise a serious question as to whether or not all these kink bands are formed indeed by the accumulation of basal dislocations, which moved on closely-spaced atomic planes, in the wall perpendicular to the basal plane. In the present study, we investigate deformation microstructures of a Mg-Al-Gd alloy containing a ternary LPSO phase after plane-strain compression deformation at room

temperature, in order to gain insight into mechanisms behind the bending of LPSO platelets during hot extrusion, excluding any high-temperature effects such as dynamic recrystallization. In our recent studies [8,9], the crystal structure of the LPSO phase in the Mg-Al-Gd system has been elucidated to be based on the stacking of 6-layer structural blocks (18*R*-type) as described with the order-disorder (OD) theory [8-10], possessing generally a one-dimensional disordered nature along the stacking direction.

An ingot of a Mg-Al-Gd ternary alloy with a nominal chemical composition of Mg - 1.5 at.%Al - 6.5 at.%Gd was produced by high-frequency induction-melting mixtures of high-purity Mg, Al and Gd in a carbon crucible with a lid in vacuum. The ingot was heat-treated at 525 °C for 64 hours. Rectangular specimens for plane-strain compression tests measuring a dimension of 2.2 mm × 2.3 mm × 5.5 mm (in *x, y z* directions respectively in Fig. 1(a)) were cut from the heat-treated ingot by electric discharge machining, and were polished mechanically and then electrolytically in a solution of perchloric acid, n-butyl alcohol and methanol (1 : 30 : 130 by volume) with 0.2 M of LiCl at -55 °C. Plane-strain compression tests were performed at a nominal strain rate of 5.5×10^{-4} s^{-1} at room temperature with a channel die similar to that designed by Chin et al. [12]. As seen in Fig. 1(a), the compression axis was set parallel to the *z* axis, while the channel die was placed to suppress specimen deformation in the *y* axis, allowing specimen extension only in the *x* direction. Compression deformation was made until the reduction measured in the *z*-axis reaches about 15 % with the specimen wrapped in polytetrafluoroethylene (PTFE) film to reduce friction with the channel die and compression jig. Deformation microstructures were examined by scanning electron microscopy (SEM), TEM and scanning transmission electron microscopy (STEM). Specimens for TEM/STEM observations were cut perpendicular to the *y*-axis of the specimen, polished mechanically and then ion-milled with 5 (initial) or 2 (final) keV Ar ions.

A SEM backscattered-electron image of Fig. 1(b) depicts a deformation structure of a Mg grain that is located on the cross section perpendicular to the *y* direction in Fig. 1(a). For this particular Mg grain, the compression axis is approximately parallel to $[25\bar{5}\,\overline{20}\,1]$, which is almost parallel to the basal plane. The *x, y* and *z* axis directions are indicated in Fig. 1(b). Many thin platelet precipitates of the LPSO phase, which appeared as bright bands in Fig. 1(b) with platelet faces being parallel to the basal plane of Mg, are observed to be bent frequently, as observed as deformation kinks after hot extrusion [4]. It is noteworthy in Fig. 1(b) that the bending angles observed for LPSO platelets are almost identical. Orientation mapping by electron back-scattered diffraction (EBSD) clearly indicates that the bending of LPSO platelets is formed as a result of the passage of deformation bands, as seen in Fig. 1(c). There are two sets of deformation bands; one with the boundary parallel to $(11\bar{2}1)$ and the other with the boundary parallel to $(\overline{11}21)$. Each of these two stes of deformation bands has an identical orientation, exhibiting a particular orientation relationship with the rest of areas (designated arbitrarily as the matrix). Orientation analysis by EBSD for the Mg grain has revealed that the matrix and the deformation bands with boundary planes parallel to $(11\bar{2}1)$ and $(\overline{11}21)$ are both approximately in 34°-rotation relationship around $[\bar{1}100]$. This is further confirmed by TEM observations, as shown in Fig. 1(d). A TEM bright-field image of Fig. 1(d) clearly indicates that the bending of LPSO platelets is always accompanied by the passage of deformation bands with the boundary plane parallel either to $(11\bar{2}1)$ or to $(\overline{11}21)$. In addition, many dislocations are observed to be generated in the Mg phase in both the matrix and deformation bands. Diffraction analysis by TEM/STEM has indicated that deformation bands are indeed deformation twins, as described below.

Figure 2(a) shows a magnified bright-field (BF) STEM image of a region containing a deformation band with the boundary planes being macroscopically parallel to $(11\bar{2}1)$. A LPSO platelet precipitate is imaged as a dark band extending along the lateral direction in Fig. 2(a). The LPSO platelet precipitate is obviously bent by the passage of the deformation band. The

boundaries between the deformation band and the matrix are parallel to $(11\bar{2}1)$* and they are observed edge-on, since the incident beam direction is $[\bar{1}100]$. Selected-area electron diffraction (SAED) patterns taken from regions containing both the deformation band and matrix are shown in Figs. 2(b) and (d) for the Mg and LPSO phases, respectively.

Analysis of the SAED pattern of Fig. 2(b) indicates that the matrix and deformation band of the Mg phase have a common (mutual) zone-axis orientation of $[\bar{1}100]$, these two regions are rotated with respect to each other by approximately 34° about the zone-axis orientation and they are in a mirror relationship with respect to the boundary plane of $(11\bar{2}1)$, as schematically shown in Fig. 2(c). These observations obviously indicate that deformation bands that make LPSO platelet precipitates bent are deformation twins. Based on the above observations, the twinning elements for the deformation twins on $\{11\bar{2}1\}$ in the Mg phase are determined as follows,

K_1: $\{11\bar{2}1\}$, K_2: $\{0001\}$, η_1: $<\bar{1}\bar{1}26>$, η_2: $<11\bar{2}0>$ and $s = 0.616$ (1).

The same rotation relationship with a rotation angle about 34° is confirmed to occur in the LPSO phase by analyzing the SAED pattern of Fig. 2(d). The possible twinning elements for the LPSO phase of the 18*R*-type are then deduced by assuming the simplest polytype of 1*M* (space group *C*2/*m*) among the possible OD structures [8,9,11], as follows:

K_1: $\{041\}_{1M}$, K_2: $\{001\}_{1M}$, η_1: $<4\ \bar{3}\ 12>_{1M}$, η_2: $<010>_{1M}$ (2a),

K_1: $\{661\}_{1M}$, K_2: $\{001\}_{1M}$, η_1: $<1\ 3\ \bar{24}>_{1M}$, η_2: $<310>_{1M}$ (2b),

K_1: $\{\bar{6}\bar{6}5\}_{1M}$, K_2: $\{001\}_{1M}$, η_1: $<17\ 3\ 24>_{1M}$, η_2: $<\bar{3}\bar{1}0>_{1M}$ (2c).

Three different sets of twinning elements are deduced for three different orientation variants because of the monoclinic symmetry of the 1*M* polytype. These indices are referred to the 1*M* polytype with the unit cell described as follows,

$$a_{1M} \approx 2\sqrt{3}a_{\mathrm{Mg}}, b_{1M} = \sqrt{3}a_{1M}, c_{1M} \approx \sqrt{(a_{1M}/3)^2 + (3c_{\mathrm{Mg}})^2}, \beta_{1M} \approx 180° - \tan^{-1}(9c_{Mg}/a_{1M}),$$

where a_{Mg} and c_{Mg} are the lattice parameters of Mg with the hcp structure.

Figure 3 shows an atomic-resolution HAADF-STEM image taken from a twin boundary region in the Mg-Al-Gd LPSO phase projected along the $[1\bar{1}00]$. The ordered arrangement of brighter spots in the double-dagger pattern for the central four consecutive Gd-enriched layers is observed in both the matrix and twin regions. This confirms that deformation twinning occurs so that the ordered atomic arrangement in each of structural blocks in the OD structure of the Mg-Al-Gd LPSO phase, which is characterized by the periodic arrangement of Al_6Gd_8 clusters with the $L1_2$-type atomic arrangement on lattice points of a $2\sqrt{3}a_{\mathrm{Mg}} \times 2\sqrt{3}a_{\mathrm{Mg}}$ two-dimensional primitive hexagonal lattice [8,9], is preserved after deformation twinning. Of importance to note is that the twin boundary is not perfectly flat but is deviated from the symmetric position on an atomic scale and that strain contrast arising from dislocations is scarcely observed on the boundary between the deformation band and matrix. These are in contrast to what is reported for kink bands by Hagihara et al. [5] for the compression behavior of directionally-solidified (DS) LPSO phase, indicating that deformation bands observed in this study are not kink bands that are formed by a mechanism proposed by Hess and Barrett but are deformation twins [7].

In the OD structure of the Mg-Al-GD LPSO phase, the stacking of structural blocks is virtually one-dimensional disordered, which can easily be recognized by investigating the

* Since the unit cell of the LPSO phase in the Mg-Al-Gd system cannot unambiguously be determined because of the stacking disorder of structural blocks along the direction perpendicular to the close-packed planes, Miller indices to express directions and planes for the LPSO phase are referred to as those of the matrix phase of Mg with the hcp structure unless otherwise stated.

relative shifts of Gd atom potions in the outer layers of structural blocks, as described in our previous papers [8,9]. Inspection of the matrix and twin regions in the experimental HAADF-STEM image of the $[1\bar{1}00]$ incidence (Fig. 3) indicates that shifts of either 0 or 1/3 occur preferentially for both the matrix and twin regions. This observation confirms that the OD structure designated as the C_1-type in our previous papers [8,9] is formed in the twin region as in the matrix, still exhibiting the one-dimensional disorder for the stacking of structural blocks of the OD structure. The stacking sequence of structural blocks is not perfectly in mirror relation with respect to the $(11\bar{2}1)$ twin boundary plane for the matrix and twin regions. Such a change in the stacking sequence of structural blocks during deformation twinning in the LPSO phase may be closely related to the formation (nucleation and growth) mechanisms of deformation twins of this type. Mechanism for deformation twinning involving the atomic movement are currently under survey in the authors' research group.

The present results clearly suggest that deformation twinning observed in this study may occur also during the hot extrusion process of Mg alloys containing LPSO platelet precipitates, if LPSO platelets are oriented so that the platelet face is parallel to the extrusion direction. We believe that this is also the case for A-oriented specimens used in the study on the DS-processed Mg-Zn-Y LPSO phase by Hagihara et al. [5,6], when judged from relatively homogeneous shapes of deformation bands observed in compression nearly along the basal plane. Most deformation bands with relatively large rotation angles are believed to be deformation twins the same as those observed in this study. Once deformation twins are formed, the basal plane inside the twins is rotated so as to favorably oriented for the activation of basal slip, which leads to an additional increase in the rotation angle of the twin region with respect to the matrix, possibly by the kink mechanism proposed by Hess and Barrett [7]. We thus believe that large rotation angles between the matrix and deformation bands and a rather wide distribution of the rotation angles reported by Hagihara et al. [5] should be interpreted by the formation of deformation twins and the subsequent lattice rotations inside the twin region by the motion of basal dislocations.

The $\{11\bar{2}1\}$ deformation twin identified presently in the Mg matrix corresponds to the *c*-axis tension twin, which has hardly ever been reported in Mg and its alloys [13]. In the polycrystalline specimen tested in the present study under a plane-strain compression condition, deformation twins are confirmed to be activated in Mg grains with the loading axis orientation *z* and the extension direction *x* being nearly parallel to the basal plane and the *c*-axis, respectively. Since basal slip is by far the easiest slip system in Mg, it is easily imagined that this geometrical situation can be realized for many Mg grains during hot extrusion so that $\{11\bar{2}1\}$ deformation twins are formed in relevant Mg grains. In view of the fact that $\{11\bar{2}1\}$ deformation twins have hardly ever been observed in Mg and its alloys, the LPSO phase may play a decisive role in the nucleation of $\{11\bar{2}1\}$ twins. In fact, our preliminary study on the compression deformation behavior of the Mg-Zn-Y LPSO phase with the use of single-crystalline micropillars has revealed the activation of similar deformation twins with their habit planes close to $\{11\bar{2}1\}$ in the Mg-Zn-Y LPSO phase [14].

In summary, deformation microstructures of a Mg-Al-Gd alloy with LPSO platelet precipitates after plane-strain compression at room temperature were investigated. The *c*-axis tension twins on $\{11\bar{2}1\}$, which have hardly ever been reported in Mg and its alloys, were confirmed to be activated in both the Mg matrix and the Mg-Al-Gd LPSO platelet precipitates. Three different sets of twinning elements for the deformation twins in the Mg-Al-Gd LPSO phase are deduced for three different orientation variants by assuming the simplest MDO polytypes with the monoclinic symmetry (1*M* polytype) as equations (2a)-(2c).

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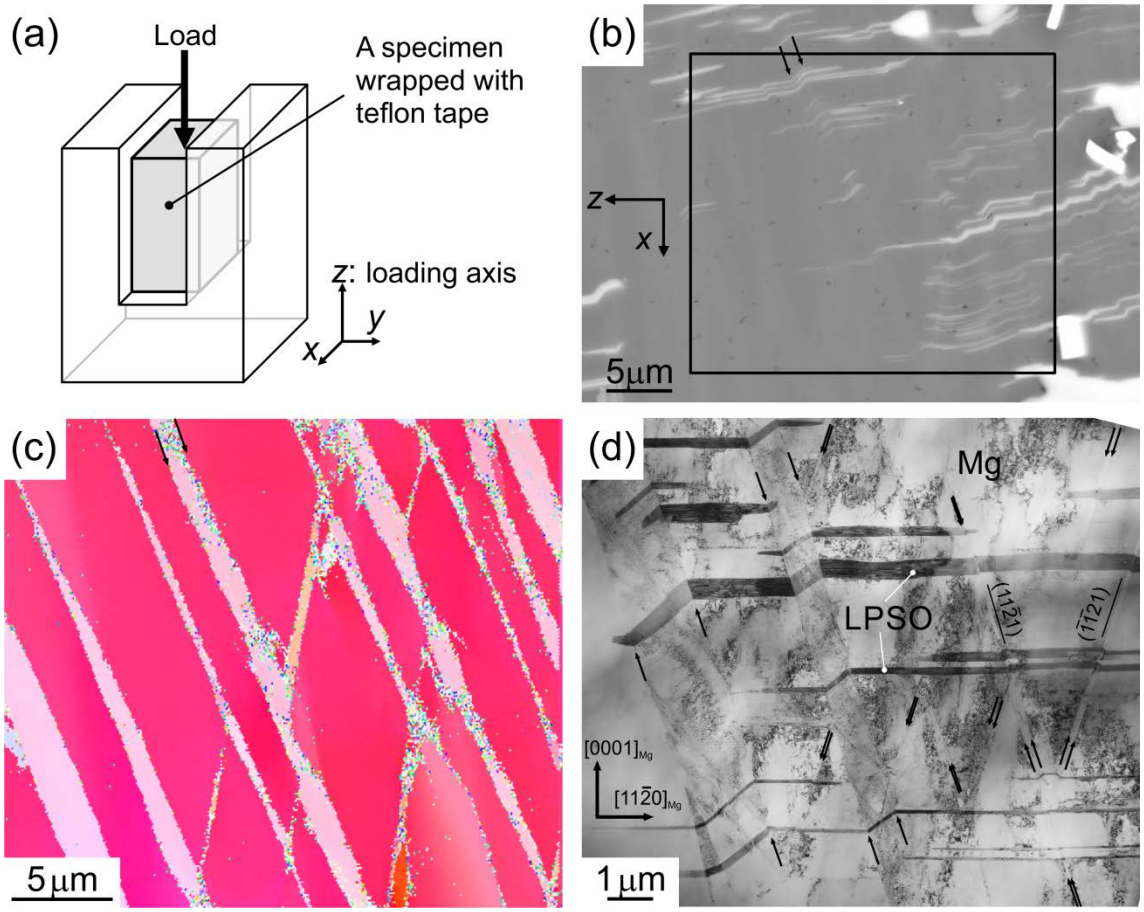

Fig. 1. (a) Geometrical configuration of plane strain compression and (b-d) deformation microstructure of Mg-Al-Gd ternary ingot deformed in plane strain compression. (b) SEM backscattered electron image, (c) EBSD orientation map and (d) TEM BF image. Arrows in (b) and (d) indicate boundaries between matrix and twinned regions.

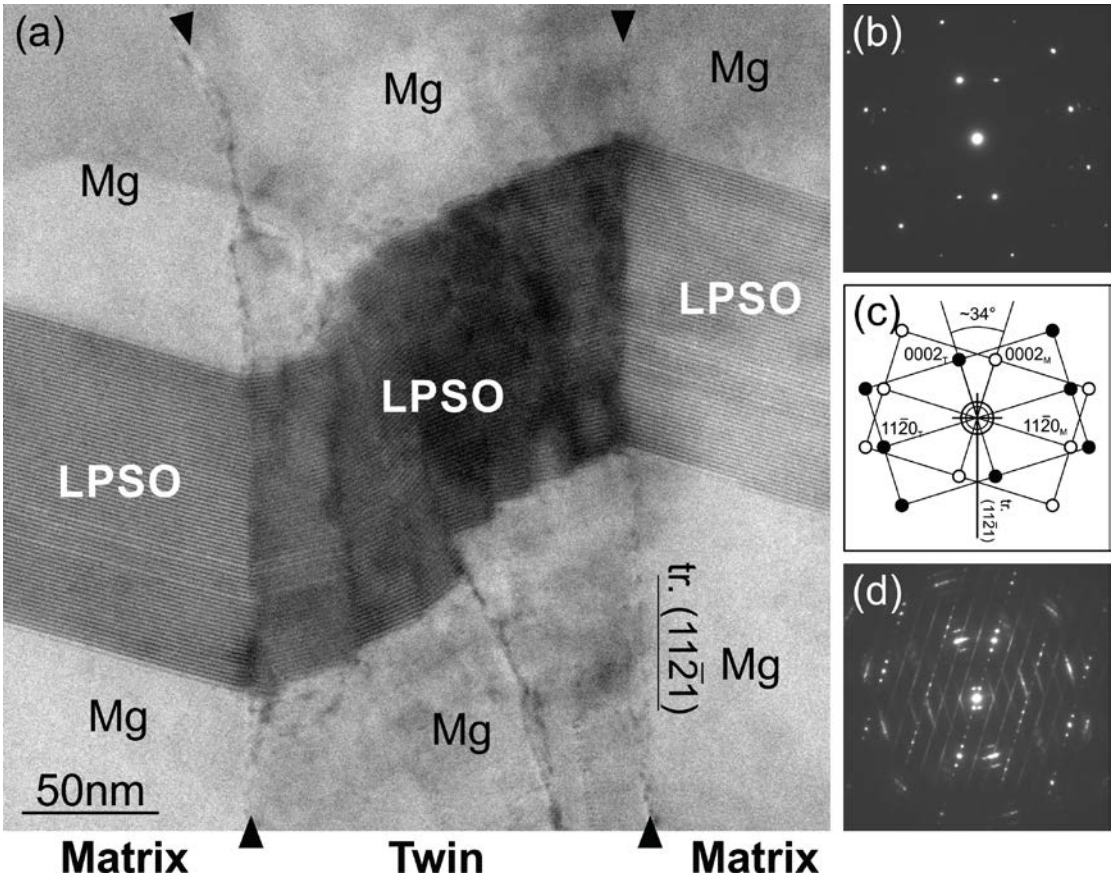

Fig. 2. (a) a BF-STEM image of a deformation twin and (b-d) SAD patterns taken from (b) Mg phase and (d) LPSO phase. (c) Schemtic illustration of the SAD pattern of (b).

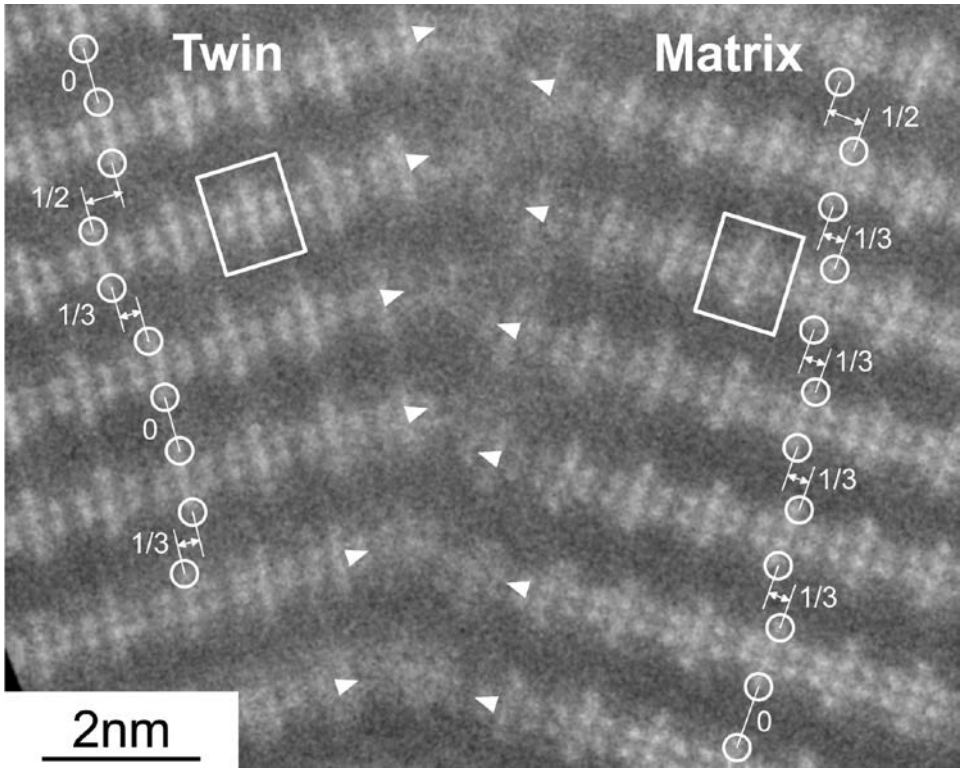

Fig. 3. Atomic resolution HAADF-STEM image of a boundary region of the deformation twin in the Mg-Al-Gd LPSO phase.