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The influence of glass transition temperature on the critical size for deformation mode transition in metallic glassy films

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The size effect on the bending deformation behaviour of magnetron-sputtered $La_{57}Al_{25}Co_{18}$, $Cu_{50}Zr_{50}$ and $Fe_{70}Y_8B_{22}$ glassy films was investigated. The transition of the deformation mode from highly localized to non-localized occurs as the film thickness reduces below the critical value, which does not exhibit a distinct dependence on Poisson's ratio. By combining the already-reported critical size for deformation mode transition in various metallic glasses, it is found that the critical size is primarily proportional to the homologous temperature.

Keywords: Metallic glassy film; Deformation mode transition; Glass transition temperature; Poisson's ratio; Size effect

The notion that smaller is more ductile was widely recognized in submicron-sized metallic glasses (MGs) [1]. The transition in the deformation mode from highly localized to non-localized was observed as the specimen size decreases to nanoscale because there is not enough elastic energy stored in the volume to feed the shear band propagation [2], although there have been a number of inconsistent results on critical size for non-localized deformation [3–11]. There have also been a number of controversies concerning yield strength: MGs became stronger by reducing the specimen size due to fewer defects for shear band initiation (suppressed shear banding) [3], but several groups revealed that no evident size dependence exists [4–10].

Recently, Kuzmin et al. [9] prepared the pillar specimens using the focused ion beam technique, and investigated the size effect of deformation behaviour in Cu–Ti–Zr–Ni–Sn–Si, Zr–Ti–Cu–Ni and Zr–Cu–Ni–Al, Al–Ni–Y MGs by in situ transmission electron microscopy (TEM) compression tests. By combining previous results for Zr–Ti–Co–Be, Zr–Cu–Ni–Al–Nb and Au–Ag–Pd–Cu–Si MGs, it was proposed that with increasing Poisson's ratio (ν), the critical size for deformation mode transition became larger. This is consistent with the observation of Wei et al. [12] on bent MGs with different values of ν, i.e. that a higher ν is found to make plastic deformation more uniform, with smaller shear-band spacing and shear offset. However, in a recent tensile experiment for Pt–Cu–Ni–P MG nanowire with a high ν of 0.42, Magagnosc et al. [11] did not observe the transition in deformation mode, even when the diameter was reduced to 100 nm. Note that in Ref. [9] the Au-based MG, possessing the highest ν and largest critical size for non-localized deformation at room temperature among the investigated MGs, actually has the lowest glass transition temperature (T_g), at ~401 K [13]. An interesting question has been raised: are there any other factors possibly influencing the critical size besides ν? It is well known that the deformation behaviour of MGs is closely associated with the homologous temperature (T/T_g) [14]. When tested at room temperature, MGs with lower T_g definitely have a stronger tendency to plastically deform in a homogeneous way, which may affect the critical size for non-localized deformation and make the judgement of the role of ν more difficult. In this work, we prepared various MG thin films with a range of ν and T_g by direct current magnetron sputtering, and the size effect of deformation mode was systematically investigated. It was revealed that the critical film

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thickness for the deformation mode transition from highly localized to non-localized mainly depends on T_g, i.e. the higher T/T_g, the larger critical thickness for non-localized deformation. A possible mechanism is also discussed.

Thin films with different thicknesses were magnetron-sputtered on the (1 0 0) Si wafer and the polished Ti substrate by using $La_{60}Al_{20}Co_{20}$ (at.%), $Cu_{50}Zr_{50}$ and $Fe_{72}Y_6B_{22}$ crystalline target. The detailed preparation method for thin films is described in Ref. [15]. The compositions of La–Al–Co and Cu–Zr thin films were determined by energy-dispersive X-ray spectroscopy, using a field-emission scanning electron microscope (Hitachi S-4800), to be $La_{57}Al_{25}Co_{18}$ and $Cu_{50}Zr_{50}$, while the composition of Fe–Y–B thin film was $Fe_{70}Y_8B_{22}$ by X-ray photoelectron spectrometry carried out on a Kratos Amicus spectrometer. The amorphous microstructure was confirmed by X-ray diffraction (XRD; PANalytical X'Pert PRO) and field-emission TEM (FEI, Tecnai G2 F20 S-TWIN). The thicknesses of thin films were measured by observing the fracture surface of films deposited on Si wafers in the scanning electron microscope after surface gold coating. Mechanical properties of thin films were measured by a nanoindenter (Agilent Nano Indenter G200). The nanoscratch test using a cube corner indenter tip was used to qualitatively measure the interfacial adhesion properties of the thin film/substrate interface. The detailed scratch parameters are described in Ref. [15]. The bars with the dimensions of $15 \times 2 \times 1$ mm^3 were cut from the deposited strips with a Ti substrate, and bent at room temperature using mandrels with a radius of 15 mm along the width of the bar by a home-made apparatus at a constant strain rate of $\sim 2 \times 10^{-3}\ s^{-1}$ [16]. For each thickness, nine specimens were tested, and the morphological evolution of thin films after bending was monitored by scanning electron microscopy (SEM) to see whether the transition in deformation mode occurs.

Cross-section SEM images of the films deposited on Si wafers are shown in Figure 1a. The thickness increases linearly with increasing deposition time, with deposition rates of 19.4 ± 0.3, 10.4 ± 0.5 and 23.2 ± 0.7 nm min^{-1} for $La_{57}Al_{25}Co_{18}$, $Cu_{50}Zr_{50}$ and $Fe_{70}Y_8B_{22}$, respectively. No diffraction peaks from the crystalline phases were detected for all the films with the thickness of 1000 nm, as shown in Figure 1b. A typical homogeneous maze contrast for glassy materials was found for the 50 nm thick films, as shown in the insets in Figure 1b. The selected area electron diffraction (SAED) pattern did not show any crystalline rings, confirming the monolithic amorphous state of the as-prepared films studied here.

The evolution of surface morphology at a location 0.2 mm away from the tensile edge, where the specimen experienced a total tensile strain of ~5% during bending, was monitored by SEM. Figure 2a shows SEM images of $La_{57}Al_{25}Co_{18}$ thin films with various thicknesses bent at $2 \times 10^{-3}\ s^{-1}$. Numerous cracks perpendicular to the direction of applied stress are clearly observed in the 568 nm thick La–Al–Co. For thinner film 558 nm in thickness, the number and length of cracks were significantly decreased, suggesting the reduction in shear localization tendency. Further decreasing the thickness to 548 nm, no shear bands and cracks were visible and deformation became morphologically banding-free, i.e. without observable shear offsets and cracks. Figure 2b depicts the morphology evolution of bent $Cu_{50}Zr_{50}$ thin films with various thicknesses. For a thickness larger than 165 nm, one can easily find cracks. Upon reducing the film thickness to 154 nm, all cracks disappear and non-localized deformation becomes the dominant mode. A similar situation was also observed in bent $Fe_{70}Y_8B_{22}$ film, as shown in Figure 2c. The critical thickness for non-localized deformation further decreases to 95 nm for Fe–Y–B, as compared with 154 nm for Cu–Zr and 548 nm for La–Al–Co. The value of ν is 0.335, 0.360 and 0.303 for $La_{57}Al_{25}Co_{18}$, $Cu_{50}Zr_{50}$ and $Fe_{70}Y_8B_{22}$ glassy specimens, respectively [13]. If ν were the only factor influencing the critical size for non-localized deformation, it would be quite interesting why the critical thickness for La–Al–Co, having the middle value of ν among the investigated compositions, was the largest. Actually, the critical thickness for non-localized deformation and T_g seems to be inversely proportional since $T_g = 477$ K for La–Al–Co, 733 K for Cu–Zr and 856 K for Fe–Y–B, respectively [13,17].

To answer this question, the condition required for the formation of shear band in MGs is firstly considered. By analogy with Griffith's crack-propagation criterion, a shear band can form and propagate only when the elastic energy relief associated with the propagation is larger than surface energy increase due to shear band formation. This method was used by Volkert et al. [2] and other

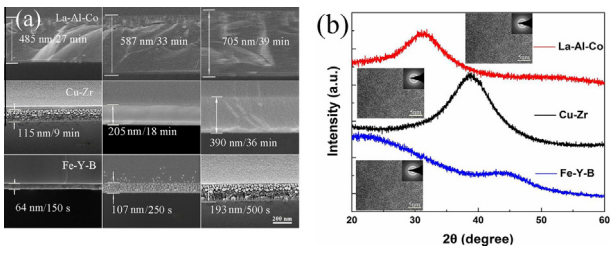

Figure 1. Cross-section SEM images (a) of as-sputtered $La_{57}Al_{25}Co_{18}$, $Cu_{50}Zr_{50}$ and $Fe_{70}Y_8B_{22}$ thin films with different thicknesses/deposition times. The scale bar is 15 μm. Typical XRD, high-resolution TEM and SAED patterns (b) of La–Al–Co, Cu–Zr and Fe–Y–B thin films.

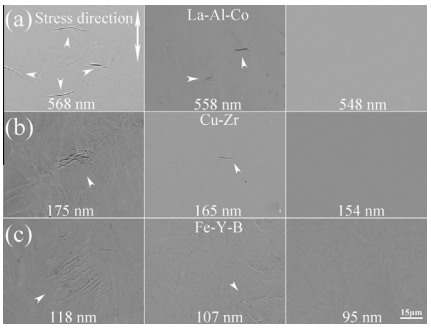

Figure 2. Surface morphology of (a) La–Al–Co, (b) Cu–Zr and (c) Fe–Y–B MG thin films with different thicknesses, deformed to a tensile strain of 5% at $2 \times 10^{-3}\ s^{-1}$ and 298 K. The cracks are indicated by white arrows.