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Cite as: Appl. Phys. Lett. **96**, 011905 (2010); https://doi.org/10.1063/1.3290246
Submitted: 21 October 2009 . Accepted: 16 December 2009 . Published Online: 06 January 2010

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Characteristic length scales governing plasticity/brittleness of bulk metallic glasses at ambient temperature

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(Received 21 October 2009; accepted 16 December 2009; published online 6 January 2010)

In this letter, we propose a unified theory for the size-dependent plasticity of bulk metallic glasses (BMGs) at room temperature. Based on the principle of energy balance and the shear-banding kinetics, two characteristic length scales are derived. One is a sample-geometry dependent variable related to the elastic energy released to drive shear-band propagation and the other is a material-dependent constant related to the internal resistance to brittle fracture. It is shown that this unified theory is effective in explaining many unusual deformation and fracture behaviors of BMGs. © *2010 American Institute of Physics*. [doi:10.1063/1.3290246]

The sample size effect on the room-temperature (RT) plasticity has recently become the research of intense interest for bulk metallic-glasses (BMGs).[1–5] The primary reason is that BMGs usually display limited ductility at RT because of catastrophic shear-band fracture and it has been a major hindrance to the structural use of BMGs. However, the experimental results reported recently, clearly demonstrate a strong size dependence of the RT ductility for BMGs. Through the reduction in sample size, many BMGs exhibit a brittle-to-ductile transition,[1–5] which implies that the deformation mechanism and fracture behavior of BMGs are fundamentally different from those of crystalline materials.

There have been numerous attempts to understand such an unusual size effect in BMGs.[4–7] For instance, it was hypothesized that there may exist a critical shear offset, which needs to be exceeded before a shear band develops into a shear crack;[5] on the other hand, assuming the cracklike behavior of a shear band, it was suggested that the size effect should originate from the compliance of a testing machine[4] or be related to the varying fracture toughness of metallic glasses.[7] As mentioned in Refs. 8 and 9 the underlying physics of a shear band differs from that of a shear crack; therefore, it is unlikely that a shear band in a BMG would follow the same physical law as a shear crack does in a brittle material. Furthermore, it is still physically unclear what determines the critical shear offset in a BMG.

Other than the aforementioned models,[4,5] it was argued that the size effect arises because of the varying free-volume contents, which result from the change in cooling rates when casting samples of different size.[6] However, recent microcompression experiments still reveal the similar size effect on the BMG micropillars cut from the same bulk material.[10] In that case, the free-volume contents are expected to be identical in the micropillars of different sizes, which implies that the free-volume effect, at least, cannot be used alone to explain the size effect in BMGs.

At RT, the ductile behavior of BMGs is characterized by serrated load-displacement curves. Recent experiments show that the load serrations are related to the intermittent propagation of individual primary shear bands,[10–12] which results from the interplay between the processes of shear-induced materials softening and subsequent structural relaxation.[10,12,13] As a shear band is nucleated, the atoms in the shear-banded region are enforced to migrate from their original positions, weakening the atomic clusters nearby and thus resulting in local materials softening;[14,15] however, the migrating atoms could also form stable clusters with their new neighbors within a short migration distance.[10,12,13] In doing so, the whole structure is relaxed to a lower energy state, which leads to the shear-band arrest and, thus, the plastic flow serrations.

As pointed out in Refs. 16 and 17 such a structural relaxation mechanism stems from the polymorphism intrinsic to a glassy solid. As such, the apparent brittle fracture is the consequence of the dynamic instability of shear banding rather than just the local materials softening induced by shear bands. Since the structural relaxation is a diffusion process, its effect relies on the waiting time between two successive atomic migration events. If the atom migration occurs very fast, the atomic-cluster reconstruction due to structural relaxation may not catch up with the atomic-cluster destruction due to shearing, thus resulting in the dynamic instability of shear banding.

Based on the above discussions, it can be envisioned that, at a given strain rate and temperature, there should exist a critical kinetic energy, above which the shear-induced disordering outweighs the shear-induced structural relaxation. This critical kinetic energy at the atomistic level is ultimately transferred into heat and corresponds to critical plastic energy dissipation over the whole plane of a shear band. In quasistatic mechanical experiments, the atom migration events are transient and related to the kinetic energy acquired from the elastic energy released upon yielding. Based on the principle of energy balance, the overall energy dissipated by such atomistic migration events is as follows:

$$\Gamma A_{SB} = \Delta U = \left(\frac{\Delta U}{U}\right) U, \tag{1}$$

where Γ = the plastic energy dissipation rate on a shear plane; A_{SB} = the area of the shear plane; ΔU = the released bulk elastic energy and U = the total elastic energy storage.

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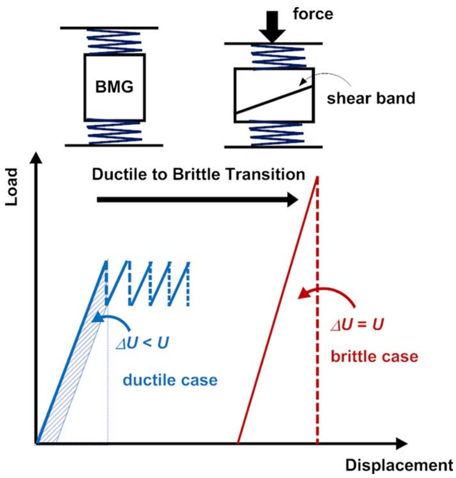

FIG. 1. (Color online) The sketch of the typical load-displacement curves for the ductile and brittle BMGs compressed by compliant fixtures.

From Fig. 1, it can be seen that the plasticity/brittleness transition in metallic glasses corresponds to $\Delta U/U=1$ and, therefore, the energy balance equation can be rewritten as $\Gamma_c A_{SB}=U$ at the transition point, where Γ_c denotes the critical plastic energy dissipation rate. *In situ*ations when $\Gamma_c A_{SB} < U$, a shear catastrophe, rather than a shear-band mediated plastic deformation process, develops upon yielding. It is worthwhile mentioning here the energy variable Γ_c, indicative of the critical energy consumption that a shear band demands to become a runaway defect during its propagation, which is physically different from the fracture toughness used as the critical energy consumption to initiate crack propagation. In other words, the brittle-to-ductile transition in metallic glasses is here treated as an issue related to the propagation rather than initiation of flow defects.

At the yielding point, $U=1/2E\varepsilon_y^2 V$, where E=the Young's modulus; ε_y=the yield strain of a BMG ($\sim 2\%$), and V=the elastically stressed volume. Note that the elastic energy release comes from two sources; one is from the stressed sample itself and the other from the elastic energy transfer from the compliant fixtures (as shown by the springs in Fig. 1). For a cylindrical compression specimen, we can derive the following:

$$V=\frac{\pi}{4}(D^2H+\alpha D^3), \tag{2}$$

where D and H denote the diameter and height of a BMG sample, respectively; and α is a dimensionless factor accounting for the effect of machine compliances.

Before proceeding, let us have a brief discussion on the dimensionless factor α. Through the dimensional analysis, it can be easily shown that α depends on the elastic and size mismatch between the sample and the fixture. For a rigid fixture, $\alpha=0$, which means that no elastic energy can be transferred onto the shear band across the sample/fixture contract from the surroundings. For an infinitely large fixture relative to the testing sample size, $\alpha \propto E/E_{\mathrm{f}}$, where E_{f} denotes the elastic modulus of the fixture. In the ideal case, the Young's modulus of the fixture is set much higher than that of a sample and, therefore, the machine compliance effect can be ruled out. However, in real experiments, the magnitude of α is finite and on the order of unity.

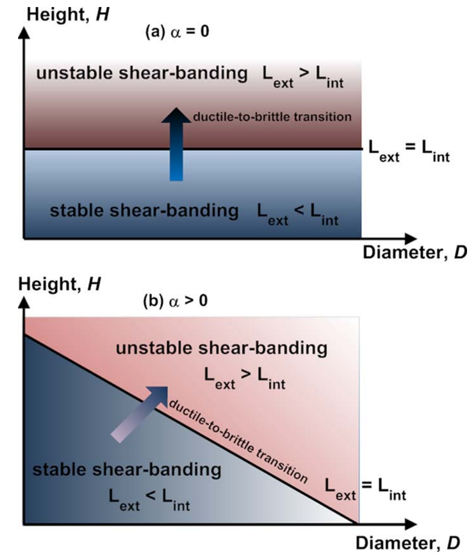

FIG. 2. (Color online) The sketches of the deformation mechanism map for quasistatic mechanical experiments at (a) $\alpha=0$ and (b) $\alpha>0$.

Based on the above discussions, the criterion of brittle fracture in BMGs can be cast as follows:

$$U \geq \Gamma_c A_{SB}. \tag{3}$$

Combining Eqs. (2) and (3), we arrive at a simple geometrical relation for the ductile-to-brittle transition

$$H+\alpha D \geq \frac{2\Gamma_c}{E\varepsilon_Y^2 \sin\theta}, \tag{4}$$

where θ denotes the angle of a shear band to the loading axis. The left-hand side of Eq. (4) thus defines an extrinsic length scale, $L_{\mathrm{ext}}=H+\alpha D$, which corresponds to the elastic energy release; whereas the right-hand side of Eq. (4) defines an intrinsic length scale, $L_{\mathrm{int}}=2\Gamma_c/\mathrm{E}\varepsilon_y^2 \sin\theta$ which can be viewed as the internal resistance of a BMG to brittle fracture and is independent of sample size. The materials will exhibit the brittle fracture if $L_{\mathrm{ext}} \geq L_{\mathrm{int}}$.

Based on Eq. (4), the deformation mechanism map for BMGs in quasistatic mechanical experiments can be constructed. For the ideal case of a rigid fixture ($\alpha=0$), the ductile-to-brittle transition, which is related to the occurrence of the unstable shear-band propagation with the minimum elastic energy input, is solely dependent on the sample height [Fig. 2(a)]; for cases of $\alpha>0$, the ductile-to-brittle transition depends on both the height and diameter of a BMG sample [Fig. 2(b)]. The general trend is that a BMG tends to display plasticity with both dimensions being shortened, which agrees with all the experimental data reported so far.[2,4–6,18] In contrast, the shear-band stability index, which was derived by assuming the cracklike behavior of a shear band, predicts a somewhat reverse trend, i.e., shortening the sample height tends to reduce the overall plasticity.[4] The difference between the theory and experiments implies that simply treating the shear band as a crack may not be appropriate during plastic deformation in BMGs.

On the other hand, the extrinsic length scale, L_{ext}, can be viewed as another shear-band stability index. As L_{ext} is kept at a level less than L_{int}, the stability of shear bands is attained, giving rise to an overall plastic deformation behavior. As shown in Fig. 3, the ductility measured for the Zr-based

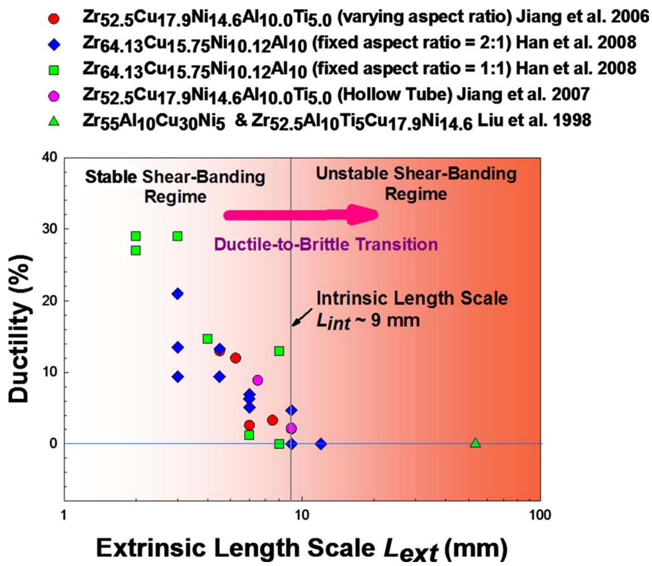

FIG. 3. (Color online) The correlation between the measured ductility of the Zr-based BMGs in the compression experiments at RT and the estimated extrinsic length scales.

BMGs using different sample geometries[4,18–20] shows a very good correlation with the extrinsic length scale ($\approx H+D$), which is estimated simply by assuming $\alpha=1$. Due to the lack of detailed information for α, the experimental results show considerable scattering at the ductile-to-brittle transition. Taking $L_{\mathrm{int}}\approx 9$ mm, $\varepsilon_y=0.02$, $E=100$ GPa, and $\theta\approx 42°$ for the Zr-based BMG, the estimated Γ_c is about 1.2×10^5 J/m^2. Such a high plastic energy dissipation rate is sufficient to cause materials melting during the fracture event, as manifested by the vein patterns observed on the sample fracture surface.[19] In contrast, the stability of shear-band propagation manifests itself as the shear striations on the shear plane.[2,10–12]

Before conclusion, it is worthwhile discussing the factors which may change the magnitude of the intrinsic length scale of a BMG. According to Johnson *et al.*,[21] the nucleation of a shear band is essentially a strain controlled process and the yield strain of about ~2% can be regarded as universal for all BMGs. In that case, the ratio of Γ_c/E plays the vital role. As discussed above, shear banding in BMGs entails the breakage and formation of atomic bonds. If the atomic structure of a BMG possesses a high density of loosely-bonded regions, i.e., the shear transformation zones (STZs), the whole system becomes more "flexible" in accommodating the shear-induced structural alteration by means of structural relaxation. Meanwhile, its elastic modulus, as closely related to the atomic bonding strength and STZ configurations,[22] decreases, both of which contribute to a higher Γ_c/E ratio and, thus, a greater ductility in BMGs than otherwise. Conversely, a higher Young's modulus signifies a more rigid configuration of atoms and less flexibility in structural relaxation, which leads to the seeming brittleness in the BMGs with a high modulus, such as the Fe-based BMGs.[23–25]

In summary, we provide a unified theory in this letter to understand the unusual behavior of ductile-to-brittle transition in BMGs from the perspectives of energy balance and shear-banding kinetics. The theoretical prediction is in good agreement with the available experimental data and a shear-band stability index, i.e., the extrinsic length scale, is herein provided. By linking sample geometries with this extrinsic length scale, our theory shows the potential of rationalizing various geometry-induced ductilization phenomena in BMGs.

Y.Y. and C.T.L. acknowledge the financial support for this work provided by the Hong Kong Polytechnic University for newly recruited academic staff. J.L. acknowledges the financial support from GRF with the Grant No. of 7/CRF/08.

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