Rate-dependent inhomogeneous-to-homogeneous transition of plastic flows during nanoindentation of bulk metallic glasses: Fact or artifact?

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There has been considerable controversy over the "apparent" rate-dependent transition from inhomogeneous-to-homogeneous flow during nanoindentation of bulk metallic glasses (BMGs) at room temperature: whether it arises from the existence of homogeneous-flow regime in BMG deformation map or is an artifact due to the instrumental blurring at high rates. To provide a clue to address this dispute, the authors performed nanoindentation experiments on a Zr-based BMG with two geometrically self-similar indenters. The results are discussed in terms of the discrete plasticity ratio, which is a useful parameter in analyzing the contribution of inhomogeneous plasticity to the total plastic deformation. © *2007 American Institute of Physics*. [DOI: 10.1063/1.2742286]

Over the past decades, unique plastic deformation behavior of bulk metallic glasses (BMGs) at room temperature (plastic strain is highly localized into very narrow zones, so-called shear bands) has attracted much scientific interest and thus has been studied both theoretically and experimentally. Recent research in the field has accelerated with advances in nanoindentation techniques that make it possible to investigate the mechanical response during the entire loading sequence.[1,2] In the early 2000s, Wang *et al.*[3] and Wright *et al.*[4] reported a few small and discrete steps (so-called pop-ins) in the loading part of load-displacement (*P*-*h*) curves recorded during nanoindentation of Zr-based BMGs. While a single or only a few pop-ins were thought to indicate the transition from elastic to plastic deformation (i.e., yielding) as in crystalline materials,[5] another interesting feature of inhomogeneous deformation during BMG nanoindentation was reported by Golovin *et al.*[6] and Greer *et al.*:[7] continuous serrations (serial pop-ins) in *P*-*h* curve. Similar discrete flow during nanoindentation has now been observed in various BMGs including Pd-, La-, Zr-, Mg-, Cu-, Ce-, and Au-based ones,[8–16] and now it is well accepted that the serrations are associated with shear band nucleation and/or propagation. Schuh *et al.* demonstrated extensively[2,8–10] that the discrete flow was strongly dependent not only on the chemical composition of the BMGs but also on the indentation loading rate; that is, the serrations in the *P*-*h* curve are more pronounced at lower loading rates and gradually disappear with increasing rate. Then, they showed an "apparent" rate-dependent inhomogeneous-to-homogeneous transition flow at room temperature.

There have been two major hypotheses on the reasons for this transition. First, Schuh and Nieh[8] proposed that the absence of pop-in events in fast indentation with a Berkovich indenter was caused by kinetic limitations on the nucleation of shear bands: in the low-rate regime, a single shear band operates in isolation, while in the high-rate regime multiple shear bands tend to operate together simultaneously.[8] Later, Schuh *et al.* additionally proposed the existence of a homogeneous-flow region at room temperature in the deformation map for BMGs:[9,10] at sufficiently high deformation rate, flow homogenization can occur not only in time but also in space. Second, Greer *et al.*[11] and Jiang and Atzmon[12] suggested that the apparent absence of serrations at high indentation rate is just an artifact due to the lack of instrumental resolution (instrumental blurring) including the limited data-acquisition rate as well as the limited response time of the indentation equipment. Thus the reason for the rate-dependent transition for homogenization of flow is still controversial: in short, the transition is fact or artifact? The purpose of this letter is to report our recent experimental results, which might yield an important clue in this dispute.

Nanoindentation experiments were made on $Zr_{60}Cu_{30}Al_{10}$ BMG (prepared by arc melting followed by drop casting) using a nanoindenter-XP (MTS Corp., Oak Ridge, TN). In addition to Berkovich indenter (with centerline-to-face angle of 65.3°) which has commonly been used in this research area, a cube-corner indenter with the angle of 35.3° was employed since a cube-corner indenter is expected to produce much higher stresses[17,18] and thus making the observation of the serrations more clear. The loading rates (dP/dt) were varied, while thermal drift was maintained below 0.05 nm/s. More than five indentation tests under each testing condition were carried out at a fixed peak load of 100 mN on the sample, which was mechanically polished to a mirror finish.

Figure 1(a) overlaps typical load-displacement (*P*-*h*) curves recorded during nanoindentation with a Berkovich indenter and a cube-corner indenter at various loading rates. In the Zr–Cu–Al BMG examined here, the total plastic deformation does not change with varying indentation rate. The sharper cube-corner indenter produces a larger peak-load displacement and a greater proportion of permanent plastic deformation after unloading than the Berkovich indenter. Figure 1(b) exhibits the *P*-*h* curves at relatively high and low rates (5 and 0.05 mN/s) with the two indenters (the *P*-*h* curve for 0.05 mN/s has been shifted for clarity in presentation). The figure suggests three tendencies in the change in

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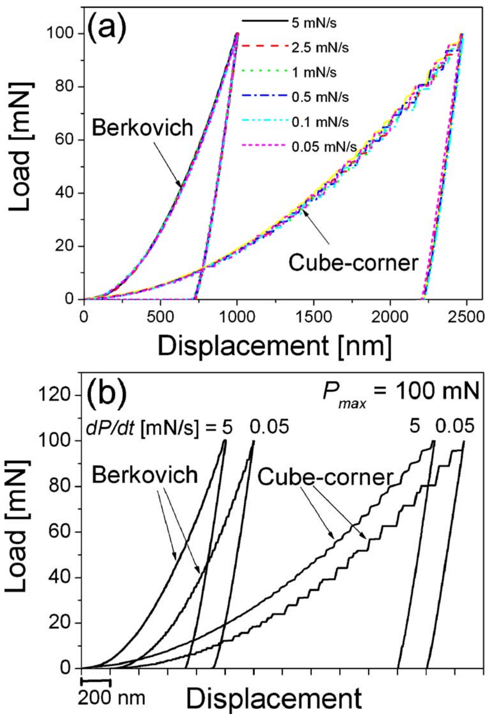

FIG. 1. (Color online) Typical load-displacement curves obtained from nanoindentation with Berkovich and cube-corner indenters at various loading rates, dP/dt: (a) superposition of all curves; (b) the curves for $dP/dt=5$ and 0.05 mN/s, separated for clarity.

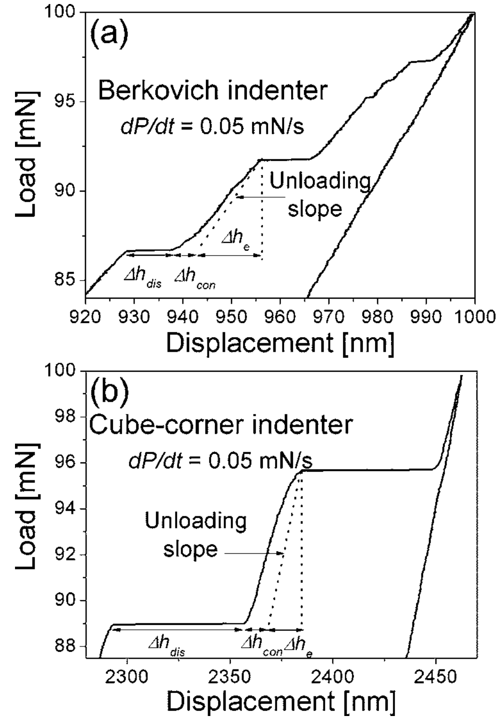

FIG. 2. How to measure a discrete portion of plastic deformation (Δh_{dis}), a continuous portion of plastic deformation (Δh_{con}), fan elastic portion of deformation (Δh_e): (a) Berkovich indentation and (b) cube-corner indentation. The curves are enlarged from Fig. 1(b).

inhomogeneity with indentation testing parameters. First, indeed, the serrated flows are strongly dependent on the indentation rate. As the loading rate increases, the discrete events gradually disappear in the low-load regime or weaken in the high-load regime. Second, the displacement discontinuities increase in size with increasing load or displacement. This might be caused by either (1) the nature of the geometrically self-similar sharp indenter (i.e., the amount of strain accommodation by each pop-in is fundamentally independent of load level) or (2) high indentation strain rate at early stages of the indentation under constant loading rate.[19] We will return to this issue later. Third, the sharper cube-corner indenter produced more pronounced discrete horizontal steps than the Berkovich indenter, because sharper indenters induce greater stresses and strains in the BMG due to the larger volume of material displaced.[17,18] Collectively, enhanced resolvability of the serrations can be achieved during indentations made at lower rate and higher load with a sharper indenter. Note that a clear observation of the serrations is essential in examining the inhomogeneous-to-homogeneous transition phenomena.

Estimating the contribution of the serrations (pop-ins) to the total plastic deformation in the BMG is very helpful in analyzing the rate-dependent transition phenomena, as pointed out in previous research.[2,8–11,13] How we estimate the contribution here is illustrated schematically in Fig. 2. The total displacement (Δh_{tot}) recorded during nanoindentation is roughly of three sorts: a discrete portion of plastic deformation (Δh_{dis}), a continuous portion of plastic deformation (Δh_{con}), and an elastic portion of deformation (Δh_e) which is expected to be recovered during unloading. Thus, the contribution of discrete plasticity to the total plasticity (here called "discrete plasticity ratio") is simply given as $\Delta h_{\mathrm{dis}}/(\Delta h_{\mathrm{con}}+\Delta h_{\mathrm{dis}})$.[13] The advantage of this calculation over that described in Ref. 8 is that one can determine the discrete plasticity contribution for each pop-in and omit regions, where the pop-ins disappear or cannot be resolved reliably.[13]

As elaborated above, the extent of the displacement burst due to a pop-in event increases with indentation load. Figure 3(a) shows the change in the discrete plasticity ratio at the lowest rate applied here (i.e., 0.05 mN/s at which the

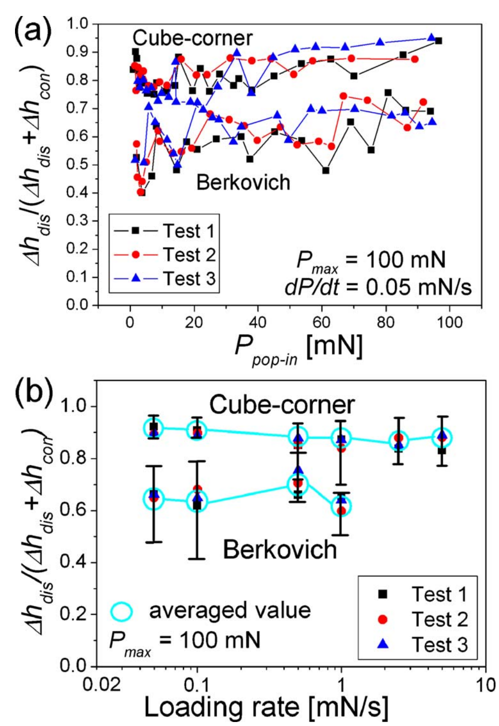

FIG. 3. (Color online) Variation in the discrete plasticity ratio with (a) the pop-in load and (b) the loading rate.

most pronounced serrations were observed) as a function of pop-in load. At this rate, the discrete plasticity ratio is about 0.75–0.95 for cube-corner indentation and around 0.4–0.7 for Berkovich indentation. The higher ratio in cube-corner indentation than in Berkovich indentation may be attributed to the much higher stress level and larger plastic zone size in the former case than in the latter.[17,18] Interestingly, the discrete plasticity ratio shows no clear tendency to change with the pop-in load level for both indenters, though the step size increases with indentation load [see Fig. 1(b)].

Since the discrete plasticity ratio does not change significantly, one can choose a pop-in at any load level in evaluating the rate dependency on the ratio. We measured the discrete plasticity ratio using the relatively large pop-ins (observed over a pop-in load of 80 mN) from at least three indentation tests made under the same conditions. Figure 3(b) shows the variation in the discrete plasticity ratio as a function of loading rate. The ratios corresponding to the Berkovich indentations at 2.5 and 5 mN/s were not reliably measured and are thus omitted in Fig. 3(b). Surprisingly, for both indenters, the ratios were independent of the rate and remained almost constant. As the total plasticity is independent of indentation loading rate (see Fig. 1), the rate-independent discrete plasticity ratio and the rate-dependent intensity of the inhomogeneities (i.e., more pronounced serrations during slower indentation) suggest together that with decrease in loading rate, the number of pop-ins decreases and the pop-in size increases.

Because the discrete deformation is attributed to shear band formation and/or propagation (as generally accepted), the increase in pop-in number and decrease in pop-in size with rate partly supports the suggestion of Schuh *et al.*[2,8–10] that a fundamental change in plastic flow characteristics occurs with indentation rate. It is also consistent with the AFM work of Jiang and Atzmon,[12] who reported higher shear band density near hardness impression after faster indentation. Nevertheless, the results in the present work do not completely support the argument of Schuh *et al.*[2,10] that at higher rates homogenized flow can occur both in time and space simply because flow is still discrete in a very fast cube-corner indentation (in which resolvability of the serrations was strongly enhanced compared with Berkovich indentation). Although we agree that the nature of shear banding (especially its scale) can change with increasing rate, we could find no solid evidence for a "transition" from inhomogeneous flow to homogeneous flow. It is thus concluded that "instrumental blurring" contributes significantly to the homogenized *P-h* curve obtained during fast indentation of BMGs.

Another homogenized-appearing curve can be found in the early part of the *P-h* curve, especially from fast indentation [e.g., the low-load regime of *P-h* curve from Berkovich indentation at 5 mN/s in Fig. 1(b)]. Since the indentation strain rate, $d\varepsilon_i/dt=h^{-1}(dh/dt)$, is diminishing during indentation testing under constant loading rate (dP/dt),[19] one might argue that the homogenization in the early part is due to a very high indentation strain rate. Very recently, Yang and Nieh[15] showed that the serrations tend to decrease linearly with the logarithmic increase in the strain rate. However, according to their calculation based on a free volume model, the critical indentation strain rate for the transition of inhomogeneous-to-homogeneous flow is extremely high ($1700\ s^{-1}$ for Au-based BMG they examined). If a steady-state value of hardness is reached (i.e., $dH/dt=0$), the indentation loading rate can be converted to indentation strain rate as $dP/dt=2P(d\varepsilon_i/dt)$.[19] Note that this is only a rough conversion equation for BMG, since there are some hardness fluctuations and dH/dt is not zero.[15] According to the above conversion equation, in reality, 1700 (or a number of similar order) s^{-1} cannot be experimentally achieved using currently existing commercial indentation equipment. Schuh and Nieh demonstrated the homogenized *P-h* curve of a Pd-based BMG that was obtained from amazingly fast indentation (330 mN/s).[8,9] However, even in this case, such a high strain rate (in order of $1000\ s^{-1}$) can be achieved only at a very low-load regime (below 165 μN.) Thus, some instrumental blurrings should contribute to their *P-h* curve homogenized up to 10 mN. Consistently, in Fig. 3(a) of the present work, the discrete plasticity ratio did not decrease significantly with decreasing indentation load. This implies that the increase in the serrations with indentation load occurs simply because the amount of strain accommodation by each pop-in is fundamentally independent of load level for a geometrically self-similar indenter.

Collectively, we reach at the conclusion that even at extremely high indentation rates, the plastic deformation of BMGs during room-temperature indentation is still inhomogeneous, which is consistent with the recent "compression" test result of Jiang *et al.*[20] The homogenized-looking indentation curve (obtained with fast indentation or low-load indentation) might arise mainly from very fine scale of the serrations, which cannot be resolved by common indentation equipments.

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