Research Article

Scratch-induced work hardening of an atomically flat bulk metallic glass by stress-driven structural ordering

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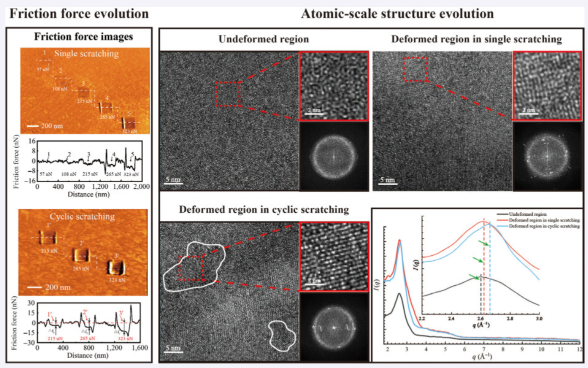
ABSTRACT: Although enhanced structural relaxation is generally believed to be an important contributor to the work hardening of metallic glasses subjected to a triaxial stress state, direct observation of the relaxation process in response to work hardening has not been achieved in metallic glasses. Here, we show that nanoscratching an atomically flat bulk metallic glass surface, a small atomic force microscopy tip with a radius of ~10 nm, results in a high hydrostatic stress within the stressed volume. This enables densifying plastic flow via enhanced structural relaxation and leads to work hardening behavior, as evidenced by an obvious decrease in friction force signals within scratched regions. Further examination of the atomic structure beneath the scratched surface via high-resolution transmission electron microscopy reveals a relaxed structural configuration, which is indicated by dispersed clusters of medium-range order scale in the case of line scratching and nucleated nanocrystals in the case of cyclic scratching. This study provides compelling evidence for stress-driven structural relaxation, greatly deepening the understanding of the work-hardening mechanism in metallic glasses.



KEYWORDS: metallic glasses; structural relaxation; scratching; work hardening

1 Introduction

Unlike crystalline metals, where work hardening typically prevails in deformation by dislocation interactions [1], metallic glasses (MGs) demonstrate deformation physics of strain softening by shear localization [2, 3]. Under applied stress, the disordered structure of MGs accommodates strain through local rearrangement of atoms, leading to the formation of shear bands (SBs). As the SBs operate, the deformation proceeds locally at ever-higher rates, resulting in runaway failure [4]. In fact, the shear process involves a dynamic competition between the rate at which the free volume is generated through shear dilation and the rate at which the free volume is annihilated through structural relaxation [5, 6]. Because diffusional relaxation is thermally activated, at low temperatures, the rate of free volume accumulation easily exceeds that of annihilation, causing net dilatation of the free volume and thus strain softening [7]. If it were possible to enhance the structural relaxation during deformation at room temperature, strain hardening could be achieved, which is highly desirable for the applications of MGs.

It has been demonstrated that MGs are capable of work hardening under conditions where the stress is constrained with high triaxiality [8, 9] or where the size of the tested samples is limited at the nanoscale [10, 11]. The hardening mechanism is primarily based on stress-driven structural relaxation and is regarded as genuine structural hardening, as it suppresses shear localization and enables deformation to continue at high strains and stresses [12, 13]. The high hydrostatic stresses induced by constrained stress states or small deformation scales can significantly increase the atomic mobility and promote the structural reordering by enhancing free volume annihilation, allowing deformation to operate in a densifying flow.

Under conditions where the stress is constrained by high triaxiality and the deformation scale is located at the nanoscale, the nanoscratching is expected to readily render the hardening of MGs, similar to dislocation-mediated frictional hardening, as is known in conventional crystalline metals. However, hardening is rarely observed in most nanoscratch cases [14–17]. Shear-induced softening is still prevalent since profuse shear bands are clearly observed along the scratched grooves [15]. This is because the

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stressed volume induced by random surface asperities interacting with the slider is too large to suppress shear banding events. Therefore, preparing a very flat MG surface is important for reducing the stressed volume during deformation and potentially achieving hardening via nanoscratching. We have attained atomically flat bulk metallic glass surfaces via thermoplastic imprinting, which feature a roughness of ~0.05 nm within individual terraces [18, 19]. With such surfaces, it becomes feasible to deform bulk metallic glasses in a well-defined manner, at the nanoscale, and even at the atomic scale, as a function of the slider apex size.

On these attained atomically flat surfaces, we performed nanoscratching tests by atomic force microscopy (AFM) under a series of normal loads, with substantial plastic deformation. The scratch-induced deformation is homogeneous, without any shear banding, and hardening is captured by friction force changes within scratched regions. High-resolution transmission electron microscopy (HRTEM) was employed to examine the structural evolution beneath the scratched surface, offering definite evidence of stress-driven structural relaxation for hardening behavior.

2 Materials and methods

The composition of the Pt-based bulk metallic glass (Pt-BMG) used in this research was $Pt_{57.5}Cu_{14.7}Ni_{5.3}P_{22.5}$, which was selected because of its high glass-forming ability and outstanding oxidation resistance. A cylindrical ingot of Pt-BMG was produced by casting and then thermoplastically formed onto a $SrTiO_3$ substrate that displayed atomically flat surface features. Afterwards, the molded Pt-BMG surface replicated the “atomic flatness” of the $SrTiO_3$. For more details on the thermoplastic imprinting process, see Ref. [18]. The structure of the imprinted sample was confirmed to be fully amorphous using an X-ray diffractometer (XRD; Empyrean Malvern Panalytical, The Netherlands). The broad hump peak in the XRD patterns verified the amorphous nature of the imprinted Pt-BMG (Fig. S1 in the Electronic Supplementary Material (ESM)). The nanoscratching experiments were carried out by an atomic force microscope (Seiko SPI3800N, Japan) with a diamond-coated silicon cantilever with a tip radius of ~10 nm (Adama AD-40-SS, USA). The nanoscratching tests were conducted under line-scanning mode to identify the deformation characteristics in terms of groove features and under raster-scanning mode over a selected area to confirm the hardening behavior in terms of friction force signals. During the scratching process, the topography and lateral force images were recorded simultaneously. The friction force image was calculated using $F_f = (F_{l,fwd} - F_{l,bwd})/2$ [20], where $F_{l,fwd}$ and $F_{l,bwd}$ are the average lateral force images recorded in the forward and backward directions, respectively. Both the characterization of the atomically flat surface of the Pt-BMGs by thermoplastic imprinting and the nanoscale frictional experiments were performed under ambient conditions (T = 23 °C, relative humility (RH) = 35%). The sliding velocity was 400 nm/s, and the applied loads ranged from elastic to plastic. The cross-sectional profiles of the scratched regions were prepared via a focused ion beam (FIB; Carl Zeiss Crossbeam 540, Germany) and further examined via a high-resolution transmission electron microscope (FEI Tecnai G2 F20, USA) to observe the changes in the atomic structure upon nanoscratching.

3 Results

To examine the replication quality of thermoplastic imprinting, a quantitative analysis of the surface roughness of the imprinted Pt-BMG surface was carried out by tapping-mode AFM. The corresponding results, which are shown in Fig. 1, reveal that the topographical morphology of the imprinted surface exhibits a series of terraces with widths of 500±100 nm separated by steps of 0.39±0.02 nm in height, which matches well with the height of a single unit cell of the $SrTiO_3$ (STO) template. The conformity of the imprinted Pt-BMG surface characteristics with those of the STO crystals demonstrated perfect reproduction of the atomically flat surface from the STO template in thermoplastic imprinting. Within individual terraces, the root-mean-square (RMS) roughness was quantified as 0.10 nm or less; for example, a 250 nm × 250 nm area within the black box had an RMS roughness of 0.057 nm, which is identical to typical RMS values measured on freshly prepared STO molds with the same-sized area. The availability of such a virtually “atomically flat” sample surface now paves the way for high-resolution investigations of the nanoscale deformation behavior of Pt glass via scratching.

Figure 2 shows the topographical and corresponding friction force images after scratching the Pt-BMG surface under loads ranging from 48 nN (row 1) to 274 nN (row 8). In the case of low load range (48–80 nN), there are no signs of wear observed in the topographical and friction force images, indicating a purely elastic deformation after scratching. When the load reached 145 and then 177 nN (row 4), no visible change was discerned in the topographical image (Fig. 2(a)), akin to the case of other lower loads. In contrast, an obvious decrease in the friction force was detected, as indicated by a visibly darker scratched trace (Fig. 2(d)). To corroborate the reproducibility of these observations, we carried out a total of six independent tests. Two cases with relatively poor quality are shown in Fig. S2 in the ESM. These tests confirmed that there was always a regime where lower friction was

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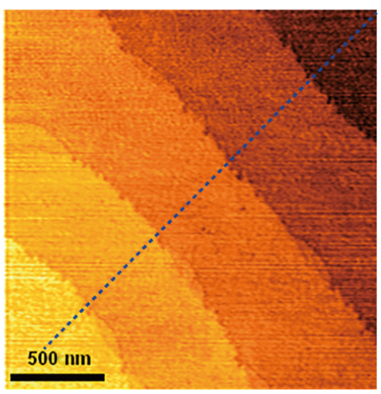


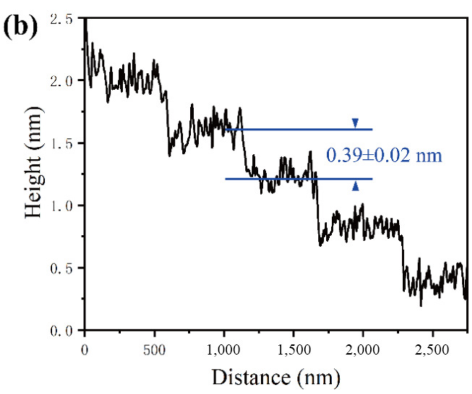
(b)


Fig. 1 (a) Atomic force microscopy characterization of an imprinted Pt-BMG surface with a size of 2 μm × 2 μm. (b) A height profile along dashed blue line indicated in (a).

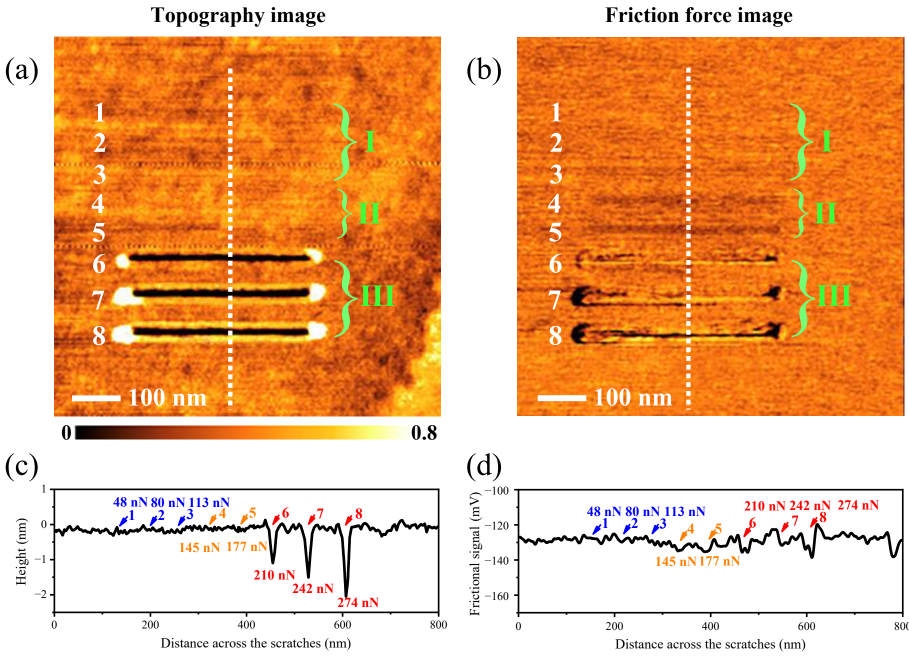


Fig. 2 (a) Topography image of Pt-BMG surface area subjected to AFM scratching tests. Color transitions from dark to bright while covering 0.8-nm-height difference. (b) Map of frictional forces that have been recorded simultaneously with normal forces of topographical image (a). (c) Height profile extracted along dashed white line highlighted in panel (a). (d) Frictional forces along dashed white line in panel (b), revealing effects of increasing normal loads on friction force.

found along the scratch trace without causing visible topographical changes; the force regime during which this was the case fluctuated from 106 to 131 nN. We speculate that the surface of the metallic glasses has been locally hardened as a result of nanoscratching. Akin to the case of this work, the apparent friction force decreased, but no visible wear loss was observed in the nanotribological tests of the graphene-coated Cu substrates [21], which supports the viewpoint that the decrease in friction force observed in Fig. 2 is a result of surface hardening. For scratching carried out under an applied normal load of 210 nN or greater (rows 6–8), wear begins to occur, as indicated by the evident material pile-ups along the scratched grooves. The uniform material pile-ups suggest that the plastic flow upon nanoscratching must have been homogeneous in nature. No signs of shear bands were observed on the scratched surface, suggesting that the formation of shear bands may be entirely suppressed during scratch-induced deformation. Note that if shear bands have formed on the atomically flat surface, their offsets can be detected owing to the high spatial resolution of AFM.

Attempts to validate the hardening behavior in line scanning mode in terms of friction force changes are hampered by the narrow width of grooves and evident pile-ups, which significantly affect the signal acquisition of friction force [20]. Therefore, we perform a series of scratching tests under raster-scanning mode over an area of 200 nm × 200 nm within individual atomically flat terraces. As shown in Fig. 3, the wear depth increased with increasing load from 57 to 323 nN, which is consistent with the results shown in Fig. 2. More importantly, compared with that in the unscratched area, the friction force sign within the scratched area was distinctly lower, especially in the scratches performed under high loads, as indicated by darker colors in the scratched regions and an evident decrease in the friction force profile. It has been demonstrated that the work hardening induced by scratched deformation is manifested by a decrease in the friction force sign obtained via the AFM tapping process. The decrease in the friction force sign became more pronounced after cyclic scratching, suggesting a greater degree of work hardening. Taking the load of 323 nN as an example, the friction force decrease is measured to be ~3 nN for line scratching and ~16 nN for cyclic scratching. It was shown that cyclic scratching or a high applied load favors hardening. However, higher loads unavoidably lead to pronounced pile-ups, which would result in unsatisfactory image quality of friction force signals. Consequently, we perform scratching tests under a higher load of 754 nN over a larger area of 1,000 nm × 1,000 nm to induce possible greater work hardening. As predicted, a greater friction force decrease is observed (Fig. S3 in the ESM) than that in Fig. 3.

Figures 4(a)–4(c) show the variation in the friction force versus the number of displacement loops as a function of the number of scratch cycles under a series of normal loads. The friction force is calculated from friction force microscopy (FFM) signals [22]. The frictional coefficient is then calculated from the friction force loops and plotted against the number of cycles, as shown in Fig. 4(d). For a fixed normal load, with increasing number of cycles, the friction force loops decreased, suggesting the occurrence of work hardening during nanoscratching. This is because upon successive scratching cycles, the worn surface repeatedly hardens, reducing the contact area between the counterbodies and resulting in a decrease in the friction force, similar to dislocation-mediated work hardening in crystalline alloys. Another interesting phenomenon is that the reduction trend appears to be more pronounced at early cycles. Taking 215 nN as an example, when the number of cycles increased from 1 to 2, the friction force reduction was approximately 14 nN, which was much greater than that of 7.5 nN when the number of cycles was further increased from 2 to 5. For a fixed number of cycles, a similar trend was observed for the variation in the frictional coefficient versus the normal load, suggesting a strong dependence of work hardening on both the number of scratch cycles and the normal load.

As suggested in connection with free volume evolution theory [23], we can speculate that the observed work hardening is a result of enhanced relaxation of the glass structure. If so, the subsurface atomic structure of the scratched regions should reflect a relaxed structure. To verify this, HRTEM was used to reveal how the

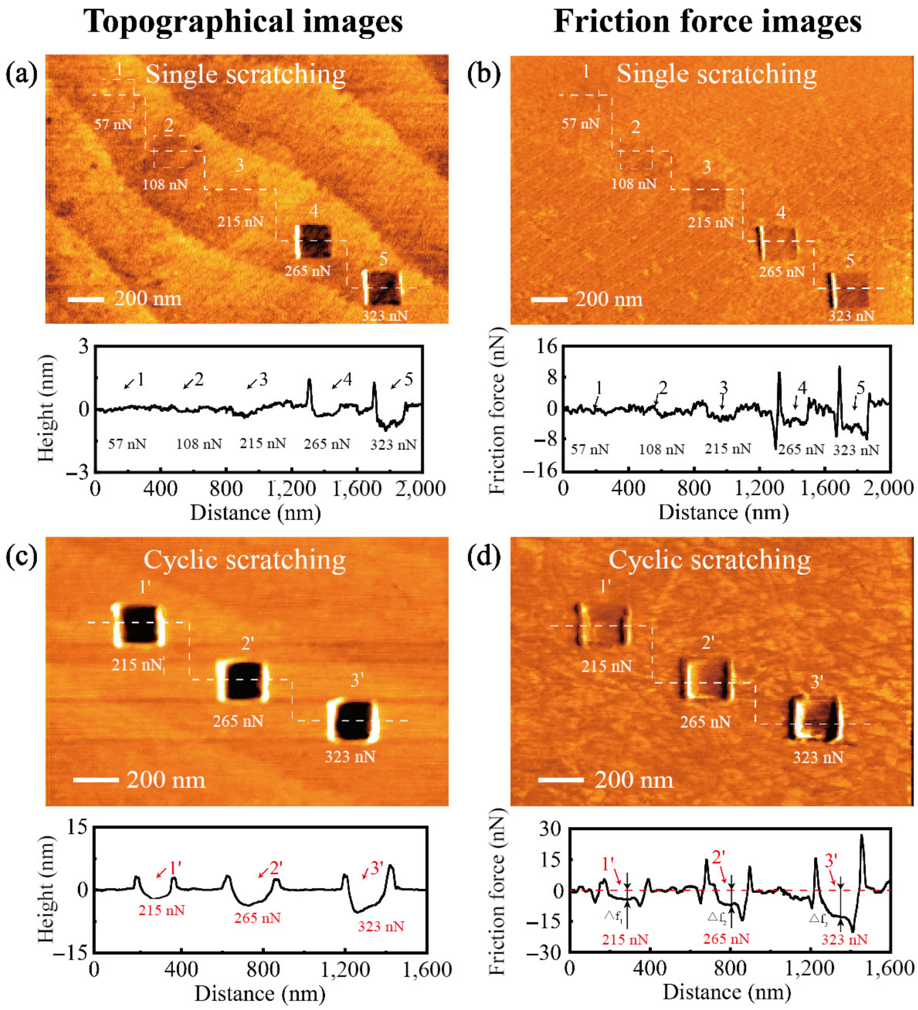


Fig. 3 (a, c) Topographical images and height profiles taken along dashed lines labeled 1–5 in (a) and 1'–3' in (c) within atomically flat terraces on Pt-BMG surface subjected to (a) line scratching and (c) cyclic scratching for 5 times. (b, d) Friction force images recorded simultaneously with normal forces of topographical images (a, c) and profiles of friction force along dashed lines indicated by 1–5 in (b) and 1'–3' (d).

structure evolves by sectioning the scratched regions via FIB, as shown in Fig. 5. The scratches labeled I and II corresponded to line scratches under a load of 513 nN and cyclic scratches under the same load, respectively. With cyclic scratching, more worn debris formed and accumulated outside the scratched region (scratch II, in Fig. 5(a)). Detailed observations revealed that the atomic structure of the debris was partially crystallized, possibly because of the high temperature generated by extensive plastic deformation during the debris formation process. The high temperature facilitates sufficient relaxation of the glass structure and leads to the crystallization and oxidation of the debris, as confirmed by the energy dispersive X-ray spectroscopy (EDS) mapping in Fig. 5(c). Upon scratching, the initially fully amorphous structure transformed into a relaxed (ordered) structure configuration, where dispersed ordered clusters of ~2 nm were extensively distributed in the amorphous matrix, as verified by the bright diffraction spots in the selected area electron diffraction (SAED) patterns (Fig. 5(d)). Upon cyclic scratching, a superlattice-like layer formed along the scratching direction, suggesting a greater degree of structural relaxation (Fig. 5(e)). The superlattice layer began at a distance of ~7 nm below the surface. The locally magnified images revealed a crystallized layer with a lattice spacing of ~0.15 nm.

It has been reported that MGs exhibit fast surface dynamics [24, 25]. The atoms near the surface exhibit faster atomic flow than those in the bulk and possibly self-assemble into a more relaxed structure when subjected to annealing [26]. To rule out the possibility that the structural ordering that occurred in scratch-induced deformation was a result of surface dynamics rather than stress-induced relaxation, we further examined the structure in the interior region, which was far from the surface but still within the stressed volume. Figures 6(a)–6(c) show HRTEM images of the undeformed and deformed regions of scratches I and II in the interior. In the undeformed region, a homogeneous maze-like pattern was observed, demonstrating the amorphous nature of the structure, as confirmed by the fast Fourier transform (FFT) patterns, which featured a broad diffraction halo with no point maxima. Upon scratching, extensive ordered regions were clearly observed. Local magnified images further revealed local ordering characteristics similar to those of medium-range order (MRO) in annealed Pd-based MGs [27]. Note that the ordering degree has not yet reached the crystal-like periodicity. This implies that the energy induced from scratch-induced deformation is not enough to induce full crystallization but is adequate to favor local atomic ordering in sheared regions on a medium-range scale, as evidenced by the emergence of bright spots on the broad halo in the FFT pattern. In the interior region of scratch II, 5–15 nm nanocrystals nucleated dispersedly in the amorphous matrix, suggesting a greater degree of structural relaxation. It has been demonstrated that structural ordering is a result of free volume

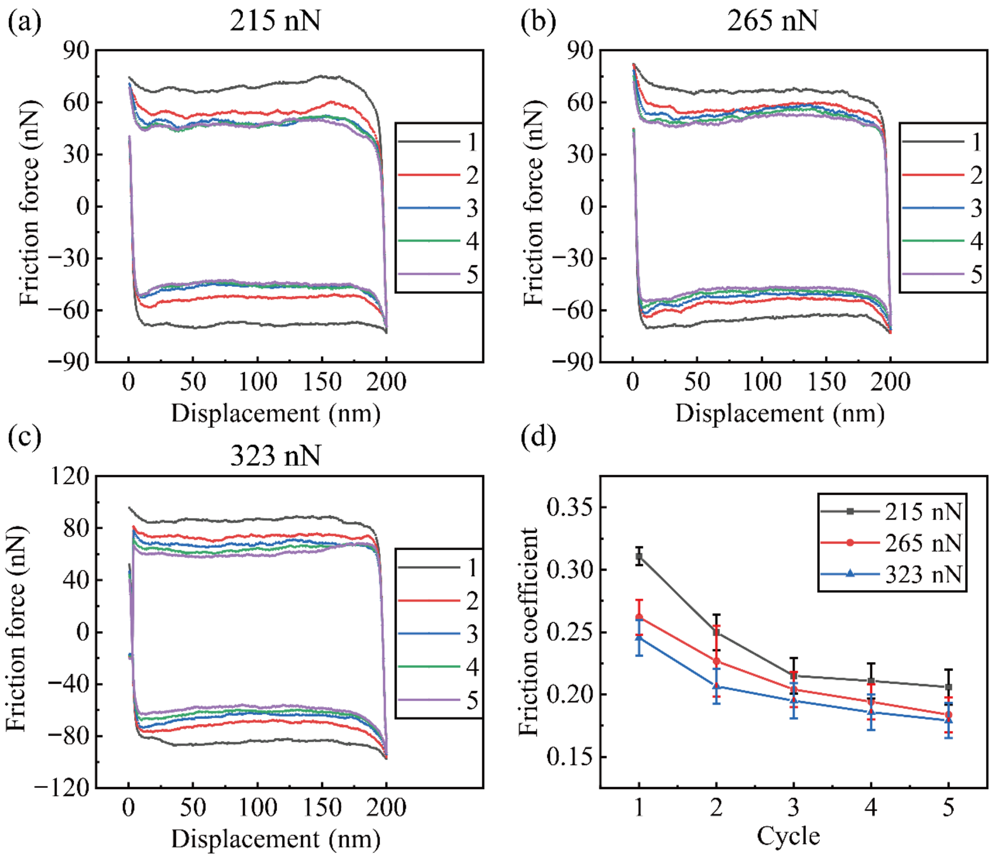


Fig. 4 Friction force versus displacement loops as a function of number of scratch cycles carried out at normal loads of (a) 215 nN, (b) 265 nN, and (c) 323 nN. (d) Friction coefficient calculated from (a–c) is plotted against number of scratch cycles to quantitatively show a decreasing trend.

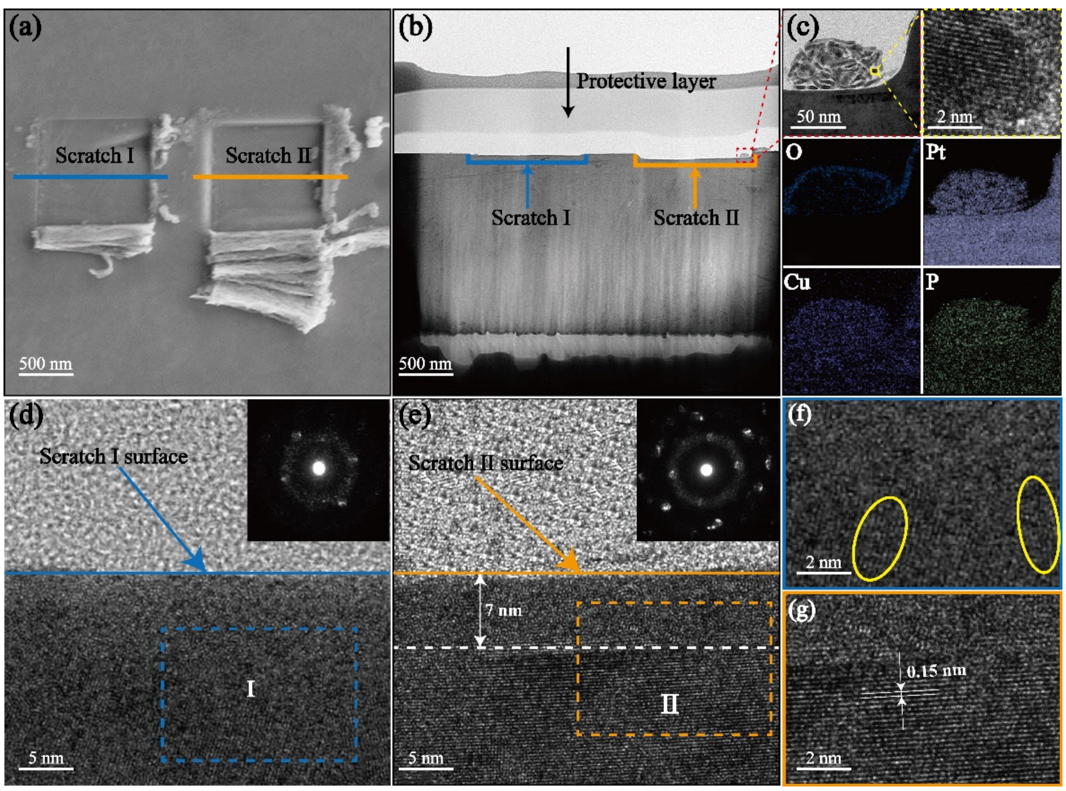


Fig. 5 (a) SEM image of scratches with different loads and (b) HRTEM image of cross-sectional profile of scratched regions. (c) Local magnification and EDS mapping of worn debris, as marked by red rectangle in (b). (d, e) HRTEM images of scratches I and II near surface. Insets are SAED patterns of regions I and II in (d) and (e), respectively. (f, g) Local magnification of regions I and II, showing an ordered cluster and superlattice-like structure formed by scratch-induced deformation in subsurface.

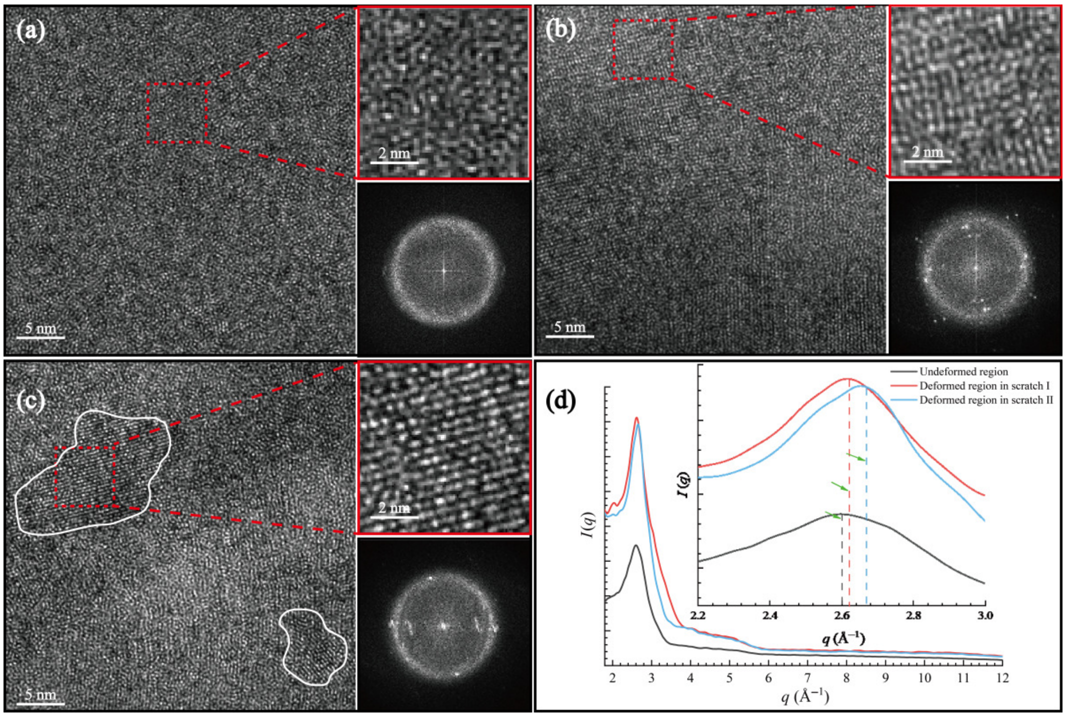


Fig. 6 HRTEM images of (a) undeformed region and (b, c) deformed regions in interiors of scratches I and II, respectively. (d) Azimuthally averaged scattered intensity, $I(q)$, as a function of the scattering vector magnitude, q, of SAED pattern. Inset: close-up of position q of main halo.

reduction through structural relaxation. To investigate the free volume evolution qualitatively, we calculate the azimuthally averaged scattered intensity I_q as a function of the scattering vector q that is computed from FFT patterns for different regions via eRDF analyzer software [28], as shown in Fig. 6(d). Upon nanoscratching, the position of the first halo is shifted to a higher q than that of the undeformed halo, which is associated with a smaller average atomic volume. This finding is in good agreement with the HRTEM observations.

4 Discussion

Previous studies have reported that deformation can lead to work hardening in Zr-based MGs loaded under triaxial stress states [8, 12]. They confirmed the hardening behavior by the hardness increase in the notched region after deformation, the rationale of which is similar to the friction force changes in this work. Although hardening behavior is confirmed in both works, the structural changes in response to deformation differ from each other. Wang et al. [8] observed no visible changes in the atomic structure after deformation via TEM analysis. In this work, however, distinct structural ordering can be clearly observed in the atomic structure upon nanoscratching. The observed structural changes in this work, but not in theirs [8], may be due to different stress levels during deformation, which lead to different degrees of structural relaxation. The maximum failure stress of the notched sample reported by Wang et al. [8] is approximately 2.8 GPa, which is slightly greater than the critical stress σ_c, at which the rates of free volume annihilation and production are in balance. Based on our previous work that established the relationship between the tip radius and yield stress [28], the yield shear stress that occurs in scratch I is estimated to be ~11 GPa, which is much greater than the critical stress σ_c. The much higher stress strongly promotes atomic diffusional relaxation, thus giving rise to a more ordered (relaxed) structural configuration.

Caron et al. [16] previously presented a similar study on the nanoscale friction and wear of Pt-based metallic glasses and reported the formation of shear bands along scratched grooves, implying that deformation is intrinsically mediated by shear bonding. The tip radius in their work is ~100 nm, which is much larger than that of ~10 nm in this work. Correspondingly, the yield shear stress can be estimated to be ~4.8 GPa for the 100-nm tip and ~11 GPa for the 10-nm tip according to the roughly linear relationship developed by Yu et al. [29]. By using the free volume evolution model proposed by Steif et al. [30] and then modified by Wang et al. [8], the free volume is associated with net gain in the 100 nm case but with considerable net loss in the 10 nm case, as shown in Fig. 7. This means that the deformation in this work is dominated by the free volume annihilation process, resulting in a densifying atomic flow. The observation of shear band formation in the work of Caron et al. [16] but not in our work can be understood in the sense that the stress is too small to induce a free volume annihilation-dominated regime, such that the formation of shear bands cannot be suppressed entirely.

Another unique phenomenon is that although local crystallization is observed in metallic glass, the crystallized structure is very different in morphology from the surface to the interior. Near the surface, a superlattice layer with nanoscale thickness formed along the scratching direction, whereas the nanocrystals were dispersed in the amorphous matrix. This means that the atomic dynamics may differ from the surface into the interior. Chen et al. [26] also reported similar superlattice-like nanostructures in the surface regions of several MGs subjected to annealing treatment, but the formation mechanism for the superlattice-like structure has not yet been fully explored. We speculate that the time scale for structural relaxation is the main reason for the difference in the crystallized nanostructure. It has been widely reported that the local heat generated by atomic flow contributes to diffusional structural ordering due to enhanced atomic mobility [31, 32]. Near the surface, the generated local heat can be rapidly dissipated from sheared regions by thermal conduction. As a result, the time scale for structural relaxation is extremely short, which may be insufficient for crystal nucleation

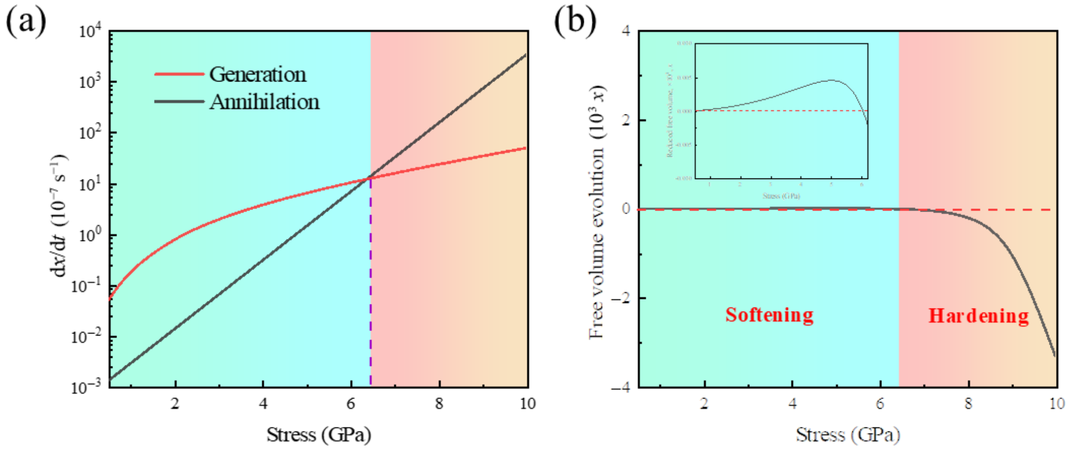


Fig. 7 (a) Free volume generation and annihilation rates as a function of stress level in Pt-based metallic glass under a compressed triaxial stress state. There is a critical stress σ_c, where rates of free volume annihilation and production are in balance. Beyond σ_c, the rate of free volume annihilation significantly exceeds that of free volume production, resulting in a net loss of free volume. (b) Net free volume evolution, x, is obtained by subtracting the free volume annihilation rate from generation rate extracted in (a), where x = (generation rate) – (annihilation rate).

but adequate for crystal growth, since the activation energy for crystal nucleation is greater than that of crystal growth in MGs [33]. In the interior, the time scale for diffuse atomic flow is longer because of the long diffusion distance, which provides sufficient energy for crystal nucleation and enables disordered atoms to self-assemble into nanocrystals.

From these results, the following mechanism for work hardening can be proposed. Upon nanoscratching, the atomic mobility within the stressed volume is significantly increased because of the high hydrostatic stress, which accelerates the diffusional relaxation of the glass structure by rapidly annihilating the shear-induced free volume. This leads to a unique densifying structural configuration in which the atoms self-assemble into an ordered structure with a lower free volume. The densifying plastic flow effectively suppresses strain localization, enabling deformation to develop in a more homogeneous way. Upon cyclic scratching, the structural ordering process is further enhanced, allowing local clusters of MRO to evolve into a crystallized structure.

5 Conclusions

In this work, the scratch-induced deformation and work hardening behavior of an atomically flat Pt-BMG that was thermoplastically imprinted by the replication of a $SrTiO_3$ single-crystal template were investigated via AFM. Upon nanoscratching, the deformation of the Pt-based metallic glass results in homogeneous plastic flow, as indicated by the continuous pile-ups along the grooves. In response to scratch-induced deformation, work hardening behavior is also identified, manifested by an obvious decrease in the friction force within scratched regions. Further examination via high-resolution transmission electron microscopy of the atomic structure beneath the scratched surface revealed a relaxed structural configuration after scratching, as indicated by the ordered clusters in the case of line scratching and the crystallized nanostructure in the case of cyclic scratching. This study provides direct observations of the ordering process in response to work hardening, which has not been achieved in metallic glasses.

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Availability of data and materials

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Declaration of competing interest

The authors have no competing interests to declare that are relevant to the content of this article.

Electronic Supplementary Material

Supplementary material is available in the online version of this article at https://doi.org/10.26599/FRICT.2025.9441100.

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