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## Single shear-band plasticity in a bulk metallic glass at cryogenic temperatures

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At cryogenic temperatures bulk metallic glasses can sustain higher plastic strains than at room temperature. This is generally believed to result from an intrinsic shear-band nucleation rate that increases with decreasing temperature. Here we report on inhomogeneous flow operating via a single shear band even at cryogenic temperatures, challenging the presupposition of increased shear-band activity. The results provide a new interpretation of non-serrated flow and explain, via a simple viscosity law, the correspondingly observed strength increase with decreasing temperature.

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Bulk metallic glasses (BMGs) belong to a class of materials that do not contain crystalline order and therefore do not admit dislocation structure and activity as seen in their crystalline counterparts—the fundamental microscopic mechanism of plasticity that underpins our understanding of materials strength and toughness of experimentally realizable metals in relation to the theoretical strength of a perfect metallic crystal. Despite the strong atomic-scale disorder of BMGs, these materials exhibit a high elastic limit and high fracture strength, good fracture toughness, soft-magnetic properties, and superplastic formability near the glass transition [1–4]. However, because of their lack of a dislocation network, they are unable to strain harden or to be cold-worked at temperatures well below their glass transition temperature  $T_g$ , which has so far hampered our ability to understand the underlying physics of their plastic deformation.

A major hurdle hindering room-temperature engineering applications of BMGs is their pronounced brittleness under tensile loading and their limited compressive ductility, which at low homologous temperatures  $(T/T_g)$  is carried by highly localized flow defects, called shear bands, which are confined to a thickness of only a few tens of nanometers [5,6]. A tremendous international effort is underway to improve the room-temperature ductility of

At cryogenic temperatures, however, a clear picture of enhanced strength and ductility at a stable stress level has been reported with decreasing testing temperature [14–19]. A testing series as a function of decreasing temperature generally comprises a strain-rate- and temperature-dependent transition from serrated flow at room temperature to smooth, non-serrated flow at sub-ambient temperatures [20–23]. More specifically, this transition is linked to the shear-band velocity at the chosen testing temperature [24]. When passing from room temperature to cryogenic temperatures, a consistent increase in shear-band density has been reported both in tension and compression, and this also provides the basis for rationalizing the increased ductility [14–16,18,25]. The phenomenon of a higher shear-band density at lower temperatures has been widely attributed to a reduction

BMGs, both via intrinsically modifying the glassy structure or by processing composite structures [7–9]. The resulting studies have shown that the increase in shearband density is the dominating factor for increasing the ductility, because the applied plastic strain can be distributed over a large number of shear bands, i.e. a larger fraction of the sample volume [10,11]. Further, the spread in strain at failure in compression testing at ambient conditions can be correlated with the boundary conditions of the test, showing that non-uniform compressive deformation at room temperature yields a higher shear-band density and consequently a larger strain prior to failure [12,13].

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of (i) the mobility of free volume and (ii) the propagation rate of the shear bands, both yielding an increase in the number of sites of shear-band nucleation [5,15,16,18,19,26]. Given that low-temperature deformation is intrinsically related to a higher shear-band nucleation rate, and thus shear-band density, resulting in a greater ductility, BMGs have been classed as highly promising materials for cryogenic applications. Due to the proposed increased shear-band nucleation rate with decreasing temperature, it has hence been concluded that deformation in the non-serrated regime (low temperature) should not occur on a single shear band.

In this paper we uncover the opposite, i.e. the BMG specimens deform on one shear band only, even at cryogenic temperatures but with immediate strain softening occurring upon yielding. Our results allow us to develop a new interpretation of non-serrated flow in terms of shear-band nucleation rate and shear-band propagation. Using this approach we are also able to explain the significant strength increase of BMGs with decreasing temperature.

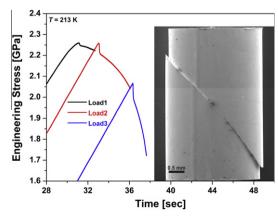
Uniaxial compression tests were conducted using cylindrical samples of glassy Zr<sub>52.2</sub>Ti<sub>5</sub>Cu<sub>17.9</sub>Ni<sub>14.6</sub>Al<sub>10</sub> (Vit105) specimens of 3 mm in diameter with an aspect ratio of 5/3. The samples were prepared by suction casting in an arc-melter, and carefully polished to ensure plane-parallel ends. Displacement-controlled tests were conducted in the temperature range of 77–298 K and at a strain rate of 10<sup>-3</sup> s<sup>-1</sup>. An extensometer bridging the upper and lower compression platens was used to record displacement, and a piezoelectric load cell was employed to measure the load during the tests. Scanning electron microscopy (SEM, Hitachi SU-70) was used to image the surface morphologies.

Figure 1 displays three successive stress–time curves for a specimen tested at 213 K, where the reload was shifted in time such that a continuous curve is formed. Once unloaded, the sample was immediately reloaded. For instance, at the end of load 1, the drive direction of the cross-head was reversed at an arbitrary moment after flow had begun until close to zero force was measured. Without delay the subsequent reloads were conducted in a similar fashion. Each loading cycle exhibits a transient yield phase with a stress overshoot before the stress decays in a smooth manner. Such stress overshoots are known from the homogeneous flow of BMGs, and are typically associated with a non-Newtonian flow regime, where the internal structural relaxation processes are not fast enough to keep up with the external loading rate [27]. Each loading cycle with the transient stress overshoot followed by non-serrated flow corresponds to the initiation of shear-band propagation, where shear-band arrest remains absent due to the high applied strain rate relative to the shear-band velocity at that temperature [28]. Since the stress overshoot magnitude increases with each reload from 13 to 49 MPa, eventually reaching 80 MPa, it may be concluded that the relative degree of structural relaxation increases as a function of accumulated damage created during each of the three unloading/loading cycles. This result thus directly demonstrates shear-band structure relaxation, leading to a shear-initiation stress upon each reloading that exceeds the previous unloading stress.

In addition to this unexpected flow response, SEM investigations reveal that the sample deformed via only a single shear band that was reactivated during each reloading cycle (see inset of Fig. 1). The fact that only a single shear band was activated places non-serrated flow, regarded as a flow regime with a shear-band nucleation controlled mechanism [5,14–16], in a new light. The observation that non-serrated flow can occur with a single flowing shear band does not agree with the view that the applied strain rate cannot be relieved quickly enough into the surrounding matrix leading to multiple shear-banding events and thus a high shear-band nucleation rate, producing a higher shear-band density. From the presented data it becomes clear that all strains can be imparted into a single shear band at a forced rate. Another remarkable difference between what is generally found in the literature on cryogenic plastic flow of BMGs and the curve seen in Figure 1 is the absence of a stable stress level with increasing deformation.

The demonstrated flow response shown in Figure 1 was also reproduced at 158 and 233 K. The present work thus demonstrates that an understanding of inhomogeneous deformation in BMGs at cryogenic temperatures is contained in the physics of a single flowing shear band. However, due to the material's high sensitivity to the testing boundary conditions [12,13], only a few samples showed this flow response, because of multiple shear-band initiation at the sample-anvil interface. In order to improve the number of samples deforming on a single shear band, defined 50 µm root radius notches were introduced 1 mm below the top surface; this applies in the remainder of this document. These notches serve as first shear-band initiation sites only, whereas subsequent flow again underlies the evolving test dynamics, as in the un-notched case.

To further investigate the flow response of a single shear band at cryogenic temperatures, additional tests were conducted. Compression curves obtained at 123 K (curve I), 173 K (curves II–IV) and 298 K (curve V) are summarized in Figure 2. Of particular interest is the direct comparison of stress–strain curves obtained at the same strain rate  $(10^{-3} \, \text{s}^{-1})$  and temperature (173 K) stemming from (a) the conventional case of a dense shear-band morphology (curve IV, as-cast specimen



**Figure 1.** Three successive loading cycles of a specimen tested in the non-serrated regime at 213 K. The inset demonstrates that the sample deformed via a single shear band.

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