

## Shear bands in metallic glasses are not necessarily hot

Stephanie K. Slaughter,<sup>1</sup> Felicitée Kertis,<sup>1</sup> Erin Deda,<sup>1</sup> Xiaojun Gu,<sup>2</sup>  
 Wendelin J. Wright,<sup>2,3</sup> and T. C. Hufnagel<sup>1,4</sup>

<sup>1</sup>*Department of Materials Science and Engineering, Johns Hopkins University,  
 3400 N Charles St, Baltimore, Maryland 21218, USA*

<sup>2</sup>*Department of Mechanical Engineering, Bucknell University, One Dent Drive,  
 Lewisburg, Pennsylvania 17837, USA*

<sup>3</sup>*Department of Chemical Engineering, Bucknell University, One Dent Drive,  
 Lewisburg, Pennsylvania 17837, USA*

<sup>4</sup>*Department of Mechanical Engineering, Johns Hopkins University, 3400 N Charles St,  
 Baltimore, Maryland 21218, USA*

(Received 23 July 2014; accepted 2 September 2014; published online 10 September 2014)

We have used the fusible tin coating method to detect shear band heating in amorphous  $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$  loaded under quasi-static uniaxial compression. High-rate load data allowed a precise determination of the duration of shearing events and final fracture. When loading was halted prior to fracture we saw no evidence of melted tin despite the presence of shear offsets up to  $6\text{ }\mu\text{m}$  on some shear bands. Samples loaded to fracture showed evidence of tin melting near the fracture surface. We attribute the difference to the duration of the events, which is much longer for shear banding (milliseconds) than for fracture (microseconds). © 2014 Author(s). All article content, except where otherwise noted, is licensed under a Creative Commons Attribution 3.0 Unported License. [<http://dx.doi.org/10.1063/1.4895605>]

From the earliest investigations of metallic glasses the observation of apparently molten patterns on metallic glass fracture surfaces suggested that high temperatures (exceeding the glass transition temperature) could be achieved during fracture,<sup>1</sup> a view corroborated by measurement of the temperature of particles ejected during fracture of a Zr-based glass.<sup>2</sup> Whether similarly high temperatures can be achieved in a shear band during plastic deformation at moderate rates without fracture is a more difficult question to answer due to the spatial and temporal scales involved.<sup>3,4</sup> The key spatial dimension is the thickness of the shear band; evidence from electron microscopy<sup>5,6</sup> suggests a thickness on the order of 10–20 nm, so thin that heat conduction away from the band would preclude significant temperature increases.<sup>3</sup> It is not clear whether such measurements are directly applicable to the case of shear bands formed in bulk specimens, however. If the shear band thickness in a bulk specimen was on the order of  $10\text{ }\mu\text{m}$  then temperature increases of several hundred kelvins would be possible,<sup>7</sup> and there is some evidence to suggest that fully developed shear bands in bulk specimens can be this thick.<sup>8,9</sup> Direct infrared thermographic observations during plastic deformation of Zr-based glasses<sup>10,11</sup> show modest temperature increases ( $<20\text{ K}$ ), but the limited spatial resolution of this technique ( $\sim 10\text{ }\mu\text{m}$ ) means that deducing the actual temperature inside the operating shear band requires an assumption about the true shear band thickness.

Lewandowski and Greer<sup>12</sup> developed a clever “fusible coating” method with spatial and temporal resolutions ( $\sim 100\text{ nm}$  and  $\sim 30\text{ ps}$ , respectively) sufficient to estimate the heat evolved during shear band operation. By coating metallic glass specimens with a thin layer of tin and observing melting near shear bands on the surface, they directly demonstrated temperature rises of at least  $200\text{ K}$  (sufficient to reach the melting point of Sn,  $505\text{ K}$ ) over distances of  $\sim 1\text{ }\mu\text{m}$ . From the width of the melted region and an estimate of the duration of the shear banding event, they arrived at an upper-bound estimate of the temperature of the shear band of several thousand kelvins. Later, Georgarakis and co-workers<sup>13</sup> explored the effect of the time scale assumption and calculated a range of possible maximum temperatures from  $3400\text{--}8600\text{ K}$  for fast shear times ( $\sim 10\text{ ns}$ ) to an insignificant rise for

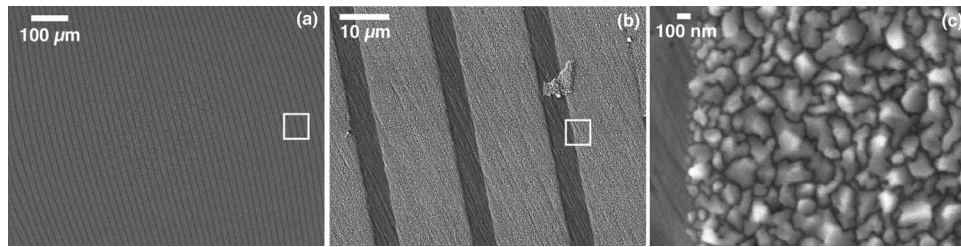


FIG. 1. Scanning electron micrographs of deposited tin lines on a metallic glass specimen, prior to compression testing. Outlines in (a) and (b) indicate areas shown in (b) and (c), respectively.

slow shear times (100 ms). Later work from Zhang and co-workers<sup>14</sup> and Miracle and co-workers<sup>15</sup> further considered the effect of the magnitude of slip offset and concluded that above a critical value (dependent on the sample size but  $\sim 1 \mu\text{m}$ ) significant heating can occur. In both cases<sup>14,15</sup> it was assumed that the shear time must be short to be consistent with the observed melting of the tin coating, discounting the possibility that shearing could occur over longer times.

On the other hand, Ketov and Louzguine-Luzgin<sup>16</sup> noted that in the original Lewandowski and Greer experiments the samples where melted tin was observed had been subjected to fracture (although the shear bands examined were well away from the fracture surface). The significance of this is that fracture occurs over much shorter time scales than shear band operation during quasi-static loading.<sup>17</sup> This rapid unloading creates opportunities for pre-existing shear bands to operate in reverse<sup>1</sup> at high rate and for new shear bands to form due to elastic waves propagating through the sample.<sup>18</sup> Furthermore, a much larger amount of elastic strain energy can be dissipated during fracture than during a shear banding event. These considerations suggest that significant heating of shear bands may only be associated with fracture, and that during ordinary plastic deformation (at least at low-to-moderate strain rates) the heating may be modest.

To investigate these issues in more detail we have performed fusible coating experiments on metallic glass specimens loaded in uniaxial compression, with two new features relative to the prior work. First, we lithographically patterned the tin coating of our specimens into narrow lines to allow a direct determination of the shear offset on each shear band. Second, we use high-rate data acquisition to directly measure the duration of individual shear banding events. Some of our specimens were loaded to fracture while for others the loading was halted prior to fracture.

We prepared rectangular prism bulk metallic glass specimens of  $\text{Zr}_{57}\text{Ti}_5\text{Cu}_{20}\text{Ni}_8\text{Al}_{10}$  (compositions in atomic percent) with dimensions  $10.0 \text{ mm} \times 5.0 \text{ mm} \times 4.5 \text{ mm}$  using techniques described in detail elsewhere.<sup>19</sup> We polished two adjacent sides to a mirror finish and, using a negative tone photolithography process and sputtering, produced tin lines ( $15 \mu\text{m}$  wide with  $5 \mu\text{m}$  spacing) parallel to the loading direction on these two sides (Figure 1). The thickness of the tin coating was  $50 \text{ nm}$  as determined by profilometry. We performed compression tests at a constant displacement rate corresponding to a nominal strain rate of  $10^{-4} \text{ s}^{-1}$  using a high stiffness, low-bending testing machine described in detail elsewhere.<sup>17,20</sup> Load data were acquired using a  $150 \text{ kN}$  Instron load cell and a Kistler piezoelectric load cell at  $100 \text{ kHz}$ . Bending was minimized by use of a subpress, and the stiffness of the machine ensured minimal energy transfer from the system to the samples.

Shear band operation is manifested as stress drops (“serrations”) in the stress-time or stress-strain data from a compression test. Figure 2 shows the largest serration we observed from among six samples for which loading was halted prior to fracture. The duration of this shearing event is  $\sim 6 \text{ ms}$ , of the same order as the time scale of shear banding events observed previously.<sup>17,21</sup> For thirteen stress drops for this sample the durations of the shear events ranged from  $2.3 \text{ ms}$  to  $6 \text{ ms}$  (with an average of  $4.6 \text{ ms}$ ) while the stress drops ranged from  $0.39 \text{ MPa}$  to  $3.02 \text{ MPa}$  (with an average of  $1.17 \text{ MPa}$ ). Although we cannot uniquely associate a given stress drop with a specific shear band observed on the sample surface, it seems reasonable to assume that the largest stress drops are associated with the largest slip offsets. Note that this assumes that each shear band observed on the specimen is due to a single slip event.

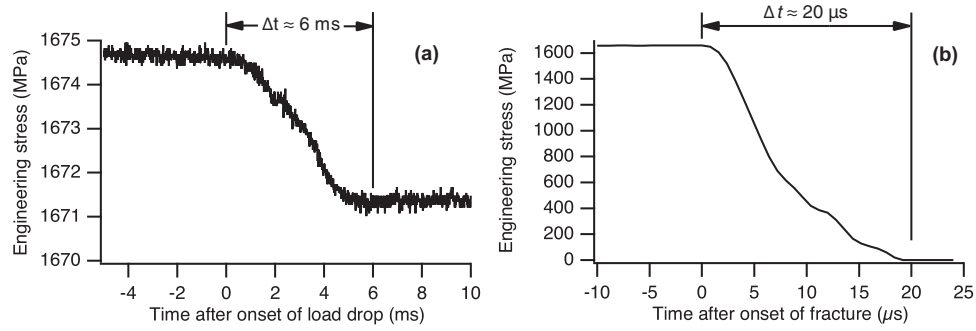


FIG. 2. Stress versus time data showing (a) a stress drop related to a shearing event in a non-fractured sample and (b) the fracture event in a fractured sample.

To calculate the temperature increase associated with a shearing event we assume that all of the elastic strain energy in the specimen released during the stress drop is converted into plastic work which heats the shear band. The change in elastic strain energy density  $\Delta h$  of a specimen that experiences a stress drop of  $\Delta\sigma$  at a flow stress  $\sigma$  is

$$\Delta h \simeq \left(\frac{\sigma}{E}\right) \Delta\sigma, \quad (1)$$

where  $E$  is Young's modulus.<sup>13</sup> Because high-speed cinematography shows that most of the slip occurs simultaneously across the entire band and not as a single concentrated front<sup>17</sup> we treat the shear band as a planar heat source. With  $V$  as the volume of the specimen,  $A$  the area of the shear band, and  $\Delta t$  the duration of the shear event, the average thermal flux out of the shear band is

$$\begin{aligned} q &\approx \Delta h \times \left(\frac{V}{A \Delta t}\right) \\ &\approx \Delta h \times \left(\frac{l}{\sqrt{2} \Delta t}\right), \end{aligned} \quad (2)$$

where  $l$  is the length of the specimen and the factor of  $\sqrt{2}$  comes in because the shear band plane is inclined at approximately  $45^\circ$  to the loading axis. Finally, the temperature rise  $\Delta T(x, t)$  can be found using a simple one-dimensional solution of the diffusion equation:<sup>22</sup>

$$\Delta T(x, t) = \frac{q}{2c_p\alpha} \left( \sqrt{\frac{4\alpha t}{\pi}} \exp\left(\frac{-x^2}{4\alpha t}\right) - x \operatorname{erfc}\left(\frac{x}{\sqrt{4\alpha t}}\right) \right), \quad (3)$$

where  $c_p$  is the constant pressure heat capacity,  $\alpha$  is the thermal diffusivity,  $t$  is the time elapsed since the onset of shear, and  $x$  is the distance from the shear band (assumed to have an infinitesimal thickness).

To calculate the temperature rise with this model in addition to material parameters such as the modulus, heat capacity, and thermal diffusivity we need to know the flow stress  $\sigma$ , the magnitude of the stress drop  $\Delta\sigma$ , and the duration of the slip event  $\Delta t$ . From our load-time data we can determine all of these. Using data from the largest stress drop in our data (shown in Fig. 2(a)), the calculated maximum temperature increase at the center of the shear band (i.e., at  $x = 0$ ) is shown as the bottom curve in Fig. 3. Although we know the duration of the shear event to be  $\approx 6$  ms from the stress drop data, we plot the temperature for a wide range of shearing times to highlight the effect of the duration on the temperature increase. Clearly, large temperature increases are only possible with shear times on the order of microseconds or shorter. But both our results and similar prior work<sup>17,21</sup> show that the characteristic time for a shear banding event is on the order of milliseconds, which results in a negligibly small temperature rise ( $\Delta T < 1$  K). Observation of the non-fractured samples

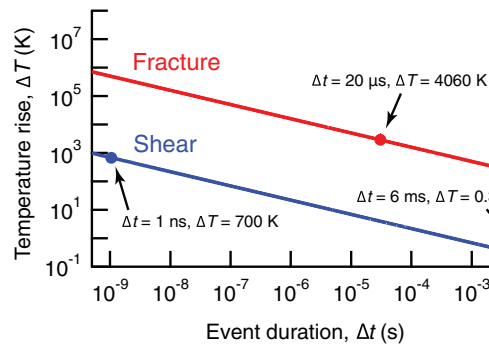


FIG. 3. Calculations of maximum temperature rise versus time at the center of a shear band. The lower curve represents a shear band event with a measured event duration (6 ms) and a hypothetical duration of 1 ns. The upper curve represents a fracture event with an experimentally measured fracture time ( $\approx 20 \mu\text{s}$ ). (Both curves calculated using thermophysical properties for  $\text{Zr}_{41}\text{Ti}_{14}\text{Ni}_{10}\text{Ni}_{12.5}\text{Be}_{22.5}$  from Ref. 15.)

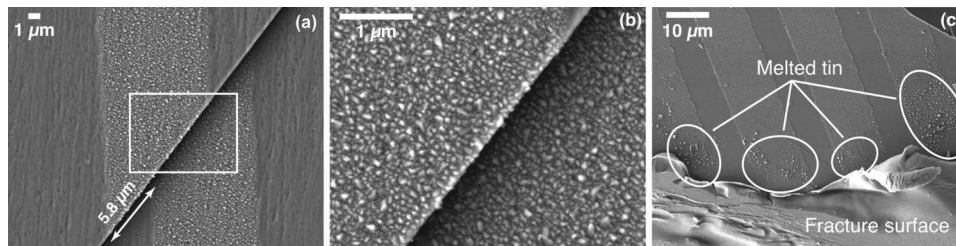


FIG. 4. Scanning electron micrographs of tin lines after testing. Samples that did not fracture showed no melted tin (a) and (b). Only for samples that fractured is any evidence of melting observed (c).

(Figures 4(a) and 4(b)) corroborates these calculations. No melted tin was observed in any of our samples that did not fracture, even those for shear bands with offsets as large as  $\sim 6 \mu\text{m}$ .

We do observe melted tin in some cases, but only on those samples that fractured. As shown in Figure 2(b) the duration of fracture events is on the order of a few tens of microseconds (or less; see also Refs. 17 and 21). Because the stress drop is also much larger, in principle a larger amount of plastic work is converted to heat in a shorter time during fracture than during shear banding. Using the same model as above the predicted maximum temperature rise for this fracture event is shown in Figure 3 (upper curve). Using stress drop duration for fracture of  $20 \mu\text{s}$  (Fig. 2(b)) the predicted maximum temperature increase is  $\Delta T \approx 4060 \text{ K}$ . This should be viewed only as an approximation because it is not necessarily true that all of the elastic strain energy released during fracture goes into melting the fracture surface; on the other hand, an unknown amount of strain energy from the load frame may be dissipated during fracture. Furthermore, the duration of the fracture event from Fig. 2(b) is an upper-bound estimate because video evidence suggests that fracture is complete before the load cell reading drops to zero.<sup>17</sup> Nevertheless, the calculation suggests that large temperature rises should be expected during fracture, and indeed in our specimens that fractured we do observe evidence that the tin coating melted (Fig. 4(c)).

Our observations are consistent with several other reports that conclude that heating in shear bands during plastic deformation can be modest,<sup>10,16</sup> but we note that heating is affected by several extrinsic factors such as the strain rate and the stiffness of the testing machine. Several groups have examined the effect of machine stiffness on the stability of inhomogeneous flow in metallic glasses, with the common observation that stable (serrated) flow is favored for small samples and stiff machines.<sup>23,24</sup> Cheng and co-workers<sup>23</sup> developed a simple kinematic model that takes into account the rate at which the sample unloads during a shear band event (which is related to stiffness of the sample and testing machine) and the rate at which the flow stress in the shear band drops due to increasing temperature. They predicted a transition from cold shear bands with self-limiting

stick-slip behavior to runaway shear bands with significant heating. They quantified this in terms of a parameter  $\lambda = L(1 + S)$  where  $L$  is the sample length and  $S = \kappa_S/\kappa_M$  is the ratio of the sample and machine stiffnesses.<sup>24</sup> A critical value  $\lambda_{\text{crit}}$  determined by experiment separates sample size/machine stiffness combinations with self-limiting (“cold” shear band) behavior ( $\lambda < \lambda_{\text{crit}}$ ) from those with runaway (“hot”) behavior ( $\lambda > \lambda_{\text{crit}}$ ). For our experiments  $\kappa_M \simeq 3.0 \times 10^8 \text{ N m}^{-1}$  and  $\kappa_S = E(A/L) \simeq 2.2 \times 10^8 \text{ N m}^{-1}$  (where  $E$  is Young’s modulus, and  $A$  and  $L$  are the cross-sectional area and length of the specimens, respectively) so  $L(1 + S) \simeq 17 \text{ mm}$ , somewhat larger than the  $\lambda_{\text{crit}} = 12 \text{ mm}$  determined by Cheng and co-workers for a metallic glass of similar composition ( $\text{Zr}_{64.13}\text{Cu}_{15.75}\text{Ni}_{10.12}\text{Al}_{10}$ ) from data in Ref. 24. This, together with the fact that we observe only a few serrations in each test (as compared to the large numbers observed in stable serrated flow in Refs. 24 and 23), suggests that significant heating might be expected under our experimental conditions.

Our observations clearly indicate that softening in shear bands under quasistatic loading is not driven by an increase in temperature due to plastic work, as indeed Lewandowski and Greer also concluded.<sup>12</sup> We suggest that observations of shear bands using the fusible coating method can be confounded by fracture of the specimens, even if the shear bands examined are well away from the fracture surface. During fracture new shear bands may form and there is an opportunity for existing shear bands to operate in reverse (when the imposed deformation is suddenly released) at strain rates much higher than the original quasistatic loading, allowing significant heating to occur.

We gratefully acknowledge support from the National Science Foundation for this work: S.K.S., F.K., E.D., and T.C.H. from NSF Grant No. DMR-1107838; and X.G. and W.J.W. from NSF Grant No. DMR-1042734.

- <sup>1</sup> C. A. Pampillo, *J. Mater. Sci.* **10**, 1194 (1975).
- <sup>2</sup> C. J. Gilbert III, J. W. Ager, V. Schroeder, R. O. Ritchie, J. P. Lloyd, and J. R. Graham, *Appl. Phys. Lett.* **74**, 3809 (1999).
- <sup>3</sup> C. A. Schuh, T. C. Hufnagel, and U. Ramamurty, *Acta Mater.* **55**, 4067 (2007).
- <sup>4</sup> A. L. Greer, Y. Q. Cheng, and E. Ma, *Mater. Sci. Eng. R: Rep.* **74**, 71 (2013).
- <sup>5</sup> P. E. Donovan and W. M. Stobbs, *Acta Metall.* **29**, 1419 (1981).
- <sup>6</sup> J. Li, F. Spaepen, and T. C. Hufnagel, *Philos. Mag. A* **82**, 2623 (2002).
- <sup>7</sup> C. T. Liu, L. Heatherly, D. S. Easton, C. A. Carmichael, J. H. Schneibel, C. H. Chen, J. L. Wright, M. H. Yoo, J. A. Horton, and A. Inoue, *Met. Mat. Trans. A* **29A**, 1811 (1998).
- <sup>8</sup> H. Guo, J. Wen, N. M. Xiao, Z. F. Zhang, and M. L. Sui, *J. Mater. Res.* **23**, 2133 (2011).
- <sup>9</sup> J. Pan, Q. Chen, L. Liu, and Y. Li, *Acta Mater.* **59**, 5146 (2011).
- <sup>10</sup> K. M. Flores and R. H. Dauskardt, *J. Mater. Res.* **14**, 638 (1999).
- <sup>11</sup> B. Yang, P. K. Liaw, M. Morrison, C. T. Liu, R. A. Buchanan, J. Y. Huang, R. C. Kuo, J. G. Huang, and D. E. Fielden, *Intermetallics* **13**, 419 (2005).
- <sup>12</sup> J. J. Lewandowski and A. L. Greer, *Nature Mater.* **5**, 15 (2006).
- <sup>13</sup> K. Georgarakis, M. Aljerf, Y. Li, A. LeMoulec, F. Charlot, A. R. Yavari, K. Chornokhvostenko, E. Tabachnikova, G. A. Evangelakis, D. B. Miracle, A. L. Greer, and T. Zhang, *Appl. Phys. Lett.* **93**, 031907 (2008).
- <sup>14</sup> Y. Zhang, N. A. Stelmashenko, Z. H. Barber, W. H. Wang, J. J. Lewandowski, and A. L. Greer, *J. Mater. Res.* **22**(2), 419 (2007).
- <sup>15</sup> D. B. Miracle, A. Concustell, Y. Zhang, A. R. Yavari, and A. L. Greer, *Acta Mater.* **59**, 2831 (2011).
- <sup>16</sup> S. V. Ketov and D. V. Louzguine-Luzgin, *Sci. Rep.* **3**, 2798 (2013).
- <sup>17</sup> W. J. Wright, R. R. Byer, and X. Gu, *Appl. Phys. Lett.* **102**, 241920 (2013).
- <sup>18</sup> V. Z. Bengus, E. D. Tabachnikova, S. E. Shumilin, Y. I. Golovin, M. V. Makarov, A. A. Shibkov, J. Miskuf, K. Csach, and V. Ocelik, *Int. J. Rapid Solidification* **8**, 21 (1993).
- <sup>19</sup> X. Gu, T. Jiao, L. J. Kecskes, R. H. Woodman, C. Fan, K. T. Ramesh, and T. C. Hufnagel, *J. Non-Cryst. Solids* **317**, 112 (2003).
- <sup>20</sup> W. J. Wright, M. W. Samale, T. C. Hufnagel, M. M. LeBlanc, and J. N. Florando, *Acta Mater.* **57**, 4639 (2009).
- <sup>21</sup> J. Antonaglia, W. J. Wright, X. Gu, R. R. Byer, T. C. Hufnagel, M. LeBlanc, J. T. Uhl, and K. A. Dahmen, *Phys. Rev. Lett.* **112**, 155501 (2014).
- <sup>22</sup> H. S. Carslaw and J. C. Jaeger, *Conduction of Heat in Solids*, 2nd ed. (Oxford University Press, 1959).
- <sup>23</sup> Y. Cheng, Z. Han, Y. Li, and E. Ma, *Phys. Rev. B* **80**, 134115 (2009).
- <sup>24</sup> Z. Han, W. F. Wu, Y. Li, Y. J. Wei, and H. J. Gao, *Acta Mater.* **57**, 1367 (2009).