

## Full length article

# Hierarchical heterogeneity and an elastic microstructure observed in a metallic glass alloy



Peter Tsai <sup>a</sup>, Kelly Kranjc <sup>a</sup>, Katharine M. Flores <sup>a, b, \*</sup>

<sup>a</sup> Washington University in St Louis, Institute of Materials Science & Engineering, One Brookings Drive, St Louis, MO 63130, USA

<sup>b</sup> Washington University in St Louis, Department of Mechanical Engineering & Materials Science, One Brookings Drive, Campus Box 1185, St Louis, MO 63130, USA

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## ABSTRACT

Heterogeneities in a Zr-based bulk metallic glass (BMG) were investigated using dynamic modulus mapping on a nanoindentation platform, revealing a complex elastic microstructure consisting of interpenetrating locally stiff and compliant regions with characteristic feature lengths on the order of 100 nm. The unique microstructures were observed in as-cast, annealed and laser-processed materials. Surprisingly, at various mapping locations across the cast sample cross sections, the elastic microstructures displayed directional alignment of the features with the mold surface. The results introduce an unprecedented spatial regime of elastic variations in monolithic BMGs that may contribute towards a more complete understanding of structure-property relationships in these materials, especially with regards to global deformation behavior.

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## 1. Introduction

Understanding structure-property relationships has long been at the heart of alloy development and design. For crystalline alloys, depending on the complexity of the overall microstructure, variations in structural features could span length scales across many orders of magnitude. Due to the periodic nature of atomic packing in even the most complex crystalline alloys, their underlying structures are readily characterized by conventional techniques such as electron microscopy and x-ray diffraction. In contrast to crystalline alloys, the atomic structure of monolithic metallic glass is amorphous, devoid of long range translational symmetry, although it is known that short and medium range ordering persist [1–3]. For this reason, metallic glasses appear deceptively homogeneous and isotropic when investigated with conventional characterization techniques, without the typical microstructural features that account for complexity and uniqueness in crystalline alloys. Despite the apparent microstructural similarity of metallic glasses, however, their deformation behavior can vary drastically

with both alloy composition and processing conditions [4–6], implying the existence of a complex underlying structure that is not yet fully understood.

On the atomic length scale, the structure of a monolithic metallic glass is by definition heterogeneous, possessing a statistical spread of different atomic cluster configurations, randomly orientated and distributed throughout its volume. Structural heterogeneities extending beyond the size of atomic clusters are less obvious, especially considering the high atomic packing efficiency of bulk amorphous alloys [7]. Recent experimental and molecular dynamics studies have reported nanoscale heterogeneous structure in metallic glasses [8–13]. The presence of such heterogeneities carries profound implications with respect to macroscopic mechanical behavior [14]. In simulated Cu-Zr binary glasses, Ma et al. demonstrated that elastically-soft regions densely populated with unstable cluster motifs are more susceptible to local shear transformations [12,13,15]. The authors proposed that increasing the population of soft spots in a glass would encourage profuse shear banding, resulting in higher fracture toughness and enhanced plastic behavior, both of which are rare among known bulk metallic glasses (BMGs). On the experimental front, using atomic force microscopy (AFM) techniques, nanoscale fluctuations of elastic modulus and energy dissipation have been measured in several amorphous alloys [8–11], but important details such as the

\* Corresponding author. Washington University in St Louis, Institute of Materials Science & Engineering, One Brookings Drive, St Louis, MO 63130, USA.

E-mail addresses: [peter.tsai@gmail.com](mailto:peter.tsai@gmail.com) (P. Tsai), [floresk@wustl.edu](mailto:floresk@wustl.edu) (K.M. Flores).

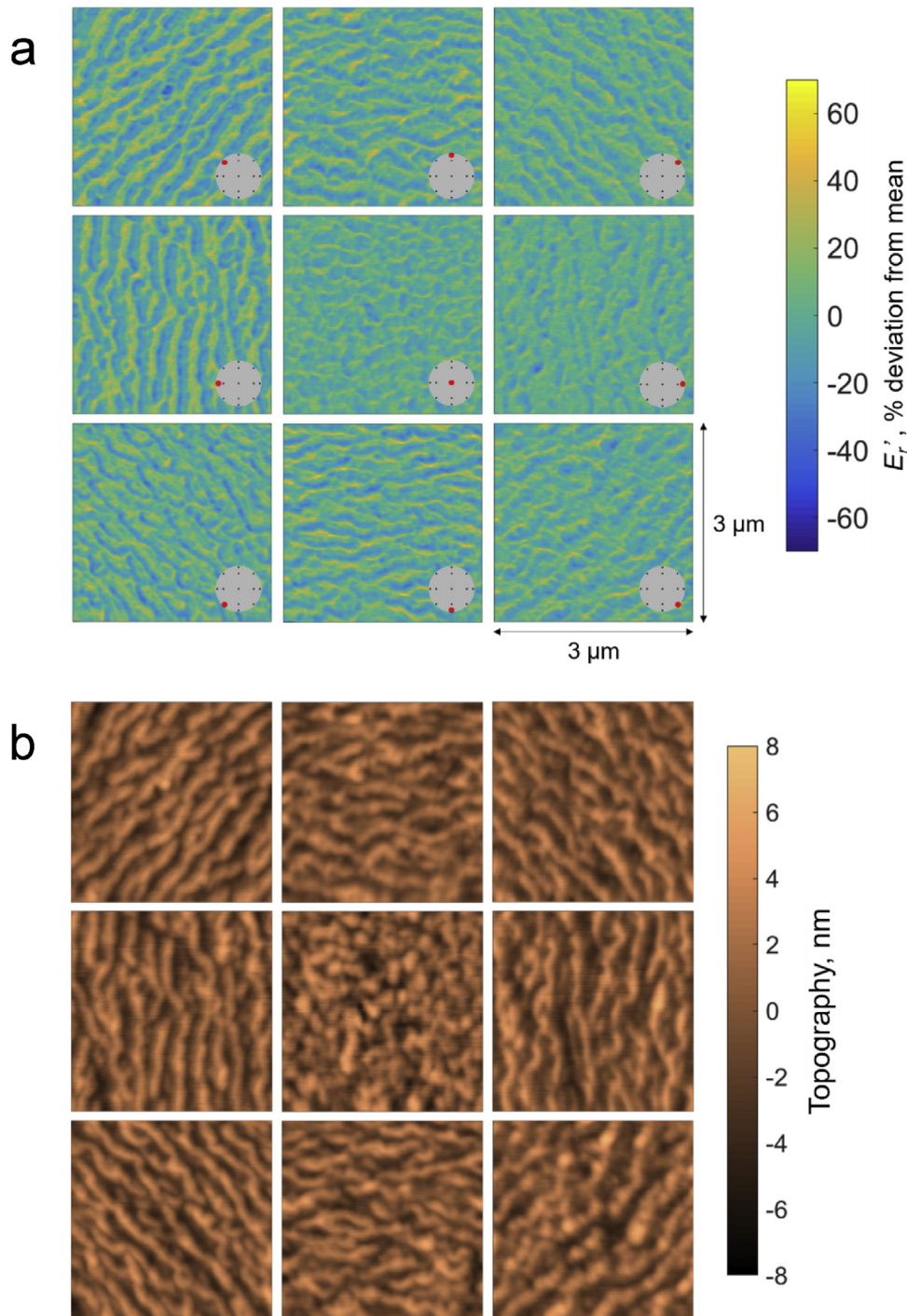
characteristic size of the heterogeneous features, their morphology and spatial distribution, and their sensitivity to thermal processing history remain ambiguous.

In this work, we apply dynamic modulus mapping (DMM) on a nanoindentation platform, to explore spatial fluctuations of mechanical properties in  $\text{Zr}_{58.5}\text{Cu}_{15.6}\text{Ni}_{12.8}\text{Al}_{10.3}\text{Nb}_{2.8}$ , a BMG with centimeter-scale critical casting thickness [16]. The collected data reveal mechanical heterogeneities on a 100 nm length scale, the characteristics of which have never been reported before. We further explore the influence of laser-pulse melting and thermal

annealing on the observed heterogeneous microstructures, and based on the results, discuss possible origins and the implications of their existence on macroscopic deformation behavior.

### 1.1. Experimental methods

The  $\text{Zr}_{58.5}\text{Cu}_{15.6}\text{Ni}_{12.8}\text{Al}_{10.3}\text{Nb}_{2.8}$  BMG specimens used for this study were prepared by arc melting pure elements with minimum purities of 99.9 at% in an argon atmosphere. The resulting ingot was flipped and remelted 3 times to ensure homogeneity prior to



**Fig. 1. Elastic heterogeneities observed by dynamic modulus mapping. (a)** Selected 3  $\mu\text{m}$   $\times$  3  $\mu\text{m}$  storage modulus ( $E''$ ) maps for the BMG in the as-cast condition, collected from various locations (lower right insets) around the cross section of the cylindrical specimen. **(b)** Corresponding surface topography maps.

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