

Structural and mechanical signatures of maximally amorphous transition metal alloys

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We demonstrate that thin metallic glasses of V_xZr_{100-x} , deposited by direct current magnetron sputtering, closely approximate the structural characteristics of theoretically predicted maximally amorphous materials. Experimentally, x-ray reflectometry reveals atomically flat films with well-defined thickness and low surface roughness, while Rutherford backscattering spectrometry confirms V/Zr stoichiometry within 1 at.% and a compact, systematically varying density with composition. Transmission electron microscopy shows no evidence of columnar growth, consistent with fully amorphous and homogeneous films. Using *ab initio* stochastic quenching to generate representative maximally amorphous configurations, we find excellent agreement with experiment in terms of mass density and pair distribution functions. The calculated structures are mechanically stable, elastically isotropic, and exhibit high Poisson ratios and Pugh ratios indicating a characteristically ductile response. The combination of compactness and ductility suggests potential for applications in mechanically resilient coatings and for hydrogen storage, where deformation tolerance and structural integrity are critical. The agreement between simulation and experiment across structural and compositional descriptors supports the conclusion that sputtered V–Zr metallic glasses can be viewed as experimental realizations of statistically maximally amorphous states within the framework of the random valley approximation. We propose such structures can serve as well defined benchmarks for the maximal degree of amorphousness.

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I. INTRODUCTION

The restrictions imposed by crystal symmetries are naturally lifted in amorphous materials, enabling the design of solids with unique physical properties. By alloying elements in nonstoichiometric ratios, one can modify the average interatomic spacing, coordination number, and local chemical environment to tune mechanical [1,2], electrical [3], and magnetic [4,5] properties.

Amorphous metals can be understood as liquids frozen in space, yet with structural motifs distinct from crystalline phases. Although the absence of long-range order gives them unique physical properties, it makes their structure difficult to classify. A number of approaches have been developed to probe amorphous structure, including x-ray scattering, electron microscopy, and computational models. In practice, these methods converge on a picture where amorphous states are characterized by dense local packing, diffuse scattering features, and short-range order characteristic of the glassy state. Previous work has highlighted the richness of this spectrum. Braeckman and Depla [6] examined sputtered thin-film alloys,

showing how deposition conditions drive density and stress variations while retaining amorphous character. Mu *et al.* [7] applied independent component analysis–assisted pair distribution function analysis to resolve distinct local bonding environments in complex amorphous nanophases. Hilke *et al.* [8] used fluctuation electron microscopy to track the reorganization of medium-range order during plastic deformation of bulk metallic glasses. These studies demonstrate that amorphous structures are not monolithic but span a continuum, with disorder manifesting at multiple length scales.

What remains absent is a reproducible way to situate different systems along this continuum. Here we propose such a scale, anchored to stochastically quenched reference states that represent maximally amorphous configurations. As described by Wallace and co-workers [9,10], the potential energy landscape of a disordered solid contains a vast number of degenerate local minima configurations that exhibit no long range order and the forces on all the atoms are zero. These configurations represent statistically saturated states of structural disorder and two such configurations would have the same pair distribution function and bond angle distribution. The central idea is that many observables are similar across these random minima, which may be viewed as a statistical family of mechanically stable disordered states, each kinetically arrested by an effective energy barrier.

It follows that for sufficiently large systems, one representative configuration can provide a useful description of the amorphous structure. The stochastic quenching method [9] approximates this ensemble by placing atoms randomly in a cell and relaxing the structure using realistic quantum

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mechanical forces to its nearest energy minimum, offering a practical computational route to probe these amorphous configurations. The structures that are created in this way correspond to the most amorphous phase that is possible under the constraint that the forces on all the atoms are zero [9].

This raises a natural and testable question: Can such maximally amorphous structures be realized experimentally? In this study, we investigate amorphous thin films of the binary alloy V_xZr_{100-x} , chosen for both practical and conceptual reasons. Vanadium and zirconium exhibit large differences in their atomic radii and large melting temperatures, making them ideal candidates for a well behaved amorphous structure. V–Zr alloys have attracted interest for their large hydrogen uptake capacity [11–13], use in hydride batteries [14], and neutron-moderating properties in nuclear applications [15]. Yet, surprisingly little is known about their glassy counterparts. While amorphous V–Zr has previously been fabricated via vacuum evaporation [16], ball milling [17], and magnetron sputtering [18], none of these studies directly addressed the concept of maximal amorphicity or the nature of the resulting short-range order. Eickert *et al.* [18] suggested that kinetic constraints during growth might stabilize amorphous structures across an unusually broad composition range, a concept closely aligned with Wallace’s random valley framework. Here, we use DC magnetron sputtering under optimized sputter gas purity and pressure conditions to mimic stochastic atomic placement under ultrafast quenching conditions ($\sim 10^{12}$ K/s), minimizing surface diffusion and promoting structural arrest. The comparison to stochastic quenching is therefore meaningful given the similarities of the quenching rates and the kinetic arrest that happens in both cases. The resulting films are assessed using x-ray reflectometry (XRR), pair distribution function (PDF) analysis, transmission electron microscopy (TEM), and Rutherford back-scattering spectrometry (RBS). These experimental observations are directly compared with first-principles stochastic quenching simulations, allowing us to test the correspondence between theoretical and real-world realizations of maximally amorphous materials.

By comparing the measured and simulated pair distribution functions to assess short-range order, mass densities to assess compactness, and x-ray reflectivity patterns to assess medium-range order, we demonstrate that sputtered V–Zr films can closely replicate the structural signatures predicted by stochastic quenching.

II. EXPERIMENTAL DETAILS

A. Sample growth and characterization

The V_xZr_{100-x} films were grown via DC magnetron sputtering under ultrahigh vacuum conditions with a base pressure 3×10^{-10} Torr. Before sputtering, the substrates [amorphous SiO_2 or crystalline sapphire Al_2O_3 (1120)] were annealed at 623 K for 30 min for the purpose of removing surface contaminants. Ar with 6N purity, further purified by a Nupure Omni 40 PF purifier, was introduced into the growth chamber at 2.5 mTorr. The V_xZr_{100-x} layers were co-sputtered from V (99.95%) and Zr (99.99%) pure targets at room temperature to obtain high quenching rates. The power of the magnetrons was kept as low as possible to minimize the deposition rates.

Either a thin layer of metallic Zr was deposited at the very end of the process and allowed to oxidize in air, or an RF sputtered Al_2O_3 layer, to protect the V_xZr_{100-x} layer from further oxidation and degradation. The sample stage was under constant rotation (60 RPM) at all times to minimize composition gradients and ensure even distribution of atoms.

RBS measurements were done at the 5-MV 15SDH-2 Tandem accelerator at Uppsala University [19], employing 2 MeV 4He primary ions. The angle between the sample normal and beam direction was 5° and the RBS passivated implanted planar silicon detector was positioned at a scattering angle of 170° . The elemental composition and areal densities were extracted by fitting the data with the SIMNRA code [20], using SRIM [21] to determine the energy loss. The stopping cross section for 2 MeV 4He ions in vanadium was adapted according to [22], and the value for SiO_2 was corrected by -4% .

A Bruker D8 diffractometer, using $CuK\alpha$ radiation, equipped with a Göbel mirror, was used to measure XRR. From the XRR scans, the thickness and surface roughness were determined via full profile fitting using the GENX [23,24] software package. Grazing incidence diffraction (GIXRD) measurements were performed following the procedure of Bylin, Kapaklis, and Pålsson [25]. The absence of crystallites beyond the detection limit was confirmed using additional GIXRD with finer step size over the entire accessible angular range.

A cross-sectional lamella was prepared from the sample using a Zeiss FIB/SEM Crossbeam 550 dual-beam system. The lamella thickness was controlled within the range of 30–50 nm to ensure electron transparency suitable for TEM analysis. TEM characterization was performed using a Titan Themis 200 microscope operated at 200 kV, located at Uppsala University. During imaging, the lamella was aligned such that the zone axis of the sapphire substrate was oriented parallel to the electron beam direction to optimize crystallographic contrast and resolution.

B. First-principles calculations

The amorphous systems were simulated *ab initio* following the stochastic quenching procedure [9] in conjunction with density functional theory [26,27] (DFT) calculations with a generalized gradient approximation (GGA) correlation-functional of Perdew, Burke, and Ernzerhof [28].

For the stochastic quenching method, the atomic coordinates of the initial $16 \text{ \AA} \times 16 \text{ \AA} \times 16 \text{ \AA}$ super cells were randomly distributed under the constraint that no pair of atoms were placed within one averaged atomic radii distance to each other until a given atomic density (see Table I) was reached. The ionic positions were thereafter iteratively relaxed with the Vienna *Ab initio* Simulation Package (VASP) [29,30] using the conjugate gradient algorithm until an energy and atomic force convergence criterion of 10^{-6} eV and 20 meV/Å, respectively, were collectively reached. The calculations were performed with an energy cutoff of 350 eV and a Γ k -point sampling, which proved to be sufficient when tested for energy self-consistency due to the large size of the supercell. The elastic constants determined from uniform deformations in x , y , and z directions, whence the elastic constants were extracted from the linear stress vs strain relation.

TABLE I. Computed mass densities in comparison to the experimental values derived from RBS and XRR results, and values of thickness and roughness parameters from fitting a slab model to the XRR data. All samples had an approximately 15 Å thick layer of Zr between the ZrO capping and the sample, except the sample with 85% vanadium.

	ρ_m [g/cm ³] (DFT)	ρ_m [g/cm ³] (RBS/XRR)	d [Å]	σ_{sub} [Å]	σ_{surf} [Å]
V ₈₅ Zr ₁₅	6.2	6.06 ± 0.12	1208	5	0
V ₈₀ Zr ₂₀	6.2	6.19 ± 0.12	1217	9	7
V ₆₇ Zr ₃₃	6.3	6.14 ± 0.12	1231	9	8
V ₅₀ Zr ₅₀	6.3	6.37 ± 0.13	1229	4	14
V ₃₃ Zr ₆₇	6.4	6.44 ± 0.13	1253	9	8
V ₂₅ Zr ₇₅	6.5	6.43 ± 0.13	1251	8	6

To avoid strain or artificial clustering, the relaxed equilibrium densities and bulk modulus were determined via a fitting procedure of the Birch-Murnaghan equation of state to determine the average of several configurations as a function of the supercell volume [31].

The collective and partial pair distribution functions were determined by forming radial histograms of relative atomic separations, averaged over several independently quenched

amorphous structures for different compositions at the optimal density.

To characterize the local atomic environments in the stochastic-quenched VASP structures, we analyzed each relaxed snapshot with `pyscal3` using a Voronoi-based neighbor definition under periodic boundary conditions [32]. For every atom, we computed the full Voronoi index and identified the most frequent motifs across the ensemble. Using the same Voronoi neighbor network, we also evaluated the Steinhardt bond-order parameters q_4 and q_6 together with their neighbor-averaged counterparts $\bar{q}_4$ and $\bar{q}_6$, and assembled species-resolved distributions for V- and Zr-centered environments. No tiny-face or minimum-area filtering was applied before constructing the Voronoi indices, so the reported motif diversity reflects the full unfiltered Voronoi tessellation of the quenched amorphous structures.

III. RESULTS

A. Pair distribution functions

First, GIXRD with small step size was performed to search for sharp Bragg peaks, which were found to be absent over the entire accessible angular range. Subsequently, a full pair distribution function was determined using the technique of Bylin, Kapaklis and Pálsson [25] as shown in Fig. 1. The

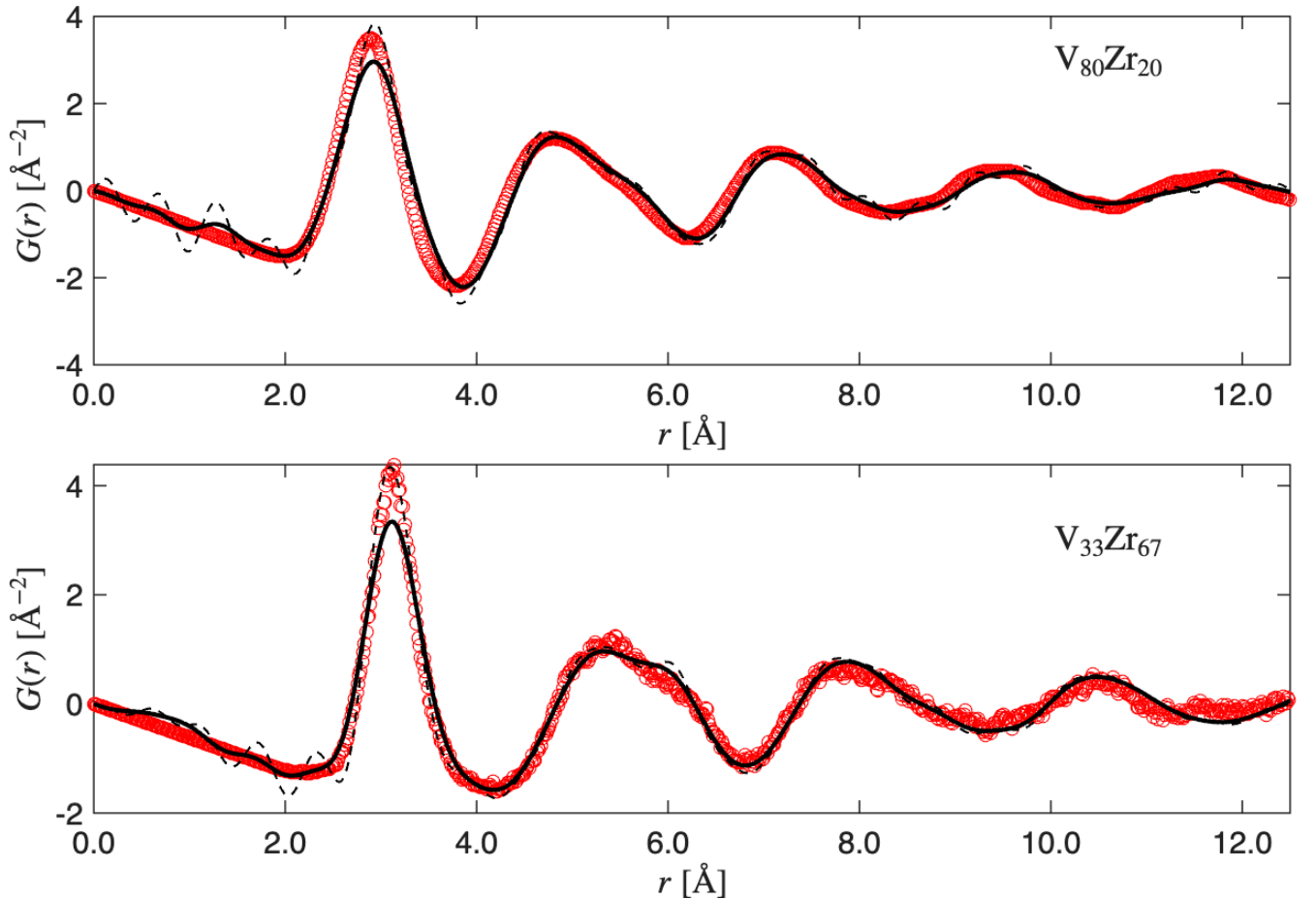


FIG. 1. Experimental (black solid and dashed) and stochastic quenching (red) for V₈₀Zr₂₀ (top) and V₃₃Zr₆₇ (bottom). The experimental pdfs are shown with and without a Lanczos filter (dashed line) applied to truncate termination ripples. The filter suppresses artificial ripples but broadens the peaks.

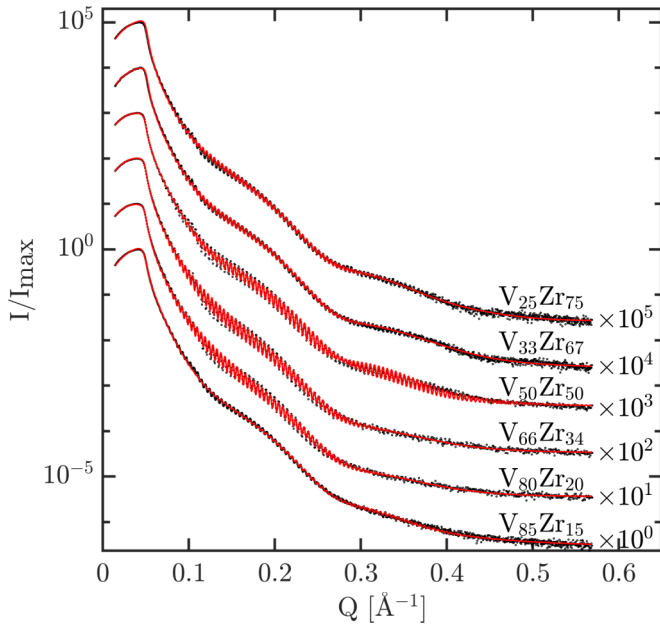


FIG. 2. X-ray reflectivity pattern of amorphous V_xZr_{100-x} . The red solid line is the result of fitting a two slab model to the data. The samples exhibit low roughness and a high density, indicating that the sample is close packed. The curves have been scaled for clarity by the factor shown in the figure.

measured intensity from the Al_2O_3 substrate can be considered negligible because the choice of incidence angle and stage rotation completely evades the Bragg reflections of the Al_2O_3 crystal throughout the complete $2\theta \in [5^\circ, 150^\circ]$ scan. The diffraction intensity of the noncrystalline protective top layer can also be neglected as the illuminated scattering volume of the V_xZr_{100-x} layer is about 20 times larger, and the scattering probability scales as the square of the atomic form factors. The figure shows the experimental pair correlation function of $V_{80}Zr_{20}$, which correspond to an inverse Fourier transform of the static structure factor either with (black solid line) or without (black dashed line) a Lanczos filter applied. The Lanczos filter dampens oscillations associated with the abrupt termination of the structure factor in reciprocal space, but broadens the peaks. By not applying the filter, one obtains a higher amplitude of the first peak but suffers artificial ripples as a consequence. In particular, the weak shoulder near $r \approx 6 \text{ \AA}$ in the unfiltered dashed curve is strongly suppressed after Lanczos filtering, indicating that it is most likely a termination artifact from finite $Q_{\max}$ rather than a robust structural signature. Moreover, the filtered second-shell feature in the 4–6 $\text{\AA}$ range remains clearly asymmetric rather than broad and rounded, consistent with a disordered glassy structure. The stochastically quenched counterpart is shown in red and with no adjustable parameters, fitting, or scaling, we conclude that the agreement is excellent.

B. X-ray reflectometry

Figure 2 shows the x-ray reflectivity of the V_xZr_{100-x} glass for the different compositions used in this study. The interference oscillations (Kiessig fringes) arise from the x-ray interference from the film/substrate interface and the top sur-

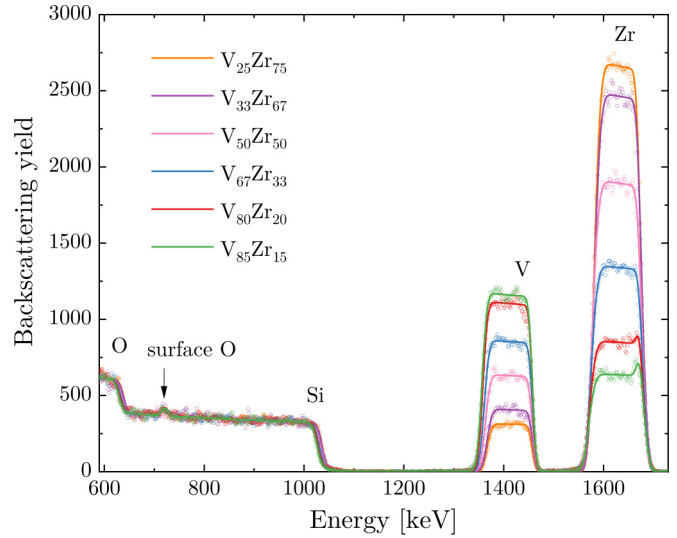


FIG. 3. RBS spectra acquired using 2 MeV ^4He ions backscattered from amorphous V_xZr_{100-x} films. The number of incident particles is normalized across all measurement to 6.8×10^{10} particles per steradian ($\cdot \text{sr}$). The symbols represent the experimental data and solid lines the best fit using the SIMNRA code [20].

face, and carry information about the thickness of the layer, the roughness of the interfaces, and the density. The further out in reciprocal space these can be observed, the flatter and more well defined the two interfaces are. As can be seen in the figure and as confirmed by the model, the roughness is less than 11 $\text{\AA}$. Considering that the substrate roughness is specified as $<5 \text{ \AA}$ by the manufacturer and films being more than 120 nm thick promoting roughness buildup, the film flatness approaches the atomic level. The results from the fits are shown in Table I.

C. Rutherford backscattering spectrometry

Figure 3 shows the RBS spectra of the V_xZr_{100-x} samples. Deviations from the intended stoichiometry are below 1%, whereas the RBS spectra reveal oxygen only at the surface (ZrO), while the V_xZr_{100-x} films exhibit no measurable signal, implying very low oxygen content. Using SIMNRA, the areal densities of each constituent are extracted. By combining the areal densities with film thicknesses determined from x-ray reflectivity, the volumetric densities are obtained. Figure 4 presents the resulting atomic density (in atoms/cm^3), which are in excellent agreement with the theoretical estimates from amorphous stochastically quenched configurations. Both trends have extrapolated values below their respective crystalline counterpart, providing a measure of consistency to the approach. The films are only marginally less compact than optimally placed atoms in a bcc crystal assuming lattice constants of $a = 3.03 \text{ \AA}$ for α -V (bcc), $a = 3.58 \text{ \AA}$ (bcc) for β -Zr, and $a = 3.23 \text{ \AA}$, $c = 5.15 \text{ \AA}$ for α -Zr (hcp). Table I summarizes both the theoretical and experimental mass densities (g/cm^3) of the maximally amorphous stochastically quenched configurations and the experimental values. We see that the density changes systematically from 6.2–6.5 g/cm^3 as the amount of Zr is increased.

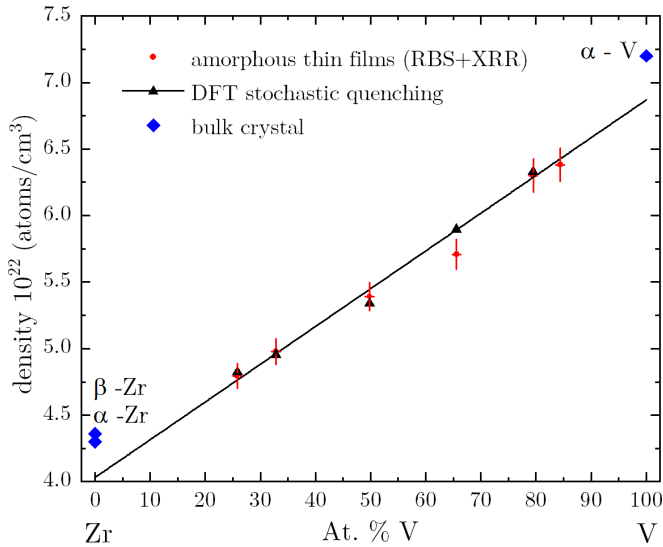


FIG. 4. Atomic density (in atoms/cm³) of amorphous V_xZr_{100-x} films as a function of vanadium content. Experimental values, obtained from RBS-derived areal densities and x-ray reflectivity thickness measurements, are compared to theoretical stochastically quenched amorphous configurations.

D. Transmission electron microscopy

High-resolution transmission electron microscopy (HRTEM) was employed to investigate the local atomic structure of the synthesized material. As shown in Fig. 5(a), the HRTEM image reveals a complete absence of long-range periodic features typically associated with crystalline order. No lattice fringes or nanocrystalline domains are observed, even upon close inspection across multiple regions of the sample. In the high-resolution TEM image, Fig. 5(b), of the $Al_2O_3/V_{80}Zr_{20}$ interface, a bright band with apparent crystalline features is observed within the VZr layer just above the interface. As shown in Supplemental Material (Fig. S1) [33], this contrast does not originate from real lattice ordering in the V-Zr but from image delocalization, where phase contrast from the crystalline Al_2O_3 substrate is displaced

into the adjacent amorphous region by lens aberrations and defocus. To reduce ambiguity from phase-contrast delocalization in HRTEM at the interface, we additionally examined the same region using HAADF-STEM (Fig. S2 in the Supplemental Material) [33], which shows homogeneous contrast in the V-Zr layer and no indications of local ordering. The diffuse FFT signature (described in the next section) of V-Zr confirms its amorphous nature, and the crystalline-like contrast should therefore be regarded as an imaging artifact rather than evidence of structural ordering. This observation strongly suggests a fully amorphous structure.

The corresponding fast Fourier transform (FFT) in Fig. 5(c) of the HRTEM image region shown in Fig. 5(b), exhibits a broad and diffused ring without any sharp spots or sharp concentric rings. This is a characteristic feature of amorphous materials, indicating that the material lacks any significant translational symmetry or periodicity on the nanometer scale. To further quantify this, a radial intensity profile of the FFT was obtained by azimuthally averaging the FFT image, shown in the inset of Fig. 5(c). The profile displays a single broad peak, and no secondary peaks were detected. This diffuse and featureless profile is qualitatively consistent with the pair distribution function as obtained by GIXRD in Fig. 1. In fact, the position of the first maxima is in full harmony with the reciprocal value obtained from x-ray scattering. On its own, transmission microscopy does not offer a unique indicator of a maximally amorphous structure. However, taken together, the absence of lattice fringes in the HRTEM image, combined with the diffused halo in reciprocal space and the lack of sharp features in the FFT profile, confirm that the material is fully amorphous as seen by both a local probe (HRTEM, HAADF-STEM) and a probe that averages over the entire sample (GIXRD).

E. Elastic constants and stability

The elastic constants (see Table II) calculated from small deformations of the stochastically quenched amorphous V-Zr alloys provide key insight into their mechanical stability and structural integrity. Despite the absence of long-range order,

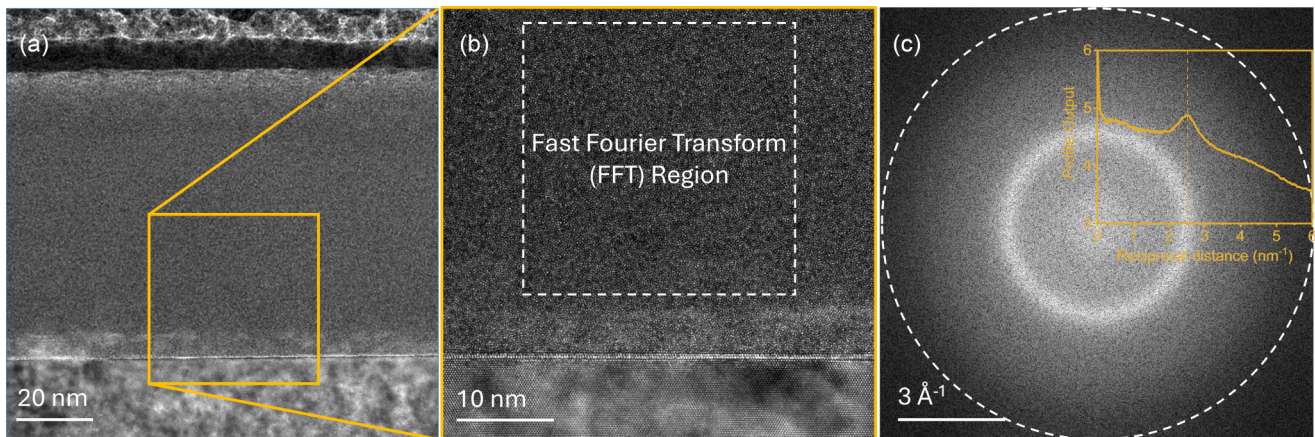


FIG. 5. Cross-section TEM image of the $V_{80}Zr_{20}$ film on Al_2O_3 substrate (a), high-resolution TEM image at the film-substrate interface showing the Al_2O_3 crystalline substrate and the amorphous $V_{80}Zr_{20}$ film (b), and a fast Fourier transform (FFT) image of a film's region with the inset showing a radial intensity profile of the FFT (c).

TABLE II. Elastic constants from stochastically quenched structures (GPa). Right block: engineering constants—bulk B , Young's Y , shear G (all GPa), and Poisson's ratio μ (dimensionless). Uncertainties are standard deviations from isotropic averaging.

	C_{11}	C_{12}	C_{44}	B	Y	G	μ
$V_{80}Zr_{20}$	178(3)	122(2)	28(2)	141(1)	78(4)	28(1)	0.408(5)
$V_{67}Zr_{33}$	159(1)	113(5)	23(3)	128(4)	65(7)	23(2)	0.41(1)
$V_{50}Zr_{50}$	137(2)	96(1)	20(2)	110(1)	57(3)	20(1)	0.414(5)
$V_{33}Zr_{67}$	123(4)	85(5)	19(3)	97(4)	55(8)	19(2)	0.41(2)
$V_{25}Zr_{75}$	122(3)	82(2)	20(1)	95(2)	55(3)	19.8(8)	0.403(6)

the isotropic moduli derived from the second-rank stiffness tensor—namely the bulk modulus (B), shear modulus (G), and Young's modulus (Y)—demonstrate that all compositions are mechanically stable, residing in well-defined local minima of the potential energy landscape. Notably, both B and G remain positive across the compositional series, satisfying the fundamental thermodynamic criteria for resistance to uniform compression and shear deformation, respectively. The high Poisson's ratios ($\mu \approx 0.41$) and large B/G (Pugh's) ratios ranging from 4.8–5.6 are well above the classic ductile/brittle heuristic of 1.75 indicating a characteristically ductile response. This is typical of densely packed amorphous metals, and suggests the presence of short-range order that supports shear without shear band localization.

Beyond confirming mechanical stability, these elastic constants offer a proxy for assessing the degree of amorphicity in the structure. The elastic constants show an overall reduction with increasing Zr content, with the decrease becoming less pronounced and approaching a plateau at high Zr concentration (notably between $V_{33}Zr_{67}$ and $V_{25}Zr_{75}$ within uncertainty). This behavior remains consistent with systematic changes in atomic packing density and local bonding environments, in line with the measured densities and surface roughness. Furthermore, the consistency of the elastic moduli across independently quenched structures indicates that the amorphous state is not only energetically metastable but also dynamically robust, without soft vibrational modes or mechanical instabilities that would signal crystallization.

IV. DISCUSSION

The dense, uniformly amorphous character of the V-Zr films can be understood as the outcome of a kinetically dominated growth window in which ultraclean, low-pressure sputtering and energetic arrival of species promote atomic mixing and densification while simultaneously suppressing nucleation and long-range ordering. At the low homologous temperature of room-temperature deposition, the thermal diffusion length is negligible, whereas crystallization requires either substantial surface mobility or time for nuclei to form. Instead, a high arrival flux of V relative to Zr, combined with very low Ar pressure, produces ballistic transport of sputtered atoms and enhanced ion/neutral energies at the substrate that drive athermal mixing. This locks atoms into a chemically homogeneous, glasslike structure before they can reorder into crystals. This picture is consistent with Thornton's structure-zone framework [34], where reducing working pressure and

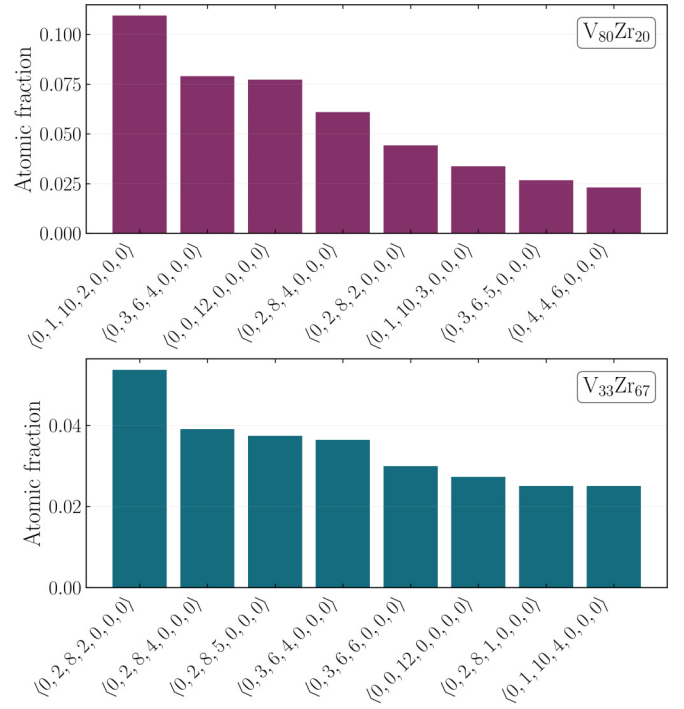


FIG. 6. Comparison of the dominant Voronoi motifs in independently quenched V-Zr structures. The Voronoi vectors span a broad range of amorphous motifs, including the icosahedral and some composition-induced changes. No evidence of crystal motif domination is observed.

increasing particle energy shift films toward densification even at low substrate temperature. Thus, for V-Zr the conjunction of low Ar pressure, ultrahigh vacuum, and an energetic plasma environment provides the necessary athermal mobility and sufficient atomic mixing to yield a maximally amorphous film: dense, chemically uniform, and devoid of crystallites.

To gain further insight into the atomistic nature of the quenched structures, we have performed Voronoi analysis as shown in Fig. 6 and bond-order statistics in Fig. 7. The Voronoi analysis resolves the amorphous structure into its dominant local coordination polyhedra and shows that both alloys ($V_{80}Zr_{20}$, $V_{33}Zr_{67}$) are governed by motifs rich in pentagonal faces, including $(0, 1, 10, 2)$, $(0, 2, 8, 2)$, and the icosahedral $(0, 0, 12, 0)$ class. These are recurring motifs found in other transition-metallic glasses [35,36]. This is consistent with the fact that the glass is a densely packed amorphous network built from fivefold-symmetry-rich clusters that are incompatible with periodic crystal tiling. At the same time, the motif spectrum remains distributed over several related polyhedra rather than one unique environment, which points to substantial local topological diversity. In compositional terms, $V_{80}Zr_{20}$ appears to show a stronger preference for a few dominant icosahedral or distorted-icosahedral motifs, whereas $V_{33}Zr_{67}$ exhibits a broader motif population and thus a more heterogeneous short-range topology. This interpretation is broadly consistent with the picture of Holmström and Wallace of a “maximally amorphous” structure: within the single-random-valley framework, the relevant amorphous states lack long-range order but may still retain

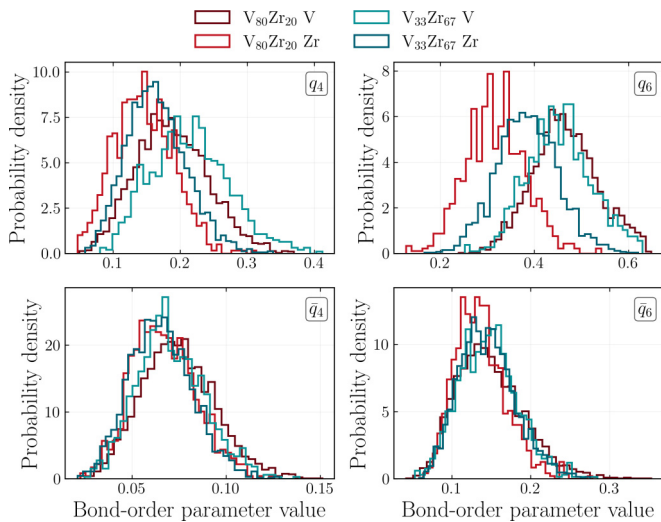


FIG. 7. Species-resolved distributions of the bond-order parameters q_4 , q_6 , $\bar{q}_4$, and $\bar{q}_6$ for V- and Zr-centered environments in amorphous $V_{80}Zr_{20}$ and $V_{33}Zr_{67}$. The broad distributions confirm the absence of crystalline order, while the systematic shift of the V-centered environments toward higher q_6 relative to Zr indicates stronger short-range orientational ordering around V atoms, with the substantial overlap in $\bar{q}_4$ and $\bar{q}_6$ suggesting more similar medium-range order across species and compositions.

well-defined short- and medium-range order, so amorphous character does not preclude the presence of recurring local structural motifs.

The bond-order analysis shown in Fig. 7 supports this picture by showing broad, mostly unimodal distributions of q_4 , q_6 , $\bar{q}_4$, and $\bar{q}_6$, which is the expected signature of a translationally disordered glass containing a range of local environments rather than narrow crystalline fingerprints. The largest separation occurs in q_6 , implying that the two compositions differ mainly in the nature or abundance of short-range orientational order, while the stronger overlap in the averaged parameters suggests that their medium-range organization is more similar. These results indicate that the amorphous phase is built from related families of fivefold-rich local motifs whose relative populations shift with composition, rather than from nanocrystals or sharply distinct competing phases. The data are consistent with an amorphous state that is characterized by local motif statistics and broad orientational-order distributions, not by emergent long-range periodicity.

V. CONCLUSIONS

We have provided experimental evidence in terms of XRR, RBS, GIXRD, HRTEM, and HAADF-STEM, that magnetron-sputtered metallic glasses of V_xZr_{100-x} exhibit structural signatures of maximally amorphous materials. This interpretation is substantiated by the close agreement with the physical properties predicted by DFT-based stochastic quenching simulations. XRR reveals well-defined thicknesses and interface roughness approaching the atomic limit. Combined with RBS, the experimental densities are in excellent agreement with the compact, maximally amorphous structures produced by the stochastic quenching method. Moreover,

experimentally obtained pair distribution functions are close to identical with the maximally amorphous counterpart. HRTEM/FFT together with complementary HAADF-STEM provide a local validation of amorphicity and interfacial homogeneity. Together, these observations confirm that magnetron sputtering, when operated under optimized conditions, can reproduce structural signatures from the atomic scale to the microscale of the disordered atomic topologies generated by rapid quenching simulations.

Furthermore, the elastic tensor becomes not merely a measure of mechanical resilience, but a meaningful descriptor of configurational saturation—supporting the notion that these films approach the theoretical limit of “maximally amorphous” materials as envisioned in the random valley approximation. These structures, and more generally the approach proposed, can serve as much needed benchmarks for other types of disordered materials. These specific structures have been shown to harbor hydrogen concentrations up to 1.6 hydrogen per metal atom without structural degradation [37], and are found to be thermally stable up to 900 K even upon hydrogen incorporation [38], highlighting the mechanical and thermal stability of such compact amorphous structures. Furthermore, owing to their large hydrogen capacity and lack of crystalline structure, these films have been used to determine stopping powers of ^{15}N ions for H, V, and Zr improving the accuracy of hydrogen detection via resonant nuclear reaction analysis [39].

The experimental films and theoretical structures presented in this study offer a valuable platform for future investigations into the properties and applications of amorphous materials. The films, characterized by their high density, compositional uniformity, and structural isotropy, can serve as reference systems for probing transport phenomena, corrosion resistance, and hydrogen-metal interactions in the absence of crystalline defects. Likewise, the stochastic quenching configurations, validated against experimental data, provide a reproducible and physically grounded model of the maximally amorphous state. These structures can be used to benchmark machine-learned interatomic potentials, calibrate spectroscopic interpretations of amorphous matter, or initialize molecular dynamics simulations that require statistically relaxed glassy configurations. Together, the theory-experiment correspondence established here provides a foundation for systematic exploration of structure-property relationships in disordered alloys and may accelerate the design of functional metallic glasses for energy, coating, and electronic applications.

A scoring system could be developed based on the likeness of the experimental pair distribution function and the mass density to maximally amorphous structures. The metric is inherently tied to the chosen DFT implementation, but that dependence can be leveraged: by documenting and reusing the same setup, the score enables consistent comparisons, method tuning, and transparent progress over time.

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DATA AVAILABILITY

There are no publicly available research data or software supporting this manuscript. Requests for further information or data should be sent to the authors.

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