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Prediction of Spontaneous Plastic Deformation of Irradiated Metallic Glasses due to Thermal Spike-Induced Plasticity

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Using large-scale molecular dynamics simulations, we predict that knock-on damage in irradiated metallic glasses yields spontaneous anisotropic deformation, i.e. shape change in the absence of externally applied loads. The root cause of this behavior is anisotropic plastic deformation around non-spherical thermal spikes. Such thermal spike-induced plasticity (TSIP) does not depend on electronic excitations and is distinct from the ‘ion hammer’ effect. Macroscale TSIP is predicted to occur under unidirectional heavy ion and neutron irradiation. The consequences of TSIP for potential applications of metallic glasses under irradiation are discussed.

Keywords: Metallic Glasses, Radiation Effects, Molecular Dynamics

Metallic glasses have attracted attention due to their good mechanical properties,[1] corrosion resistance,[2] and formability.[3] They also exhibit markedly different responses to radiation than crystalline solids. For example, crystalline alloys swell without limit under continuous irradiation,[4] whereas amorphous materials may expand or contract following irradiation.[5] Irradiated crystalline alloys embrittle [6] while some irradiated amorphous alloys may become more ductile.[7–9]

We performed large-scale molecular dynamics (MD) simulations to better understand the behavior of metallic glasses under irradiation. Our investigation leads us to predict an unexpected new radiation response mechanism of metallic glasses: spontaneous plastic deformation induced by inter-nuclear scattering. This phenomenon is distinct from the well-known ‘ion hammer’ effect [10] and suggests that some amorphous metals are unsuitable for use as structural materials under neutron irradiation, e.g. in certain nuclear reactor designs.

High-energy neutrons create energetic primary knock-on atoms (PKAs) in solids, which subsequently dissipate their kinetic energy in a cascade of secondary collisions before coming to rest.[11] This process occurs over lengths of nanometers (nm) and times of picoseconds (ps).[12] No experimental methods are currently capable of such spatiotemporal resolution. However, MD

simulations are ideally suited to these length and time scales.

We investigate the effect of neutron irradiation on amorphous metals by simulating 475 keV niobium (Nb) PKAs passing through $\sim 1/2$ billion-atom models of an amorphous alloy of copper (Cu) and Nb (*a*-CuNb). The PKA energy is at the upper end of the range where inter-nuclear—rather than electronic—scattering dominates its dissipation to the environment in metals.[13] Thus, we carry out our simulation using classical inter-atomic potentials.[14] Models with differing preparation histories and compositions ranging from 25% Cu to 75% Cu were investigated. The outcomes of these simulations were qualitatively similar. Therefore, we focus on results from a 475 keV PKA in *a*-Cu₅₀Nb₅₀ synthesized by rapid quenching from the liquid.

Simulations are conducted in LAMMPS [15] using an embedded atom method potential with Ziegler–Biersack–Littmark short-range interactions.[14] The *a*-Cu₅₀Nb₅₀ model is constructed by quenching a 474,353,318-atom, 4,000 K Cu₅₀Nb₅₀ liquid at 10^{13} K/s to 300 K in 25 K decrements using a velocity rescaling thermostat, constant number of particles (N), pressure (P), and enthalpy (H) Nosé-Hoover barostat, and periodic boundary conditions. Model properties match those of a previous study on a $\sim 50,000$ atom *a*-Cu₅₀Nb₅₀

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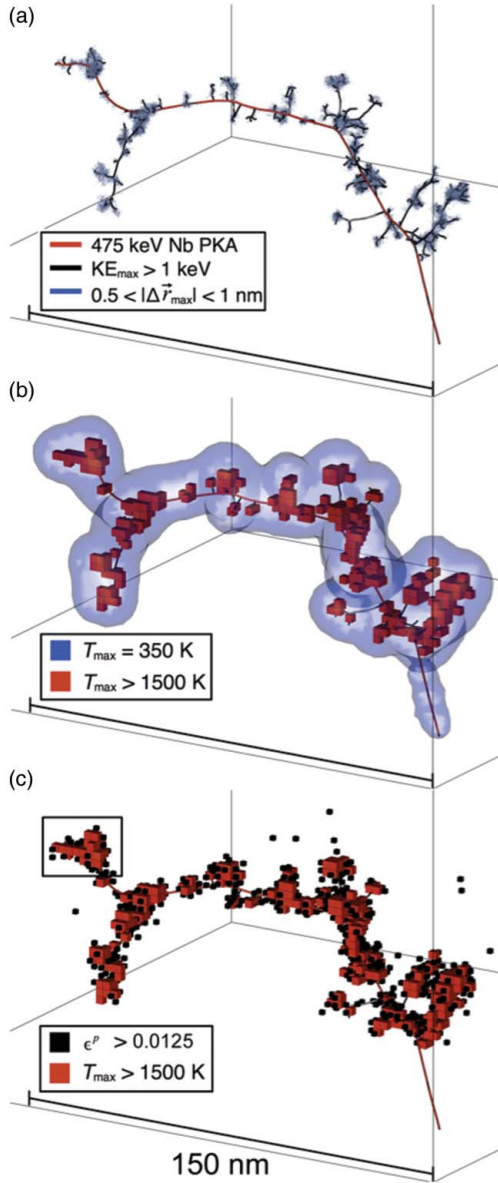


Figure 1. 475 keV Nb PKA passing through a $\sim 1/2$ billion-atom model of $a\text{-Cu}_{50}\text{Nb}_{50}$. (a) Trajectory of PKA plotted in red, >1 keV KAs in black, and atoms displaced by 0.5–1 nm in blue. (b) Temperature fields due to inter-nuclear collisions. Red voxels have $T_{\max} > T_G = 1,500$ K, [16] blue contour is for $T_{\max} = 350$ K. (c) TS-induced plastic deformation. Black voxels have $\epsilon^p > 0.0125$ at $t = 12$ ps. The boxed TS is analyzed in detail in Figures 2 and 3.

system. [16] During the collision cascade, constant number of particles (N), volume (V), and total energy (E) ensemble equations of motion are integrated using adaptive time steps to ensure energy conservation. The simulation cell size is greater than the projected range of 475 keV Nb ions in $a\text{-Cu}_{50}\text{Nb}_{50}$ computed using a version of SRIM [17] modified to exclude electronic stopping. [18]

Figure 1(a) shows a 475 keV Nb PKA collision cascade in $a\text{-Cu}_{50}\text{Nb}_{50}$. The PKA (red trajectory) loses more than 90% of its energy through collisions that

transfer more than 1 keV, yielding higher order knock-on atoms (KAs, black trajectories) that terminate in spatially disconnected regions with numerous displacements of 1 nm or less (blue vectors). Rectilinear, collision-free KA trajectories connect these zones, demonstrating that inter-nuclear scattering does not generate ion tracks. Over distances of hundreds of nanometers, the distribution of PKA-induced collisions in Figure 1(a) is comparable to those in crystalline alloys. [19]

To characterize radiation response at lower length scales, we analyze our simulations on a grid of cubic volumes (voxels) with an edge length of 2.6 nm. These voxels are large enough to be representative volume elements of the material. We compute densities, potential energies, and stresses in each voxel by averaging per-atom quantities over voxel constituent atoms; [20] voxel temperatures by fitting the kinetic energy distribution of voxel atoms to the Maxwell–Boltzmann distribution, [21] with temperature as the fitting parameter; time-dependent voxel diffusivities at time t from a linear fit to voxel mean-squared displacement [$\text{MSD}(t)$] in 2 ps time intervals centered at t ; and Lagrangian total strains from the best-fit uniform deformation gradient between an initial ($t = 0$ ps) and final voxel atom configuration. [22] Plastic strains are computed by subtracting voxel elastic strain ϵ_{ij}^e from total strain, with $\epsilon_{ij}^e = S_{ijkl}\sigma_{kl} + \alpha\Delta T\delta_{ij}$, where σ_{kl} is the voxel stress, S_{ijkl} is the compliance tensor, α is thermal expansion, and ΔT is the voxel temperature change. Our model is elastically isotropic with $\lambda = 140.8$ GPa, $\mu = 50.1$ GPa, and $\alpha \approx 9 \times 10^{-6}$ 1/K.

We find that the kinetic energy deposited in regions of dense atomic displacements causes the temperature there to rise rapidly. In some voxels, shown as red cubes in Figure 1(b), the maximum temperature $T_{\max}$ rises above the glass transition temperature of $a\text{-Cu}_{50}\text{Nb}_{50}$ ($T_G = 1500$ K [16]) within ~ 1 –2 ps and then quenches below T_G at rates approaching 10^{14} K/s. Consistent with previous studies, [23,24] structural analysis reveals no crystallization: all voxels return to an amorphous, vitrified state, although the rapid quenching reduces the short-range icosahedral order initially present. [16]

Within voxels where $T_{\max} > T_G$, the temperature dependence of density and diffusivity matches that of uniform liquid Cu–Nb, demonstrating that these voxels are not merely superheated, but rather contain genuine Cu–Nb liquid, albeit only for times of ~ 10 ps. Connected voxels with $T_{\max} > T_G$ form spatially isolated ‘thermal spikes’ (TSs) approximately 2–12 nm in diameter. TSs are extensively studied in crystalline solids due to their role in the production of radiation-induced point defects. [12,19,25,26] However, TSs have a distinctly different effect on atomic structure evolution in amorphous metals. [27–29]

By computing voxel-level strains, we find that the material immediately adjacent to TSs undergoes permanent plastic deformation. The tensile work-equivalent

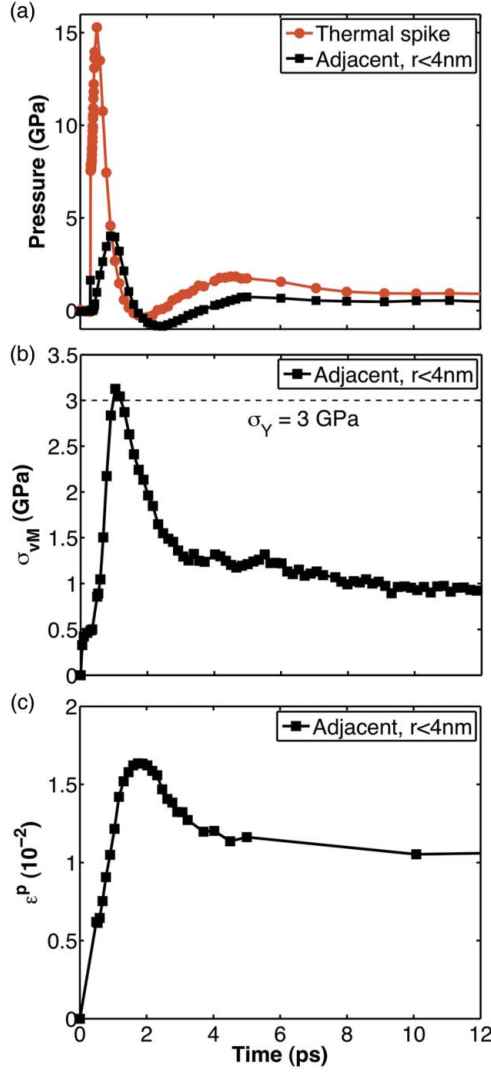


Figure 2. Stresses and strains inside and adjacent to the TS boxed in Figure 1(c). (a) Pressure versus time in the TS and in adjacent material. Von Mises stress (b) and tensile work-equivalent plastic strain (c) versus time, averaged over all voxels within 4 nm of the TS.

plastic strain ϵ^P of black voxels in Figure 1(c) exceeds 1.25%. This plasticity originates with TSs. When the material inside a TS melts, its zero-pressure density decreases to that of an equilibrium liquid. However, the liquid is constrained by the surrounding unmelted material, causing the pressure inside the TS to rise sharply. As in crystalline solids,[19,25] this pressure pulse initiates a stress wave that propagates away from the TS at the longitudinal speed of sound (see supplementary material). Unlike in crystalline solids, close to the TS, the deviatoric component of the stress wave exceeds the yield stress and initiates plastic flow.

We illustrate this process on the TS boxed in Figure 1(c). Figure 2(a) shows that the pressure of the liquid inside the TS shoots up to >15 GPa and drops back to near zero within ~ 2 ps. A pressure pulse of ~ 4 GPa

is seen with a ~ 0.5 ps delay in a 4 nm-thick shell of solid immediately adjacent to the TS. By comparing with the analytical solution for stresses around a transiently pressurized inclusion in an isotropic material,[30] we confirmed that the pressure pulse in the liquid initiates the succeeding pulse in the solid. Figure 2(b) shows that the deviatoric stress in the material adjacent to the TS exceeds the yield stress, computed in a uniform α -Cu₅₀Nb₅₀ model at the corresponding temperature and strain rate. Concurrently, the deviatoric plastic strain rises to just under 2% and falls back to a steady value of $\sim 1\%$ (likely due to elastic back stresses), as shown in Figure 2(c).

If TSs in irradiated metallic glasses were spherical, then the local plasticity in their vicinity would occur equally in all directions, resulting in no net shape change of the material. However, the TSs in our simulations are typically prolate ellipsoids with major axes along the direction of the KA that initiates them, e.g. the TS boxed in Figure 1(c) and shown close up in Figure 3(a). Under the influence of a high internal pressure, these prolate ellipsoids deform to become more spherical, as illustrated in Figure 3(b), i.e. they expand perpendicular to their major axis and contract parallel to it. Therefore, unlike spherical TSs, plasticity around prolate ellipsoid TSs is not equal in all directions and leads to a net anisotropic strain in the material.

In a cylindrical coordinate system aligned with the TS major axis, the deformation illustrated in Figure 3(b) may be described by a tensile hoop strain $\epsilon_{\theta\theta}$ and compressive radial and axial strains, ϵ_{rr} and ϵ_{zz} , in the material adjacent to the TS. We compute these strains for the TS in Figure 3(a) and plot them as a function of position along its major axis in Figure 3(c), confirming that $\epsilon_{\theta\theta} > 0$ and $\epsilon_{rr}, \epsilon_{zz} < 0$.

We repeat this analysis for the seven largest TSs in our simulation and find that they are also prolate ellipsoids with major axes along KA directions and with cylindrically symmetric plastic strain distributions, similar to those in Figure 3(c). The voxel average plastic strains for each TS, shown in Figure 4(a), satisfy $\bar{\epsilon}_{rr} \leq \bar{\epsilon}_{zz} < \bar{\epsilon}_{\theta\theta}$ and are nearly independent of TS energy. We attribute this fact to the linear dependence of TS volume on deposited knock-on energy, shown in Figure 4(b). The average TS volume per unit knock-on energy is nearly constant at $\langle V_{TS}/E_{TS} \rangle = 14.2 \pm 0.5 \text{ nm}^3/\text{keV}$, giving rise to a narrow distribution of maximum pressures inside TSs (average of 13 GPa) and a similarly narrow distribution of maximum stresses in the adjacent solid material (average of 2.5 GPa).

To enable comparisons of these modeling insights with experiments, we calculate the net plastic strain per unit fluence due to all TSs in our simulation. The voxels that undergo plastic deformation are adjacent to TSs and have maximum temperatures that fall in the range $350 < T_{\text{max}} < 1500$ K. We identify these voxels, average the plastic strains in them, and scale the result by the fraction

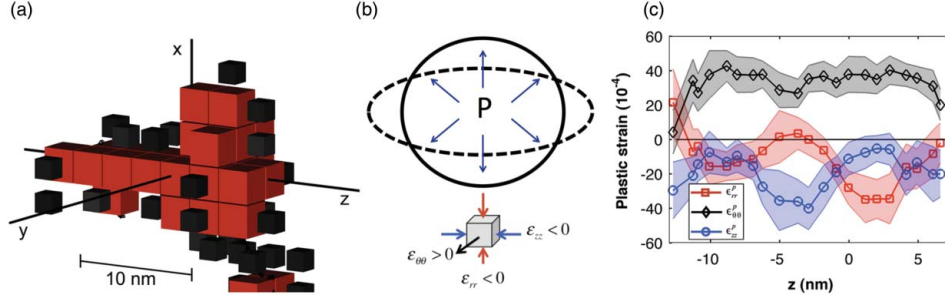


Figure 3. (a) Close-up view of the TS boxed in Figure 1(c) with a cylindrical coordinate system defined along the TS major (z) axis and voxels with $\epsilon^p > 0.0125$ in black. (b) An initially ellipsoidal TS (dashed line) deforms into a more spherical shape (solid line) when internally pressurized, yielding the illustrated plastic strains in surrounding material. (c) Diagonal components of plastic strain computed at $t = 12$ ps in cylindrical coordinates as a function of position along the major axis of the TS in (a). Shaded bands indicate uncertainties.

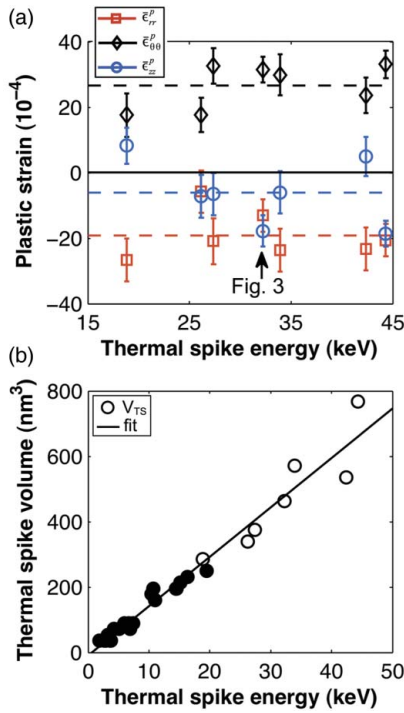


Figure 4. (a) Voxel average plastic strain components in material adjacent to the seven largest TSs as a function of the TS energy. The arrow indicates the TS analyzed in Figures 2 and 3. (b) TS volume V_{TS} versus deposited knock-on energy E_{TS} . Linear fit: $V_{TS} = (15.1 \pm 0.6 \text{ nm}^3/\text{keV})E_{TS} - (8.9 \pm 11.2 \text{ nm}^3)$. Open symbols for TSs analyzed in (a).

of the volume of the model that these voxels occupy ($f = 1.12\%$). We find that the PKA causes compressive plastic flow along the cascade direction ($\epsilon_{\text{casc}} = -2.8 \times 10^{-6}$), tensile flow in one lateral direction ($\epsilon_{l1} = 4.4 \times 10^{-6}$), and compressive flow in the other ($\epsilon_{l2} = -0.8 \times 10^{-6}$). In a material subjected to numerous collision cascades, the lateral strains are expected to average to a single value of net expansion in the lateral directions. The deformation includes a hydrostatic component $\epsilon_H^p = 0.2 \times 10^{-6}$, which is small compared with the tensile work-equivalent

strain $\epsilon^p = 4.3 \times 10^{-6}$ and consistent with shear-induced dilatation.[1]

Thus, we predict that TS-induced plasticity leads to a net contraction along the incident ion beam direction and expansion perpendicular to it. Barring any recovery or devitrification that may occur over longer times, the radiation-induced plastic strain per fluence along the beam direction is $A = \epsilon_{\text{casc}}/\phi \approx -10^{-15} \text{ cm}^2/\text{PKA}$, where $\phi = 1 \text{ PKA/SA} \approx 2.6 \times 10^9 \text{ PKA/cm}^2$ is the effective fluence in our simulation (SA is the model cross-sectional area). To predict the magnitude of TS-induced plasticity under irradiation by a specific high-energy particle, X , A must be further scaled to account for the mean free path between PKAs, λ_{PKA} , and the mean energy of PKAs, E_{PKA} : $A_X = A \times (196 \text{ nm}/\lambda_{\text{PKA}}) \times (E_{\text{PKA}}/475 \text{ keV})$, where 196 nm is the length of our model and 475 keV the PKA energy in our simulation.

As an example, we employ the strain per fluence from MD to compute the radiation-induced plastic strains expected under neutron irradiation. Using a hard-sphere neutron scattering cross section of $\sim 3b$ (representative of transition metals [31]), the measured fast neutron energy spectrum at the High Flux Isotope Reactor at Oak Ridge National Laboratory,[32] and a neutron energy range of 0.1–20 MeV, we calculate [11] $\lambda_{\text{PKA}} \approx 5.3 \text{ cm}$ and $E_{\text{PKA}} \approx 94 \text{ keV}$ (see supplementary material), yielding a neutron-induced strain per fluence of $A_n \approx -7.3 \times 10^{-22} \text{ cm}^2/\text{n}$. Thus, for unidirectional neutron beams with fluences of order 10^{20} n/cm^2 , we predict strains of $\sim 7\%$.

Our findings suggest that TS-induced plasticity (TSIP) will give rise to spontaneous plastic deformation of amorphous metals under neutron irradiation. TSIP is distinct from the ‘ion hammer’ effect,[10] wherein irradiation of amorphous solids with swift, heavy ions, e.g. 360 MeV Xe ion irradiation, also leads to plasticity. Ion hammering is due to viscoelastic relaxation induced by electronic excitations in continuous ion tracks.[33,34] It is not expected under fission neutron irradiation because maximum PKA energies for neutrons are below the

threshold for ion track formation. Whereas ion hammering is due to intense ionization tracks, TSIP arises from plastic deformation around isolated TSs initiated by inter-nuclear collisions and therefore is to be generally expected in amorphous metals under neutron irradiation. A fraction of ion-induced plasticity hitherto attributed to ion hammering may in fact also be due to TSIP, especially under low energy ion bombardment.[35]

We have shown that non-uniform, anisotropic plastic deformation around non-spherical TSs may produce pronounced anisotropic shape changes in metallic glasses irradiated with unidirectional ion or neutron beams. In thermal fission reactors, where the directions of transmutation-induced recoils are close to isotropic, the superposition of all recoil-induced strains is expected to cancel. However, anisotropic neutron fluxes may arise in fast neutron or fusion reactors. TSIP may limit the utility of metallic glasses as structural materials in such cases, despite their other, attractive properties.[7–9,36] TSIP may also enable new applications of amorphous materials in radiation responsive devices, e.g. in sensors where plastic strain is used to monitor dose or in radiation-driven actuation. Moreover, under controlled, unidirectional ion beams, TSIP may provide novel routes for radiation-assisted processing of metallic glasses, such as contactless forming-at-a-distance of amorphous components.

Supplementary online material In the supplementary material, we discuss the velocity of stress pulses emitted by TSs and demonstrate how to relate the thermal spike induced plasticity (TSIP) seen in MD to strains expected under experimental conditions. Supplementary material is available at <http://dx.doi.org/10.1080/21663831.2014.916760>.

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