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Crossover from stochastic activation to cooperative motions of shear transformation zones in metallic glasses

W. Jiao,[1] B. A. Sun,[1] P. Wen,[1] H. Y. Bai,[1] Q. P. Kong,[2] and W. H. Wang[1,a)]
[1]*Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*
[2]*Institute of Solid State Physics, Chinese Academy of Sciences, Hefei 230031, China*

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We observe that the stress relaxation of metallic glasses below glass transition temperatures presents a universal double power-law decay behavior, indicating that relaxation dynamics transforms from a fast mode to a slow mode. This is attributed to a crossover from the stochastic activation to the self-organized cooperative motion of localized shear transformation zones. The phenomenon is further confirmed via strain recovery experiments and illustrated by a stochastic model. The results demonstrate that the plastic deformation exhibits the hallmarks of critical phenomenon, and offer a picture on the onset of deformation and evolution of relaxations in metallic glasses. © *2013 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4819393]

Full understanding of the relaxation dynamics of glasses and supercooled liquids remains one of the major unsolved problems in condensed matter physics.[1–9] There are two types of relaxation process: the fast mode which persists from supercooled liquid to glassy state and the slow mode frozen below the glass transition temperature (T_g) or in glassy state.[1,4] Despite extensive studies, the physical origin of the relaxation modes is not yet completely understood.[4,8] Due to the sluggish relaxation rate in glassy solid, the information of the relaxation dynamics in glassy state obtained from the experiments is quite limited, and only the fast relaxation mode has been detected in glassy state via dynamic mechanical spectroscopy.[10–12] The latest advancement in glassy polymers shows that the molecular mobility is accelerated up to a factor of 1000 under uniaxial tensile creep[13,14] indicating that the relaxation dynamics can be altered by stress. The questions are then raised: Can the slow relaxation mode appear in the glassy state? If it can happen, when and how does it vary with temperature and time?

The relaxation process is found to correlate with the deformations in glasses,[3,8] and the revelation helps to understand the puzzling issue of the plastic deformation mechanism in metallic glasses (MGs).[15–17] Recently, the relaxation dynamics of MGs is investigated by X-ray photon correlation spectroscopy,[18] inelastic neutron scattering,[19] and nuclear magnetic resonance[20] under static state. The dynamic mechanical spectroscopy was also carried out on relaxation in MGs but only under infinitesimally weak perturbations (~0.02%).[10,12] Up to now, there is little experimental work and systematic description on relaxation dynamics in glassy state under a strong external drive and the concomitant inelastic deformation. The process of activation and evolution of the deformation units, which is crucial for understanding of the deformation mechanism and relaxations in glasses, remain unclear yet.

In this letter, we investigate the stress decay at a constant strain within the nominal elastic region and the followed strain recovery after unloading the applied strain. The stress relaxation dynamics is found to transform from a fast mode associated with the stochastic activation of the localized shear transformation processes to a slow mode related with the cooperative motion of these deformation units. The relaxations show the robust power-law relation which gives direct evidence that the onset of the plastic deformation is a critical phenomenon. Our results offer a clear picture on the onset of the permanent deformation and evolution of relaxation dynamics in glass.

The tensile stress relaxation experiment and following strain recovery measurements were performed on Dynamic mechanical analyzer (DMA) model Q800 obtained from TA Instruments. We applied a constant strain of 0.7% (in the apparent elastic region of the MG) and to record the stress decay (stress relaxation) with the loading time. Figure 1 shows the representative stress relaxation experiment of $Zr_{52.5}Ti_5Cu_{17.9}Ni_{14.6}Al_{10}$ (vit105) at 540 K with the loading time of 75 000 s. One can see the stress decrease with the loading time. To study the characteristics of the stress relaxation, we unloaded the applied strain, and to record the residual strain recovery behavior subsequent to the stress relaxation processes. The process is called strain recovery and marked in shadow region in Fig. 1. The strain recovery experiment is carried out on various samples which have previously been stress relaxed at the strain of 0.7% for different times (say, 120 s and 75 000 s).

The representatively scaled stress relaxation data of vit105 at 540 K and strain of 0.7% in the whole regime cannot be fitted by the Kohlrausch–Williams–Watts (KWW) function [see Fig. 2(a)], and the KWW function can only fit the short time regime (<~300 s). At long time regime, the variation of scaled stress with time is well fitted by the power-law function. The delayed appearance of the relaxation in the power-law form implies that it is frozen in the glassy state at the beginning of the relaxation process, and only operates after sufficient gestation time. The result indicates that there are two relaxation modes in the glass under loading state: the fast relaxation mode in the stretched exponential form, and the slow relaxation mode which decays in the power-law fashion. It is more obvious via analyzing the differential of scaled stress against time in the log-log

a)Author to whom correspondence should be addressed. Electronic mail: whw@aphy.iphy.ac.cn

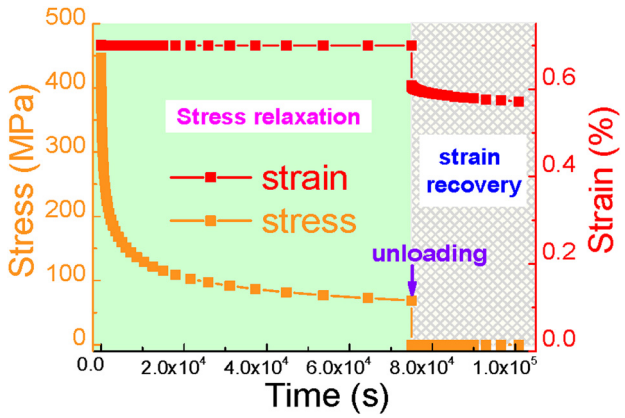

FIG. 1. A representative experiment curve of stress relaxation and strain recovery of Vit105 at 540 K. After the stress relaxation process lasts for some time at the strain of 0.70%, the applied loading is removed (marked with arrow). The stress (or strain) recovers after the unloading the applied strain (marked in shadow region).

fashion as shown in Fig. 2(b). We can see that the stress response can be divided into two distinct power-law decays. The index of the former part associated with the fast relaxation is $n = 0.471 \pm 0.05$, and index of the latter corresponding to the slow relaxation is $m = 1.19 \pm 0.11$. A clear crossover (at 277 s) determined based on the intersection of two power-law fitting marks the boundary of the two different stress relaxations, which has also been observed in the relaxation of dielectric materials.[21] The double power-law decay of the stress is also found in other MGs suggesting that the behavior is a universal phenomenon.

To characterize the relaxation features of the two relaxation modes, we analyze the strain recovery behavior subsequent to the stress relaxation processes. The representative strain recovery after the operation of stress relaxation for 120 s, which is ahead of the appearance of crossover is shown in Fig. 3(a). The residual strain is almost fully recovered after the applied load is removed indicating that the glass only deforms an elastically in the fast relaxation process, and the recovered strain upon time can be fitted by the KWW function. In elastic regime of MG, it is widely accepted that the individually microscopic deformation units are activated, and the localized shear transformation events are fully recoverable.[15] The stretched exponential fashion of the recovery behavior indicates that the deformation units are not uniform in MGs, and the detailed distribution of deformation units in size and energy barrier can be obtained via stress relaxation,[22] creep experiments,[23] and the molecular dynamical simulation.[24–26] All these imply that the observed fast relaxation corresponds to the stochastic activation of deformation units, and thus leads to the anelasticity in MGs.

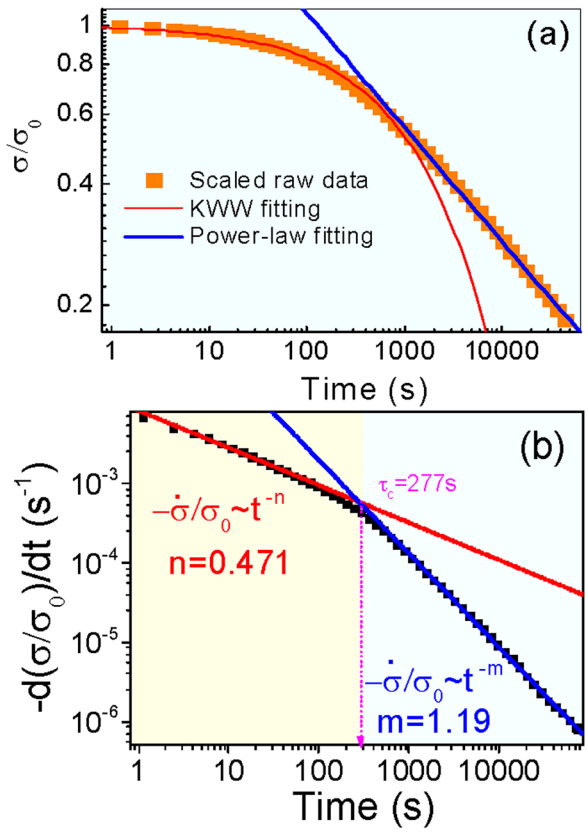

FIG. 2. (a) The plot of scaled stress against time can only be fitted by the KWW function in short time region and the power law function at long time scale. (b) The log-log plot of scaled stress reduction rate against time. The curves show two parts of distinct power-law decay, corresponding to the fast and slow relaxation mode individually. The power law index n and m equal to 0.471 ± 0.05 and 1.19 ± 0.11, respectively. A crossover with crossover time τ_c can be clearly seen.

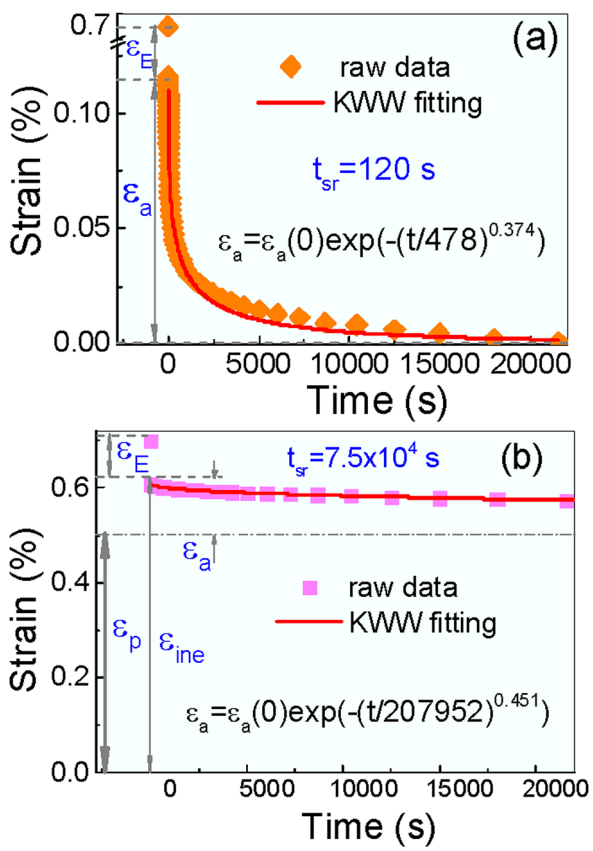

FIG. 3. (a) The strain recovery process after the stress relaxation process lasts for 120 s (before the appearance of crossover, and to see the recovery process of fast relaxation.) The recovered strain ε_a fitted by the KWW function with the residual strain ε_p approaching to zero, indicating the strain is almost recoverable and the glass only deforms an elastically in the fast relaxation process. (b) The strain recovery process after the stress relaxation process lasts for 75 000 s (after the appearance of crossover, to see the recovery process of slow relaxation). The ε_a fitted by the KWW function with ε_p approaching to 0.494%, indicating the appearance of permanent deformation.

At long time scales, the visco-elasticity or permanent deformation appears and only partial applied strain recovers as exhibited in Fig. 3(b). For an applied strain of 0.7% lasting for 75 000 s, the residual strain ε_p is up to 0.572% after recovery for 26 000 s. Obviously, this represents irreversible atomic rearrangements and permanent deformation associated with the operation of slow relaxation mode. During the stress relaxation, the applied elastic strain is converted to inelastic strain.[26] The appearance of permanent deformation, concomitant with the slow relaxation mode, indicates that the localized deformation units self-organize and operate in a cooperative manner. The power law associated with the slow relaxation mode in temporal scale is due to the cooperative organization of the deformation units. The fractal geometry feature of the local atomic arrangements in spatial scale via simulation further supports the intensive interaction of deformation units in the plastic flow region.[6,25]

The crossover and the two power law decays of stress response can be further substantiated by a stochastic model. The macroscopic inelastic deformation is essentially an accumulation of local strains generated via the operation of deformation units.[15] According to the constitutive relations, the local inelastic strain produced by deformation units and treated as viscous liquid[15] satisfies: $d\varepsilon_{ine}/dt = \sigma/\eta$, therefore $\sigma = \sigma_0 \exp(-t/\tau_D) = \sigma_0 \exp(-bt)$. In the extreme case, as all surrounding deformation units have already been shear transformed in the loading direction and cannot find any pathway to further accommodate the applied strain, the investigated deformation units is constrained by the mechanical compatibility. Hence, for each deformation unit to further relax its local stress, the cooperation with the surrounding deformation units becomes the only way.[27,28] Thus, the probability that the ith deformation unit has not transformed until time t is

$$\Pr(\theta_i \geq t | \beta_i = b,\ \max(\eta_1, ..., \eta_{i-1}, \eta_{i+1} ..., \eta_N) = s) = \begin{Bmatrix} \exp(-bt) & t<s \\ \exp(-bs) & t \geq s \end{Bmatrix}, \quad (1)$$

where s is the maximum relaxation time of all the surrounding clusters, θ_i, η_i, and β_i refer the time needed for changing the orientation, structure renormalization, and the transformation rate of the ith deformation units, respectively.[29] Hence, the scaled stress relaxation function is

$$\frac{\sigma(t)}{\sigma_0} = \phi(t) = \lim_{N\to\infty} \Pr(A_N \min(\theta_1, \ldots, \theta_N) \geq t). \quad (2)$$

According to the Levy-stable law, through a series of ratiocination, one can get[30]

$$\phi(t) = \exp\left[-\frac{1}{\kappa}\int_0^{\kappa(Bt)^\alpha}\left[1-\exp\left(-\frac{1}{s}\right)\right]ds\right], \quad (3)$$

the resultant parameter α $(0<\alpha<1)$ refers to the stability index of the distribution function of β_i, k $(k>0)$ denotes the speed of the structural reorganization of deformation units, $B = 1/\tau_M$, τ_M is the most possible time needed for transforming orientation of deformation units. The response function for the short- and long-time limits are obtained[30]

$$f(t) = -\frac{d\phi(t)}{dt} \approx \begin{cases} C_1(Bt)^{-n} & \mathrm{Bt} \ll 1 (\text{short time limit}) \\ C_2(Bt)^{-m} & \mathrm{Bt} \gg 1 (\text{long time limit}) \end{cases}, \quad (4)$$

where $n = 1-\alpha$, $m = 1+\alpha/k$, C_1 and C_2 are constants. The result of the stochastic model is in line with the experimental result. Based on the obtained crossover time and the experimental power law indexes, the parameters of Eq. (3) are obtained: $\alpha = 1-n = 0.529 \pm 0.05$, $k = \alpha/(m-1) = 2.116 \pm 0.31$, $B = 1/\tau_c = 0.0025 \pm 0.0018$. The resultant relaxation function of the stochastic model shown in Fig. 4 is well consistent with the experimental one. This further indicates the physical picture of the stochastic model conforms well to the microscopic process of the stress relaxations: In the initial period, the deformation units, occurs stochastically and is fully recoverable after removing the strain. When $k \to 0$ (indicating the interaction among deformation units can be ignored), Eq. (3) takes the form of the KWW function of $\phi(t) = \exp[-(Bt)^\alpha]$.[30] This explains why the fast relaxation mode can be well described by the KWW function. After the crossover time, the deformation units move cooperatively to further relax the local stress and result in permanent deformation. Computer simulations in MGs[24] also show that most activated configurations are localized in the initial state, whereas, at long time scales, the deformation units exhibit displacement cascades that percolate through the sample associated with plastic deformation.

Our observed two-step relaxations of MG clearly show that the slow mode preserved in the glassy state could be activated at long time scale with the assistance of stress. The underlying reason might be that the atom mobility is accelerated under deformation, similar to the molecular behavior in polymer glasses under stress.[13] The crossover of the relaxation dynamics associates with a transition from a weak to a strong coupling of the localized shear transformation process. The resultant power-law-distributed phenomenon is in strong resemblance to the scaling behavior near a critical

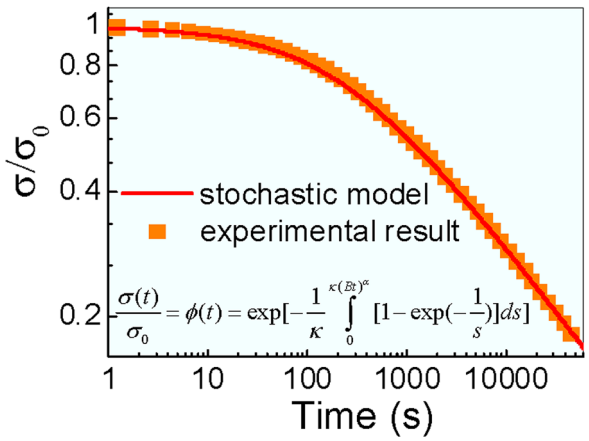

FIG. 4. The plot of the relaxation function of the stochastic model well consistent with the experimental result, where the parameters (α, k, B) are obtained from two forms of power-law decay.

point, reflecting the existence of an underlying nonequilibrium critical points in the plastic flow process.[26,31] The independent activated deformation units in the initial state self-organize in a highly cooperative fashion near the critical point,[32] and results in the formation of shear band or the global plastic flow. The critical phenomenon indicates that not only the power-law relation appears temporally but also in spatial scale where the fractal geometry feature of the local atomic arrangements might appear. Indeed, the fractal feature is obtained via recent simulation in the macroscopic plastic flow,[26] and the pattern of shear band and the fracture surface morphology also presents the fractal nature which indirectly illustrate that the deformation units can evolve into a self-organized critical point.[33,34]

In summary, we experimentally show a crossover of relaxation dynamics from fast mode associated with stochastic localized shear transformation process to slow mode associated with the cooperative motion of STZs in metallic glasses. Our results demonstrate that the onset of plastic deformation exhibits the hallmarks of a critical phenomenon which can be well illustrated by the proposed stochastic model. The results give a clear picture on the onset and evolution of plastic flow and relaxations in metallic glasses.

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