

## Predicting Complex Relaxation Processes in Metallic Glass

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(Received 17 April 2019; published 6 September 2019)

Relaxation processes significantly influence the properties of glass materials. However, understanding their specific origins is difficult; even more challenging is to forecast them theoretically. In this study, using microseconds molecular dynamics simulations together with an accurate many-body interaction potential, we predict that an  $\text{Al}_{90}\text{Sm}_{10}$  metallic glass would have complex relaxation behaviors: In addition to the main ( $\alpha$ ) relaxation, the glass (i) shows a pronounced secondary ( $\beta$ ) relaxation at cryogenic temperatures and (ii) exhibits an anomalous relaxation process ( $\alpha_2$ ) accompanying  $\alpha$  relaxation. Both of the predictions are verified by experiments. Computational simulations reveal the microscopic origins of relaxation processes: while the pronounced  $\beta$  relaxation is attributed to the abundance of stringlike cooperative atomic rearrangements, the anomalous  $\alpha_2$  process is found to correlate with the decoupling of the faster motions of Al with slower Sm atoms. The combination of simulations and experiments represents a first glimpse of what may become a predictive routine and integral step for glass physics.

DOI: [10.1103/PhysRevLett.123.105701](https://doi.org/10.1103/PhysRevLett.123.105701)

Compared with crystals, glasses inherently feature diverse relaxation dynamics over a wide range of temperature and timescales. These relaxation processes significantly influence properties of glass materials and are related to a number of crucial unresolved issues in glassy physics [1–5]. Understanding how the atomic rearrangements govern these processes represents an outstanding issue in glass physics [1,3,4,6,7].

Usually, the most prominent relaxation process is the primary ( $\alpha$ ) relaxation, which is responsible for the vitrification of glass-forming liquid. Its falling out of equilibrium indicates the glass transition phenomenon. Processes occurring in addition to the  $\alpha$  relaxation at shorter timescales or lower temperature are referred to as secondary ( $\beta$ ) relaxations. Studies over the past few decades have established that the  $\beta$  relaxation could have important consequences on the mechanical properties for metallic and polymeric glasses, as well as thermal stability of glassy pharmaceuticals and biomaterials, and thus attract considerable attention [8–13]. Moreover, recent studies have discovered there might be more relaxation processes in addition to the  $\alpha$  and  $\beta$  relaxations in glasses [7,14–21]. Even the structurally simplest metallic glasses (MGs) (compared to molecular and polymeric glasses) could exhibit multiple relaxations, indicating a far richer than expected scenario for glass relaxation. For example, Wang *et al.* [17] and Kuchemann and Maass [15] identified a new relaxation process that is faster than the  $\beta$  relaxation in MGs, which was named  $\beta'$  and  $\gamma$  relaxations. Wang *et al.* [22] also illustrate that the  $\beta'$  relaxation might be correlated

with the initiation of plastic deformation in MGs. Luo *et al.* [16] reported that the nonequilibrium  $\alpha$  relaxation would split into two processes in a deep glassy state, which causes early decay in the stress-relaxation experiments [23]. While these results illustrate that MGs possess complex relaxation phenomena and have consequences on properties, it is difficult to understand their specific origins because of the lack of microscopic data of these processes.

In principle, molecular dynamics (MD) simulation is a powerful tool to investigate detailed atomic rearrangements in the glass relaxations at the microscopic level. However, the relaxations in the glassy states are extremely complicated, as they are sensitive to chemical compositions [24] and thermal histories [25,26]. The related relaxation timescales (e.g., milliseconds to seconds) are usually several decades longer than the current available computational timescales (picoseconds to nanoseconds). It is therefore challenging to model the relaxation dynamics of realistic glass materials under experimental conditions. Moreover, for large-scale MD simulations of MGs, the force field which describes the many-body interactions (i.e., empirical potentials) is of vital importance [27–29]. Although there are a few potentials that can reproduce some static structural features and thermodynamics of MGs, their capability of describing dynamical processes is mostly unknown.

Recently, a realistic interaction potential for the study of  $\text{Al}_{90}\text{Sm}_{10}$  MG was developed [30]. It correctly describes the glass structure [31], complex devitrification behaviors [32,33], and crystal growth [34] in the Al-Sm systems.

It also brings insight to the competition of crystallization and glass formation [35,36]. Therefore, it provides a model system to investigate the relaxation mechanism in the realistic MG.

In this work, relying on this accurate interaction potential, we simulate the dynamical mechanical spectroscopy (DMS) of  $\text{Al}_{90}\text{Sm}_{10}$  MG in the timescale up to  $10 \mu\text{s}$ , which almost reaches the limit of state-of-the-art computational power. With such a slow frequency, we find that an anomalous relaxation process (noted as  $\alpha_2$ ) decouples from  $\alpha$  relaxation. The MG also exhibits a strongly pronounced  $\beta$  relaxation even on the MD timescale. The behaviors predicted from the MD simulations are verified with DMS experiments at cryogenic temperatures. The detailed atomic motions that lead to the relaxation processes are revealed from the MD trajectories. The feasibility that atomic simulations could discover new relaxation processes in MGs and elucidate their underlying mechanisms would be useful for understanding the dynamics and properties as well as the design of glass materials.

*Simulation.*—MD simulations were carried out based on a Finnis-Sinclair potential [30], using the GPU (graphics processing units)-accelerated LAMMPS code [37–39]. The  $\text{Al}_{90}\text{Sm}_{10}$  glass model, containing 32 000 atoms, was obtained by the continuous cooling with a cooling rate of  $10^8 \text{ K/s}$ . The MD simulations of DMS [40] were performed during the cooling process, covering a wide temperature range from deeply undercooled liquid to low-temperature glass. In MD DMS, a sinusoidal strain was applied with an oscillation period  $t_\omega$  (related to frequency  $f = 1/t_\omega$ ) and a strain amplitude  $\epsilon_A$ . The resulting stress  $\sigma(t)$  and phase difference  $\delta$  between stress and strain were measured and fitted by  $\sigma(t) = \sigma_0 + \sigma_A \sin(2\pi t/t_\omega + \delta)$ . The storage and loss moduli were calculated by  $E' = \sigma_A/\epsilon_A \cos(\delta)$  and  $E'' = \sigma_A/\epsilon_A \sin(\delta)$ , respectively. A strain amplitude  $\epsilon_A = 0.6\%$  was applied in all MD DMS, which ensured deformations in the linear response regime.

*Experiments.*—The  $\text{Al}_{90}\text{Sm}_{10}$  MG was prepared by spinning-quenching techniques (see Supplemental Material [41] for details). The relaxation dynamics of the MG was studied by a dynamical mechanical analyzer using liquid nitrogen for temperature control, which allows us to reach the cryogenic temperature (down to 150 K). The measurements were conducted during a temperature ramping of 3 K/min together with a film tension oscillation using the discrete testing frequencies of 0.5, 2, 4, 8, and 16 Hz. The storage ( $E'$ ) and loss ( $E''$ ) moduli were recorded for subsequent analysis and compared with MD DMS results.

*Predictions by simulations.*—Figure 1 shows MD DMS results by plotting the storage ( $E'$ ) and loss moduli ( $E''$ ) as a function of temperature under different oscillation periods. The temperature-dependent loss modulus curves are fitted with a series of Gaussian peaks which correspond to different relaxation processes. With the longest oscillation period  $t_\omega = 1 \mu\text{s}$  in Fig. 1, the loss modulus exhibits a

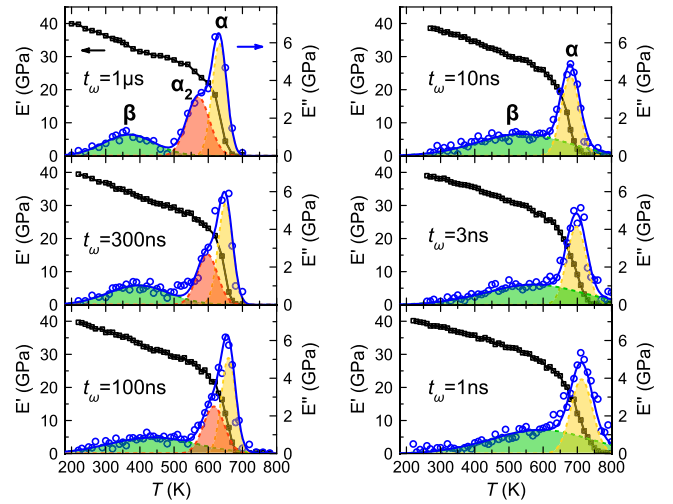


FIG. 1. Relaxation behaviors by simulations. Storage  $E'$  and loss  $E''$  moduli of  $\text{Al}_{90}\text{Sm}_{10}$  MG at different oscillation periods  $t_\omega$  ranging from  $1 \mu\text{s}$  to 1 ns. The loss moduli are fitted by multiple Gaussian functions.

broad peak in the temperature range from 200 to 500 K, which corresponds to the typical  $\beta$  relaxation. We note that there exists no such pronounced peak of  $\beta$  relaxation in any previous MD simulations where only shoulderlike or excess wings were observed. At higher temperature, the dominant primary ( $\alpha$ ) relaxation peak shows a strong shoulder in the temperature range 500–600 K, which needs an extra peak function for the fitting. Considering that this process decreases in amplitude as frequency increases (or  $t_\omega$  decreases), which is consistent with behavior of  $\alpha$  relaxation in general, this new peak is named an  $\alpha_2$  process.

As shown in Fig. 1, with the decrease of oscillation period, all three peaks,  $\alpha$ ,  $\alpha_2$ , and  $\beta$ , shift towards higher temperature. The  $\alpha_2$  peak gradually collapses in the  $\alpha$  peak so that one can hardly differentiate them when  $t_\omega$  is smaller than 10 ns. With a very short oscillation period (e.g.,  $t_\omega = 1 \text{ ns}$ ), the  $\beta$  peak also largely overlaps with the  $\alpha$  peak. Thus, the simulations predict a complex relaxation scenario in the  $\text{Al}_{90}\text{Sm}_{10}$  MG: at suitably long timescales (e.g.,  $1 \mu\text{s}$ ) it has a pronounced  $\beta$  peak and an anomalous  $\alpha_2$  process in addition to the  $\alpha$  relaxation.

*Experimental verification.*—We next validate these computational predictions by DMS experiments for the as-quenched  $\text{Al}_{90}\text{Sm}_{10}$  samples. Even though our longest simulation period reaches  $1 \mu\text{s}$ , it is still about 5–6 orders of magnitude faster than the typical DMS experiments in which probing timescales are 0.1–10 s. Extrapolating the temperature-time relation for the  $\beta$  relaxation from simulations to the experimental timescale leads to a characteristic temperature about 200 K [inset of Fig. 2(a)].

Figure 2(a) shows the experimental  $E''(T)$  for the MG from a cryogenic DMS measurement at the testing frequency of 8 Hz. Remarkably, it clearly shows a pronounced  $\beta$  relaxation peak at about 220 K, consistent with the

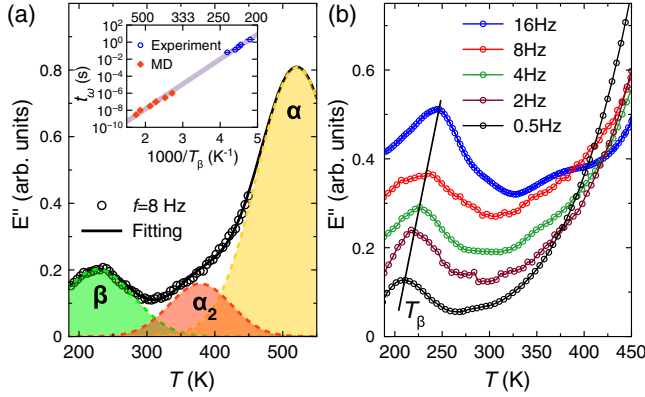


FIG. 2. Experimental DMS for  $\text{Al}_{90}\text{Sm}_{10}$  MG. (a) DMS measured at the frequency 8 Hz and (b) at different frequencies as indicated. The inset in (a) shows the Arrhenius fitting of  $\beta$  relaxation peaks and DMS periods from both simulations and experiments. The fitting of  $\alpha$  relaxation peak is to guide the eye. It is based on the extrapolation of simulation data at the experimental temperature; see details in Supplemental Material [41].

extrapolation of MD simulations. Figure 2(b) shows  $E''(T)$  for more different testing frequencies. The peak temperature of the  $\beta$  relaxation increases with higher frequency (or shorter time period), which also quantitatively agrees with the MD simulations, as shown in the inset of Fig. 2(a).

Additionally, a close examination of the  $E''(T)$  curve in Fig. 2(a) indicates that there is notable excess contribution to the  $\alpha$  relaxation around  $T = 380$  K, which corresponds well with the  $\alpha_2$  relaxation found from MD DMS (Fig. 1). This  $\alpha_2$  process can also be discerned from different testing frequencies in Fig. 2(b). For example, it is more evident on the curve with the frequency of 16 Hz. Because of close coupling with  $\alpha$  relaxation, the  $\alpha_2$  process is more difficult to resolve than the pronounced  $\beta$  relaxation. Nevertheless, its time-temperature relation can still be determined and compared favorably with MD simulations (see Supplemental Material [41]). Therefore, the presence of the  $\alpha_2$  process in the MG could be ascertained with the guidance of MD simulation. Unfortunately, because of the occurrence of a strong devitrification process in the current MG system [32], one cannot fully access the  $\alpha$  relaxation peak at experimental timescales, resulting in the termination of experimental data at 450 K.

**Mechanism for  $\beta$  peak.**—The above experiments validate the predictions from MD simulations. Now we are in position to investigate the mechanisms of these complex relaxation processes. Recently, the structural rearrangements governing the  $\beta$  relaxations have been investigated in a model  $\text{Ni}_{80}\text{P}_{20}$  MG [45] and a  $\text{Y}_{65}\text{Cu}_{35}$  MG [46], which suggests stringlike motions might be the origin of  $\beta$  relaxation. However, these MGs do not show such clear  $\beta$  relaxation peak as the  $\text{Al}_{90}\text{Sm}_{10}$  at MD accessible timescales. One feature about stringlike motions is that a particle jumps to a position that was occupied previously by another particle [45]. Structurally, this would result in a multippeak curve for the distribution  $p(u)$  of atomic

displacements  $u$  during the deformation period, which is clearly observed in the present MG, as shown in Fig. 3(a) (the mathematical definitions of displacement  $u$  and stringlike motion are provided in Supplemental Material [41]). The fact that the second and third peaks of  $p(u)$  match exactly the first and second peaks of the pair distribution function  $g(r)$  at various  $\beta$  relaxation peaks in Fig. 3(a) indicates that atoms prefer to jump to the position that is previously taken by another atom in its nearest or secondary neighbors, which further evidences stringlike motions. Figure 3(b) shows that the strings can propagate in a rather large spatial range and form different types of geometries such as aggregations, loops and long-range chains. The string size  $\xi$  is defined by the number of atoms involved in the string. Figure 3(c) shows that the probability of the atoms forming a string follows an exponential function with the string size that can span up to 70 atoms. While similar stringlike motions were also observed in Lennard-Jones liquid model [47] and other systems [48,49], the string size in the current  $\text{Al}_{90}\text{Sm}_{10}$  is much longer than other systems. For example, the longest reported string in  $\text{Ni}_{80}\text{P}_{20}$  contains 12 atoms [46], which is smaller by a factor of 6 than current MG. Such long-range and large-scale stringlike motion is a unique feature of the present MG,

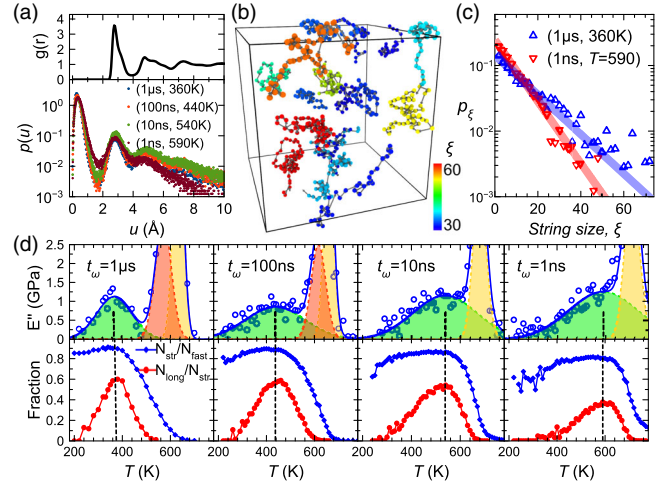


FIG. 3. Stringlike motion and  $\beta$  relaxation. (a) Upper panel shows the pair correlation function of  $\text{Al}_{90}\text{Sm}_{10}$  MG at 300 K from MD simulation, while lower panel shows the probability of atomic displacement  $u$  at the condition of  $(t_\omega, T_\beta)$ , where  $T_\beta$  is the peak temperature of  $\beta$  relaxation with the oscillation period  $t_\omega$ . (b) String configurations at  $T_\beta = 360$  K with  $t_\omega = 1 \mu\text{s}$ . The atoms in the strings with size  $\xi < 30$  are removed for clarity. (c) The probability of stringlike-moving atoms involved in the different sizes of strings at  $T_\beta$  with  $t_\omega = 1$  ns and  $1 \mu\text{s}$ , respectively. The lines indicate exponential fitting. (d) Relationship between  $\beta$  relaxation and string motion under four different oscillation periods from  $1 \mu\text{s}$  to 1 ns. The lower panel shows the fraction of stringlike-moving atoms in the fast-moving atoms ( $N_{\text{str}}/N_{\text{fast}}$ , blue) and the fraction of atoms in the long stringlike motions in the total stringlike-moving atoms ( $N_{\text{long}}/N_{\text{str}}$ , red).

which could be the reason for the uniquely pronounced  $\beta$  peak.

Figure 3(d) quantifies the fraction of stringlike-moving atoms to the total number of fast-moving atoms ( $N_{\text{str}}/N_{\text{fast}}$ ), as well as the fraction of atoms involved in the long-string motions to the total number of stringlike-moving atoms ( $N_{\text{long}}/N_{\text{str}}$ ) as a function of temperature. The long-string motion is defined by  $\xi \geq 10$  here. The  $T$ -dependent loss moduli  $E''$  are also plotted for comparison. One can see that the peak of  $N_{\text{long}}/N_{\text{str}}$  well matches the peak of the  $\beta$  relaxation over all the studied oscillation periods  $t_\omega$ . While the curve of  $N_{\text{str}}/N_{\text{fast}}$  reaches a maximum plateau at the peak temperature of the  $\beta$  relaxation  $T_\beta$ ,  $N_{\text{long}}/N_{\text{str}}$  manifests as a pronounced peak, whose position and width quantitatively agree with those of  $\beta$  relaxation peaks. Note that this correlation does not change with the choice of the long-string motion threshold (see Supplemental Material [41]). These results suggest that long-string motions contribute more to the  $\beta$  relaxation than shorter ones, which emphasizes the cooperative nature of  $\beta$  relaxation.

*Mechanism for  $\alpha_2$  process.*—To grasp the microscopic origin of the  $\alpha_2$  process, we analyze the probabilities  $p(u)$  of atomic displacements  $u$  for Al and Sm atoms, respectively. As shown in Fig. 4(a), at a temperature  $T = 500$  K lower than the peak of  $\alpha_2$  relaxations (about 560 K), the peaks of  $p(u)$  for Al and Sm separate from each other. While at a temperature higher than the  $\alpha_2$  process, the  $p(u)$  peaks of Al and Sm well overlap with each other. This comparison implies that decoupling of the motions of Al and Sm atoms occurs when the temperature crosses the  $\alpha_2$  peak.

To further quantify this behavior, Fig. 4(b) plots the most probable displacement, i.e., the peak position  $u_p$  of  $p(u)$  as a function of temperature. When the temperature increases from the lower regime,  $u_p$  of Al and Sm atoms first increases separately until reaching a transition point where two curves merge to one. When comparing  $u_p$  with loss

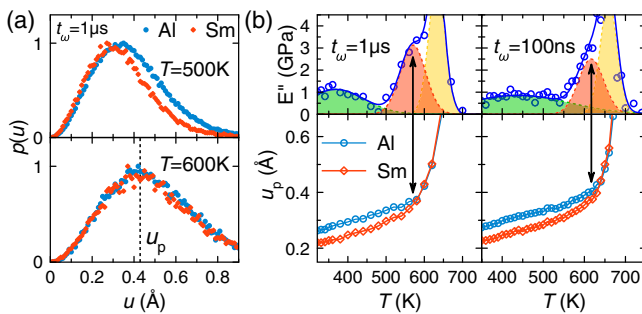


FIG. 4. Atomic motion decoupling and  $\alpha_2$  relaxation. (a) The probability of Al and Sm displacement with the oscillation period  $t_\omega = 1 \mu\text{s}$  at 500 and 600 K, respectively. The dashed line indicates the peak position  $u_p$ . (b)  $u_p$  of Al and Sm atoms and loss moduli as a function of temperature for  $t_\omega = 1 \mu\text{s}$  and 100 ns, respectively. The arrows highlight the transition points and the peak positions of  $\alpha_2$  relaxations.

moduli in Fig. 4(b), we find that the transition point coincides with the peak position of  $\alpha_2$  relaxation over all the studied oscillation periods. Therefore, the  $\alpha_2$  relaxation well correlates with the dynamical transition from coupling to decoupling motions of Al and Sm atoms. It indicates that the asynchronous freezing of fast and slow motions could be the key factor leading to this process.

In Fig. 5, we summarize all the studied processes in a relaxation map over a wide range of temperature and timescales. One can see that the  $\alpha_2$  relaxation and the decoupling between Sm and Al atoms follow the same temperature-time relation, suggesting an intrinsic correlation between them. Meanwhile, the  $\beta$  relaxation and the long stringlike motions ( $\xi \geq 10$ ) agree with each other. Hence, the atomistic simulations not only predicted the complex relaxations in the MGs, but also elucidate the underlying mechanisms for them.

Figure 5 also reveals that the  $\text{Al}_{90}\text{Sm}_{10}$  MG is a typical glass system in which the solute and solvent elements show dramatically different dynamical behaviors: the  $\alpha$  relaxation time calculated based on the intermediate scattering function (ISF) of Sm atoms is orders of magnitude longer than the Al atoms (see details in Supplemental Material [41]). Moreover, the global  $\alpha$  relaxation determined from MD DMS correlates only with the  $\alpha$  relaxation time from the ISF of Sm atoms, implying that it is controlled predominately by the slowest process. Previous simulations [50–52] suggested that the large atomic size ratio disparity might cause more than one glass transitions in model systems. In a recent theoretical work, Cui *et al.* [53] pointed out that the dynamical decoupling between constituents with wide mass disparity might lead to a separated relaxation process and suggested it to be a  $\beta$  relaxation. The identified  $\alpha_2$  process here might be an experimental evidence for these scenarios

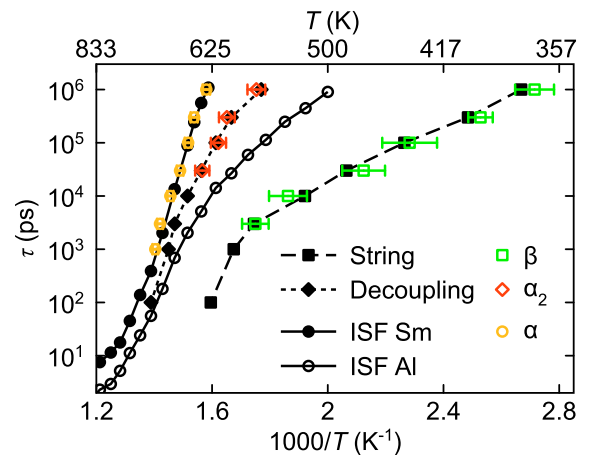


FIG. 5. Relaxation map of  $\text{Al}_{90}\text{Sm}_{10}$  MG. ISF Sm/Al: The  $\alpha$  relaxation time of Sm/Al atoms based on the Sm/Al ISF spectra. Decoupling: The transition temperature from coupled to decoupled most probable motions  $u_p(T)$  of Al and Sm atoms. String: The most probable temperature to form the long-string motions.

in real glasses. Moreover, it indicates that the related process can be an additional primary process instead of  $\beta$  relaxation. Finally, we note that the  $\alpha_2$  process might not be unique to the Al<sub>90</sub>Sm<sub>10</sub> MGs: in a recent work Xue *et al.* [54] reported the relaxation processes in a series of LaGa-based MGs. Although not discussed explicitly, their data indeed exhibit a discernable  $\alpha_2$ -like process, which may also be related to the mobility decoupling between fast Ga and slow La atoms.

We have shown that with atomic simulations one could predict complex relaxation processes in MGs at the laboratory timescales and clarify their microscopic origins. A MG system with a previously unidentified  $\alpha_2$  relaxation process due to the mobility decoupling and strong  $\beta$  relaxations caused by long-string motions has thus been predicted by simulations and verified by experiments. The combined experiments (validations) and simulations (predictions and clarification of mechanisms) represent a first glimpse of what may become a routine and integrated step in the study of glass relaxation. With the above interpretations, one would expect abundant  $\alpha_2$  relaxations, or even more relaxation processes in glass states. It then suggests that efforts aimed at a quantitative theory to predict glass relaxation would be desirable. The results presented above thus open new challenges and opportunities for furthering our understanding of glass relaxations.

We thank Ms. Xiao-Hui Qin for experimental help. The work at HUST is supported by National Science Foundation of China (NSFC 51601064) and the Fundamental Research Funds for the Central Universities (2018KFYXKJC009). Work at Ames Laboratory was supported by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences, Materials Science and Engineering Division, under Contract No. DE-AC02-07CH11358, including a grant of computer time at the National Energy Research Supercomputing Center (NERSC) in Berkeley, CA.

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