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The excess wing of bulk metallic glass forming liquids

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Abstract

We performed the dynamic mechanical relaxation measurements in typical metallic $Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5}$ and $Ce_{70}Al_{10}Cu_{20}$ supercooled liquids close to their glass transition temperature T_g . An obvious excess wing is observed both in temperature- and frequency-dependent loss modulus curves by the calculation of the relaxation time of α -relaxation in supercooled liquids using the Kohlrausch–Williams–Watts equation in combination with the Vogel–Fulcher–Tamman equation. The results support that the slow β -relaxation process exists in the metallic liquids and the excess wing is the high-frequency flank of the β -relaxation. The existence of β -relaxation in the metallic supercooled liquids affirms that the β -relaxation process arises from the small-scale translational motions of atoms which are hindered in its metastable atomic solid-like islands. © 2006 Elsevier B.V. All rights reserved.

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1. Introduction

In the supercooled liquids (SLs) some characteristic relaxation processes can be observed [1]. The α -relaxation reflecting structural rearrangement in SLs, is characterized by a broad and asymmetric absorption spectrum, along with deviation from an Arrhenius dependence of relaxation time on temperature [2–7]. For many SLs, a second relaxation (or slow β -relaxation), is found at frequencies higher than the α -process, and above glass transition temperature T_g both relaxations tend to bifurcate [8,9]. The slow β relaxation is considered to be a near universal feature of glass-forming liquids at low temperatures [8,9]. However, many SLs, especially for rather strong SLs, do not exhibit the well-resolved β -process, but an excess contribution to the high-*f* power law of the α -peak [1,5–7]. This wing can be described by a second power law [1]. Based on the Nagel

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Scaling the excess wing was proposed to be an intrinsic feature of the α -relaxation and had nothing to do with the slow β -relaxation [5]. On the other hand, Ngai [10] proposed that the excess wing is just the high-frequency flank of the slow β -relaxation whose low frequency part is fully hidden by the dominant α -peak. Both the opposite models are supported by some experiments results [11,12], and there has so far been no consensus on this issue even on a qualitative level. The physical origin of the excess wing is commonly considered as one of the great mysteries of glass physics.

The origin of the slow β -relaxation is another problem [2,3]. The β -relaxation was originally seen to indicate local diffusion, both rotational and translational, in loosely packed isolated regions [8,9]. So it represents the motion of 'island'. Another view based on the dipolar correlation function is that the slow β -relaxation may be attributed to faster, small angle reorientational motion of all molecules [13]. It is still unclear if this process has an intermolecular or intramolecular origin, or whether it reflects small or relatively large-angle jumps. Another issue is that whether all molecular contribute to the relaxation, or only those residing in sidergroups.

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Due to the broad dynamic range accessible, dielectric spectroscopy is a main tool to study the molecular dynamics of glass-forming liquids [1]. However, it is difficult to understand the nature of the β -relaxation only by this method because the reorientational and translational motions of molecular in molecular SLs both contribute to the dielectric relaxation and cannot be distinguished [1-3]. The dynamical mechanical relaxation arising from the elastic dipole reorients after the removal an external stress has been found to be available to the dynamics of the nonmolecular SLs [2,14]. The dynamic mechanical relaxation measurements in a SL with an assembly of equal-size or different-size could provide more direct evidences for the underlying physics of the excess wing and slow β -relaxation, since in the system reorientation of elastic dipole can not occur by the reorientation of the spheres about their axis [14]. Metallic liquid is the very systems with atoms which interact through metallic bonding, and its structure is close to a dense random packing of spheres [15,16]. However, metallic SLs are normally quite unstable with respect to crystallization. Recently, by careful selection of composition the glass-forming stability of some metallic SLs can be cast into bulk metallic glasses (BMGs) at low cooling rates ($\leq 10 \text{ K s}^{-1}$) [15,16]. The availability of these alloys offers a wide experimental temperature and time windows for investigating the dynamical characteristics in metallic SLs and for verifying that if exists the excess wing and how universal it is in the SLs.

In this paper, the dynamical mechanical relaxation measurements on typical metallic $Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5}$ (SL region is from 603 K to 688 K with available experimental time scale) and $Ce_{70}Al_{10}Cu_{20}$ (SL region: 343– 400 K) SLs close to their T_g were performed. An obvious excess wing is observed both in temperature and frequency dependent loss modulus curves, and the nature of the excess wing is discussed. Our result offers new evidence for the existence of the slow β -relaxation in metallic SLs.

2. Experimental

The Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni₁₀Be_{27.5} (vit4) and Ce₇₀Al₁₀Cu₂₀ BMGs were prepared from a mixture of the elements of purity (99.99 at.%) by arc melting, and subsequently sucked into a copper mold [15,16]. High-precision dynamical mechanical relaxation measurements were performed in a thermal mechanical analyzer (DMA) using a TA DMA2970. The temperature and frequency dependent modulus measuring accuracies for the DMA were better than 2%. The experimental frequency range was from 10^{-2} Hz to 200 Hz. Specimen in the shape of rectangular sheet was tested in a single-cantilever bending configuration and protected in an argon atmosphere. The low value of $T_{\rm g}$ and stable SL state of the alloys offer a wide available temperature and time windows for investigating the dynamical characteristics. To avoid the partial crystallization during measurement, we kept the measuring as short as possible, especially at higher temperatures in SL region.

3. Results

Fig. 1(a) exhibits the typical storage (G') and loss (G'')moduli of vit4 determined from 473 K to 750 K at a heating rate of 3 K/min and a frequency (f) of 1 Hz. In glassy state G' and G'' are almost independent of T-increasing from 300 K to 500 K. From 500 K to the onset temperature of glass transition (603 K), both G' and G'' increases slowly and gradually due to the physical aging occurring during heating. In the SL region, an obvious asymmetrical peak characterized by a smaller slope at low-T side can be found in the G''-curve, while the G' decreases to zero at higher T, exhibiting a typical liquid characteristic. In Fig. 1(a) the occurrence of the crystallization is also exhibited with an abrupt increasing of G' at 725 K. Fig. 1(b) shows that G'and G'' as function of f at 653 K. An asymmetrical peak is also exhibited well in the G''-curves. G' increases from zero to a constant value with the increasing f. The change of the G' and G'' in Fig. 1(a) and (b) has an inverse tendency.

Fig. 2 shows the *f*-dependence of G' and G'' of metallic vit4 SL determined isothermally by DMA at different temperatures (from 608 K to 678 K). The dynamical characteristic of the SL is exhibited well by the G''-curves. The values of $\log_{10} f_{\rm P}$ ($f_{\rm P}$ is the peak frequency) directly indicate the frequencies of the molecular fluctuation which are relevant for the susceptibilities. The T-dependence of $\log_{10} f_{\rm P}$, usually called as molecular mobility, is relevant for the dynamical relaxation [2]. Fig. 3 shows $\log_{10} f_{\rm P}$ versus reciprocal peak temperature $1/T_{\rm P}$ of G"-curves determined from the isothermal and continuous heating processes. The consistence in the molecular mobility between isothermal and continuous heating processes affirms the correlation between them, and the isothermal process is the limitation of the continuous heating process. The T-dependent $f_{\rm P}$ of the G''-curves is fitted by the Vogel–Fulcher–Tamman



Fig. 1. The storage G' and loss moduli G'' of vit4 determined in a heating process (a) and isothermal process (b). The measurement accuracy is better than 2%.



Fig. 2. The *f*-dependence of G' and G'' of metallic vit4 SL determined isothermally by DMA at different temperatures. The solid lines are the fits using the KWW equation. The measurement accuracy is better than 2%.



Fig. 3. The plots of $\log_{10} f_{\rm P}$ versus $1000/T_{\rm P}$ of the G"-curves in vit4 supercooled liquid determined in isothermal processes and continuous heating processes.

(VFT) expression: $f_{\rm P} = f_0 \exp\left(\frac{-B}{T-T_0}\right)$ [3]. The solid line in Fig. 3 is the best fit, and the parameters of f_0 , B and T_0 for vit4 are determined to be 4.6559×10^{14} Hz, 10134.6 K and 352 K, respectively. The fragility m [17] $\left(m = \log_{10}e \cdot \left[\frac{BT_g}{(T_g - T_0)^2}\right]\right)$ of the vit4 liquid is then calculated to be 38. The values of B, T_0 and m are almost same as those determined from the T-dependent viscosity [18]. The vit4 is a rather 'strong' liquid in terms of fragility [17].

The loss or imaginary component carries the entire information regarding the microstructural relaxation. Fig. 4 exhibits the G'' as functions of f determined at 613 K, 633 K, 653 K, 673 K, and 683 K for vit4. Fig. 5 shows the G'' as a function of frequency for CeAlCu SL at 343 K, 353 K, 363 K and 373 K. All spectra of G'' for the two alloys dominated by a single and asymmetrical peak shifting through the f-axis with temperature



Fig. 4. The frequency dependence of the G'' of vit4 supercooled liquid determined isothermally by DMA at different temperatures. The solid lines are the fits using the KWW equation.



Fig. 5. The frequency dependence G'' determined at temperatures of 343 K, 353 K, 363 K and 373 K. All the G'' data are normalized by the maximum G'' value G''_p . The solid lines are the fits using the KWW equation. The measurement accuracy is better than 2%.

exhibit a typical relaxation behavior, which has been considered to represent mainly the α -relaxation process [5,10]. Thus, we fit the data with the Kohlrausch-Williams-Watts (KWW) form [6]: The Fourier transforms of $d\left\{\Delta G \exp\left[-\left(\frac{t}{\tau}\right)^{\beta_{\text{KWW}}}\right]\right\}/dt$, where $\Delta G = G(f = \infty) - C(f = \infty)$ G(f=0), β_{KWW} a stretched coefficient, and τ the averaged relaxation time. The originally and purely phenomenological KWW form is used because the KWW function has been theoretically derived from a system with a distribution of relaxation times, and can describe the α -relaxation well which represents the process with a distribution of relaxation time [2,19,20]. We also tried with the Havriliak-Negami function [2] which is convenient for dynamic susceptibility acquired in f-domain but found that the KWW form is better. To fit the data well with the KWW form, we decrease the value of β_{KWW} with the decreasing T. Even then, it can be clearly found in Figs. 4, 5 that a departure or excess wing at high-f flank of the α -peak exists and becomes more obvious with decreasing T. The departure from the KWW form has also been observed in dielectric measurements on the molecular SLs without obvious slow β -peak [1.11.12.21]. Fig. 6 shows the comparison of our data with the dielectric relaxation loss data of a small molecule glassformer called MTHF at two temperatures [22]. We choose MTHF because it has obvious excess wing in the dielectric relaxation loss curve and the same β_{KWW} as vit4. The data are shifted to coincide with our data. One can see that the excess wing in vit4 over and above the KWW fit is larger than that of MTHF, but otherwise the feature is qualitatively the same. The larger excess wing of metallic SL is likely due to the translational of the atoms in metallic alloys in contrast to the small angle rotation of the MTHF molecules that contribute to the excess wing. The excess wing in ZrALCu metallic SL was also detected by a double-paddle oscillator method [23].

In order to exhibit the *T*-dependent G''-curves of vit4 we plotted $G''(f)/G''(f_P)$ against f/f_P for three temperatures in Fig. 7. The data collapse into one curve for $f < f_P$, while the data for $f > f_P$ are *T*-dependent. Obviously, the dynamics of the metallic SL does not obey the *time-temperature superposition* (TTS) principle [1,2]. This is different from that of the metallic liquid above model-coupling tempera-



Fig. 6. Comparison with the dielectric relaxation loss data [22] of a small molecule glass former-MTHF (has the same β_{KWW} as the vit4 metallic glass) at two same temperatures (638 K and 648 K, black open circles).



Fig. 7. $G''(f)/G''(f_P)$ versus f/f_P for vit4 SL at different temperatures. The data do not overlap for $f/f_P > 1$.

ture $T_{\rm C}$ (\cong 1.2 $T_{\rm g}$ [2]) where the liquid has been found to obey the TTS [24-26]. Because the α -relaxation always obevs TTS, the violation of TTS has been explained as the interference from an additional minor relaxation process, viz. the slow β -relaxation often faster than the dominant α -relaxation [2]. Recent experimental results [11,27] successfully affirmed the suggestion. The experiments showed a well-resolved slow β -relaxation peak at a low-T after the long-time annealing of the sample below T_g in the materials without a well-resolved slow β -relaxation in SLs. The similar phenomena in Fig. 7 are also found in many systems without obvious β -relaxation [28]. Therefore, the single and asymmetrical peak in Figs. 4, 5 should be the evidence of containing slow β -relaxation in addition to the α -relaxation. Meanwhile, this implies that the fit of the KWW form with the *T*-dependent β_{KWW} in Figs. 4, 5 is not appropriate for the whole data. The dynamics of the SLs related to 'cage effect' should become more pronounced as the density increases at lower T [2]. However, the model-coupling theory providing quantitative relation of motion of the 'cage' only describes well the relaxation of the liquids above $T_{\rm C}$. The fails to explain the dynamics of the SLs with the model-coupling theory was suggested to be due to the large size characteristic dynamic length of SLs which is beyond the 'cage' dimension ($\cong 0.5 \text{ nm}$) [2]. The current investigation has exhibited that the metallic SLs is dynamic heterogeneous and cannot be represented by the simple 'cage' effect.

Therefore, we tried to simulate the G"-curves of the metallic SLs using the superposition of an α - and a β -peaks. Satisfactory agreement between the data and the fit is obtained. Fig. 8 shows the example for the fit of vit4 (the solid lines). The dash lines are the fits of the α -relaxation with $\beta_{\rm KWW} = 0.65$ which is related to the invariable slope (0.91) of the curves for $f < f_{\rm P}$ in Fig. 7. It is noted that the constant $\beta_{\rm KWW}$ obeys the empirical correlation of $m = m_{\rm o} - s \beta_{\rm KWW}$, with $m_{\rm o} = 250 \pm 30$ and s = 320 [29].



Fig. 8. Frequency-dependent G'' of vit4 alloy for three temperatures. The solid lines are the sum of two KWW form fits. The dash lines are fits for the α -relaxation with β_{KWW} equal to 0.65. The dash dot lines are fits for the β -relaxation, whose β_{KWW} decreases with increasing temperature.

The function has connected successfully the characteristics of the α -relaxation with the dynamical characteristic of the glass-forming liquids. The dashed dot lines in Fig. 8 show the underlying β -curves resulting from the fits. The β curves can also be described well with the KWW form rather than with the Havriliak–Negami form. The β -curve width decreases with the increasing temperature. The values of β_{KWW} of the β -curves are 0.45, 0.49 and 0.51 for 628 K, 643 K, and 668 K, respectively. At the same time, the peak height of the β -curve decreases with the increasing temperature, and the T-dependent f_p of the β -relaxation is a non-Arrhenius behavior. Above results indicates that the β -relaxation in the metallic SL possesses the nature of α relaxation, which is consistent with the Nagel scaling [5]. With Nagel scaling, the loss elastic modulus within wide T- and f-regions should be collapsed into one master curve indicating that the excess wing has an intrinsic feature of α relaxation. Fig. 9 shows a plot of $w^{-1}\log_{10}(G''f_{\rm P}/\Delta Gf)$ versus $w^{-1}(1+w^{-1})\log_{10}(f/f_{\rm P})$, here, $\Delta G \cong G_{\infty}$ the relaxation modulus in the high-f limit, and the half-maximum width w is normalized by a Debye relaxation (1.14 decades) [5]. All of the loss modulus data determined isothermally from all temperatures and frequencies are collapsed onto a single line. However, in contrast to Ref. [5], in which all data show a *f*-independent behavior for $f < f_p$ in the scaling picture, our data show obvious f-dependent behavior. This is because our data have to be described by $G'' \sim f^a$ with a = 0.91 for $f \le f_p$, while the f-independent behavior requires $a \cong 1$. Similar result has also been observed in some low molecular weight glass-forming liquids [28]. The *f*-independent behavior at $f \leq f_p$ in the Nagel-scaling indicates that the slope of the loss susceptibility curves in f-domain is an important parameter that represents the α relaxation and is characterized by the intrinsic dynamical property of SLs.

The superposition of the α - and β -processes should be also exhibited in the G''-curves determined in the continuous heating process according to the results of Figs. 1 and 3. The G''-curves determined in the two processes are equivalent in the SL region. However, the behavior of the slow β -process exhibited above is more complex than that of the α -process because the relaxation strength and the shape of the slow β -process is T-dependent. It is not easy to calculate the T-dependent slow β -process in the metallic SL, while the α -process determined in continuous heating process is easier to simulate because the α -process obeys TTS principle near T_{g} . The α -relaxation determined by DMA represents information about the molecular cooperative motions associated with sources of elastic dissipation mechanisms. The viscosity corresponds to the fluidity related to the structural relaxation in low-f domain which is dominated by the α -relaxation. Then, the averaged relaxation time of the α -relaxation in the metallic SL can be in the form of the inverse $f_{\rm P}$ [2]. We can fit the G''-curves in T-region with the Fourier transforms of KWW. Fig. 10 exhibits the best fits for the G''-curves of vit4 determined in continuous heating processes (heating rate of 5 K/min) at 1 Hz, 4 Hz, and 16 Hz, respectively. The fitting τ has the form: $\tau = 1.6 \times 10^{-15} \exp{\left(\frac{9668.42}{T-352}\right)}$. Fig. 11 exhibits the *T*-dependence of *G''* for CeAlCu SL at the frequencies of 1 Hz, 2 Hz, and 5 Hz, respectively. In Figs. 10 and 11, the fits yield a better representation of the peak part above $T_{\rm P}$, but the low-T data display higher value than the fits, which is consistent with the results deduced from Fig. 8. The slow β -process existing in the metallic SL is faster than the α -process and appears firstly during continuous heating, and then the α -process follows. Because of the intrinsic dynamical characteristic of the metallic SLs the slow β -process is not fully decoupled from the α -process. So, similar to the observation in loss modulus curves determined isothermally no obvious slow β -peak in G''-curves can be observed and only a excess wing on the α -curve is exhibited. It should be noted that the departure should be connected within the SL region. In the glassy state the departures shown in Figs. 10 and 11 are due to the physical aging of the glass and not to the relaxation. This measurement



Fig. 9. The scaling plot for the relaxation curves of the vit4 SL determined isothermally by DMA. $w^{-1}\log_{10}(G''f_{\rm P}/\Delta Gf)$ is plotted against $w^{-1}(1 + w^{-1})\log_{10}(f/f_{\rm P})$.



Fig. 10. Temperature-dependent G'' of vit4 alloy for three frequencies. The solid lines are fits using the KWW equation with the relaxation time $\tau = 1.6 \times 10^{-15} \exp\left(\frac{9668.42}{T_{-1} \times 10^{-15}}\right)$ and β_{KWW} equal to 0.650.65.



Fig. 11. The G''-curves of CeAlCu alloys determined in continuous heating processes at the frequencies of 1 Hz, 2 Hz and 5 Hz, respectively. The heating rate is 5 K/min. All the loss modulus data are normalized by maximum G'' value G''_p in the figure. The solid lines are the fits using the KWW equation. The measurement accuracy is better than 2%.

provides a convenient way for determine excess wing in metallic SLs.

4. Discussions

The characteristic of the dynamical mechanical relaxation in the metallic SL is directly related to its microstructure. Unfortunately, the exact structural picture of SL is unavailable so far. Conceivably, we explain the observations based on the physical scenario proposed by Tanaka [9]: The slow β -process in glass-forming liquids appears only below a temperature which can be connected with the $T_{\rm C}$ of the model coupling theory. Below $T_{\rm C}$ ordinary metallic liquid is transformed into the Griffiths-phase-like state characterized by the appearance of metastable solid-like islands. The slow β -process represents the motions of atoms escaping from the solid-like islands. Above $T_{\rm C}$ the slow β -relaxation does not exist in the metallic liquid because the escaping of atoms from their cages is easy, and all atoms make full rotation. That is, the dynamical microstructure of the metallic liquid will change when temperature decreases cross over its $T_{\rm C}$ [30]. The $T_{\rm C}$ of vit4 is about 875 K [24]. Thus, the existence of metastable islands in vit4 below 875 K makes the slow β -relaxation appears. The α -relaxation of the metallic SL arises from the reorganization of metastable island [2,9]. The computer simulation based the energy landscape theory affirms that rather strong SLs have small-size cooperatively rearranging region [31]. This indicates that in rather strong SLs the motion escaping from the small-size metastable solid-like islands will be modulated strongly by the rearrangement of the islands. Therefore, the slow β -relaxation cannot be fully decoupled from the α -relaxation and possesses the α -relaxation nature in the rather strong SLs, and hence the excess wing.

The evidence of existence of the β -relaxation in the metallic SL strongly support that the β -relaxation arises from the hindered translational motion of some atoms in some distribution of local regions [8]. The microstructure of the metallic liquid is close to a system consisting of an

assembly of different-size spheres. Therefore, the β -relaxation in the metallic SL observed by DMA measurement must be only related to the translational motion of atoms because reorientations of the spheres about their axis do not attribute to the reorientation of the elastic dipole [8]. If the occurrence of short-range translational motions was combined with the premise that essentially all atoms contribute to the β -relaxation, application of an external mechanical stress would permanently deform the glass at its β -relaxation time scale. In fact, a liquid-like deformation of the metallic glass at the time scale of the β -relaxation process has not been observed in Figs. 10, 11. Therefore, the translational motions of atoms attributing to the β relaxation are small-scale and hindered by solid-like islands.

5. Conclusions

An obvious excess wing is observed in the relaxation curves of metallic $Zr_{46.75}Ti_{8.25}Cu_{7.5}Ni_{10}Be_{27.5}$ and Ce_{70} - $Cu_{20}Al_{10}$ supercooled liquids, which is considered to be connected with the high-frequency tail of the slow β -relaxation whose low-frequency part is overlapped with the high-frequency tails of the α -peak. The evidence for existence of the slow β -relaxation in the metallic SLs supports that the separation of the α - and β -relaxations is universal and the β -relaxation process arises from the small-scale translational motions of atoms that hindered by the metastable islands.

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