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MECHANISM OF ULTRASOUND-INDUCED STRUCTURAL INSTABILITY IN BULK METALLIC GLASSES

T. Ichitsubo[1], E. Matsubara[1], J. Saida[2], H. S. Chen[3], N. Nishiyama[4] and T. Yamamoto[5]

[1]Department of Materials Science and Engineering, Kyoto University, Kyoto 606-8501, Japan
[2]Center for Interdisciplinary Research, Tohoku University, Sendai 980-8578, Japan
[3](Ret.) Bell Laboratories, Lucent Technology, Murray Hill, New Jersey 07974, USA
[4]RIMCOF, R&D Institute of Metals and Composites for Future Industries, Sendai 980-8577, Japan
[5]Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan

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Abstract. This work devotes to consider an underlying mechanism of the ultrasound-induced instability phenomenon observed for some metallic glasses. Mechanical spectroscopy analysis strongly suggests that the instability is caused by that atomic motions associated with the . relaxation are resonant with the periodic ultrasonic-strain field. It is reasonable to consider that such atomic motions at temperatures lower than the kinetic freezing temperature T_g originates from relatively weakly bonded regions in an inhomogeneous microstructure of glass.

Recently, we have reported that crystallization of some bulk metallic glasses (BMGs) is much accelerated around the glass transition temperature T_g under ultrasonic (US) perturbation in the process of determining the elastic moduli of some BMGs around the glass transition temperature [1-6]. Actually, long before our works, similar phenomena on conventional metallic glasses have already been reported. Kopcewicz *et al.* showed lots of works on the effects of radiofrequency (RF) magnetic field on crystallization of some ferrous amorphous alloys [7-9]. They discussed that RF magnetostrictive vibration is responsible for the acceleration of crystallization. Similarly, Gupta *et al.* also reported that ultrasonic vibrations enhance the crystallization process of an Fe-Si-B-C amorphous alloy [10]. In a more recent work, significant reduction of T_g and T_x was also found during the ultrasonic pulse-echo measurements for a $Zr_{55}Al_{10}Ni_5Cu_{30}$ metallic glass [11]. These phenomena indicate that the structural stability of metallic glasses is deteriorated under dynamic external fields even at relatively low temperature (below T_g). As is described in the literature [12], only the b (secondary) relaxation process [13–21] remains active in the temperature range ($T < T_g$) where the system is completely frozen with respect to the α (primary) relaxation process (dynamic glass transition). Recently, Rösner *et al.* have discussed the possible origin of the b relaxation in detail [21]; they have pointed out that the β process in metallic glasses can be caused by the Johari-Goldstein mechanism (there exist local, highly mobile regions in the glassy state) [13,19,20]. It is, therefore, considered that the β relaxation is responsible for the low-temperature instability under ultrasonic perturbation. In our recent paper [5,6], we discussed a possible mechanism of the US-induced instability from the above viewpoint. Here, on the basis of our recent works, we briefly explain that the instability can be caused by that atomic motions associated with the β relaxation are resonant with the periodic ultrasonic-strain field.

Corresponding author: T. Ichitsubo, e-mail: tichi@mtl.kyoto-u.ac.jp

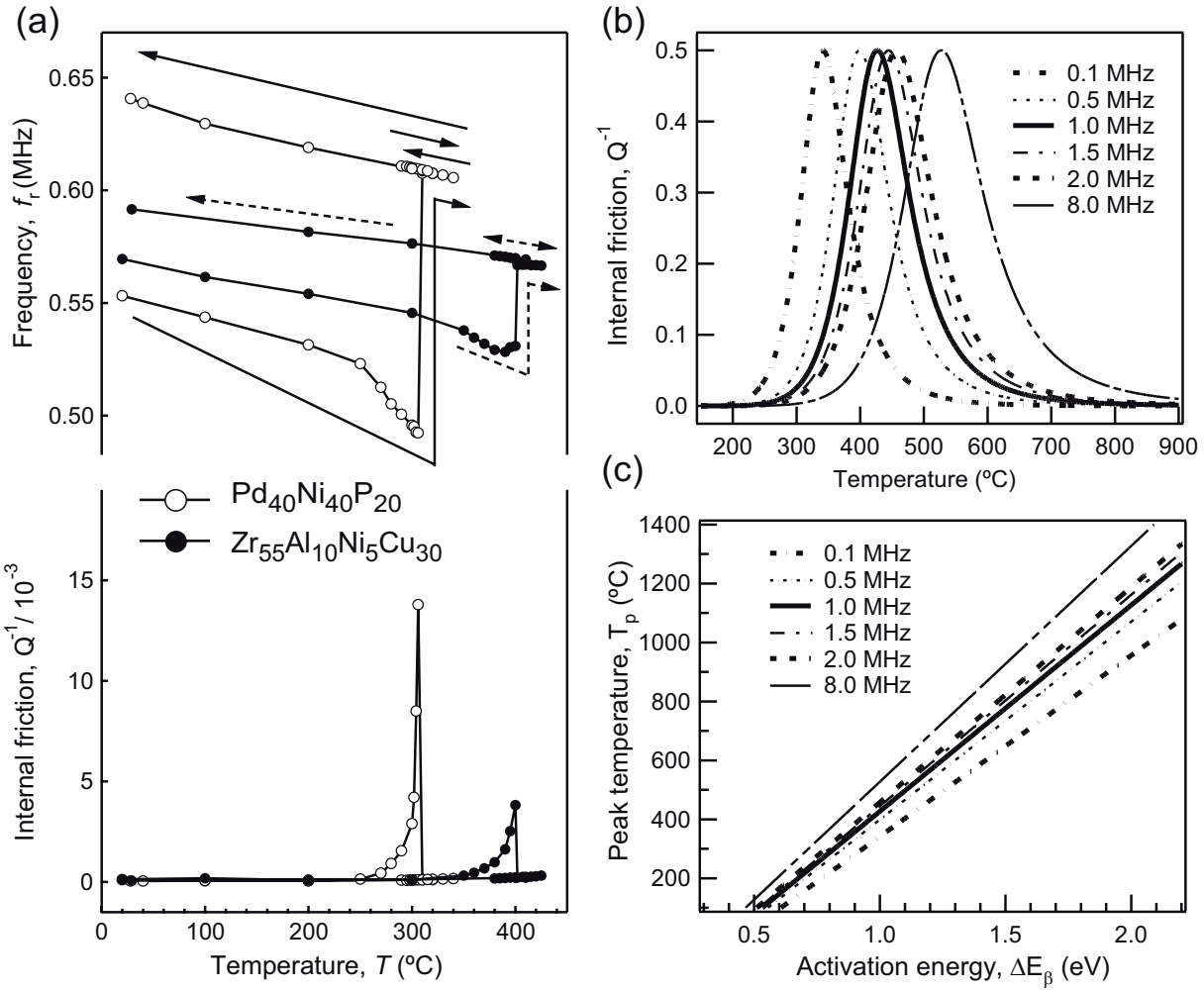

Fig. 1. (a) Internal friction and resonance frequency as a function of temperature, obtained for fully structurally-relaxed samples of Pd- and Zr-based metallic glasses. (b) Temperature dependence of the internal friction calculated for the β relaxation in a Pd-based metallic glass. Q^{-1}-T curves calculated using Eq. (1). (c) The activation-energy (ΔE_β) dependence of the peak temperature (T_p) that satisfies $\omega\tau = 1$. The figure is taken from Ref. [6] and the original data are presented in Ref. [3].

Alloy ingots of $Zr_{55}Al_{10}Ni_5Cu_{30}$ and $Pd_{40}Ni_{40}P_{20}$ (atomic%) BMGs were used in this work. At heating rates lower than 20 °C/min, T_g of each sample is about 390-400 °C for $Zr_{55}Al_{10}Ni_5Cu_{30}$ and 290-300 °C for $Pd_{40}Ni_{40}P_{20}$. To carry out annealing at elevated temperatures under US vibrations in a vacuum atmosphere, we employed the EMAR method, in which acoustic vibrations are generated by the Lorentz force mechanism [1-3]. Resonance frequencies f_r of the cylindrical samples were detected in a radiofrequency (sub/low-MHz frequency) range. Internal friction Q^{-1} was measured at each resonance frequency by the free-decay method: $A(t) = A_0 \exp(-Q^{-1}\pi f_r t)$, where A and f_r denote the amplitude and the resonance frequency, respectively.

Fig. 1a shows the changes in internal friction and the resonance frequency at around T_g of each glass. Here we take notice of the increase of the internal friction around T_g, and examine whether the increase is indeed associated with the β relaxation. The activation energy for the β relaxation is known to be far lower than that for the α relaxation in which cooperative atomic motions occur with a high activation energy [14]. Actually, the β relaxation were observed, for example, in Pd-Cu-Si [15],

La-Al-Ni [16,17], and Pd-Ni-Cu-P glasses [18]. While the a relaxation is observed around T_g at a low frequency (~ 1 Hz), the β relaxation is observed far below T_g at the same frequency[14]. In order to evaluate a temperature range of the β relaxation in the present frequencies, the $Q_\beta^{-1}(T)$ curves are calculated for a MHz frequency range by using ΔE_β~1.0 eV and τ_0~1.0·10^{-14} s (which were obtained with low-frequency internal-friction measurement for a Pd-Ni-Cu-P glass by Pelletier *et al.* [18]) with the Debye function:

$$Q_\beta^{-1} = \Delta_Q \frac{\omega\tau}{1+(\omega\tau)^2}, \quad \tau = \tau_0 \exp\left(\frac{\Delta E_\beta}{kT}\right), \qquad (1)$$

where Δ_Q is the relaxation strength, ω (= $2\pi f$) is the angular frequency (f is the frequency of oscillator), and τ is the relaxation time per event, τ_0 being the atomic/molecular-scale time, and ΔE_β is the activation energy of the event. Fig. 1b shows the calculated internal friction $Q_\beta^{-1}(T)$ for the β relaxation, and clearly indicates that the experimental $Q^{-1}(T)$ curves at the MHz frequencies around T_g in Fig. 1a corresponds to the rising edges of the calculated profile of Fig. 1b. In addition, since the relation, $\omega\tau = 1$, holds at the peak temperature T_p in Eq. (1), T_p is given by

$$_p = -\frac{\Delta E_\beta}{k \ln \omega\tau_0}. \qquad (2)$$

The peak temperature T_p is shown in Fig. 1c as a function of the activation energy ΔE_β. T_p is very sensitive to ΔE_β, and the increase of the internal friction occurring around 300-400 °C can be explained well with the ΔE_β value for the β relaxation (~ 1 eV). Thus, in the case of Pd-based metallic glasses, the β relaxation observed far below T_g at low frequencies is detected at a temperature around or higher than T_g at MHz frequencies. On the assumption that the activation energy of the β relaxation in $Zr_{55}Al_{10}Ni_5Cu_{30}$ is close to 1 eV, the same discussion is applicable. From the above viewpoint, the b relaxation is the most possible candidate for explaining the US-induced instability. Actually, the increase of internal friction in Fig. 1a means that a periodic external strain is stochastically resonant with certain atomic motions and, from Figs. 1b and 1c, it is reasonable to consider that these motions are associated with the β relaxation. In the absence of US strains atomic jumps occur repeatedly at the same sites, but in contrast, atomic jumps will become different from the ordinary ones under resonant periodic stress/strain fields, that is, atoms will move into more stable potential (energetically preferential) sites newly caused due to the strain. This situation resembles the Snoek relaxation. However, since the amorphous structure is not in thermodynamically equilibrium, repetition of such jumps will gradually change the energy landscape, eventually leading to crystallization.

In conclusion, we present a plausible underlying mechanism of ultrasound (US)-induced structural instability in metallic glasses. From the mechanical spectroscopy analysis, the typical frequency for the β relaxation of ΔE_β ~ 1 eV is found to become within a radiofrequency (MHz) range. Hence, it is considered that the radiofrequency internal-friction behavior observed around T_g and the US-induced instability are due to the β relaxation.

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