

Effect of passing electric current on the elastic property of amorphous alloys

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Abstract

The effect of passing electric current (PEC) on the low temperature elastic property of amorphous $\text{Cu}_{50}\text{Ti}_{50}$, $\text{Cu}_{50}\text{Zr}_{50}$, $(\text{Cu}_{30}\text{Zr}_{70})_{92.5}\text{Al}_{7.5}$ and $\text{Pd}_{80}\text{Si}_{20}$ (a-CuTi, A-CuZr, A-CuZrAl and a-PdSi respectively) was investigated for current densities $i_d \leq 5 \times 10^3 \text{ A cm}^{-2}$ using a vibrating reed technique with a strain amplitude $\epsilon_i \approx 10^{-6}$ and compared with the effect of ϵ_i without PEC for $\epsilon_i \leq 4 \times 10^{-3}$. For all the a-alloys f increases with increasing i_d , where f is the resonant vibrational frequency of the reed specimens. In each a-alloy specimen the feature of the i_d dependence of f observed at room temperature is very similar to that at 80 K, suggesting that the increase in f due to PEC is revealed through an athermal process. The strength of the increase in f due to PEC increases in the order of a-PdSi, a-CuZrAl, a-CuTi and a-CuZr. These features are very similar to those for the ϵ_i dependence of f without PEC observed in these a-alloys after scaling between the ϵ_i and i_d axes. An atomic model for the effect of PEC is discussed.

1. Introduction

Recently we found that both the structural relaxation (SR) and crystallization processes in amorphous $\text{Cu}_{50}\text{Ti}_{50}$ (a- $\text{Cu}_{50}\text{Ti}_{50}$) can be anomalously enhanced under passing electric current (PEC) of current density $i_d \approx 10^3 \text{ A cm}^{-2}$, where the progress of these processes was investigated by means of resistivity R measurements [1]. Later we improved the set-up for the R measurements using boron nitride substrates with a good thermal conductivity of $1 \text{ W cm}^{-1} \text{ K}^{-1}$ to minimize the effect of Joule heating under PEC and found that for a- $\text{Cu}_{50}\text{Zr}_{50}$ and a- $(\text{Cu}_{30}\text{Zr}_{70})_{92.5}\text{Al}_{7.5}$ both the SR and crystallization processes can be anomalously enhanced, but the effect of PEC reported in ref. 1 has been somewhat overestimated [2]. A detailed study on a- $(\text{Cu}_{30}\text{Zr}_{70})_{92.5}\text{Al}_{7.5}$ [2] suggested that under a PEC of $4.5 \times 10^3 \text{ A cm}^{-2}$ the pre-exponential frequency factor $\nu_{\text{SR},0}$ and activation energy E_{SR} for the SR process show an increase by about one decade and an increase by about 0.05 eV respectively and the activation energy E_x for the crystallization shows a decrease by about 0.07 eV. A recent study on a- $\text{Cu}_{50}\text{Zr}_{50}$ [3] again suggested a strong increase in $\nu_{\text{SR},0}$ and a slight increase in E_{SR} as well as a slight decrease in E_x under PEC, where the decrease in E_x is suggested to be associated with an enhancement of the nucleation process of crystallites under PEC.

On the other hand, recent diffusion studies on various a-alloys suggest that a diffusion process via a probable

collective motion involving many atoms can be predominant when the diffusion process associated with large free volume is suppressed [4–10]. From the diffusion studies we surmised that the increase in $\nu_{\text{SR},0}$ under PEC reflects some changes in a probable collective motion of many atoms under PEC in a-alloys [2, 3]. If this is the case, one may expect that the elastic property of a-alloys can be modified under PEC. For the elastic property of a-alloys without PEC, recently we found that in both a- $\text{Cu}_{50}\text{Ti}_{50}$ [11, 12] and a- $\text{Cu}_{50}\text{Zr}_{50}$ [13] the Young modulus M below room temperature (RT) shows a strong increase at first and tends towards saturation with increasing strain amplitude ϵ_i below about 4×10^{-3} . That is, with increasing ϵ_i the decreased M in a-alloys increases towards M in their crystalline counterparts, suggesting that the constituent non-linear elastic strain ϵ_i in ϵ_i increases, showing saturation. In refs. 11–13 we surmised that ϵ_i reflects a reversible localized shear deformation in a small cluster of atoms (LSD in a deformable unit). From the ϵ_i dependence of ϵ_i we further surmised that the number density of deformable units which can undergo LSD between ϵ_i and $\epsilon_i + d\epsilon_i$ shows a gaussian distribution at around $\epsilon_i = 0$ but for $\epsilon_i \geq 0$, and the LSD attained in each deformable unit remains unchanged for ϵ_i below $(3\text{--}4) \times 10^{-3}$. These results may predict that the probable LSD in the deformable units can be a useful probe to provide insight to the anomalous effect of PEC on a-

alloys. We investigated the elastic property under PEC as well as that under increased ϵ_i for various a-alloys.

2. Experimental procedures

Amorphous $\text{Cu}_{50}\text{Ti}_{50}$, $\text{Cu}_{50}\text{Zr}_{50}$, $(\text{Cu}_{30}\text{Zr}_{70})_{92.5}\text{Al}_{7.5}$ and $\text{Pd}_{80}\text{Si}_{20}$ (a-CuTi, a-CuZr, A-CuZrAl and a-PdSi respectively) tapes were prepared by melt spinning in a high purity Ar gas atmosphere. As-quenched tapes were mechanically polished by emery paper in water to remove the surface layer. The size of as-polished specimens was about $20\ \mu\text{m}$ thick and 1–1.5 mm wide. Figure 1 shows the measuring set-up. An a-alloy specimen is clamped at one end to form a vibrating reed. To allow PEC to the specimen, a thin Cu tape about $20\ \mu\text{m}$ thick and 0.4 mm wide is soldered at the free end of the specimen. The excitation and measurement of the resonant flexural vibration of the specimen were made electrostatically, where the vibrational frequency f and internal friction Q^{-1} were measured with $\epsilon_i \approx 10^{-6}$. At first, without PEC, the specimens were prestabilized by a thermal cycle between 80 and 400 K in a vacuum of 10^{-4} – 10^{-3} Pa and then f and Q^{-1} without PEC were measured during a further thermal cycle as the reference state. The measurements of f and Q^{-1} under PEC were carried out in an He gas atmosphere of 10^2 Pa to minimize the effect of Joule heating, while the increase in the specimen temperature T under PEC was measured separately. After the measurements of f and Q^{-1} , a thin thermocouple $25\ \mu\text{m}$ in diameter was attached to the specimen surface at a distance of 2–3 mm from the clamped end and then changes in T during the resonant vibrations under PEC in 10^2 Pa He were measured. Similar measurements were also made using a tape $20\ \mu\text{m}$ thick and 2 mm wide of crystalline nickel–chromium alloy, where the increase in T estimated from a change in f under PEC shows good agreement with that in T measured separately under PEC, and the increase in T under PEC is found to be proportional to i_d^2 . Q^{-1} measured in a vacuum

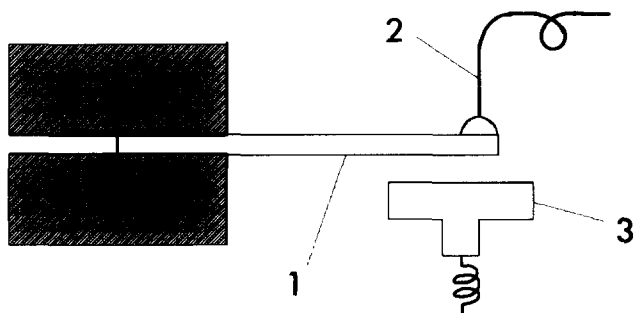


Fig. 1. Schematic drawing of the measuring set-up: 1, vibrating reed specimen; 2, Cu lead wire; 3, electrode.

mainly reflected that in the Cu lead wire and Q^{-1} in 10^2 Pa He the damping due to the gas atmosphere. Therefore the results for Q^{-1} are not given here. Without PEC we further investigated the ϵ_i dependence of f for $\epsilon_i < 3 \times 10^{-3}$ using the large ϵ_i vibration excited electromagnetically (see refs. 11–13 for a description of the measuring apparatus), where the effect of increased ϵ_i on f without PEC is compared with the effect of PEC on f at $\epsilon_i \approx 10^{-6}$.

3. Results and discussion

Figure 2 shows examples of the i_d dependences of f observed in a-CuZr at RT and 80 K, where the measurements were made with $\epsilon_i \approx 10^{-6}$ in an He gas atmosphere of 10^2 Pa. In Fig. 2 a PEC of 1 A corresponds to an i_d of about $3.7 \times 10^3\ \text{A cm}^{-2}$. The increase in T due to Joule heating under a PEC of 1 A was about 70 K at RT and about 80 K at 80 K. We carried out measurements for PEC values below 1 A, where the i_d dependence of f showed good reproducibility for repetition of measurements with various PEC values.

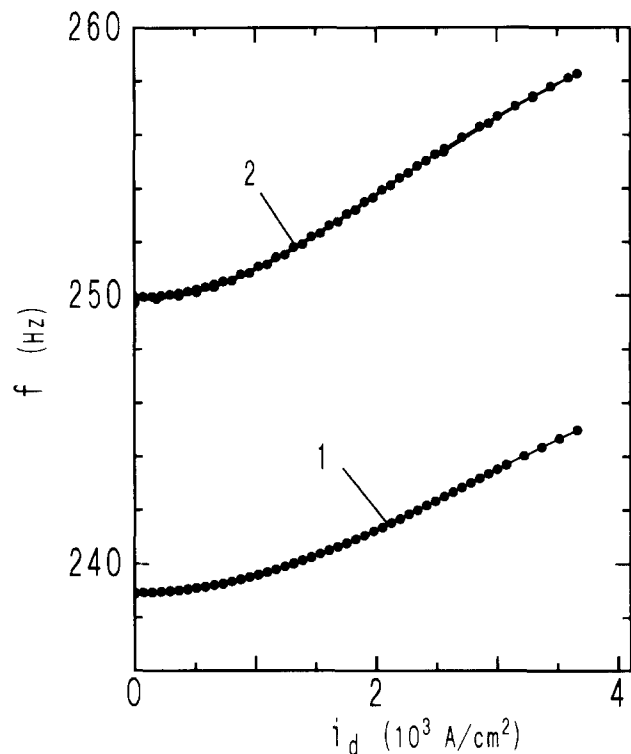


Fig. 2. Resonant frequency f of an a- $\text{Cu}_{50}\text{Zr}_{50}$ specimen observed with a strain amplitude ϵ_i of 10^{-6} under PEC vs. current density i_d , where an He gas atmosphere of 10^2 Pa was used during the measurements. Data 1 and 2 were observed at RT and 80 K respectively. Note that the observed changes in f are composed of a decrease in f due to Joule heating and an increase in f due to PEC without Joule heating (see Fig. 3); here the latter is predominant (see text).

That is, the changes in f shown in Fig. 2 are reversible and are composed of a decrease in f due to Joule heating and an increase in f due to PEC without Joule heating, where the latter increase in f is predominant at both RT and 80 K. In Fig. 2, reflecting the usual temperature change in f , f at 80 K shift to the higher f side as a whole.

We determined the increase in f due to PEC ("PEC" will be used hereafter to refer to the effects other than Joule heating) from the data shown in Fig. 2 after subtraction of the decrease in f due to Joule heating (see Section 2). Similar measurements were made for various a-alloys with the results shown in Fig. 3, where the i_d dependences of f under PEC observed for a-CuZr, a-CuTi, a-CuZrAl and a-PdSi are depicted together with that observed for a crystalline Ni-Cr alloy specimen. For the data shown for the crystalline Ni-Cr alloy the small decrease in f with increasing i_d does not suggest a change in the elastic property due to PEC, but is due to the experimental error in the T measurements under PEC and to a certain distribution of T along the long axis of the specimen under PEC. For the data for the a-alloys a similar experimental

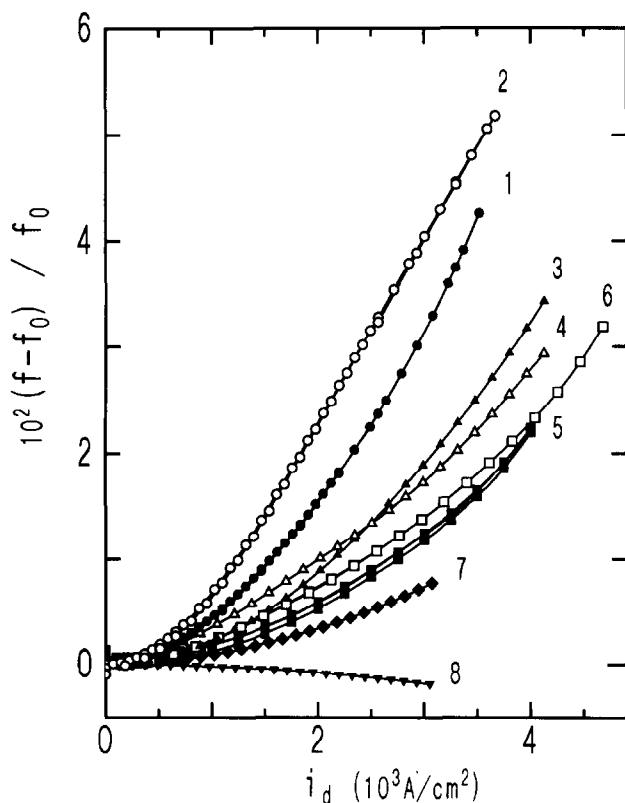


Fig. 3. Changes in f due to PEC observed after subtraction of the decrease in f due to Joule heating found at $\epsilon_t \approx 10^{-6}$ for various specimens, where f_0 found without PEC is taken as standard: 1 at RT and 2 at 80 K for a-Cu₅₀Zr₅₀; 3 at RT and 4 at 80 K for a-Cu₅₀Ti₅₀; 5 at RT and 6 at 80 K for a-(Cu₃₀Zr₇₀)_{92.5}Al_{7.5}; 7 at RT for a-Pd₈₀Si₂₀; 8 at RT for crystalline Ni-Cr alloy (see text).

error can be expected, but always contributes the minor part of the observed change in f . For all the a-alloys here an increase in f due to PEC is observed, suggesting that it reflects a common property in a-alloys. On the contrary, the magnitude of the change in f under PEC varies among the various a-alloys, *i.e.* it increases in the order of a-PdSi, a-CuZrAl, a-CuTi and a-CuZr. In each a-alloy the i_d dependences of f observed at RT and 80 K are very similar to each other, suggesting that the process responsible for the increase in f under PEC is an athermal one.

The ϵ_t dependence of f without PEC reported for a-CuTi [11, 12] and a-CuZr [13] suggested that it is revealed through an athermal process and the strength of the ϵ_t dependence of f is larger in a-CuZr than in a-CuTi. The preliminary results for the ϵ_t dependence of f found for a-CuZrAl and a-PdSi here are very similar to those reported for a-CuTi and a-CuZr, except that the strength of the ϵ_t dependence of f is smaller in both a-CuZrAl and a-PdSi than in a-CuTi (see Fig. 4 for a-CuZrAl). That is, the features found for the i_d dependence of f with $\epsilon_t \approx 10^{-6}$ are very similar to those observed for the ϵ_t dependence of f without PEC when the effect of i_d on the elastic property is assumed to be similar to the effect of ϵ_t . Figure 4 shows the ϵ_t dependence of f without PEC for a-CuZrAl observed

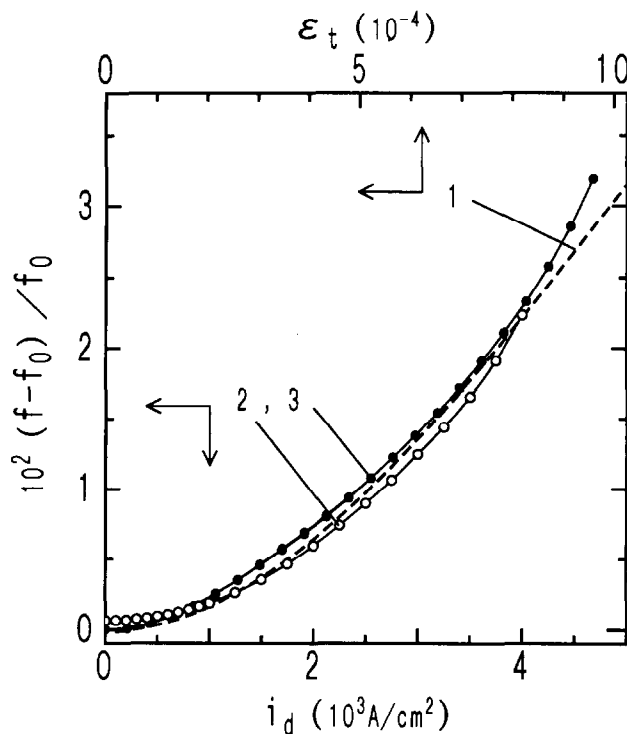


Fig. 4. Curve 1 denotes the ϵ_t dependence of f found for ϵ_t below 10×10^{-4} without PEC observed at RT for a-(Cu₃₀Zr₇₀)_{92.5}Al_{7.5}. Curves 2 and 3 are respective redrawings of curves 5 and 6 shown in Fig. 3, *i.e.* the i_d dependence of the increase in f due to PEC found at $\epsilon_t \approx 10^{-6}$ for a-(Cu₃₀Zr₇₀)_{92.5}Al_{7.5} (see text).

at RT together with a redrawing of the i_d dependence of f for a-CuZrAl shown in Fig. 3 after scaling between the ϵ_t and i_d axes. In Fig. 4 one can see that the general character of the ϵ_t dependence of f without PEC and that of the i_d dependence of f are very similar to each other. It is noted that the ϵ_t range shown is too low to observe the saturation behaviour of f in the ϵ_t dependence of f . Very similar results for the scaling between the ϵ_t and i_d axes can be seen for all the a-alloys (not shown here). This fact suggests that LSD in the deformable units can proceed under PEC. On the other hand, the R measurements for the effect of PEC on SR [2, 3] suggested that a probable collective motion involving many atoms can be modified under PEC. Combining these views, one can surmise that LSD in the deformable units can be proceed through the enhancement of a collective motion involving many atoms under PEC. Although application of the usual electromigration model to the anomalous effect of PEC found for a-alloys is not strictly valid [1–3], it is employed as the first step in the discussion below.

Using the scaling between the ϵ_t and i_d axes in Fig. 4, one may find an effective charge Z^* of the order of 10^5e . On the other hand, when the enhancement of SR under PEC [2, 3] is assumed to be due to a decrease in E_{SR} , one may also find Z^* of the order of 10^5e . That is, the enhancement of atomic processes due to PEC is comparable between these processes. To clarify these issues, however, further work is needed.

4. Conclusions

The effect of passing electric current on the low temperature elastic property of amorphous $\text{Cu}_{50}\text{Ti}_{50}$, $\text{Cu}_{50}\text{Zr}_{50}$, $(\text{Cu}_{30}\text{Zr}_{70})_{92.5}\text{Al}_{7.5}$ and $\text{Pd}_{80}\text{Si}_{20}$ was investigated for current densities i_d below about $5 \times 10^3 \text{ A cm}^{-2}$ using a vibrating reed technique with $\epsilon_t \approx 10^{-6}$ as well as the effect of the strain amplitude ϵ_t without PEC for $\epsilon_t \leq 4 \times 10^{-3}$. After subtraction of the decrease in f due to Joule heating, f shows an increase with increasing i_d for all the a-alloys, where f is the resonant vibrational frequency of the a-alloy specimens. In each a-alloy specimen the character of the i_d dependence

of f observed at RT is very similar to that observed at 80 K, suggesting that the increase in f due to PEC is revealed through an athermal process. The magnitude of the increase in f due to PEC increases in the order of a-PdSi, a-CuZrAl, a-CuTi and a-CuZr. Very similar features are also observed for the ϵ_t dependence of f without PEC for the a-alloys after scaling between the ϵ_t and i_d axes. From these results we surmise that reversible local shear deformation in the deformable units, which was suggested in a previous study on the ϵ_t dependence of f without PEC, can proceed through the enhancement of a collective motion involving many atoms under PEC.

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References

- 1 H. Mizubayashi and S. Okuda, *Phys. Rev. B*, **40** (1989) 8057.
- 2 H. Mizubayashi and R. Takemoto, *Defect Diffus. Forum*, **95–98** (1993) 1187.
- 3 R. Takemoto and H. Mizubayashi, *Mater. Sci. Eng. A*, in press.
- 4 E.C. Stelter and D. Lazarus, *Phys. Rev. B*, **36** (1987) 9545.
- 5 J. Horvath, J. Ott, K. Pfhler and W. Ulfert, *Mater. Sci. Eng.*, **97** (1988) 409.
- 6 F. Faupel, P.W. Huppe and K. Ratzke, *Phys. Rev. Lett.*, **65** (1990) 1219.
- 7 P.W. Huppe and F. Faupel, *Phys. Rev. B*, **46** (1992) 120.
- 8 F. Faupel, K. Ratzke and P.W. Huppe, *Defect Diffus. Forum*, **95–98** (1993) 1175.
- 9 S.K. Sharma, M.-P. Macht and V. Naundorf, *Acta Metall. Mater.*, **39** (1992) 609.
- 10 S.K. Sharma, M.-P. Macht and V. Naundorf, *Defect Diffus. Forum*, **95–98** (1993) 1145.
- 11 H. Mizubayashi, S. Okuda and Y. Ikeda, *Phys. Status Solidi A*, **116** (1989) 633.
- 12 H. Mizubayashi, S. Hoshina and S. Okuda, *J. Non-Cryst. Solids*, **117–118** (1990) 203.
- 13 H. Mizubayashi, M. Kaida, S. Otsuka and S. Okuda, *Mater. Sci. Eng. A*, in press.