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Mechanical and Dielectric Energy Loss Related to Viscous Motion and Freezing of Domain Walls

Y.N. Wang, Y.N. Huang, H.M. Shen and Z.F. Zhang

National Laboratory of Solid Microstructures, Nanjing University, Nanjing 210093, China

Abstract: Three dielectric loss (D) and/or three mechanical loss (Q^{-1}) peaks (P_1, P_2, P_3) and corresponding susceptibilities for ferroelectric KDP and TGS, and ferroelastic LNPP crystal and Cu-Al-Zn-Ni alloy have been investigated in the kHz range. P_1 and P_2 almost overlap for first order phase transitions. P_1 , a very narrow peak, is attributed to the dynamic domains due to fluctuations near T_C . P_2 is recognized to be related to domain walls (DWs). The formulas of Q^{-1} and D caused by the viscous motion of DWs are derived, being fairly coincident with the experimental data of P_2 . The viscous coefficient, pinning force constant of DW and the spontaneous shear strain have been calculated for Cu-Al-Zn-Ni. The susceptibility, ϵ' or $\Delta J/J$, always shows a high plateau below T_C and drops down rapidly nearly consistent with the low temperature side of P_3 peak for poly-domain TGS, KDP and Cu-Al-Zn-Ni. P_3 is thought to originate from the freezing of DWs of high density due to a collective pinning of randomly distributed defects, similar to the pinned-vortex glass model suggested for high T_C superconductors.

1. INTRODUCTION

Often three energy loss peaks below T_c appear in internal friction or dielectric measurements for ferroelastic and ferroelectric materials. Peak P_1 just at T_c is very narrow and difficult to measure, and for 1st order phase transitions, P_1 and P_2 always overlap. So, in many cases only two separated peaks may be observed. The dielectric loss (D) or mechanical dissipation (Q^{-1}) peak P_2 just below T_c is commonly attributed to domain walls [1-8]. The mobility of DWs plays an important role in ferroelectric memory, the shape memory effect, and even in high damping capacity. If a correlation between D (or Q^{-1}) and the amount as well as mobility of domain wall can be found, the memory effect can be studied by using the dynamic method. Snead and Welch [8] have proposed a theory of Q^{-1} in A15 alloys on the basis of the movement of DWs. The result is in accordance with the experimental data at the high temperature side of the peak in pure A15 alloy, but not with other results. Firstly we need to find out experimentally the relation between the loss D (or Q^{-1}) and the DWs. $La_{1-x}Nd_xP_5O_{14}$ (LNPP) undergoes an $mmmF2/m$ 2nd order ferroelastic transition at 141 °C. Thus, in a LNPP single crystal, phase interfaces are absent, only twin DWs occur in the ferroic phase, and there is a simple domain structure (all walls are parallel planes often crossing the whole sample) making the above-mentioned investigation possible. From studies on LNPP, we obtained a relation between Q^{-1} and the amount of DWs [9]. To investigate the mobility of DWs, the frequency dependence of the energy loss peak P_2 for KDP, TGS, Cu-Al-Zn-Ni and Mn-Cu has been investigated by both experimental and theoretical analysis [10-11], from which the viscous coefficient and the pinning force constant of DWs can be acquired [12]. These parameters are important for studies on problems in memory devices, such as fatigue life or stabilization and switching rate.

A third peak P_3 often occurs at a lower temperature than of P_1 and P_2 , which is also related to DWs. The dielectric loss peak P_3 in KDP showing freezing effect of DWs has been studied in detail [13-23]. The dielectric constant (ϵ') parallel to the ferroelectric axis is anomalously high as compared to the value predicted by the phenomenological Landau theory [13-18]. On cooling, ϵ' shows a plateau-like

temperature dependence below T_c , and then at a lower temperature T_f abruptly decreases down to its phenomenological value, forming a 'kink'. The peak P_3 covers the temperature range of the 'kink'. The anomalously large dielectric constant in the temperature interval $T_f < T < T_c$ (the "plateau" region) is believed to be due to the motion of the domain walls. The abrupt decrease of the dielectric constant is then attributed to the freezing of domain walls. At least three models to describe this freezing effect have been proposed: by those (1) Bornarel [14,18,22], (2) Fedosov and Sidorkin [15] and (3) Kuramoto and Nakamura [16,19,21,23]. These models can explain some experimental results, but some questions are still open. The Vogel-Fucher relation $\tau = \tau_0 e^{U/(T-T_0)}$ [24], the criterion for relaxation due to the freezing process, has been measured only in the high-frequency range (10^6 Hz) [16]. The low frequency range (10^3 Hz) reveals a static hysteresis character [17], although ϵ' goes down rapidly at T_f . In this paper, we give a detailed results about the frequency dependence of P_3 in the kilo-Hertz range, and propose a new model—collective pinning of DWs.

As we know, there always appears a large peak at a lower temperature than of the phase transition peak (P_1+P_2) in shape memory alloys. Only the peak in Mn-Cu alloy [25] has been confirmed to be attributed to the twin walls, but those in NiTi [26] and Cu-Zn-Al [27] are usually suggested to be caused by dislocations together with point defects. In view of the large peak height and peak width, we speculated it likely originate from the DWs interacting with point defects. In this paper, the primary results on P_3 with the possible freezing effect for Cu-Al-Zn-Ni will be reported.

2. EXPERIMENTS AND SPECIMENS

The internal friction and elastic constant in the low kilo-Hertz range were measured by a two-node clamped-reed vibration technique with electrostatic drive and detection device, and measurement in the 50-150kHz range was carried out on a Marx three-component resonator. The dielectric loss (ϵ'') and the dielectric constant (ϵ') were measured on a GR1615-A Capacitance bridge with sample size about $10 \times 10 \times 1 \text{ mm}^3$. A torsion pendulum was used for low frequency measurements. The samples reported in this paper are listed in the table:

	chemical formula or composition	type of phase transition	order of phase transition	temperature of phase transition
LNPP	$\text{La}_{1-x}\text{Nd}_x\text{P}_5\text{O}_{14}$	ferroelastic	2nd order	141 °C
TGS	$\text{NH}_2(\text{H}_2\text{COOH})_3 \cdot \text{H}_2\text{SO}_4$	ferroelectric	2nd order	322K
LATGS	α -alanine TGS	ferroelectric	2nd order	323K
KDP	KH_2PO_4	ferroelectric	weak 1st order	122K
Cu-Al-Zn-Ni	Cu-28.76at%Al-4.76at%Zn-2.33at%Ni	thermoelastic martensitic	1st order	$A_s = 60^\circ\text{C}$ $A_f = 95^\circ\text{C}$
Cu-Zn-Al-Mn	Cu-22.5at%Zn-9.3at%Al-1at%Mn	thermoelastic martensitic	1st order	$A_s = 50^\circ\text{C}$ $A_f = 76^\circ\text{C}$
Mn-Cu	Mn-20wt%Cu	thermoelastic martensitic	1st order	

3. EXPERIMENTAL RESULTS AND ANALYSIS

3.1 Energy Loss Spectrum for LNPP, KDP, TGS, LATGS, Cu-Zn-Al-Ni and Cu-Al-Zn-Ni

3.1.1 $\text{La}_{1-x}\text{Nd}_x\text{P}_5\text{O}_{14}$ Crystal

Fig. 1. Density of DWs N and Q^{-1} vs T for LNPP

Fig. 2. Q^{-1} vs T before (●) and after (○) removing the 'frozen domains in LNPP'

The LNPP samples were cut along the c -axis from a single crystal of composition $x=0.9$. Measurements in the 50-150 kHz range were carried out in a special furnace in which domains of a sample can be observed in situ with a polarization microscope during Q^{-1} measurements [9]. Two Q^{-1} peaks ($f \approx 50 \text{ kHz}$) at $\sim 134^{\circ}\text{C}$ (P_2) and 141°C (P_1), are shown in Fig. 1(b) [9]. X-ray analysis and measurement of the elastic constant C_{55} [9] show that the peak temperature of P_1 is located just at T_C (141°C), whereas no structural change was detected at the peak temperature of P_2 . In situ observation of DWs during the Q^{-1} measurement show that the density of DWs, N , increases with T , nearly proportional to $(1-T/T_C)^{-1}$ [Fig. 1(a)], so-called domain coarsening (or thinning). The peak temperature of P_2 corresponds to the temperature where N begins to increase rapidly ($N=50 \pm 10 \text{ mm}^{-1}$). Both P_2 and the N - T curve show remarkable thermal hysteresis. The thermal hysteresis and the abrupt formation of a new domain, just like a martensite plate, indicate the coarsening (or thinning) of domains has the character of first-order phase transitions. This is also confirmed by the heat capacity results [11].

The peak P_1 that appears at 141°C has no thermal hysteresis within the resolution of 0.1 K . This means that it must originate from a second-order phase transition. However, among the conventional theories of energy dissipation of second-order phase transitions, such as the Landau-Khalatnikov theory of relaxation of the order parameter and the theory of fluctuation dissipation [9], it is generally predicted that Q^{-1} is too small to be detected in the kilohertz range. So, the peak P_1 at low frequency has been attributed to the stress-induced preferred orientation of domains of the dynamic new phase [1,9] due to fluctuation near T_C .

Another peak P_3 occurs at temperatures below that of P_1 and P_2 (Fig. 2). If a small stress is applied to a LNPP sample at a higher temperature, the coarsening is frustrated, i.e. a large number of domains are frozen down to a lower temperature [9]. As a result, P_2 is suppressed, while P_3 becomes much larger. So, P_3 is still considered to be induced by domain walls, but with a different mechanism from P_2 . At higher temperatures, the domain walls cannot be pinned by point defects, so the dissipation of P_2 may come from the interaction between DWs and the lattice. By contrast, P_3 , at a lower temperature, which is sensitive to defects, may be ascribed to the thermal assistant motion of pinned DWs.

3.1.2 KDP and TGS Crystal

The dielectric constant (ϵ') and dielectric loss (ϵ'') as functions of temperature for KDP, DKDP and TGS crystals were measured at various frequencies. Fig. 3 shows the results for TGS and LATGS, with three loss peaks P_1 , P_2 and P_3 . P_1 which occurs just at T_C , often failed to appear in the measurements due to its extreme narrowness. Similarly to that in LNPP, it is attributed to the dynamic domains of the new phase due to fluctuation near T_C . P_2 and P_3 in TGS were recognized to be related to DWs, as they disappear in monodomain LATGS, but the mechanisms are still open. The peaks P_2 and P_3 in KDP and DKDP, similar to that in Fig. 3 are also confirmed to originate from domain walls (Figures omitted here).

Fig. 3. Dielectric measurements for TGS: (a) ϵ' vs T at various frequencies; (b) ϵ'' vs T at various frequencies

120°C for 40 min., then cooling down. A sharp peak (P_1, P_2) is due to martensitic transformation, and a broad peak P_3 appears at a lower temperature.

As mentioned in the introduction, a peak similar to P_3 appears in many shape memory alloys, which needs confirmation that it originates from domains. To obtain various domain structures, different thermo-mechanical treatments for Cu-Al-Zn-Ni sample were used. Firstly, the sample was heated at 1123 K for 15 min, followed by quenching into oil at 393 K for 45 min, then by cooling to room temperature in air. The internal frictions after such treatment is shown by curve I in Fig. 5, revealing peak P_3 . Afterward, we stretched the sample by 3% at room temperature. Then it was heated first at 873 K for 1.5 hr, second at 1123 K for 1.5 hr, followed by the same cooling process as that for curve I.

The dielectric constant ϵ' vs T curves for KDP, DKDP and TGS show the same characteristics: a high plateau in a rather large temperature range, rapid drop at a certain lower temperature (T_f), forming a kink, below which ϵ' remains unchanged. The kink region corresponds closely to the low temperature side of the peak P_3 . It is evident that the peak P_3 is associated with a freezing effect.

Because KDP crystal is both ferroelectric and ferroelastic, the internal friction must have the same characteristics as the dielectric measurements. Q^{-1} for KDP, measured at 2 kHz, does exhibit three peaks similar to the dielectric peaks [10].

3.1.3 Cu-Zn-Al-Mn and Cu-Al-Zn-Ni Alloy

Thermal-elastic martensitic transformation in alloys such as Cu-Zn, Cu-Al, Ni-Ti, Mn-Cu and etc. possesses almost all the characteristics of a ferroelastic transition except that the thermal hysteresis is generally larger than that in oxide crystals.

Fig. 4 shows the internal friction spectrum for a Cu-Zn-Al-Mn alloy. The sample was heated at 850°C for 10 min., followed by quenching in oil at

Q^{-1} after the 2nd treatment is shown by curve II (Fig. 5). Compared to curve I, the peak height decreases much. Metallographic observation showed that the domain size is much smaller (therefore more domain walls) for the first treatment of the sample than for of the second. It may be considered this peak is also associated with DWs.

These results indicate that there generally exist two relaxation processes connected with domain walls: P_2 occurs near the phase transition temperature T_C and drops rapidly with temperature closely up to T_C ; P_3 appears near the freezing temperature T_f , dropping to zero when the temperature goes to T_f . It is reasonable to speculate that P_2 is caused by an interaction of DWs with the lattice and P_3 originates from the interaction of DW with defects. However, the detailed mechanisms need further study as presented below.

3.2 P_2 Peak and Viscous Motion of Domain Wall

Fig. 4. Q^{-1} and f^2 vs T for Cu-Zn-Al-Mn after treated as described in text. $f=0.76\text{Hz}$

Fig. 5. Q^{-1} vs T (peak P_3) for Cu-Al-Zn-Ni: I. thin domain; II. coarse domain. $f=1\text{Hz}$

3.2.1 KDP and TGS: To get more information about the mechanism of the P_2 peaks, the dependence of both Q^{-1} and the dielectric loss on frequency was surveyed further. It was discovered that the high temperature sides of the peaks are almost independent of frequency, but the low temperature sides are frequency dependent and the positions of the peaks move to higher temperature as the frequency increases (Fig. 6,7), but the relaxation time does not obey the Arrhenius relation [10]. The similar figures for TGS were published in [10]. This behavior is quite different from that of a thermally activated peak. The peaks may be considered to be related to a complicated viscous movement of DWs.

3.2.2 Cu-Al-Zn-Ni and Mn-Cu Alloy:

The Cu-28.76at%Al-4.76at%Zn-2.33at%Ni sample was kept at 830°C for 10 minutes, followed by air quenching to room temperature. The characteristic temperatures on heating are $A_s=60^\circ\text{C}$ and $A_f=95^\circ\text{C}$. An internal friction peak appears at 78°C , which is found to be independent of the measuring frequency (0.7 kHz and 1.98 kHz) [12], similar to the results of the low frequency internal friction associated with the martensitic transformation, revealing a mechanism of static hysteresis type. Moreover, the results from 50 to 150 kHz [Fig. 8] show an unsymmetrical internal friction peak at the same temperature as that measured near 1 kHz. Fig. 9 shows the results for Q^{-1} and $\Delta E/E$ for a Mn-Cu alloy which was heated at 950°C for 20min and quenching in water. The peak temperature (T_p) in Fig. 8 and Fig. 9 increases with measuring frequency and the relationship between $\ln f$ and T_p^{-1} disobeys the Arrhenius relationship, which indicates that the internal friction peak is neither a static hysteresis loss nor a thermally activated Debye relaxation, very similar to those of the internal friction peak P_2 in KDP and TGS crystals.

3.3 Theoretical Analysis

As mentioned above, P_2 is thought to be caused by the interaction between DWs and the lattice. There are two mechanisms: one is due to static friction, just as the Peierls force on the dislocation; the other is dynamic friction due to the viscous motion of DWs in lattice. Here we consider the latter case only, because the loss is frequency dependent.

Fig. 6. Q^{-1} vs T for various f in KDP.
Theor. (—) with $A/k=5.4 \times 10^{-2}$ s, $B=9.5$ K.

Fig. 7. D vs T for various f in KDP.
Theor. (—) with $A/k=1.3 \times 10^{-2}$ s, $B=9.5$ K.

There are five forces acting on domain walls [10]: (1) the configuration force F_C by the external field, (2) the interaction force between the nearest-neighbor DWs F_I , (3) the viscous force $-\Gamma \dot{x}$ as DWs move in the lattice, where Γ is the viscosity coefficient, (4) the recovering force $-k_0 x$ due to the pinning defects, where k_0 is the force constant, and (5) the Peierls force which, although existing, is relatively small and can be neglected here.

Letting M be the effective mass per unit area of DWs and x be the displacement of the wall under the applied stress, the equation of motion of a DW is

$$M\ddot{x} + \Gamma \dot{x} + k_0 x - F_I = F_C \quad (1)$$

It is easily deduced from the energy change due to the displacement of the interfaces under the applied force that [10]

$$F_C = 2\varepsilon_{ij}^{(s)} \sigma_{ij} \quad (2)$$

$$F_I = -2C_{ijij} N [\varepsilon_{ij}^{(s)}]^2 x = -k' x \quad (3)$$

$$\varepsilon_{ij}^{(d)} = \varepsilon_{ij}^{(s)} x / d \quad (4)$$

where $\varepsilon_{ij}^{(s)}$ is the spontaneous shear strain, C_{ijij} is the corresponding shear modulus, σ_{ij} is the effective shear stress which can drive the DWs to move, $\varepsilon_{ij}^{(d)}$ is the shear strain caused by the displacement x of DW under an applied stress, N is the average number of parallel interfaces per unit perpendicular distance. So, $N=d^{-1}$ where d is the distance between the nearest-neighbour interfaces.

When $\sigma_{ij} = \sigma_0 e^{i\omega t}$ with $\omega^2 \ll k/M$, where $k=k_0+k'$, Q^{-1} is

$$Q^{-1} = \frac{2N [\varepsilon_{ij}^{(s)}]^2}{Jk} \frac{\omega \tau}{1 + \omega^2 \tau^2} \quad (5)$$

where $\tau = \Gamma/k$ is the relaxation time, and J is the compliance.

The expression for Γ obtained by Combs and Yip[28] with constants A and B is

Fig. 8 Q^{-1} vs T for Cu-Al-Zn-Ni at different f . — Curves drawn for $A/k=7.0 \times 10^{-4}$ sec., $B=130$ K.

Fig.9. Q^{-1} vs T for Mn-Cu at different f . — Curves for $A/k=1.5 \times 10^{-4}$ sec., $B=110$ K.

$$\Gamma = Ae^{-B/(T_c-T)} \quad (6)$$

Similarly, the dielectric loss D ($=\epsilon''/\epsilon'$) can be deduced to be

$$D = \frac{2NP_s^2/k}{[\chi_{mm}(\chi_{mm} + 2NP_s^2/k)]^{1/2}} \frac{\omega\tau}{1 + \omega^2\tau^2} \quad (7)$$

where P_s is the spontaneous polarization, χ_{mm} is the susceptibility of monodomain crystal along the direction of P_s .

For a second order, or a weak first order, phase transition, $N \approx N_0/(T_c - T)$, P_s^2 [$(\epsilon_{ij}^{(s)})^2$] $\propto (T_c - T)$. So, NP_s^2 and $N[\epsilon_{ij}^{(s)}]^2$ are nearly independent of T . If we can measure N and P_s (or $\epsilon_{ij}^{(s)}$) at a temperature within the peak, we can get k from (5) or (7). By the computer simulation of Q^{-1} or D curves, A/k and B are easily obtained, thus Γ can be calculated. Theoretical curve shown in Figures 6, 7, 8 and 9 are fairly close to the experimental data. A/k and B for KDP, Mn-Cu and Cu-Al-Zn-Ni are given in the captions. The value of B for KDP obtained from the Q^{-1} data is the same as from the dielectric loss curve, and the theoretical curves are in good agreement with the data measured at various frequencies, indicating that the mechanism of viscous motion of DW is reasonable. The high temperature side of peak P_2 drops rapidly to zero at T_c , which is easily explained. The spontaneous strain or polarization tends to zero as $T \rightarrow T_c$, so the difference between the domain wall and the domain interior becomes small, thus the scattering by phonons to DWs tends to 0, then the viscous coefficient, $\Gamma \rightarrow 0$ and the relaxation time $\tau = (\Gamma/k) \rightarrow 0$ rapidly [see formula (6)], as does Q^{-1} (or D).

For Mn-Cu and Cu-Al-Zn-Ni the agreement between theory and experiment is fairly good on the low temperature side, but not on the high temperature side. The reason may be that the change of N on high temperature side is not so simple as for KDP due to the 1st order phase transition. However, the starting points are nearly at the same temperature A_f for different frequencies, quite different from the thermal activated relaxation peak, for which the starting point should shift in parallel with the peak temperature. The other important point is that the Q^{-1} peak of Mn-Cu, as well as Cu-Al-Ni alloys, near

1 kHz (see Ref. [12], Fig. 1) is much narrower than at ~ 100 kHz. That means the variation of N is concentrated in temperature range near the peak temperature T_P . The broadening of the low temperature side of peak P_2 at ~ 100 kHz is due to the viscous motion of DWs. So, we can use the data in this region to calculate Γ and k . As an example, Γ and k for Cu-Al-Zn-Ni have been calculated as follows [12]. The average density of DWs N is counted to be $1.2 \times 10^5/\text{m}$. The elastic modulus is calculated to be $1.5 \times 10^5 \text{ MPa}$ from the resonant frequency. The spontaneous strain was obtained by using the results of x-ray diffraction in Cu-Al-Zn-Ni. The structure of the orthorhombic r_1' martensite is Cu_3Ti type with lattice parameters, $a=0.4382 \text{ nm}$, $b=0.5356 \text{ nm}$, $c=0.4222 \text{ nm}$ and the structure of the parent phase is DO_3 type with lattice parameter $a_0=0.5836 \text{ nm}$, which is similar to the results obtained by K. Otsuka et al [29]. The martensitic transformation belongs to the $m3mFmmm(ss)$ ferroelastic phase transition, that means the principal axes in the r_1' martensitic phase are parallel to the directions $[101]$, $[010]$, $[10\bar{1}]$ of the β_1 cubic phase. According to the Neumann principle and the symmetry relationship between the β_1 phase and the r_1' phase, the strain states caused by a martensitic phase transformation can be calculated. There are only 4 nonzero components of strain, i.e. $\varepsilon_{11}=\varepsilon_{33}$, ε_{22} and ε_{13} , and six effective strain states $X(S_k)$ ($k=1-6$).

Using Aizu's definition [30] for the spontaneous strain tensors,

$$P(S_i) = X(S_i) - \frac{1}{6} \sum_{k=1}^6 X(S_k) \quad (8)$$

where $P(S_i)$ ($i=1-6$) denote six effective spontaneous strain states, the spontaneous strain tensors can be calculated, from which the spontaneous shear strain for martensitic transformation should be $\varepsilon_{ij}^{(s)} = 2\varepsilon_{13}$.

In the principal axes coordinate system of the r_1' orthorhombic phase, we can obtain the strains due to martensitic phase transformation based on the results of x-ray diffraction experiments.

$$\begin{aligned} \varepsilon_{11}' &= (a - \frac{\sqrt{2}}{2}a_0) / \frac{\sqrt{2}}{2}a_0 = 0.061, & \varepsilon_{22}' &= (b - a_0) / a_0 = -0.082 \\ \varepsilon_{33}' &= (c - \frac{\sqrt{2}}{2}a_0) / \frac{\sqrt{2}}{2}a_0 = 0.022, & \varepsilon_{ij}' (i \neq j) &= 0 \end{aligned}$$

Because the principal axes in r_1' phase are parallel to the directions $[101]$, $[010]$, $[10\bar{1}]$ of β_1 cubic phase respectively, the spontaneous shear strain in the β_1 cubic phase coordinate system is

$$\varepsilon_{ij}^{(s)} = 2\varepsilon_{13} = 2 \sum_{i,j=1}^3 a_{1i} a_{3j} \varepsilon_{ij}' = 0.0388.$$

where $a_{11} = a_{31} = a_{33} = \sqrt{2}/2$, $a_{13} = -\sqrt{2}/2$, $a_{12} = a_{32} = 0$. Using the values of A/k and B given under Fig. 9, we obtain

$k=1.6 \times 10^{15} \text{ N/m}^3$, $k_0=1.54 \times 10^{15} \text{ N/m}^3$ and $\Gamma=11.2 \times 10^{11} \exp[-130/(363-T)] \text{ Kg/m}^2\text{s}$.

3.4 P_3 Peak and Freezing Effect (or Glass Transition) of Domain Walls

The criterion of a freezing effect is the Vogel-Fulcher relation, $\tau = \tau_0 \exp[E/(T-T_f)]$. Taking the data of ε' and ε'' of the P_3 peak for TGS (Fig. 3), the V-F plot and Arrhenius plot is shown in Fig. 10. It indeed obey the V-F relation, the same as for KDP (Figure omitted).

For Cu-Al-Zn-Ni which was heated at 830°C for 10min and quenched in water, Q^{-1} and $\Delta J/J$ were measured at 500 Hz (Fig. 11). $\Delta J/J$, corresponding to ε' , manifests a large kink near P_3 . That means the modulus M shows a hardening process near P_3 , imply the freezing of DWs. Tests of the V-F relation are in progress.

Three mechanisms have already been proposed to explain the freezing phenomenon. The first Bornavel's[14], is based on quasidislocations of edge type that are known to exist in the DW. He suggests that the critical shear stress to move the quasidislocations depends strongly on temperature. The second, proposed by Fedosov and Sidorkin [15] gives two possible configurations of the DW, T_f being the temperature of transition from one to the other. The structure of a DW at $T < T_f$ is the same as paraelectric phase, i.e. $P_s = 0$. The third, proposed by Kuramoto[16] is that the dipole reversal in the DW has a certain size of the cooperative region (cluster), the restriction for the dipole reversal becomes rigid as the spontaneous polarization reaches its saturated value, leading to the freezing of DWs. None of these three has been brought to satisfactory explanation. As Kubinec put it in 1995 [31], "although the domain freezing phenomenon has been known for a long time and was intensively studied in recent years, the problem has not been solved yet".

A new model is needed that must take account of: (1) T_f increases with increasing defect density on irradiation [17], (2) T_f increases with decreasing thickness t of sample [22], i.e. with the increasing of the density of DWs N , due to $N \propto t^{-1/2}$ [32]. Therefore the new model must involve not only pinning of defects but also interactions between DWs. Inspired by the model of the collectively-pinned flux glass [33] we propose a model of collectively-pinned domain wall glass. At low temperature, there exist the collective-pinning to DWs between random distribution defects, i.e. the pinning of defects in a certain spatial range is correlative; at the same time, due to the interaction between DWs, an effective pinning region ΔS forms, i.e. the pinning in ΔS is correlative or a local order forms. The collective pinning energy U_{CP} is much larger than the individual pinning energy U_{IP} , leading to the freezing of DWs or a glass transition at T_f . At the temperature much higher than T_f , the individual pinning becomes more and more and then the thermal activated Arrhenius mechanism may contribute to the relaxation.

Fig. 10. τ vs $1/T$ and τ vs $1/(T-T_f)$ (inset) for P_3 of TGS, data taken from Fig.3.

Fig. 11. Q^{-1} and $\Delta J'/J'$ of CuAlZnNi as functions of T at $f=500\text{Hz}$.

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