INTERNAL FRICTION ASSOCIATED WITH DOMAIN WALLS AND FERROELASTIC PHASE TRANSITION IN LNPP

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Abstract – Two internal friction peaks (50 - 100kHz) have been measured in LNPP near its transition temperature. The peak at $T_C=141°C$ is associated with the ferroelastic transition where the elastic constant C_{55} vanishes and the peak at a lower temperature is attributed to the motion of domain walls. The variation of domain walls and its relation to Q^{-1} were obtained by in situ measurements. The mechanisms of the peaks are discussed.

I - INTRODUCTION

Like martensitic phase transition, ferroelastic transition is also associated with shear strains that can be re-oriented by applied stress. Several experimental results /1,2/ show that the internal friction in the first order ferroelastic phase transition is also similar to that of martensitic transition. The idea that the internal friction under isothermal condition is due to the stress-induced motion of coherent boundaries /3,4,5,6/ has been widely accepted. But so far, there is no report on the correlation, especially on the quantitative correlation between Q^{-1} and boundaries, nor the investigation on the internal friction associated with second order ferroelastic transition

In $La_{1-x}Nd_xP_5O_{14}$ (LNPP), there is a phase change from mmm to 2/m symmetry at 140±2°C. According to Aizu /7/, it is a pure ferroelastic transition with $\pm e_{13}$ as the order parameters /8/. More detailed studies /9,10,11,12/ show that the characteristics of LNPP are of second order. There are two types of domains in the ferroic phase of LNPP crystal: a- and b-domains with their walls perpendicular to c and a axis respectively. Generally, only domains of a-type that traverse all the crystal are present /10/, so the domain configuration is much simpler. This, togather with the fact that coherence is higher in ferroelastic substances than in martensite, makes possible the quantitative investigation of the correlation between internal friction and domain walls, that will be helpful for the clarification of mechanisms of internal friction in martensitic transition as well as in high damping.

II - EXPERIMENTAL PROCEDURE

Marx three-component-resonator method was used to drive and detect the longitudinal vibration along the c axis of samples that had been cut from single crystals to fit the frequency of 50, 86 and 100kHz respectively. Q^{-1} measurements were carried out in a special self-made furnace in which domains can be observed in situ with a polarization microscope during heating and cooling. C_{55} was measured with pulse-echo -overlap method. All measurements were carried out on $La_{o.1} Nd_{o.9} P_5 O_{14}$ samples.

III - RESULTS AND ANALYSIS

Experimental results show that domains increase proportionally to $(T_C-T)^{-1}$ (Fig. 1). On heating, domains increase with temperature and internal friction increases simultaneously, which leads to the ascription of the internal friction peak P_2 to newly appeared domains. When domains begin to increase rapidly and become nearly periodically distributed in the crystal (40-80 walls per mm), the internal friction reaches a peak (P_2) value with a minimum of the resonant frequency f_r, and then decreases as domains continue to increase with temperature. The number of domains becomes larger and larger as temperature approaches T_C, and goes beyond the resolution of the microscope. Domains finally disappear at T_C. A very narrow internal friction peak (P_1), as well as very rapid changes of resonant frequency can be measured at T_C, corresponding to the minimum of C_{55} (Fig. 2). Because the width of P_1 is only about 0.1°C, it is very difficult to make an accurate measurement of the peak height. On cooling, there is a 2-3°C hysteresis for P_2 but no hysteresis for P_1 within the experimental accuracy (also see Fig. 2). Results are the same in other samples. It is essential that there is no anomaly in C_{55} except for the rapid mode softening at T_C (Fig. 2a), which suggests that P_2 is not associated with the ferro-elastic phase transition. In order to study the mechanism of P_2, a small stress was applied on sample 1 on heating. Consequently, a large number of domains was "frozen" at room temperature in most portion of the crystal. As a result, P_2 almost disappeared, and the corresponding minimum of f_r totally disappeared while a broad internal friction peak P_3 was measured at a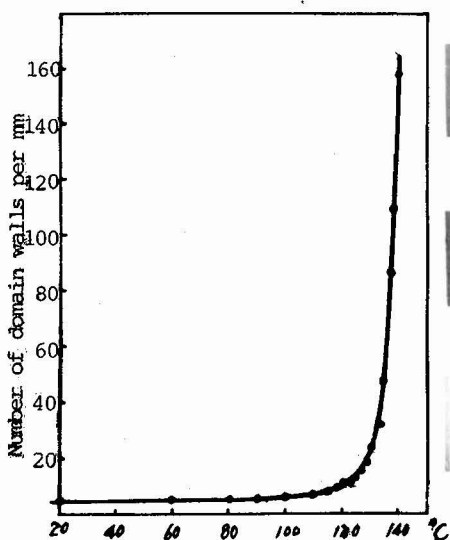bout 60°C (Fig. 3). After removing the "frozen" domains with a shear stress, P_2 and f_r became the same as before whereas P_3 was essentially eliminated (see curve b Fig. 3). Obviously, P_1 remains unchanged as can be seen in the three curves in Fig. 3.

In addition, the measurement made at $3f_r$ on sample 1 and at different heating rate on sample 2 (86kHz) show no significant change either in peak height or in peak temperature. On the basis of these facts, it is reasonable to consider that P_2 is a static hysteresis loss due to newly appeared domains, and P_3 is due to the "frozen" domains, the mechanism of which is not yet studied. It should be noted that though P_2 is attributed to the motion of domain walls, it is quite different from martensitic transition, because the peak does not correspond to the minimum of the soft mode. The minimum of f_r is due to the modulus defect.

The reason why the internal friction reaches a peak value instead of varying monotonously with the number of domains may be
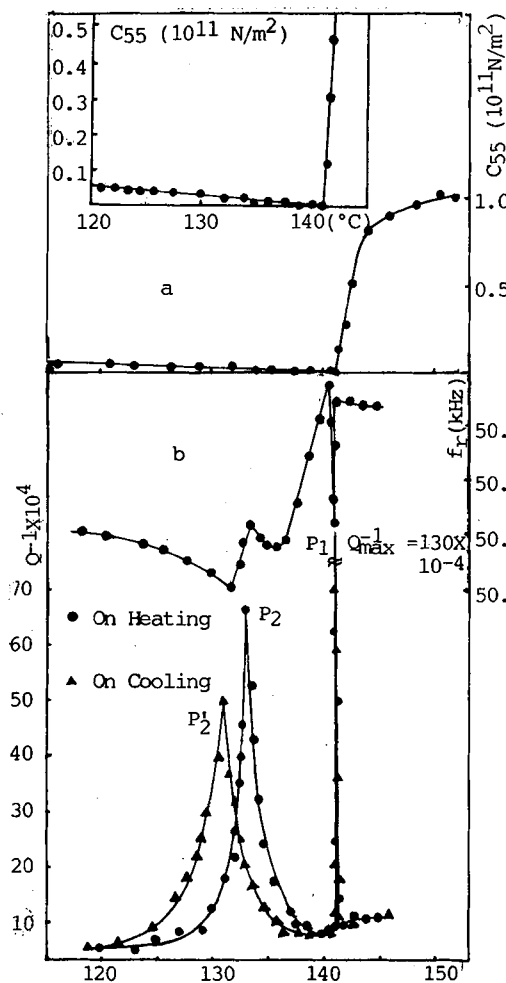

Fig. 1 Number of domain walls per mm vs. temperature. The magnification of the microscope is 40X.

Fig.2 The variation of the elastic constant C_{55} (a), Q^{-1} and the resonant frequency f_r of sample 1 (b) with temperature.
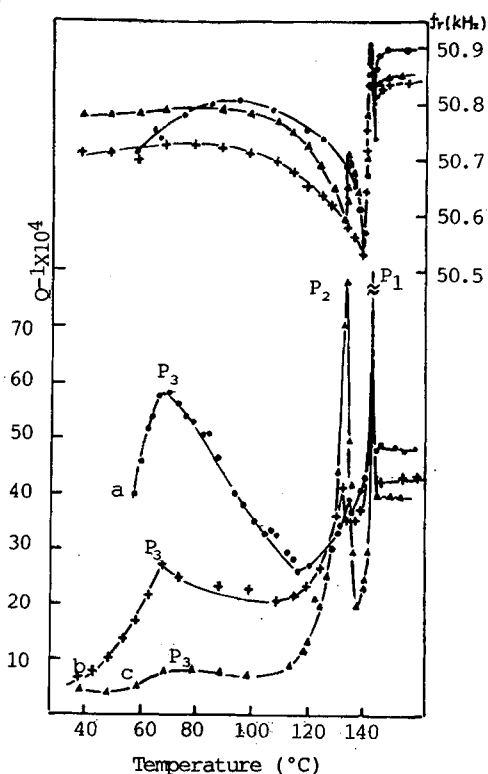
Fig. 3 Q^{-1} and f_r of sample 1 vs. temperature. Curves a and c: before and after removing the "frozen" domains. Curve b: "frozen" domains are introduced again.

Fig. 4 Lateral motion of domains a and b due to the formation of domain c.

interpreted in terms of the interaction between domain walls. Domains are more mobile when the distance between two walls is large enough that the interaction between them is negligible, however, when the distance becomes smaller so that the strain field induced by the walls partly overlap, the mobility of the walls is then reduced by the interaction. That accounts for the decrease of Q^{-1}. It has been observed in LNPP that domains move laterally as a result of the squeeze by the formation of new domains as illustrated in Fig. 4, at a relatively larger domain density. The phenomenon has not been observed at smaller domain density. The reduction of domain wall mobility due to interaction has also been observed in some other ferroelectric substances /13, 14/.

IV - DISCUSSION

1) In LNPP, the number of domains is proportional to $(T_C-T)^{-1}$, that is different from martensitic phase transition in which twinns and variants increase with temperature lowered. The correlation between domains and temperature in LNPP can be

understood by considering the surface energy of the ferroic phase. According to /11/, the presence of ferroelastic domains causes deformation of crystal surfaces, that leads to the increase of surface energy, a process somewhat similar to the cases of ferroelectricity /15/ and ferromagnetism /16/. The increment of the elastic surface energy is given by /17/:

$$E=(K/N)\,\eta^2 \tag{1}$$

where K is a constant, N is the number of domain walls per unit volume and η is the order parameter which is equal to $\pm\,e_{13}$ for the present case. Taking into account the surface and wall energy, the total free energy should be witten as:

$$F=(a/2 + K/N)\,\eta^2 + b/4\,\eta^4 + \sigma\,N \tag{2}$$

where σ is the energy of a single wall which can be calculated with the commonly used method /15/. Since the assumption that the coefficient before η^2 should be proportional to $(T-T_C)$ is still valid, it should also should be true that $(a/2+K/N)=a'(T-T_C)$, that gives $N=A/(T_C-T)$ with $A>0$. More detailed calculation is underway. 2) The appearance of P_l at T_C is in conflict with the fact that in second order transition, the internal friction can be measured only in MHz range. The possible explanation is that C_{55} changes so rapidly (it can hardly be followed at T_C) that it can be treated as a sudden change. According to Eshelby /18/, an energy loss is incurred by the sudden change of elastic constant(s):

$$E=(1/2)\int(C'_{ijkm}- C_{ijkm})\,e'_{ij}\,e_{km}\,dv \tag{3}$$

where C_{ijkm} , e_{km} and C'_{ijkm} , e'_{ij} are the elastic constants and strains before and after the sudden change respectively. Actually, a small discontinuity with 0.2°C is not ruled out yet /12/.

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