Dissipation function of the first-order phase transformation in solids via internal-friction measurements

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Reconstruction and displacement of crystal structure and motion of the phase interface induces dissipation of energy, and latent heat appears during a first-order phase transition (FOPT) in solids. In this series of investigations, we first express the energy dissipation as a function of four physical parameters. Since there are more unknowns than the number of equations, we introduce four more equations describing the dynamics of the system on which internal friction (IF, the dissipation of vibration energy) measurements are conducted. Via IF measurements during FOPT, we can then calculate the relevant four parameters and hence the dissipation function. We have completed the first step in establishing a phenomenological theory to describe FOPT in solids.

I. INTRODUCTION

It is well known that the behavior of interfaces (phase interface and domain wall) under an applied field is, in the broad sense, a generalized first-order phase transformation (FOPT) and is an important aspect for both basic physics and practical applications. A lot of physical properties of materials, such as coercivity, susceptibility, and hysteresis of ferromagnetic material or ferroelectric material, hysteresis of phase transformations (PT's), shape memory effect, superelasticity of thermoelastic martensitic transformations (MT's), etc., are closely associated with the moving characteristics of the interface.

During the FOPT, two phases coexist initially. In the process of a FOPT, migration of the phase interfaces occurs and the process is completed within a limited region when the phase interface disappears.[1,2] A phenomenological theory of phase interface dynamics should be fruitful for understanding the common crucial features of the FOPT. In going through a typical FOPT in solids, there is a complete lattice reconstruction because the symmetry relationship between new and parent phases may not exist in general for a FOPT in solids; consequently, the order parameter is not defined generally in such a transition. On the other hand, in a second-order phase transition, the order parameter is well defined. As a consequence, mean-field theory cannot be applied in general to the FOPT. It is interesting to note that even up to very recently, though there are attempts to apply the Landau phase transition theory (including classical Ginzburg-Landau theory) to certain phase transitions (see, e.g., Refs. 3–10), it is generally recognized that such a theory cannot function as a general theory for the FOPT in solids. Moreover, the crucial characteristics of the FOPT are (i) the dissipation of energy, usually in the form of acoustic emission, occurring in the lattice reconstruction process; (ii) the existence of a latent heat during the transformation; (iii) the coexistence of new and parent phases, which leads to various hystereses. All these crucial features signify that the FOPT is an irreversible thermodynamical process which is very difficult to deal with experimentally and mathematically. For this reason, we believe much effort has been expended to investigating the characteristics related to nucleation and growth dynamics, rather than to analyzing the energy dissipation in the FOPT. In view of the mentioned difficulties, we attempt first in this investigation to derive an explicit phenomenological expression for the energy dissipation via an internal friction measurement, which includes a number of unknown parameters, during the FOPT. Through internal friction measurements, we were able to obtain specific values for the relevant parameters and, hence, the dissipation energy. This investigation indicates that analysis of the "dissipation function" would provide a very useful methodology in the study of the FOPT in a relatively general way under the phenomenological regime.

The measurement of internal friction (IF), Q^{-1}, during the FOPT is a powerful experimental method for the investigation of the relevant dissipation behaviors. An internal friction peak associated with the FOPT at low-frequency range has been observed in a number of FOPT systems, such as FeMn,[11–13] TiNi,[14,17] AuCd,[18,19] CuZnAl,[20,21] VO_2,[22] Co,[23] and $BaTiO_3$.[24] This IF peak is characterized by the following: (i) the peak height increases with an increasing value of $\dot{T}/\omega$ (where $\dot{T}=|\partial T/\partial t|$ and ω is the frequency of the measuring stress), but the relation is not linear; (ii) the IF is indepen-

dent of the measuring strain amplitude A_ε, which is of the order of 10^{-5}; (iii) a fairly high value of Q^{-1} can be observed only if $\dot{T}\neq 0$. In fact, it is well established that the IF curve shows a peak when T is varied. However, if Q^{-1} is measured suddenly at constant temperature ($\dot{T}=0$), the Q^{-1} value will drop to the background value Q_0^{-1} at a relatively fast rate.

Various theories have been proposed for interpreting the IF peak in the process of the FOPT. Wang *et al.* presented a theoretical model to explain the change in Q^{-1} arising from the change of the elastic constant during the martensite transition.[11] Ma and Ke indicated that IF is associated with the motion of an extended dislocation at the coherent interface.[12] Postnikov *et al.* considered that IF originates from the fluctuation[25] of certain relevant quantities in the process of a FOPT and derived an expression of IF as function of $\dot{T}$, ω, and A_ε.

Delorme *et al.* derived an explicit expression for Q^{-1} in terms of $\dot{T}$ and a function involving the volume fraction V_M of M and T.[26] Djonghe *et al.* extended the expression of Delorme *et al.* to arrive at an expression for Q^{-1} involving the stress also.[27] We would emphasize that all the stated expressions for Q^{-1} during a FOPT are associated with the volume effect during a FOPT and can be written in the simple form $Q^{-1}=C(T)\dot{T}/\omega$. Gremaud *et al.*[28] extended the work of Delorme *et al.* and proposed a new model for the IF of the FOPT, which explains the nonlinear relationship between IF and $\dot{T}/\omega$ (Ref. 28) (see also Ref. 23). Such a result of the $(\dot{T}/\omega)$ dependence was close to early experimental findings,[11,20,21,28] but significant discrepancies have been found during the past decade and the explicit phenomenological equation which is consistent with recent and more accurate experimental results is expressible as $Q^{-1}=C(T)\omega^{-2l}(\dot{T}/\omega)^n$, with $0<n,\ l<1$.[13,16,17,22,24] In this paper, we address this problem and present an equation of motion of the phase interface during a FOPT under an applied alternating stress for an IF measurement simultaneously. In the end, we are able to derive the dissipation function (i.e., resistance curve) and obtain an explicit expression for Q^{-1}. It is emphasized that the observed high value of Q^{-1} during a FOPT is associated only with the motion (dynamics) of a phase interface rather than with the volume effect (such as the volumetric fluctuations) in the process of the FOPT.

We would remark here again that IF is defined as a dissipation (or absorption) of vibration energy supplied by the IF measuring system, while the energy dissipation during a FOPT is a dissipation of heat or mechanical energy supplied by the driving system. The contribution of the present paper is to derive the energy dissipation of the FOPT from the experimental data of the absorption of vibration energy (IF).

II. EQUATION OF MOTION OF PHASE INTERFACE

A. Various forces acting on a unit moving phase interface

It has been well established that the FOPT in solids can be viewed as a moving process of the phase interface (PI) between the parent phase (P) and the new phase (N).[29–33] Such a concept is applicable even to the nucleation stage. The new phase grows and the FOPT develops when the direction of motion of the PI points toward the parent phase and the transformation is completed when the parent phase is exhausted. In addition to the PT driving force, i.e., ΔG_d, which acts on the PI, a resistive force ΔG_R (arising from the origins specified below) is also at work, impeding the motion of the PI, and the FOPT occurs only if $\Delta G_d > \Delta G_R$.

1. Effective phase transformation driving force acting on the phase interface

Now consider a unit PI in the process of a FOPT; the driving force per unit area, ΔG_d, is equivalent to the change in Gibbs free energy per unit volume between the P and N phases and it is well established,[2]

$$\Delta G_d=\Delta H\Delta T/T_0\ , \tag{1}$$

where the dimension of ΔG_d is 1 cal/cm^3 or 1 dyn/cm^2, ΔH is the change of enthalpy of the system in the process of the FOPT, and T_0 is an equilibrium temperature of the N and P phases, expressible as $T_0=(T_s+T_s')/2$, with T_s and T_s' the starting temperature of the new and parent phases of the FOPT and $\Delta T=|T-T_0|$.[34] The force ΔG_d can therefore be written as

$$\Delta G_d=A_0\dot{T}t'\ , \tag{2}$$

where $A_0, \dot{T}=$const and time $t'=0$ when $T=T_0$.

The resistance per unit area, ΔG_R, acting on the moving PI arises mainly from three origins: (i) the lattice distortion arising from the difference of the molar volume between the N and P phases; (ii) the intrinsic deformation of the new phase, such as the shear strain during the martensitic transformation; and (iii) acoustic waves emitted from the reconstruction of the lattice during the FOPT. Note that each of the three aspects is dependent on the rate of the FOPT and that of ΔT or $(\dot{T}t')$, meaning that ΔG_d is a function of ΔT or $(\dot{T}t')$ (when $t'=0$, $T=T_0$) and the effective driving force $\Delta G'$ is defined as

$$\Delta G'=\Delta G_d-\Delta G_R \tag{3a}$$

or

$$\Delta G_R=\Delta G_d-\Delta G'\ , \tag{3b}$$

which is a function of $(\dot{T}t')$. It is clear from (3b) that both ΔG_d and $\Delta G'$ are supplied by the applied PT driving system and the energy dissipated, ΔG_R, represents part of the energy supplied by the PT driving system.

In the FOPT, past experimental results indicate that the volume fraction of the new phase F is monotonically increasing in T as shown in Fig 1 (see also, e.g., Refs. 1,2). The past data also show that there exists a maximum value of dF/dT at $T>T_s$, where $T=T_s$ at $t=0$. If N_A is the total area of the moving PI and v is the average velocity of the PI; obviously $dF/dt=N_A v$. Since $dF/dt=(dF/dT)\dot{T}$, dF/dt has the same form as dF/dT. We know that all the three quantities $N_A v$, N_A, and v must vanish at the starting temperature T_s and final tem-

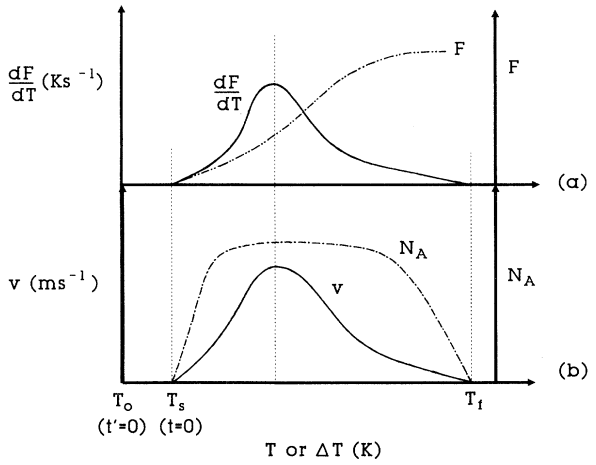

FIG. 1. Volume fraction $F, dF/dT$, total area N_A of the moving PI vs temperature T.

perature T_f of the FOPT. When the transformation occurs, many new phase nuclei appear, each of which increases in size, and so the area of the PI will be increasing around each nucleus. The total area around the N phase (i.e., around all the N nuclei) must be increased until some of the nuclei begin to tough their neighboring N phase particles. As soon as that process occurs, the total N_A starts to decrease and quickly shrinks to zero. Hence the transformation is completed as T approaches T_f.

We would remark here that experimental results of thermoelastic martensite transformations in single crystals show that N_A may remain close to a constant value during most of the temperature range (T_s, T_f) [the F-T curve has about the same shape as that shown in Fig. 1(a)].[35] For other FOPT's in polycrystalline materials, we anticipate that the total PI area of the average new phase

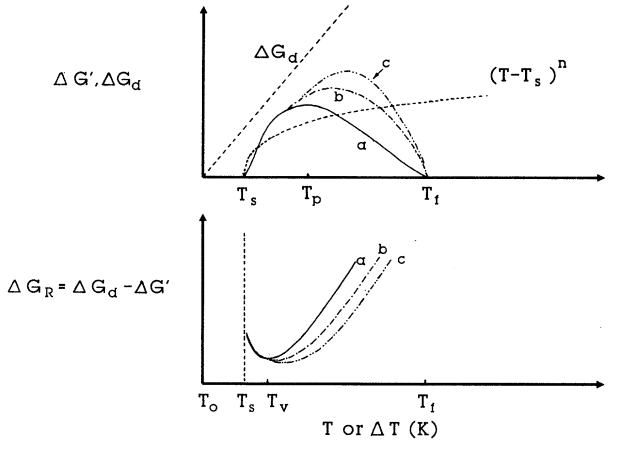

FIG. 2. ΔG_d vs T (dotted straight line), $\Delta G'$ vs T with three different $\dot{T}$ as marked by a (corresponding to $\dot{T}_1$), b (corresponding to $\dot{T}_2$), and c (corresponding to $\dot{T}_3$), where $\dot{T}_3 > \dot{T}_2 > \dot{T}_1$. For convenience in analysis, the curve $(T-T_s)^n$ vs T is also plotted. The function ΔG_R is obtained from the curves stacked on the top part.

cannot have a singular point or minimum in the range (T_s, T_f). Thus without loss of generality, N_A has the form shown in Fig. 1(b) and the velocity v also has a maximum value at T accordingly. Thus the effective driving force $\Delta G'$ must also have a maximum at $T=T_s$. In reality, the maximum may not occur at the midpoint of the range (T_s, T_f) and each curve of N_A or v may not be symmetrical. For convenience in qualitative analysis, we consider the simple situation where $\Delta G'$ has a similar shape with a maximum, as shown in Fig. 2, where each curve (a,b,c) corresponds to one value of $\dot{T}$. We also show the $(T-Ts)^n$-T relation $(0<n<1)$ in the top part of Fig. 2 for convenience in the discussion. When $T<T_s$ and $T \geq T_f$, $\Delta G'=0$ or $\Delta G_R > \Delta G_d$, and no PI can move under such a condition. For various possible $\Delta G'$-T curves as shown in Fig. 2, it is reasonable to write

$$\Delta G' = \Delta G_d - \Delta G_R = A_1(T)\Delta T^n = A_1(T)(\dot{T}t)^n , \quad (3c)$$

where $T=T_s$ at $t=0$; in Fig. 2, a,b,c pertain to $\dot{T}_1, \dot{T}_2, \dot{T}_3$, respectively. So far, $A_1(T)$ is still an unknown function having a maximum near $T=T_s$ stated before. But $\Delta G'$ does not have the same maximum as $A_1(T)$. The validity and limitations of Eq. (3c) have to be determined by experimental measurements.

2. *Harmonic force arising from forced oscillation during the IF measurement*

As mentioned in the Introduction, it is difficult to measure the dissipation of energy in the process of the FOPT directly. We try to calculate the dissipative energy in the process of a FOPT by internal friction (IF) measurement data, which represents the absorption of vibration energy supplied by IF measuring system in the low-frequency range. In order to measure IF, a time alternating stress $\sigma = \sigma_A \sin(\omega t + \vartheta)$ must be applied; here, σ_A is the vibration amplitude of the applied stress and ϑ is the initial phase. As the sample is subject to the applied stress $\sigma_A \sin(\omega t + \vartheta)$, as a result of the difference in the molar volumes of the N and P phases as a result of the distortion of the new phase, there should be an effective force acting on the PI:

$$\Delta G_a^0 = \alpha(\omega, 0)\sigma_A \sin(\omega t + \vartheta) , \quad (4)$$

where $\alpha(\omega,0)$ describes the coupling strength between the oscillating stress and static PI when the PI is not in motion $(V=0)$. The experimental measurements of IF for the materials FeMn and NiTi (Ref. 36) indicate that in a FOPT, which occurs across a range of temperature in general, IF is dependent of the frequency even under the isothermal condition such that $\dot{T}=0$; therefore, α depends on ω. Consequently, when the frequency of the applied stress is varied, the response of the PI would change accordingly and α should be a function of ω rather than a constant, since the response characteristic of the PI to the influence of stress depends on the configurations of the PI. We shall discuss further the characteristics of the ΔG_a^0-ω dependence later in this section.

3. *Force arising from the interaction of the PI motion and the applied oscillating force*

So far, we have considered the situation where the PI is not in translational motion. Our experimental investigation indicates very clearly that a peak in the Q^{-1}-T curve appears only if the following conditions are satisfied simultaneously.[31,32] Consider a system undergoing a FOPT under significant variation of temperature. Suppose we vary the temperature in steps and measure IF under isothermal conditions after the temperature has been stabilized. Experimental results reported in Refs. 13, 24, and 36 show that Q^{-1}-T peaks do occur. However, the Q^{-1} peaks are significantly diminished with respect to those cases pertaining to $\dot{T}\neq 0$. For example, the Q^{-1} peak ($\dot{T}=0$) for FeMn is only one order of magnitude lower and that for the material $BaTiO_3$ is about 30% of the value for the $\dot{T}\neq 0$ case. Such a feature occurs because in these two materials there exists a phase transition for the situation $\dot{T}\neq 0$, in addition to the phase transition under the restriction $\dot{T}=0$. However, there is still no loss of generality presented in our argument because the Q^{-1}-T peak observed under the condition $\dot{T}\neq 0$ is much more prominent than that observed when $\dot{T}=0$. Hence the conditions for the appearance of internal friction peak(s) in a FOPT during temporal variation can be summarized as (i) $\Delta T\neq 0$ when the sample is in the temperature range of the FOPT, (ii) $\sigma_A\neq 0$ when the IF measurement is carried out, and (iii) $\dot{T}\neq 0$. These "combined conditions" imply that if the PI is not moving (even though the sample is in the temperature range of the FOPT and a significant static PI is present), the dynamic IF is zero. Suppose v is the average velocity of the PI. The above deduction means that the condition $\dot{T}\neq 0$ would induce PI motion and the coupling factor α would be different for $v=0$ and $v\neq 0$ (i.e., $\dot{T}=0$ and $\dot{T}\neq 0$).

While expression (4) describes the force (due to oscillating motion) in the static condition ($\dot{T}=0$ and $\nu=0$), the coupling strength $\alpha(\omega,0)$ must be modified to depend on the average velocity v of the PI in the dynamic situation ($\dot{T}\neq 0$). Thus the harmonic force arising from forced oscillation during the IF measurement under the dynamic situation should be rewritten in the form

$$\begin{aligned}&\Delta G_a^{\rm dyn}=\alpha(\omega,v)\sigma_A\sin(\omega t+\vartheta)\ ,\\&\Delta G_a=\Delta G_a^0+\Delta G_a^{\rm dyn}\ .\end{aligned}\tag{5}$$

It is observed experimentally that when either $\alpha(\omega,v)=0$ or $\sigma_A=0$, $\Delta G_a^{\rm dyn}=0$, the dynamic IF is zero; therefore, the simplest form $\Delta G_a^{\rm dyn}$ must be expressible as a product form of $\alpha(\omega,v)$ and $\sigma_A\sin(\omega t+\vartheta)$, and not as an additive form. Since the average velocity V of the PI is determined by the effective driving force $\Delta G'$, we can write $\alpha(\omega,v)$ as $\alpha(\omega,\Delta G')$. Consider the situation where the coupling between the PI motion and harmonic driving force is weak (which can be ensured approximately by the experimental conditions where the strain amplitude is $\sim 10^{-5}$ and $\dot{T}\leq 0.1$ K sec^{-1}), we may then approximately express $\alpha(\omega,\Delta G')$ into the simple product form

$$\alpha(\omega,v)=\alpha(\omega,\Delta G')=\alpha_0(\omega,0)\alpha_g(\Delta G')\ .\tag{6}$$

In general, $\alpha_g(\Delta G')$ may be a power series of $\Delta G'$. Under the weak-coupling condition, we assume here that $\alpha_g(\Delta G')$ is only a linear function of $\Delta G'$:

$$\alpha_g(\Delta G')=C'\Delta G'\ .\tag{7}$$

In view of (4)–(7),

$$\Delta G_a^{\rm dyn}=C'\Delta G'\Delta G_a^0\ ,\tag{8}$$

where C' is an interaction coefficient and is independent of the measuring frequency, because the frequency effect has already been contained in $\alpha_0(\omega,0)$ and $\Delta G_a^0\equiv\alpha(\omega,0)\sigma_A\sin(\omega t+\vartheta)$. We may view the above expression with the following simple physical picture. When the FOPT system is subjected to an external stress, the time varying stress and the condition $\dot{T}\neq 0$ have two physical effects on the PI: (i) a force of magnitude ΔG_a^0 acts on the PI arising from the change in volume (or distortion); (ii) the PI motion would cause a change in the configuration of the PI, which in turn would modify the magnitude (and direction) of the effective driving force $\Delta G'$. We may therefore introduce an interaction force ΔG_i to describe the PI motion and the dynamic IF associated with the moving PI. This force ΔG_i comes into being only if both $V\neq 0$ (or $\Delta G'>0$) and $\Delta G_a^0\neq 0$ are satisfied. For a general description, ΔG_i may be expressed as a product of two functions $U_1(\Delta G')$ and $U_2(\Delta G_a^0)$. As we are studying the weak-coupling case, only linear terms of U_1 and U_2 are taken, giving rise to $\Delta G_i=\Delta G_a^{\rm dyn}=C'\Delta G'\Delta G_a^0$, namely, Eq. (8). Obviously, the force ΔG_i is due to (weak) nonlinear effects. The stated methodology of treating the coupling problem described by two parameters is well established (see, e.g., Ref. 37).

B. Equation of motion of the phase interface and solution

Consider the unit area of a PI moving along the x axis. Based on the derivation of Sec. II A, the equation of motion of the PI in the dynamic situation is

$$\rho\ddot{x}+\gamma\dot{x}+kx=\Delta G'+\Delta G_a^0+\Delta G_a^{\rm dyn}\tag{9a}$$

or

$$\begin{aligned}\ddot{x}+2\eta\dot{x}+\omega_0^2x&=CA_1(\dot{T}t)^n+C\alpha(\omega)\sigma_A\sin(\omega t+\vartheta)\\&\quad+CC'\alpha(\omega)A_1(\dot{T}t)^n\sigma_A\sin(\omega t+\vartheta)\\&=f(t)\ ,\end{aligned}\tag{9b}$$

where $\rho=C^{-1}$ is the average mass density of the PI, γ is the effective damping coefficient, k is the dynamic restoring coefficient arising from the interaction between the PI's, and $\omega_0^2=k/\rho$ is the square of the resonant frequency as the PI's undergo oscillatory motion, while $2\eta=C\gamma$. During the process of a FOPT, the situation of $V=0$ and $V\neq 0$ is coexistence; therefore, we need both ΔG_a^0 and $\Delta G_a^{\rm dyn}$ in Eq. (9).

Carrying out a Laplace transform on Eq. (9), note that if the coefficients η,ω_0 are constant, the analytical solution to (9) is

$$x=\exp(-\eta t)/\omega_d \int_0^t e^{-\eta t'} f(t') \sin[\omega_d (t-t')]dt' , \quad (10)$$

where $\omega_d^2=\omega_0^2-\eta^2$. If η and ω_0 are weak functions of time, we can readily obtain a very accurate numerical solution to x for known $f(t')$ using standard numerical methods of integration. As the average spatial distribution of the PI can be considered as constant before and after each IF measurement, η and ω_0 would indeed be very weak functions of time and the solution (10) is rather accurate.

We would remark here that there are different orientations of the PI's during the FOPT of solids and the shapes and directions of motion of these PI's are also quite different in general. To analyze the PI motion, we may, however, employ the method of analysis in treating the dynamics of domain walls in ferromagnetic materials.[38,39] In other words, a planar phase interphase with total area N_A (which equals the total area of the PI's in motion) moving along one direction is taken as a model to represent the motion of the PI's during a FOPT. Such a model is valid because the total energy dissipated during a FOPT is a strong function of the total (area) PI and the average speed to these moving PI's; but not a strong function of the number of PI's involved or the interactions among the PI's. The validity of such a model has already been thoroughly discussed in Ref. 39. The limitations of this model will be considered in Sec. V.

III. INTERNAL FRICTION ARISING FROM PI MOTION DURING THE FOPT PROCESS

The dissipation of vibration energy due to PI motion per cycle of oscillation per unit volume of the specimen is

$$\Delta W=N_A \int_0^p \Delta G_a^0 \dot{x}\, dt , \quad (11)$$

where p is the period oscillation, N_A is the total area of moving a PI in a unit volume, and ΔG_a^0 is the effective alternating stress applied to the PI during IF measurements. By definition, the internal friction is then

$$Q^{-1}=\Delta W/2\pi W=N_A \int_0^p \Delta G_a^0 \dot{x}\, dt/(\pi\sigma^2/\mu)$$
$$=Q_0^{-1}+Q_s^{-1}+Q_{\rm dyn}^{-1} , \quad (12)$$

where μ is the modulus associated with the particular mode of oscillation, while W is the maximum kinetic energy during vibration. The total IF of the sample can be considered to be contributed by $\dot{x}$, which in turn is determined by various driving forces in an additive manner, in view of the superposition principle. Any nonlinear effect is assumed to be included in all the forces discussed above. Under the weak-coupling condition and the low-frequency forced oscillation situation, surely $\omega_0^2 \gg \omega^2$, and $\omega_0^2 \gg \eta^2$, implying that the $e^{-\eta t}$ term in (10) can be neglected.[31] The expression of the total internal friction can be considered to be contributed by the following three parts.

A. Contribution to IF from the effective driving force $\Delta G'$

We have already obtained the explicit expression for the effective driving force per unit area $\Delta G'$ acting on the PI. Identifying $\Delta G'$ as $f(t')$ in (10), we can calculate $\dot{x}$ and substitute the result into (12), arriving at the internal friction contributed from $\Delta G'$:

$$Q_0^{-1}=(CN_A B_0 A_1 \alpha\mu/\sigma_A \omega_0^2)(\dot{T}/\omega)^n , \quad (13)$$

where $B_0 \approx 1$ is a numerical coefficient and N_A is the total area of the PI when the temperature is T.

B. Contribution to IF from the harmonic stress ΔG_a^0

In view of the fact that the internal friction varies slightly with the temperature T even when $\dot{T}=0$ during the IF measurement, the IF generated by the harmonic stress alone must be a weak function of T. From (4) and (11) such a contribution to the total IF is exactly

$$Q_s^{-1}=2CN_A(T)\eta\alpha^2(\omega)\omega\mu/\omega_0^4 . \quad (14)$$

Because Q_s^{-1} is independent of $\dot{T}$, Q_s^{-1} is therefore associated with the oscillation of the static PI.

C. Contribution to IF from the dynamic interaction force $\Delta G_a^{\rm dyn}$

Based on relations (5) and (11), it is easy to write down the contribution to IF originating from $\Delta G_a^{\rm dyn}$,

$$G_{\rm dyn}^{-1}=(CC'N_A B_m A_1 \alpha^2\mu/\omega_0^2)(\dot{T}/\omega)^n , \quad (15)$$

where B_m is a numerical number of order of unity. Comparing (13) and (15), as $\mu \sim 10^{12}$ (cgs units) for most solid materials of interest, $\sigma_A \sim 10^{-5}\mu$, $C' \sim 10^{-1}-10^{-2}$, $\alpha \sim 10^{-1}-10^{-2}$, and $Q_{\rm dyn}^{-1}/Q_0^{-1} \simeq C'\alpha\sigma_A \sim 10^{4-6}$. We shall provide more information on the estimated values of C' and α later.

We see that $Q_{\rm dyn}^{-1} \gg Q_0^{-1}$ and we can neglect Q_0^{-1} in the overall estimation of IF. We can then readily show from (14) and (15) that to a very good degree of approximation, the total IF can be expressed as

$$Q^{-1}=A(T)\alpha^2(\omega)(\dot{T}/\omega)^n+B(T)\alpha^2(\omega)\omega , \quad (16)$$

with

$$A(T)=CC'N_A(T)A_1(T)B_m\mu/(\pi\omega_0^2) , \quad (17)$$

$$B(T)=2C\eta N_A(T)\mu/\omega_0^4 . \quad (18)$$

Using the same procedure in deducing (11) from (10), the modulus defect $\Delta M/M$ originating from the interface mobility under the action of oscillating stress in the process of a FOPT is found to be[29,30]

$$(\Delta M/M)_{\rm PI}=\varepsilon''/\varepsilon'$$
$$=L\varepsilon_0/(\sigma/\mu)$$
$$=[f_1(n)N_A CC' A_1 \varepsilon_0 \mu/(\pi\omega_0^2)](\dot{T}/\omega)^n\alpha(\omega) , \quad (19)$$

where ε' and ε'' are the elastic and nonelastic strains of the interface, respectively, under the exerting oscillating stress, $L=\int_0^{p/4}\dot{x}\, dt$ is the amplitude value of the interface displacement, $f_1(n)(\sim 1)$ is a numerical coefficient

dependent on the parameter n, and ε_0 is the strain of the PT ($\sim 10^{-2}$). Comparing Eqs. (15) and (19), we have

$$Q_{\rm dyn}^{-1}/(\Delta M/M)_{\rm PI}=f(n)\alpha(\omega)/\varepsilon_0 , \tag{20}$$

where $f(n)$ ($\lesssim 1$) is also a numerical coefficient and the value of the transition strain ε_0 can be determined from experimental data.

IV. METHODOLOGY OF CALCULATING THE DISSIPATION FUNCTION FROM THE EXPERIMENTAL RESULTS OF IF MEASUREMENTS

We need to relate the resistance force ΔG_R to the experimental data of IF measurements. From Eq. (3),

$$\Delta G_R=\Delta G_d-A_1(T)(\dot{T}t)^n . \tag{21}$$

The driving force ΔG_d can be obtained from differential scanning calorimetry (DSC) measurement as indicated by Eq. (1). If the numerical exponent n is determined by IF experiments as specified by Eqs. (15) or (16), only $A_1(T)$ is needed for the determination of ΔG_R. Such a calculation can be achieved by the following procedures.

A. Determination of the coupling coefficient α for a range of frequency ω

Because the softening of the phonon mode occurs almost always during the process of the FOPT, the measured value of $\Delta M/M$ therefore describes the modulus defect originating from the moving interface and the softening of the phonon mode. More specifically, if we define $(\Delta M/M)_s$ to be the modulus defect arising from the softening of the phonon mode, the ratio $R\equiv(\Delta M/M)_{\rm total}/(Q^{-1})_{\rm total}$ can be expressed as

$$R\equiv\frac{(\Delta M/M)_{\rm total}}{(Q^{-1})_{\rm total}}=\frac{(\Delta M/M)_s+(\Delta M/M)_{\rm PI}}{Q_{\rm dyn}^{-1}+Q_s^{-1}}$$
$$\simeq\frac{(\Delta M/M)_s}{Q_{\rm dyn}^{-1}}+\frac{(\Delta M/M)_{\rm PI}}{Q_{\rm dyn}^{-1}} , \tag{22a}$$

where Q_s^{-1} is neglected since it is only 1% of $Q_{\rm dyn}^{-1}$. (If Q_s^{-1} is comparable to $Q_{\rm dyn}^{-1}$, this procedure can be done by computer.) We note that according to relations (16) and (19), both $(\Delta M/M)_{\rm PI}$ and $Q_{\rm dyn}^{-1}$ are functions of $\dot{T}^n$, so that their quotient is independent of $\dot{T}$. It has been established experimentally that $(\Delta M/M)_s$ is also independent of $\dot{T}$,[5] as expected from the basic theory of phonons in solids. Now as $Q_{\rm dyn}^{-1}$ increases with $\dot{T}$ like $\dot{T}^n$ based on Eq. (15) and $0<n<1$, it is obvious that $Q_{\rm dyn}^{-1}$ approaches an asymptotic value when $\dot{T}$ is large enough. In other words, when $\dot{T}$ takes on large enough values, R in (22a) also has an asymptotic value when the frequency ω is fixed. In view of (20) and (22a), R is relatively large when ω is large. The qualitative behavior of $(\Delta M/M)_{\rm total}/(Q^{-1})_{\rm total}$ with respect to a change of $\dot{T}$, taking ω as a parameter (high, low), is described in Fig. 3. Based on graphs shown in Fig. 3, the dependence of the coupling coefficient α on ω in Eq. (4) for the asymptotic part (R depends on $\dot{T}$ linearly) can be expressed simply as

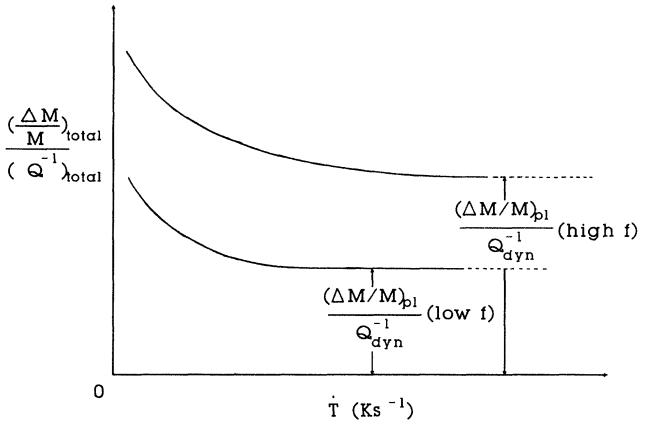

FIG. 3. Ratio $(\Delta M/M)_{\rm total}/(Q^{-1})_{\rm total}$ against $\dot{T}$ for high and low frequency of oscillation.

$$\alpha(\omega)=\alpha'\omega^{-l} , \tag{22b}$$

where l is a parameter specified by the particular phase transition concerned and α' a proportionality constant. It has been found experimentally in Refs. 17, 24, and 32 that

$$0<l<1 \tag{22c}$$

for different phase transformations; the value of l falls in the range of 0.1–0.8.

Now we can substitute (22b) into (16), giving

$$Q^{-1}=\alpha'^2A(T)\omega^{-2l}(\dot{T}/\omega)^n+\alpha'^2B(T)\omega^{1-2l}$$

or

$$Q^{-1}=A(T)\dot{T}^n/\omega^{n+2l}+B(T)\omega^{1-2l} , \tag{23a}$$

leading to

$$Q^{-1}/\omega^{1-2l}=A(T)\dot{T}^n/\omega^{1+n}+B(T) , \tag{23b}$$

where α' has been absorbed in $A(T)$ and $B(T)$. The value of the left-hand side of Eq. (23b) is now known, while n, $A(T)$, and $B(T)$ on the right-hand side are still unknown.

B. Calculation of $A(F)$ and $B(F)$

Owing to the fact that the total area of interface N_A in the process of a FOPT is dependent on the volume fraction F of the product phase and the peak temperature (T_p) of the IF during a FOPT shifts to a higher temperature when $\dot{T}$ increases, the values of N_A at the same T but different $\dot{T}$ are not the same. Therefore the values of A and B must be associated with the same F value, but not the same T. Suppose we apply the same oscillation frequency to the same specimen, but repeat the Q^{-1} measurements for two different values of $\dot{T}$ (i.e., $\dot{T}_1,\dot{T}_2$), the IF curves would look like that shown in Fig. 4, where $\dot{T}_1<\dot{T}_2$. Both the Q^{-1} IF peak heights and peak temperature T_p increase with increasing $\dot{T}$ in the Q^{-1}-T curves. It is well established experimentally that the shapes of the Q^{-1}-T curves are very similar for different

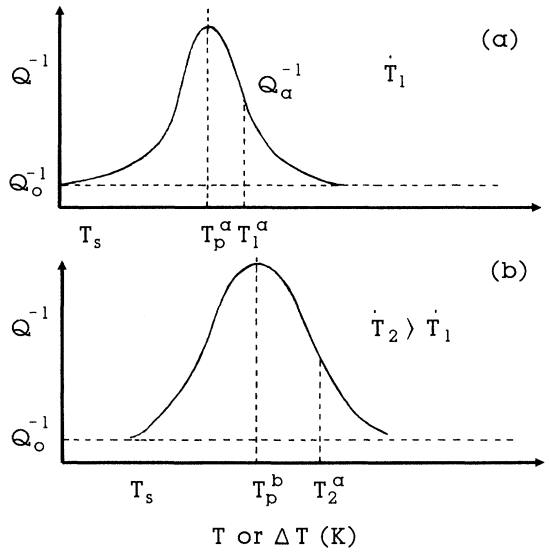

FIG. 4. Internal friction Q^{-1} as a function of temperature T for two values of $\dot{T}(\dot{T}_2 > \dot{T}_1)$.

$\dot{T}$. The area to the left of the dotted straight vertical line (passing through the maximum Q^{-1}) occupies about 42–47 % (the absolute value depends on the materials of the sample) of the total area under the Q^{-1} curve in each case of Fig. 4. We have also found out experimentally that at T_p, the fraction of the new phase is the same (i.e., a specific value within the range 42–47 % for this specimen) for various $\dot{T}$ and ω. Such a fixed value of F has been substantiated by electrical resistance measurements. Based on the stated experimental findings, we can introduce a new ratio $r \equiv Q^{-1}/Q_p^{-1}$ to measure indirectly the fraction of the new phase, where Q_p^{-1} is the peak value of IF.

In Fig. 4(a), consider the instant at which Q^{-1} has the value Q_a^{-1} and the temperature is T_1^a. The volume fraction of the sample corresponding to a certain value of $\dot{T}=\dot{T}_1$ and at a certain temperature T_a has a certain value F_a, say. Then, in Fig. 4(a), we can find the value of Q_b^{-1} and the corresponding T_b such that $r=(Q_b^{-1}-Q_s^{-1})/Q_{p,b}^{-1}=(Q_a^{-1}-Q_s^{-1})/Q_{p,a}^{-1}$. At such values of Q_b^{-1} and T_b, though $\dot{T}$ and ω may be different, the fraction of the new phase is the same, being F_a also. Note again that the value of r defined above does not equal the value of the fraction volume directly.

Having found a way to measure Q^{-1} for the same fraction of the new phase at different $\dot{T}$ and ω, we can then collect the various Q^{-1} values corresponding to different $\dot{T}$ and ω combinations, but corresponding to the same new phase fraction F. Using (23b), we can now plot $Q^{-1}/(\omega^{1-2l})$ against $\dot{T}^n/(\omega^{1+n})$ for a range of n, which is taken as the parameter for each curve. We choose the n value such that the state relation is closest to a straight line. This n value is the right numerical parameter to be used in Eq. (23b). The slope is obviously $A(F)$ and the intercept is simply $B(F)$.

C. Deduction of the dissipation function

Now we proceed to the final stage of finding the dissipation function $\Delta G_d(F)$ or $\Delta G_d(T)$. From (3), (17), and (18), we have

$$A_1(F)=(2\pi\eta/C'B_m\omega_0^2)A(F)/B(F)=k'A(F)/B(F)\ , \tag{24a}$$

$$k'=2\pi\eta/C'B_m\omega_0^2\ , \tag{24b}$$

and from (21), $\Delta G_R(F)=\Delta G_d(F)-A_1(F)(\dot{T}t)^n$, meaning that the dissipation function is acquired when k' is determined.

We would remark that the damping coefficient η depends only on the difference of crystal structure of the two phases involved and should not be a strong function of temperature. Note also that $\omega_0^2=k/\rho$ and k is the dynamic restoring coefficient arising from the interaction between the PI's and also from the difference of the crystal structure of the two phases involved. Both are weak functions of temperature. Now the time interval of each internal friction measurement takes only less than 10 sec. The maximum temperature change is only less or about 0.1 K. The parameter C' in (7) remains practically constant during each IF measurement. Thus k', in (24b), $\propto \eta/(C'\omega_0^2)$ should not be a strong function of temperature.

As indicated by the experimental evidence, the function $A(T)$ has a maximum and a shape similar to the Q^{-1}-T curve, while $B(T)$ is roughly constant for a range of materials. We have found that the dissipation function $\Delta G_R(T)$ has a variety of shapes, but they all possess a minimum in each curve (Fig. 2). At the minimum, we can set $d\Delta(G_R(T))/dT=0$ at temperature T_v and obtain the desired expression for the parameter k' in (24b):

$$k'=(\Delta H/T_0)/(A(T_v)(T_v-T_s)^n\{d[\ln A(T)]/dT+n/(T_v-T_s)\}/B)\ . \tag{25}$$

With all parameters found, we can deduce the dissipation function $\Delta G_R(T)$ from relation (3).

V. DISCUSSION

(1) The crucial difference between the present phenomenological theory and that of other theories published so far (including the joint works of J.X.Z.) is the introduction of the effective force $\Delta G_a^0=\alpha(\omega,0)\sigma_A\sin(\omega t+\theta)$ acting on the PI in the general equation of motion (9a). We first note that if the coupling factor $\alpha(\omega,0)$ becomes a (normalized) constant of unity (meaning $l=0$), our Eq. (23a) gives $Q_{\rm dyn}^{-1}\propto(\dot{T}/\omega)^n$ for the dynamic term, which is

identical to previous results reported (see Refs. 14, 15, 23, and 28–31); in particular in Ref. 29, the concept of effective force was not considered). The recent experimental work with a VO_2 sample indicates that in general the $Q_{\rm dyn}^{-1}$-$\dot{T}$ relation is different from the $Q_{\rm dyn}^{-1}$-ω^{-1} relation.[43] We are thus led by such a result to reexamine the physical concept involved in the equation of motion (9a). In fact, we would remark that the time alternating stress $\sigma=\sigma_A \sin(\omega t+\theta)$ applied during internal friction measurements acts on the specimen as a whole, but not on the PI directly. The force acting on the PI arises from the difference in the structure of the parent and new crystal phases or the difference in the distortion of the two phases. The vibration of the PI can have a direction different from that of the oscillating stress applied during experimentation. Vibrations of PI's lead to internal friction as observed. Since the PI's motions are induced by external applied stress, we must use a coupling factor $\alpha(\omega,0)$ to describe such an induction.

Before translational occurs, the effective force acting on a static PI is therefore $\Delta G_a^0=\alpha(\omega,0)\sigma_A \sin(\omega t+\theta)$ [i.e., Eq. (4)]. Experimental evidence[13,40,41] indicates that the internal friction IF is a function of the frequency of vibration applied. Such a result is expected in our theory. Vibration of the PI would induce a rearrangement of the lattice at both sides of the boundary and the rearrangement pertains to a specific relaxation time; the consequence is that the coupling factor must in general be a function of frequency $[\alpha(\omega,0)]$ even for the static case. When translational motion of the PI occurs with velocity V, the response of the lattice on both sides of the PI boundary would be different and the coupling factor should be described by the function $\alpha(\omega,V)$ and the dynamic effective driving force of the PI is then $\Delta G_a^{\rm dyn}=\alpha(\omega,V)\sigma_A \sin(\omega t+\theta)$. When $\dot{T}\neq 0$, $V\neq 0$; thus, the relation $Q_{\rm dyn}^{-1}$-$\dot{T}$ should be intuitively different from the relation $Q_{\rm dyn}^{-1}$-ω^{-1}, as described by Eq. (23). (Only if $l=0$ are the relations $Q_{\rm dyn}^{-1}$-$\dot{T}$ and $Q_{\rm dyn}^{-1}$-ω^{-1} are the same.) On the other hand, $\Delta G_a^{\rm dyn}$ is expressible in a product form $\Delta G_a^{\rm dyn}=C'\Delta G'\Delta G_a^0$. Only when both $\Delta G'\neq 0$ and $\Delta G_a^0\neq 0$, $\Delta G_a^{\rm dyn}\neq 0$, and we call $\Delta G_a^{\rm dyn}$ the interaction driving force in accordance with common terminology in physics.

We now search for more experimental evidence in published work to check the consequence of our theoretical deduction: The relations $Q_{\rm dyn}^{-1}$-$\dot{T}$, $Q_{\rm dyn}^{-1}$-ω^{-1} are different. In fact, in 1989, Bidaux, Schaller, and Benoit[23] reported such relations for the metal cobalt. In Fig. 4 of Ref. 23, when $f=0.5$ Hz, with $\dot{T}=1$ K/min and $\dot{T}=4$ K/min, the corresponding ratio of the internal friction peaks is $Q_{\rm max}^{-1}$ ($\dot{T}=4$ K/min)/$Q_{\rm max}^{-1}$ ($\dot{T}=1$ K/min)$\simeq 2.15$. In Fig. 6 of Ref. 23, consider the case when $\dot{T}=2$ K/min (a value halfway between 1 and 4 K/min). When the frequency $f=1$ and 0.25 Hz (a value halfway between 0.5 Hz, parameter of Fig. 4), the corresponding ratio of the internal friction peaks is, however, $Q_{\rm max}^{-1}$ ($f=0.25$ Hz)/$Q_{\rm max}^{-1}$ ($f=1.00$ Hz)≈ 2.55. Clearly, $Q_{\rm dyn}^{-1}$ is a stronger function of the inverse frequency than $\dot{T}$. Our corresponding theoretical consequence is in line with the experimental finding reported in Ref. 23 earlier.

Moreover, the difference between the $Q^{-1}(T)$ and $Q^{-1}(\omega^{-1})$ dependences could also have been deduced from the theoretical calculation of Gremaud, Bidaux, and Benoit[28] by taking into account that α in Eq. (3) of Ref. 23 is not a constant, but is actually $\dot{T}$ dependent. Note also that $Q_{\rm dyn}^{-1}$ in Ref. 23 is a function of the parameter A (α in Ref. 23), which is $\propto \Delta H^{-1}$. A more recent result shows ΔH increases with increasing $\dot{T}$,[42] with the result that A decreases with increasing $\dot{T}$. Consequently, the $Q_{\rm dyn}^{-1}$-$\dot{T}$ relation is significantly different from the $Q_{\rm dyn}^{-1}$-ω^{-1} relation.

(2) We have adopted a simple model to describe the FOPT in solids: A planar phase interphase with total area N_A moving along a certain direction is taken to represent the motion of the PI's during the FOPT. During the motion of the planar surface, there must be an effective frictional force ΔG_R being exerted on the representative PI, and the work done against the friction is thus

$$W=\int_{t_s}^{t_f}\Delta G_R(T)N_A V\,dt\ ,$$

where V is the average speed of the planar PI. Because $dF/dt=N_A V$, it is clear that

$$\begin{aligned} W&=\int_{t_s}^{t_f}\Delta G_R(T)\left[\frac{dF}{dt}\right]dt \\ &=\int_{T_s}^{T_f}\Delta G_R(T)\left[\frac{dF}{dt}\right]dt \\ &=\int_0^1\Delta G_R(F)dF\ . \end{aligned} \tag{26}$$

Physically, ΔG_R is the rate of energy dissipation. It is thus of prime importance to devise a method for calculating ΔG_R as a function of temperature or new phase fraction, because the total energy dissipated per unit volume can be obtained by integrating over ΔG_R. In this investigation, we therefore introduce a method for finding ΔG_R.

(3) The explicit representation of ΔG_R has been found to be

$$\Delta G_R=\Delta G_d-k'\frac{A(T)}{B(T)}(\dot{T}t)^n\ . \tag{27}$$

While ΔG_d can be deduced from DSC measurements, there are still four parameters, i.e., k', $A(T)$, $B(T)$, and n, to be determined. In other words, we still need four other equations to close the system and obtain solutions to ΔG_R in a self-consistent manner. The special feature of our methodology is to carry out internal friction measurements of the system during the FOPT. Mathematically, we have added four equations (16), (19), (22), and (24) to the equation system (3b), (3c), which now include $\dot{T}$ and ω, that can be controlled during experimentation. We would remark that although many parameters appear in our deductions, such as $B_0,B_m,\eta,\omega_0^2,\epsilon_0,C',n,\propto(\omega),A(T),B(T),\rho,k',\ldots$, yet the final explicit representations of ΔG_R [i.e., Eq. (27)] involve only the four parameters just mentioned. In particular, we emphasize that the coupling coefficients $\alpha'(\omega),l$ involved in IF measurements are canceled during the mathematical manipulation. Such a result is expected because the crucial pa-

rameters in the FOPT should not be dependent on the characteristics of the internal friction measurement.

(4) There are limitations of the simple planar one-dimensional (1D) model taken in this investigation. In reality, the curvature of the moving "averaged" PI may change during the transformation. A variation of curvature of the PI in general would alter the characteristics of the dynamics of the PI. In a realistic system, a large number of PI's exist; they migrate and grow along different directions. As a result, the "tracks" of the new phase materials may intercept. The two limitations, when relaxed, would imply that the exponent indices n and l are functions of the new phase fraction rather than constants. In other words, there is a certain degree of inaccuracy in deducing n and l using our simple planar 1D model.

(5) We have neglected the effects of long-range diffusions of atoms during the FOPT. The theory is applicable as it stands, only to polymorphic (reconstructure type), martensite (displactive type), and other nondiffusion types of transformations such as order-disorder transitions and commensurate-incommensurate transitions. However, if we include a term describing the diffusion driving force and a term pertaining to the interaction between diffusion and vibrations applied during IF measurements, our theory can be extended to analyze diffusion transformations also.

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