

Mathematical Theory of Thermodynamics in Multi-Relaxation in Solids with Discrete Relaxation Spectra

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Abstract

Multi-relaxation in physics of any kind is discussed. First, thermodynamic consideration is developed mathematically in terms of the linear algebra to yield available expressions especially to the study of point defects. Single and multiple relaxations are well distinguished by means of a conjugate pair $\{x, \xi\}$ composed of the partial potential x and the density ξ . The notion of equivalence is introduced between two conjugate pairs. There are two distinct ways to consider the relaxation; the one is based on the transient process during the relaxation through the diffusion of density and the other on the condition of the resultant state in equilibrium. These two ways yield two different expressions for the relaxation of the compliance. Secondly diffusion operator, relaxation function are introduced, to readily derive the differential equation of the observing conjugate pair $\{x, \xi\}$ in such two ways as to use the relaxation function and the diffusion equation.

1. INTRODUCTION

The elasticity is a well-known property of solid since the work of Hooke¹⁾ in the 17th century. Hooke's law is, however, not always valid even for a small stress; it occurs that strain is not uniquely determined and lags behind a periodically oscillating harmonic stress by a certain phase angle. The creep, elastic aftereffect and stress relaxation are typical quasi-static relaxations, while the internal friction, frequency dependence of the compliance and the elastic modulus (or simply modulus) are dynamic phenomena different from the elasticity. Relaxations are also observed in ferromagnetic materials through magnetic field and magnetic flux density, and in ferroelectric materials through electric field and electric flux density. These relaxations are thermodynamically irreversible process; that is, irreversible process represented phenomenologically by equations without terms of higher than and equal to the 2nd order of the general force. Zener²⁾ defined the irreversible linear process concerning the stress and the strain as anelasticity, and stimulated researchers to lead to many theoretical and experimental works³⁾. This paper constructs mathematical fundamentals of multi-relaxation with discrete relaxation spectra in solids from the ther-

modynamic point of view.

2. ELASTICITY AND ANELASTICITY

Here, let physical quantities be functions of time in class C^∞ . Two physical quantities x , ξ , the product of which has the dimension of energy density (energy per unit volume), are called conjugate variables and the pair of the variables is called a conjugate pair, writtes as $\{x, \xi\}$; x or ξ is said to be conjugate to ξ or x , respectively, and x is called a partial potential and ξ a density. Although the following discussion is general enough, teminology of the anelasticity is used for simplicity unless it leads to misunderstanding.

The elasticity is the property that the conjugate pair $\{x, \xi\}$ satisfies Hooke's law:

$$x = J\xi \quad \text{or} \quad \xi = Mx, \quad M = 1/J, \quad (2.1)$$

where J and M are constants called the compliance and the modulus, respectively. Equation (2. 1) implies that

- (1) x (or ξ) is proportional to ξ (or x).
- (2) The proportionality constants J and M are independent of time.

Condition (2) is satisfied both just at the onset of ξ (or x) to the solid and after enough time elapsed to attain an equilibrium state of x (or ξ). The load of ξ (or x) causes an instantaneous unrelaxed state represented by eqn. (2. 1) with the unrelaxed compliance J_U and the unrelaxed modulus M_U . The other case satisfying Condition (2) corresponds to the equilibrium state expressed by the relaxed compliance J_R or the relaxed modulus M_R . The time independence in Condition (2) is realized in the two extremities of the relaxation process. The process from the onset to the equilibrium is a transient process, during which the instantaneous state relaxes to an equilibrium state. If the proportionality between the partial potential and the density holds during the transient process, the property is called the anelasticity.

Let a solid be in an equilibrium state and it suffers a change in density ξ from 0 (defined as 0 for the initial equilibrium state) to a small non-zero constant value ξ_0 . The partial potential x is then decomposed into two terms according to time dependence. The time-independent term is called the elastic partial potential written as x_e , and the time-dependent term the anelastic partial potential written as x_a , such that

$$x = x_e + x_a \quad (2.2)$$

$$x_e = J_U \xi_0 \quad (2.3)$$

$$x_a = \delta J(t) \xi_0, \quad (2.4)$$

where $\delta J(t)$ is the differential of the compliance. The anelasticity then induces a time-dependent term of the partial potential x , proportional to the density ξ . For the anelasticity Condition (2) is, therefore, replaced by

- (2') The differential of the compliance δJ (similarly δM) is dependent on time, or a function of time.

3. RELAXATION

A relaxation represented by an equation with a relaxation time is called a single relaxation and one represented with k different relaxation times, a multi-relaxation of order k , or simply a relaxation of order k . Let $(n+1)$ conjugate pairs involve in a relaxation. A conjugate pair should be fixed for the use of observing the relaxation, called the observing conjugate pair and denoted by $\{x^{(0)}, \xi^{(0)}\}$. The other n conjugate pairs are $\{x^{(i)}, \xi^{(i)}\} (1, 2, \dots, n)$. In this case, eqns. (2. 2), (2. 3), (2. 4) are written as

$$x^{(0)} = x_e^{(0)} + x_a^{(0)} \quad (3.1)$$

$$x_e^{(0)} = J_U^{(0)} \xi_0^{(0)} \quad (3.2)$$

$$x_a^{(0)} = \delta J^{(0)}(t) \xi_0^{(0)}. \quad (3.3)$$

3. 1 Single Relaxation

Consider $n = 1$ for simplicity. It is assumed that the anelastic partial potential $x_a^{(0)}$ is induced by the conjugate pair $\{x^{(1)}, \xi^{(1)}\}$ through the equation:

$$x_a^{(0)} = \lambda^{(1)} \xi^{(1)}(t) \quad (3.4)$$

with a proportionality constant $\lambda^{(1)}$. The density $\xi^{(1)}$ in equilibrium, denoted by $\bar{\xi}^{(1)}$, is furthermore assumed to be proportional to $\xi_0^{(0)}$, namely,

$$\bar{\xi}^{(1)} = \mu^{(1)} \xi_0^{(0)}, \quad (3.5)$$

where $\mu^{(1)}$ is a proportionality constant.

3. 1. 1 Diffusion of density

The diffusion of the density is assumed to be represented by the equation

$$\left(\frac{d}{dt} + \frac{1}{\tau_{\xi^{(1)}}^{(0)}} \right) \xi^{(1)} = \xi_0^{(0)} \frac{\mu^{(1)}}{\tau_{\xi^{(1)}}^{(0)}}. \quad (3.6)$$

Here, $\tau_{\xi^{(1)}}^{(0)}$ is called a relaxation time at constant $\xi^{(0)}$, and there involves only one relaxation time in the relaxation accompanied with a conjugate pair other than the observing conjugate pair. The case of $n = 1$ is, thus, a single relaxation. By operating

$\left(\frac{d}{dt} + \frac{1}{\tau_{\xi^{(1)}}^{(0)}} \right)$ to both sides of eqn. (3. 4), is obtained

$$\dot{x}_a^{(0)} + \frac{x_a^{(0)}}{\tau_{\xi^{(1)}}^{(0)}} = \frac{\lambda^{(1)} \mu^{(1)}}{\tau_{\xi^{(1)}}^{(0)}} \xi_0^{(0)}. \quad (3.7)$$

This equation is the equation corresponding to that of the Voigt-model solid^{3,4)}. The solution of eqn. (3. 7) is given by

$$x_a^{(0)}(t) = \lambda^{(1)} \mu^{(1)} \xi_0^{(0)} [1 - \exp(-t/\tau_{\xi^{(1)}}^{(0)})], \quad (3.8)$$

from the conditions: $x_a^{(0)}(0) = 0$ and $x_a^{(0)}(\infty) = \lambda^{(1)} \mu^{(1)} \xi_0^{(0)}$ ($\cdot \cdot$) in eqn. (3. 7), $\dot{x}_a^{(0)}(\infty) = 0$.

The differential of the compliance $\delta J^{(0)}(t)$ defined in eqn. (3. 3) is, thus, written as

$$\delta J^{(0)}(t) = \lambda^{(1)} \mu^{(1)} [1 - \exp(-t/\tau_{\xi^{(0)}}^{(1)})], \quad (3.9)$$

and, especially, the relaxation of the compliance $\delta J_R^{(0)}$, defined as $\delta J_R^{(0)} \stackrel{\text{def}}{=} x_a^{(0)}(\infty)/\xi_0^{(0)}$, is expressed as

$$\delta J_R^{(0)} = \lambda^{(1)} \mu^{(1)}. \quad (3.10)$$

3. 1. 2 Condition of equilibrium

The equilibrium state resulted from the onset of $\xi_0^{(0)}$ is attained when the driving force of the density for diffusion vanishes, i.e., $x^{(1)} = 0$. Since $x^{(1)}$ is given by

$$x^{(1)} = -\lambda^{(1)} \xi_0^{(0)} + b_{\xi^{(0)}}^{(1)} \xi^{(1)} \quad (3.11)$$

with a constant $b_{\xi^{(0)}}^{(1)}$, the relaxation of the compliance is expressed in a form other than eqn. (3. 10) as

$$\delta J_R^{(0)} = \frac{\lambda^{(1)2}}{b_{\xi^{(0)}}^{(1)}}. \quad (3.12)$$

In fact, eqn. (3. 11) becomes, in equilibrium,

$$0 = -\lambda^{(1)} \xi_0^{(0)} + b_{\xi^{(0)}}^{(1)} \bar{\xi}^{(1)}. \quad (3.13)$$

Combined with eqns. (3. 3), (3. 4), is derived

$$\delta J_R^{(0)} = \frac{x_a^{(0)}(\infty)}{\xi_0^{(0)}} = \frac{\lambda^{(1)} \xi^{(1)}}{\xi_0^{(0)}} = \frac{\lambda^{(1)2}}{b_{\xi^{(0)}}^{(1)}}. \quad (3.14)$$

3. 2 Multi-Relaxation

Return to the general case of $(n+1)$ conjugate pairs. Two conjugate pairs are said to be equivalent, if their relaxation times are equal. Now, let k non-equivalent relaxation times involve in the relaxation. Let $\tau_{\xi^{(i)}}^{(i)}$ ($i = 1, 2, \dots, k$) be k different relaxation times at constant $\xi^{(0)}$ related to the relaxation. Initially the solid is in equilibrium and then a change in density $\xi^{(0)}$, denoted by $\xi_0^{(0)}$, is given to the solid. The load deviates the solid from the initial equilibrium. For each $\tau_{\xi^{(i)}}^{(i)}$ there are $n^{(i)}$ equivalent conjugate pairs $\{x_j^{(i)}, \xi_j^{(i)}\}$ ($j = 1, 2, \dots, n^{(i)}$), so that $n = \sum_{i=1}^k n^{(i)}$. The observing partial potential consists of the two terms: the time-independent elastic term and the time-dependent anelastic term. In the case of multi-relaxation, $x_a^{(0)}$ is expressed as

$$x_a^{(0)} = \sum_{i=1}^k x_{a,i}^{(0)} \quad (3.15)$$

$$x_{a,i}^{(0)} = \vec{\lambda}^{(i)} \cdot \vec{\xi}^{(i)}, \quad (3.16)$$

where $x_{a,i}^{(0)}$'s are anelastic terms induced by the conjugate pairs $\{\vec{x}^{(i)}, \vec{\xi}^{(i)}\}$ ($i = 1, 2, \dots, k$) with $\vec{x}^{(i)} = {}^t(x_1^{(i)}, x_2^{(i)}, \dots, x_{n^{(i)}}^{(i)})$, $\vec{\xi}^{(i)} = {}^t(\xi_1^{(i)}, \xi_2^{(i)}, \dots, \xi_{n^{(i)}}^{(i)})$, $\vec{\lambda}^{(i)} = {}^t(\lambda_1^{(i)} \lambda_2^{(i)} \dots \lambda_{n^{(i)}}^{(i)})$ with constants $\lambda_j^{(i)}$ ($j = 1, 2, \dots, n^{(i)}$) and $\vec{\lambda}^{(i)} \cdot \vec{\xi}^{(i)}$ is the inner product of the two vectors. Each component of the density vector $\vec{\xi}^{(i)}$ in equilibrium, denoted by $\bar{\xi}^{(i)}$, is assumed

to be proportional to $\xi_0^{(0)}$, namely,

$$\vec{\xi}^{(i)} = \xi_0^{(0)} \vec{\mu}^{(i)},$$

where $\vec{\mu}^{(i)}$ is the vector composed of the proportionality constants.

3. 2. 1 Diffusion of density

The temperature determines the active relaxations which occur in solid. Here, let the i -th conjugate pair $\{x^{(i)}, \vec{\xi}^{(i)}\}$ dominates the relaxation. The solid perturbed by the onset of $\xi_0^{(0)}$ approaches to an equilibrium with the diffusion of the densities $\xi_j^{(i)} (j = 1, 2, \dots, n^{(i)})$ which correspond to a relaxation of the same relaxation time $\tau_{\xi}^{(i)}$. The equation of the diffusion of density is given by

$$\left(\frac{d}{dt} + \frac{1}{\tau_{\xi}^{(i)}} \right) \vec{\xi}^{(i)} = \xi_0^{(0)} \frac{\vec{\mu}^{(i)}}{\tau_{\xi}^{(i)}}. \quad (3.17)$$

Operating $\left(\frac{d}{dt} + \frac{1}{\tau_{\xi}^{(i)}} \right)$ to both sides of eqn. (3. 16) yields

$$\dot{x}_{a,i}^{(0)} + \frac{x_{a,i}^{(0)}}{\tau_{\xi}^{(i)}} = \frac{\vec{\lambda}^{(i)} \cdot \vec{\mu}^{(i)}}{\tau_{\xi}^{(i)}} \xi_0^{(0)} \quad (3.18)$$

The relaxation of the compliance is, in this case, expressed in the form:

$$\delta J_{k,i}^{(0)} = \vec{\lambda}^{(i)} \cdot \vec{\mu}^{(i)} \quad (3.19)$$

3. 2. 2 Condition of equilibrium

In the previous paragraph, the relaxation of the compliance is derived, based on the transient process from the initial perturbed state to the final equilibrium which corresponds to the relaxation through the diffusion of density. Another formulation of the relaxation of the compliance is attainable by considering only the final equilibrium where the deriving force for the diffusion of density vanishes. Since

$$du = x^{(0)} d\xi^{(0)} - \sum_{i=1}^k x^{(i)} \cdot d\vec{\xi}^{(i)} \quad (3.20)$$

with the internal energy density u , the partial potential $\vec{x}^{(i)}$ is

$$\vec{x}^{(i)} = -\text{grad } \vec{\xi}^{(i)} u, \quad (3.21)$$

which implies that $\vec{x}^{(i)}$ is the deriving force for the diffusion of the density $\vec{\xi}^{(i)}$. In equilibrium, the deriving force is zero; that is,

$$\vec{x}^{(i)} = \vec{0}. \quad (3.22)$$

Let $\vec{x}^{(i)}$ and $\vec{\xi}^{(i)}$ be related by a linear equation

$$\vec{x}^{(i)} = -(\vec{\lambda}^{(i)}) \xi_0^{(0)} + B_{\xi}^{(i)} \vec{\xi}^{(i)}, \quad (3.23)$$

where $B_{\xi}^{(i)}$ is a symmetric $n^{(i)}$ square matrix. The condition of equilibrium (3. 22), then, yields the following expression different from eqn. (3. 19)

$$\delta J_{R,i}^{(0)} = \left\| D_{\xi^{(i)}}^{(i)-1} P_{\xi^{(i)}}^{(i)} \vec{\lambda}^{(i)} \right\|^2, \quad (3.24)$$

where $D_{\xi^{(i)}}$ and $P_{\xi^{(i)}}$ are $n^{(i)}$ square diagonal and orthogonal matrices, respectively, such that

$$B_{\xi^{(i)}}^{(i)} = P_{\xi^{(i)}}^{(i)} D_{\xi^{(i)}}^{(i)2} P_{\xi^{(i)}}^{(i)}. \quad (3.25)$$

The representation (3. 24) is available especially to the study of point defects^{3,5)}.

3. 3 Equation of Multi-Relaxation

If many relaxtions are simultaneously activated, their superposition is observed through the observing partial potential $x^{(0)}$. Each relaxation may individually develop, if the interaction of conjugate pairs is negligible. In this case the relaxation of the compliance is also expressed as (3. 19) and (3. 24) for the active relaxations and $\delta J_{R,i}^{(0)} = 0$ for the inactive, by considering that the vector $\vec{\mu}^{(i)}$ is $\vec{0}$ and the matrix $B_{\xi^{(i)}}^{(i)}$ is zero matrix for the inactive conjugate pairs at the given temperature of the solid.

The relaxation of the observing partial potential is due to the relaxation related to the conjugate pairs other than the observing conjugate pair. A direct relation of $x^{(0)}$ and $\xi^{(0)}$ is consequently derived by subntituting $\delta J_{R,i}^{(0)}$ for $\vec{\lambda}^{(i)} \cdot \vec{\mu}^{(i)}$ and expressed as the equation of multi-relaxation:

$$x^{(0)} = \left[J_U^{(0)} + \sum_{i=1}^k \delta J_R^{(i)} \{1 - \exp(-t/\tau_{\xi^{(i)}})\} \right] \xi_0^{(0)} \quad (3.26)$$

or
$$x_a^{(0)} = \left[\sum_{i=1}^k \delta J_R^{(i)} \{1 - \exp(-t/\tau_{\xi^{(i)}})\} \right] \xi_0^{(0)}. \quad (3.27)$$

4. DIFERENTIAL EQUATION OF MULTI-RELAXATION

4. 1 Diffusion Operator

Let D_τ be

$$D_\tau \stackrel{\text{def}}{=} \frac{d}{dt} + \frac{1}{\tau} I = d_t + \frac{1}{\tau} I, \quad (4.1)$$

where $d_t \stackrel{\text{def}}{=} \frac{d}{dt}$, and τ is a real positive constant; i.e., $\tau \in \mathbf{R}^+$ (\mathbf{R}^+ : the set of all positive real numbers) and I is the identity operator. Since the equation of diffusion is written as epn. (3. 17):

$$D_\tau \vec{\xi}^{(i)} = \xi_0^{(0)} \frac{\vec{\mu}^{(i)}}{\tau_{\xi^{(i)}}}, \quad (4.2)$$

the operator is, here, called diffusion operator. The following propositions are readily derived from eqn. (4. 1).

Proposition 1 The diffusion operator satisfies the following properties:

- (1) D_τ is a linear operator of C^∞ into C^∞ .

$$(2) \quad D_\tau f(t) = 0 \iff f(t) = a \exp(-t/\tau) (a \in \mathbf{R})$$

$$(3) \quad D_\tau \exp(-t/\tau) = \left(\frac{1}{\tau'} - \frac{1}{\tau} \right) \exp(-t/\tau) \quad (4.3)$$

especially for $\tau' = \tau$,

$$D_\tau \exp(-t/\tau) = 0 \quad (4.4)$$

$$(4) \quad D_\tau a = \frac{a}{\tau} (a \in \mathbf{R}) \quad (4.5)$$

$$(5) \quad \varphi_\tau(t) \stackrel{\text{def}}{=} 1 - \exp(-t/\tau), \text{ called a relaxation function}$$

$$D_\tau \varphi_\tau(t) = \left(\frac{1}{\tau'} - \frac{1}{\tau} \right) \varphi_\tau(t) + \frac{1}{\tau} \quad (4.6)$$

especially for $\tau' = \tau$,

$$D_\tau \varphi_\tau(t) = \frac{1}{\tau} \quad (4.7)$$

$$(6) \quad f, g \in \mathbf{C}^\infty$$

$$D_\tau (fg) = g D_\tau f + f D_\tau g - \frac{fg}{\tau'} \quad (4.8)$$

$$(7) \quad D_\tau (\varphi_\tau f) = \varphi_\tau D_\tau f + \left(\frac{1}{\tau'} - \frac{1}{\tau} \right) \varphi_\tau f + \frac{1}{\tau} \left(1 - \frac{\tau}{\tau'} \varphi_\tau \right) f \quad (4.9)$$

especially for $\tau' = \tau$,

$$D_\tau (\varphi_\tau f) = \varphi_\tau D_\tau f + \frac{1}{\tau} (1 - \varphi_\tau) f \quad (4.10)$$

Proof.

$$(1) \quad \text{For } \forall f_1, f_2 \in \mathbf{C}^\infty, \forall a_1, a_2 \in \mathbf{R},$$

$$\begin{aligned} D_\tau (a_1 f_1 + a_2 f_2) &= \left(d_t + \frac{1}{\tau} I \right) (a_1 f_1 + a_2 f_2) = d_t (a_1 f_1 + a_2 f_2) + \frac{1}{\tau} (a_1 f_1 + a_2 f_2) \\ &= (a_1 d_t f_1 + a_2 d_t f_2) + \left(a_1 \frac{f_1}{\tau} + a_2 \frac{f_2}{\tau} \right) = a_1 \left(d_t + \frac{1}{\tau} I \right) f_1 + a_2 \left(d_t + \frac{1}{\tau} I \right) f_2 \\ &= a_1 D_\tau f_1 + a_2 D_\tau f_2. \end{aligned}$$

(2), (3), (4) are easily proved.

$$(5) \quad \begin{aligned} D_\tau \varphi_\tau(t) &= D_\tau (1) - D_\tau \exp(-t/\tau) = \frac{1}{\tau'} - \left(\frac{1}{\tau'} - \frac{1}{\tau} \right) \exp(-t/\tau) \\ &= \left(\frac{1}{\tau'} - \frac{1}{\tau} \right) \{1 - \exp(-t/\tau)\} + \frac{1}{\tau} = \left(\frac{1}{\tau'} - \frac{1}{\tau} \right) \varphi_\tau(t) + \frac{1}{\tau}. \end{aligned}$$

$$(6) \quad \begin{aligned} D_\tau (fg) &= \left(d_t + \frac{1}{\tau} I \right) (fg) = f'g + fg' + \frac{fg}{\tau'} \\ &= g \left(d_t + \frac{1}{\tau} I \right) f + f \left(d_t + \frac{1}{\tau} I \right) g - \frac{fg}{\tau'} = g D_\tau f + f D_\tau g - \frac{fg}{\tau'} \end{aligned}$$

$$(7) \quad \begin{aligned} D_\tau (\varphi_\tau f) &= f D_\tau \varphi_\tau + \varphi_\tau D_\tau f - \frac{\varphi_\tau f}{\tau'} = \left\{ f \left(\frac{1}{\tau'} - \frac{1}{\tau} \right) \varphi_\tau + \frac{f}{\tau} \right\} + \varphi_\tau D_\tau f - \frac{\varphi_\tau f}{\tau'} \\ &= \varphi_\tau D_\tau f + \left(\frac{1}{\tau'} - \frac{1}{\tau} \right) \varphi_\tau f + \frac{1}{\tau} \left(1 - \frac{\tau}{\tau'} \varphi_\tau \right) f. \end{aligned}$$

Definition 1

$$(1) \quad \mathfrak{D} \stackrel{\text{def}}{=} \{D_\tau | \tau \in \mathbf{R}^+ \cup \{\infty\}\}, D_0 \stackrel{\text{def}}{=} I, D_\infty \stackrel{\text{def}}{=} d_t$$

(2) For $\forall D_{\tau_1}, D_{\tau_2} \in \mathfrak{D}$,

$$(D_{\tau_1}D_{\tau_2})f \stackrel{\text{def}}{=} D_{\tau_1}(D_{\tau_2}f) \quad (f \in C^\infty) \quad (4.11)$$

$$(3) \quad d_t^n \stackrel{\text{def}}{=} \frac{d^n}{dt^n}, \quad d_t^0 \stackrel{\text{def}}{=} I$$

Proposition 2 For $\forall D_{\tau_1}, D_{\tau_2}, \dots, D_{\tau_n} \in \mathfrak{D}$,

(8) $D_{\tau_1}D_{\tau_2}$ is a linear operator of C^∞ into C^∞

$$(9) \quad D_{\tau_1}D_{\tau_2} = D_{\tau_2}D_{\tau_1} \quad (4.12)$$

$$(10) \quad (D_{\tau_1}D_{\tau_2})D_{\tau_3} = D_{\tau_1}(D_{\tau_2}D_{\tau_3}) \quad (4.13)$$

$$(11) \quad D_{\tau_1}D_{\tau_2} = d_t^2 + \left(\frac{1}{\tau_1} + \frac{1}{\tau_2}\right)d_t + \frac{1}{\tau_1\tau_2} \quad (4.14)$$

$$(12) \quad \prod_{i=1}^n D_{\tau_i} = \left(\prod_{i=1}^{n-1} D_{\tau_i}\right)D_{\tau_n} = \left(\prod_{i=1}^{n-1} D_{\tau_i}\right)d_t + \left(\prod_{i=1}^{n-1} D_{\tau_i}\right)\frac{1}{\tau_n}I \\ = d_t^n + \left(\sum_{i=1}^n \frac{1}{\tau_i}\right)d_t^{n-1} + \dots + \left(\prod_{i=1}^n \frac{1}{\tau_i}\right)I = \sum_{i=0}^n \left(\sum_{\Omega_i \subset \Omega_n} \prod_{j \in \Omega_i} \frac{1}{\tau_j}\right)d_t^{n-i}, \quad (4.15)$$

where Ω_i is a subset of i elements of Ω_n , called a subset of order i of Ω_n , the summation $\sum_{\Omega_i \subset \Omega_n}$ is made over all the subset of order i of Ω_n and $\prod_{j \in \Omega_i}$ means the product over all the elements of Ω_i , and, furthermore, $\Omega_0 \stackrel{\text{def}}{=} \phi$, $\prod_{j \in \emptyset} \frac{1}{\tau} \stackrel{\text{def}}{=} 1$, $\sum_{\Omega_i \subset \emptyset} 1 \stackrel{\text{def}}{=} 1$.

Proof.

(8) D_{τ_1}, D_{τ_2} are linear operators. Then,

$$(D_{\tau_1}D_{\tau_2})(a_1f_1 + a_2f_2) = D_{\tau_1}(a_1D_{\tau_2}f_1 + a_2D_{\tau_2}f_2) = a_1D_{\tau_1}D_{\tau_2}f_1 + a_2D_{\tau_1}D_{\tau_2}f_2.$$

$$(9) \quad (D_{\tau_1}D_{\tau_2})f = \left(d_t + \frac{1}{\tau_1}I\right)\left[\left(d_t + \frac{1}{\tau_2}I\right)f\right] = \left(d_t + \frac{1}{\tau_1}I\right)\left(d_t f + \frac{f}{\tau_2}\right) \\ = d_t^2 f + \left(\frac{1}{\tau_1}d_t f + \frac{1}{\tau_2}d_t f\right) + \frac{1}{\tau_1\tau_2}f = \left[d_t^2 + \left(\frac{1}{\tau_1} + \frac{1}{\tau_2}\right)d_t + \frac{1}{\tau_1\tau_2}\right]f.$$

(10) is easily proved, similarly to (9), and (11), (12) are derived from (9), (10).

4. 2 Derivation of the Differential Equation Based on the Relaxation Function

Equation (3. 27) is written as

$$x_a^{(0)} = \left(\sum_{i=1}^k \delta J_{R,i}^{(0)} \varphi_{\tau_i^{(i)}}\right) \xi_0^{(0)}. \quad (4.16)$$

By defining operators D and D_i as

$$D \stackrel{\text{def}}{=} \prod_{i=1}^k D_i, \text{ and } D_i \stackrel{\text{def}}{=} d_t + \frac{1}{\tau_i^{(i)}}I, \quad (4.17)$$

and operating them to eqn. (4. 16),

$$Dx_a^{(0)} = \sum_{i=1}^k \delta J_{R,i}^{(0)} D(\varphi_{\tau_i^{(i)}}) \xi_0^{(0)} \\ = \sum_{i=1}^k \delta J_{R,i}^{(0)} \prod_{j=1, j \neq i}^k D_j \left(D_i \left(\varphi_{\tau_i^{(i)}} \right) \xi_0^{(0)} \right)$$

$$\begin{aligned}
 &= \sum_{i=1}^k \delta J_{R,i}^{(0)} \prod_{j=1, j \neq i}^k D_j \left(\frac{\xi_0^{(0)}}{\tau_{\xi^{(0)}}^{(i)}} \right) \\
 &= \sum_{i=1}^k \frac{\delta J_{R,i}^{(0)}}{\tau_{\xi^{(0)}}^{(i)}} \prod_{j=1, j \neq i}^k D_j \xi_0^{(0)}
 \end{aligned} \tag{4.18}$$

$$\therefore D x_a^{(0)} = \left(\sum_{i=1}^k \frac{\delta J_{R,i}^{(0)}}{\tau_{\xi^{(0)}}^{(i)}} \prod_{j=1, j \neq i}^k D_j \right) \xi_0^{(0)} \tag{4.19}$$

$$x_a^{(0)} = x^{(0)} - x_e^{(0)} = x^{(0)} - J_U^{(0)} \xi_0^{(0)} \tag{4.20}$$

$$\therefore D x^{(0)} = \left(J_U^{(0)} D + \sum_{i=1}^k \frac{\delta J_{R,i}^{(0)}}{\tau_{\xi^{(0)}}^{(i)}} \prod_{j=1, j \neq i}^k D_j \right) \xi_0^{(0)} \tag{4.21}$$

with $\prod_{j=1, j \neq i}^k D_j \stackrel{\text{def}}{=} I \quad (k=1)$ (4.22)

4. 3 Derivation of the Differential Equation Based on the Diffusion Equation

From eqns. (3. 8), (3. 19), the diffusion equation is given by

$$D_i x_{a,i}^{(0)} = \frac{\delta J_{R,i}^{(0)}}{\tau_{\xi^{(0)}}^{(i)}} \xi_0^{(0)}. \tag{4.23}$$

Then,

$$\begin{aligned}
 D x_a^{(0)} &= \sum_{i=1}^k D x_{a,i}^{(0)} = \sum_{i=1}^k \prod_{j=1, j \neq i}^k D_j (D_i x_{a,i}^{(0)}) \\
 &= \sum_{i=1}^k \prod_{j=1, j \neq i}^k D_j \left(\frac{\delta J_{R,i}^{(0)}}{\tau_{\xi^{(0)}}^{(i)}} \xi_0^{(0)} \right) = \sum_{i=1}^k \frac{\delta J_{R,i}^{(0)}}{\tau_{\xi^{(0)}}^{(i)}} \prod_{j=1, j \neq i}^k D_j \xi_0^{(0)}.
 \end{aligned} \tag{4.24}$$

4. 4 Explicit Representation of the Differential Equation of Multi-Relaxation of Order ≤ 3

Here, the observing conjugate variable $\xi^{(0)}$ is considered any function in C^∞ and formally substituted for $\xi_0^{(0)}$. The reasoning will be given in another paper⁷⁾.

4. 4. 1 Single relaxation ($k=1$)

Since $D = D_1$,

$$D_1 x^{(0)} = \left(J_U D_1 + \frac{\delta J_{R,1}^{(0)}}{\tau_{\xi^{(0)}}^{(1)}} \right) \xi^{(0)}. \tag{4.25}$$

$$\therefore \dot{x}^{(0)} + \frac{x^{(0)}}{\tau_{\xi^{(0)}}^{(1)}} = J_U^{(0)} \dot{\xi}^{(0)} + \frac{J_U^{(0)} + \delta J_{R,1}^{(0)}}{\tau_{\xi^{(0)}}^{(1)}} \xi^{(0)}. \tag{4.26}$$

4. 4. 2 Relaxation of order 2 ($k=2$)

$$D_1 D_2 \dot{x}^{(0)} = \left[J_U^{(0)} D_1 D_2 + \frac{\delta J_{R,1}^{(0)}}{\tau_{\xi^{(0)}}^{(1)}} D_2 + \frac{\delta J_{R,2}^{(0)}}{\tau_{\xi^{(0)}}^{(2)}} D_1 \right] \xi_0^{(0)}. \tag{4.27}$$

$$\begin{aligned}
 \therefore \dot{x}^{(0)} + \left(\frac{1}{\tau_{\xi^{(0)}}^{(1)}} + \frac{1}{\tau_{\xi^{(0)}}^{(2)}} \right) x^{(0)} + \frac{x^{(0)}}{\tau_{\xi^{(0)}}^{(1)} \tau_{\xi^{(0)}}^{(2)}} &= J_U^{(0)} \dot{\xi}^{(0)} + \left(\frac{J_U^{(0)} + \delta J_{R,1}^{(0)}}{\tau_{\xi^{(0)}}^{(1)}} + \frac{J_U^{(0)} + \delta J_{R,2}^{(0)}}{\tau_{\xi^{(0)}}^{(2)}} \right) \xi^{(0)} \\
 &+ \frac{J_U^{(0)} + \delta J_{R,1}^{(0)} + \delta J_{R,2}^{(0)}}{\tau_{\xi^{(0)}}^{(1)} \tau_{\xi^{(0)}}^{(2)}} \xi^{(0)}.
 \end{aligned} \tag{4.28}$$

4. 4. 3 Relaxation of order 3 ($k=3$)

$$D_1 D_2 D_3 \dot{x}^{(0)} = \left[J_U^{(0)} D_1 D_2 D_3 + \frac{\delta J_{R,1}^{(0)}}{\tau_{\xi^{(0)}}^{(1)}} D_2 D_3 + \frac{\delta J_{R,2}^{(0)}}{\tau_{\xi^{(0)}}^{(2)}} D_3 D_1 + \frac{\delta J_{R,3}^{(0)}}{\tau_{\xi^{(0)}}^{(3)}} D_1 D_2 \right] \xi_0^{(0)}. \tag{4.29}$$

$$\therefore \ddot{x}^{(0)} + \left(\frac{1}{\tau_{\xi^{(0)}}^{(1)}} + \frac{1}{\tau_{\xi^{(0)}}^{(2)}} + \frac{1}{\tau_{\xi^{(0)}}^{(3)}} \right) \dot{x}^{(0)} + \left(\frac{1}{\tau_{\xi^{(0)}}^{(1)} \tau_{\xi^{(0)}}^{(2)}} + \frac{1}{\tau_{\xi^{(0)}}^{(2)} \tau_{\xi^{(0)}}^{(3)}} + \frac{1}{\tau_{\xi^{(0)}}^{(3)} \tau_{\xi^{(0)}}^{(1)}} \right) x^{(0)} + \frac{x^{(0)}}{\tau_{\xi^{(0)}}^{(1)} \tau_{\xi^{(0)}}^{(2)} \tau_{\xi^{(0)}}^{(3)}}$$

$$\begin{aligned}
&= J_U^{(0)} \dot{\xi}^{(0)} + \left(\frac{J_U^{(0)} + \delta J_{R,1}^{(0)}}{\tau_{\xi^{(0)}}^{(1)}} + \frac{J_U^{(0)} + \delta J_{R,2}^{(0)}}{\tau_{\xi^{(0)}}^{(2)}} + \frac{J_U^{(0)} + \delta J_{R,3}^{(0)}}{\tau_{\xi^{(0)}}^{(3)}} \right) \ddot{\xi}^{(0)} + \left(\frac{J_U^{(0)} + \delta J_{R,1}^{(0)} + \delta J_{R,2}^{(0)}}{\tau_{\xi^{(0)}}^{(1)} \tau_{\xi^{(0)}}^{(2)}} \right. \\
&+ \left. \frac{J_U^{(0)} + \delta J_{R,2}^{(0)} + \delta J_{R,3}^{(0)}}{\tau_{\xi^{(0)}}^{(2)} \tau_{\xi^{(0)}}^{(3)}} + \frac{J_U^{(0)} + \delta J_{R,3}^{(0)} + \delta J_{R,1}^{(0)}}{\tau_{\xi^{(0)}}^{(3)} \tau_{\xi^{(0)}}^{(1)}} \right) \dot{\xi}^{(0)} + \frac{J_U^{(0)} + \delta J_{R,1}^{(0)} + \delta J_{R,2}^{(0)} + \delta J_{R,3}^{(0)}}{\tau_{\xi^{(0)}}^{(1)} \tau_{\xi^{(0)}}^{(2)} \tau_{\xi^{(0)}}^{(3)}} \xi^{(0)}. \quad (4.30)
\end{aligned}$$

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