Non-equilibrium thermodynamics as gauge fixing

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Received June 19, 2018; Revised August 11, 2018; Accepted August 24, 2018; Published September 27, 2018

By considering a thermodynamic force as a gauge field, we extend the constitutive equations of Onsager's [L.Onsager, Phys. Rev. **37**, 405 (1931)] non-equilibrium thermodynamics to non-linear equations. In Onsager's non-equilibrium thermodynamics, the thermodynamic force corresponds to a pure gauge, for which the constitutive equations are obtained by gauge fixing. If we extend the thermodynamic force from a pure gauge to a physical one, we obtain non-linear constitutive equations of non-equilibrium thermodynamics.

Subject Index A51, A53, A56, A58, B00

1. Introduction

Onsager's theory is the most important one in non-equilibrium thermodynamics with linear constitutive equations [1,2], in which constitutive equations for currents are derived from the minimum energy dissipation principle. Later on, this argument was supported by the path-integral representation of the probability distribution [3–7]. Onsager's theory holds in the case of linear constitutive equations, but it is not well understood in the non-linear case (for the latest research see Refs. [8,9]). Recently, Sugamoto, along with his collaborators, including the present author, pointed out that the thermodynamic force can be viewed as a gauge field (A. Sugamoto, private communication and Ref. [10]).

In this paper we discuss this statement more definitely by means of gauge fixing, and derive a non-linear constitutive equation by adding the free action of the usual electromagnetism.

The paper contains the following sections. In the next section, we review the electromagnetism in a pure gauge as an useful analogy in later discussions. In Sect. 3, non-equilibrium thermodynamics is introduced using a differential form. In Sect. 4, we examine the gauge properties of the thermodynamic force. We extend the thermodynamic force to a thermodynamical gauge field in Sect. 5. In Sect. 6, we discuss this gauge theory in the path-integral method. The final section is devoted to the discussion.

In addition, the paper contains the following appendices. In Appendix A, the meaning of the time dependence of S(a, t) is discussed. In Appendix B, we examine the restriction from the second law of thermodynamics. In Appendix C, we discuss how non-linear effects work in dimensional analysis. In Appendix D, a simple example is derived in our model.

2. Electromagnetism

First, we consider electromagnetism as a typical gauge theory. The notion of a pure gauge and its gauge transformation (local phase shift) is important in later sections in order to consider thermodynamics as a gauge theory.

As is well known, a pure gauge A_{μ} is described as

$$d\theta(x) = A_{\mu}(x)dx^{\mu},\tag{1}$$

where

$$A_{\mu}(x) \equiv \frac{\partial \theta(x)}{\partial x^{\mu}},\tag{2}$$

using phase $\theta(x)$. The gauge transformation

$$\delta A_{\mu}(x) = \partial_{\mu} \epsilon(x) \tag{3}$$

causes a phase shift

$$\delta\theta(x) = \epsilon(x). \tag{4}$$

At the pure gauge, the electromagnetic field F is zero:

$$F = dA = dd\theta = 0. \tag{5}$$

For a general gauge potential A_{μ} , F satisfies Maxwell's equations:

$$\partial_{\mu}F_{\nu\rho} + \partial_{\nu}F_{\rho\mu} + \partial_{\rho}F_{\mu\nu} = 0, \tag{6}$$

$$\partial^{\mu}F_{\mu\nu} = 0. \tag{7}$$

3. Differential form of non-equilibrium thermodynamics

A linear constitutive equation in non-equilibrium thermodynamics is expressed as

$$\dot{a}^i = L^{ij} X_j, \tag{8}$$

where a^i is an extensive quantity for entropy, X_j is the thermodynamic force for a^j , and L^{ij} is Onsager's phenomenological matrix. According to Onsager's theory [1,2], Eq. (8) is obtained by taking the variation

$$\delta\left(-\frac{1}{2}R_{ij}\dot{a}^{i}\dot{a}^{j} + X_{i}\dot{a}^{i}\right) = 0 \tag{9}$$

with respect to \dot{a}^i , where R_{ij} is the inverse matrix of L^{ij} :

$$R_{ij}L^{jk} = \delta_i^k. \tag{10}$$

As its dual version, Eq. (8) is also obtained by taking the variation

$$\delta\left(-\frac{1}{2}L^{ij}X_iX_j + X_i\dot{a}^i\right) = 0\tag{11}$$

with respect to X_i .

At first glance, thermodynamics and electromagnetism are different theories. In thermodynamics, an important quantity, entropy S(a, t), is described in terms of the extensive quantities a^i and time t.

If we describe, however, entropy S(a, t) in terms of a differential form, then quantities analogous to the Lagrangian L and gauge field A appear.¹

The differential form of entropy S(a, t) is expressed as

$$dS(a,t) = \Phi(a,t)dt + X_i(a,t)da^i,$$
(12)

with

$$\Phi(a,t) \equiv \frac{\partial S(a,t)}{\partial t},\tag{13}$$

$$X_i(a,t) \equiv \frac{\partial S(a,t)}{\partial a^i},\tag{14}$$

where $\Phi(a, t)$ is called the dissipation function.

We will take the following notation:

$$X_0 \equiv \Phi, \tag{15}$$

$$a^0 \equiv t, \tag{16}$$

and X_{μ} , a^{μ} by

$$\{X_{\mu}\} = X_0, X_1, \dots, X_N, \tag{17}$$

$$\{a^{\mu}\} = a^0, a^1, \dots, a^N.$$
(18)

Then, Eq. (12) is rewritten as

$$dS(a) = X_{\mu}(a)da^{\mu},\tag{19}$$

where

$$X_{\mu}(a) \equiv \frac{\partial S(a)}{\partial a^{\mu}}.$$
(20)

If we describe a^{μ} using parameter τ , we get the non-equilibrium thermodynamical Lagrangian L as follows:²

$$L(a, \frac{da}{d\tau})d\tau = X_{\mu}(a)\frac{da^{\mu}}{d\tau}d\tau.$$
(21)

4. Gauge transformation property of X_{μ}

In order to know the properties of the above non-equilibrium thermodynamical Lagrangian L, we have to choose an "appropriate gauge-fixing condition" so that we can get Onsager's phenomenological transfer matrix L^{ij} and its constitutive equations for X_i .

The Lagrangian L in Eq. (21) has gauge symmetry described by the transformation

$$\delta X_{\mu}(a) = \partial_{\mu} \epsilon(a). \tag{22}$$

¹ In non-equilibrium thermodynamics, the time dependence of S(a, t) is natural when we consider the path-integral formalism and Wentzel–Kramers–Brillouin (WKB) approximation; see Appendix A.

² The parameter τ is an arbitrary parameter, and it can always be introduced into a dynamical system.

The transformation (22) causes an entropy shift:

$$\delta S(a) = \epsilon(a). \tag{23}$$

Since the gauge transform (22) introduces an entropy shift (23), we want to fix this gauge. To fix the gauge, we take a gauge-fixing condition:

$$X_0(a) = -\frac{1}{2} L^{ij} X_i(a) X_j(a).$$
(24)

Here, we assume $\tau = t$ to fix the parametrization invariance.

Finally, the thermodynamical Lagrangian L becomes

$$L(a,\dot{a})dt = -\frac{1}{2}L^{ij}X_i(a)X_j(a)dt + X_i(a)\dot{a}^i dt.$$
 (25)

Since X_i is a non-dynamical field, the equation of motion for X_i produces a constitutive equation [1,2]:

$$\dot{a}^i = L^{ij} X_i(a). \tag{26}$$

Substituting Eq. (26) into Eq. (25), we obtain

$$L(a,\dot{a})dt = \frac{1}{2}R_{ij}\dot{a}^{i}\dot{a}^{j}dt.^{3}$$
(27)

5. Thermodynamical gauge theory

According to gauge theory, if we consider the thermodynamic force as a gauge potential, then Eq. (25), with Onsager's phenomenological matrix, corresponds to a pure gauge theory. On the other hand, if we introduce a physical gauge theory instead of the pure one into this thermodynamic system, then we will obtain a different equation of motion for X_{μ} , reflecting the gauge-fixing condition for the physical gauge theory. This equation of motion is similar to Maxwell's equations in electromagnetism. Furthermore, we can obtain the non-linear constitutive equation of non-equilibrium thermodynamics.

So far we define the thermodynamic gauge field F as

$$F(a) = dX(a), \tag{28}$$

but because dS(a) = X(a), F(a) is always zero:

$$F(a) = ddS(a) = 0.$$
⁽²⁹⁾

Now we will generalize this, and assume that X_{μ} is not a pure gauge field, but a gauge field in a physical gauge theory.⁴ The action

$$S_F[X] = \int d^{N+1} a F_{\mu\nu} F^{\mu\nu}$$
(30)

³ This corresponds to the Rayleigh dissipation function.

⁴ Its dimension is N + 1, which is the number of components of X.

is added to the original action with a gauge-fixing condition:

$$S_a[a, X, \lambda] = \int d\tau \left(X_\mu(a) \frac{da^\mu}{d\tau} - \lambda \left(X_0(a) + \frac{1}{2} L^{ij} X_i(a) X_j(a) \right) \right), \tag{31}$$

where λ is a Lagrange multiplier.

The equation of motion for X_{μ} is

$$\frac{da^0}{d\tau} - \lambda = \partial_\mu F^{\mu 0}(X) = \partial_i E^i(X), \qquad (32)$$

$$\frac{da^{i}}{d\tau} - \lambda L^{ij} X_{j}(a) = \partial_{\mu} F^{\mu i}(X) = \dot{E}^{i}(X) + \partial_{j} B^{ji}(X), \qquad (33)$$

where E_i and B_{ij} are

$$E_i(X) \equiv F_{i0}(X) = \partial_i \Phi(X) - \dot{X}_i$$
$$= L^{jk} X_j \partial_i X_k - \dot{X}_i, \qquad (34)$$

$$B_{ij}(X) \equiv F_{ij}(X) = \partial_i X_j - \partial_j X_i.$$
(35)

We call $E_i(X)$ and $B_{ij}(X)$ the thermodynamic electric field and thermodynamic magnetic field, respectively.

Equation (32) is solved for λ :

$$\lambda = \frac{da^0}{d\tau} - \partial_i E^i,\tag{36}$$

and we take the static gauge $\tau = t$; then Eq. (33) yields

$$\dot{a}^{i} = \left(1 + \partial_{k}E^{k}\right)L^{ij}X_{j} + \dot{E}^{i} + \partial_{j}B^{ji}.$$
(37)

From

$$\partial_i E^i = L^{il} L^{jk} \partial_l X_j \partial_i X_k + L^{il} L^{jk} X_j \partial_l \partial_i X_k - L^{ij} \partial_i \dot{X}_j, \tag{38}$$

$$\dot{E}_i = L^{jk} \dot{X}_j \partial_i X_k + L^{jk} X_j \partial_i \dot{X}_k - \ddot{X}_i, \tag{39}$$

we get a non-linear constitutive equation of non-equilibrium thermodynamics:

$$\dot{a}^i = L^{ij}X_j + \xi^i,\tag{40}$$

where ξ^i is a term from the thermodynamic gauge fields:

$$\xi^{i} = L^{ml}L^{jk}L^{in}\partial_{l}X_{j}\partial_{m}X_{k}X_{n} + L^{ml}L^{jk}L^{in}X_{j}\partial_{l}\partial_{m}X_{k}X_{n} - L^{kj}L^{il}\partial_{k}\dot{X}_{j}X_{l}$$

$$+ L^{jk}L^{il}\dot{X}_{j}\partial_{l}X_{k} + L^{jk}L^{il}X_{j}\partial_{l}\dot{X}_{k} - L^{ij}\ddot{X}_{j}$$

$$+ L^{jk}L^{il}\partial_{j}\partial_{k}X_{l} - L^{jk}L^{il}\partial_{j}\partial_{l}X_{k}.$$
(41)

This means that, by introducing the usual kinetic action of electromagnetism into the thermodynamics, the gauge field $X_i(a)$ can have additional transverse components, in addition to the longitudinal (or the pure gauge) component existing in Onsager's non-equilibrium thermodynamics. Then, the non-linear constitutive equation is obtained. The second law of thermodynamics restricts thermodynamic gauge fields to $\dot{E}^i + \partial_j B^{ji} \ge -(\frac{1}{2} + \partial_k E^k) L^{ij} X_j$.⁵

From the dimensional analysis in Appendix C, the non-linear term dominates and our model with a non-linear constitutive equation becomes effective at long relaxation times.

Thermodynamic gauge fields naturally lead to an oscillatory phenomenon.⁶

6. Path integral

Here, let us discuss this gauge theory in the path-integral representation. For the thermodynamic variable a(t), let P[a] be the probability with which a thermodynamic path will be realized. We will call the entropy for this probability the path entropy, given by

$$S[a] \equiv -k_B \log P[a]. \tag{42}$$

The probability of the transition from state (a_i, t_i) to (a_f, t_f) can be written by path integration:

$$P(a_f, t_f | a_i, t_i) = \int \mathcal{D}a P[a], \qquad (43)$$

where the path is fixed at times t_i and t_f . The path entropy can generally be described by the entropic Lagrangian:

$$S[a] = \int_{t_i}^{t_f} L(a, \dot{a}) dt, \qquad (44)$$

$$L = \dot{S} = X(a)\dot{a} + \Phi(X(a), a).$$
(45)

Here, in order to be able to regard X as a free variable, we insert

$$\int \mathcal{D}X\,\delta\left(X(t) - \frac{\partial S}{\partial a}\right) = \int \mathcal{D}X\mathcal{D}\eta\,e^{i\int dt\,\eta(X - \frac{\partial S}{\partial a})} = 1 \tag{46}$$

into the path integral. Then, the entropic Lagrangian is rewritten as

$$L = X\dot{a} + \Phi(X, a) + i\eta \left(X - \frac{\partial S}{\partial a}\right).$$
(47)

Solving Eq. (47) for X, we obtain the constitutive equation:

$$\dot{a}^{i} = -\frac{\partial \Phi}{\partial X_{i}} - i\eta^{i}. \tag{48}$$

Here, if we take Φ as

$$\Phi = -\frac{1}{2}L^{ij}X_iX_j,\tag{49}$$

the constitutive equation yields

$$\dot{a}^i = L^{ij} X_j - i\eta^i. \tag{50}$$

⁵ See Appendix B for details.

⁶ For a simple example, see Appendix **D**.

$$\dot{S} = \frac{1}{2}\dot{a}^2 + i\eta\left(\dot{a} - \frac{\partial S}{\partial a}\right) - \frac{1}{2}\eta^2,$$
(51)

and solving this equation for η , we get

$$\dot{a}^{i} = L^{ij} \frac{\partial S}{\partial a^{j}} - i\eta^{i}.$$
(52)

Substituting this expression into \dot{S} , we obtain

$$\dot{S} = \frac{1}{2} \left(\frac{\partial S}{\partial a} \right)^2 - \frac{\partial S}{\partial a^i} i \eta^i.$$
(53)

Then, the first term of \dot{S} gives the entropy-increasing law of the macroscopic irreversible process, and the second term is an effect of fluctuation.

Next, let us assume that Φ is gauge-fixed like

$$dS = X(a)da + \Phi(a)dt + B(\Phi(a) - \Phi(X(a), a))dt.$$
(54)

Then we understand that, before gauge fixing, dS is

$$dS = X(a)da + \Phi(a)dt.$$
(55)

This action before gauge fixing is invariant under gauge transformation:

$$X(a) \to X(a) + \frac{\partial \epsilon}{\partial a},$$
 (56)

$$\Phi(a) \to \Phi(a) + \frac{\partial \epsilon}{\partial t}.$$
(57)

Then, this theory is originally a gauge theory of

$$A_{\mu} = (\Phi, X). \tag{58}$$

From the above discussion, the differential form representation of non-equilibrium thermodynamics is natural from the viewpoint of the path-integral representation, and the gauge fixing is naturally introduced to obtain a specific thermodynamical physics.

7. Discussion

We have assumed that the thermodynamic force is a gauge potential and generalized this potential from a pure gauge to a physical gauge potential. By gauge fixing (24) and adding a thermodynamical gauge-field action (30), we have obtained a non-linear constitutive equation for non-equilibrium thermodynamics (41). If we choose another gauge-fixing condition, we may get another thermodynamical physics. This will be discussed elsewhere.

The viewpoint that the existing theory is some kind of gauge theory with gauge fixing may be useful to generalize the theory. One of the most interesting applications of this method is the application to classical mechanics, where a momentum may be regarded as a kind of statistic (S. Katagiri, manuscript in preparation).

The entropy of a thermodynamic path, which is essential for the differential form representation of non-equilibrium thermodynamics, is also key to the detailed fluctuation theorem [11]. Then, the gauge-fixing method and the gauge principle may help understand the second law of thermodynamics.

All arguments so far are in the realm of classical theory. Reconsideration of our discussion in the context of quantum theory is important. In the latter case, the quantum fluctuation may be added to Eq. (53).

For other applications, application to phase transition phenomena can be considered. In timedependent Ginzburg–Landau theory [12], the free energy is expressed in terms of a complex-order parameter field and has the gauge symmetry of this parameter's phase shift. By applying our gaugefixing method to time-dependent Ginzburg–Landau theory, we may obtain understanding between these two gauge symmetries.

Finally, optics and non-equilibrium thermodynamics may have a closer relationship than previously thought, such as the invariance of the line element of light under the gauge transformation. We will investigate such a relationship in future work.

Acknowledgements

I am deeply grateful to Akio Sugamoto for discussions that clarified important points on thermodynamic forces. I am indebted to Ken Yokoyama and Shiro Komata for reading this paper and giving useful comments. I would also like to thank Jun Katagiri, Naoki Fujimoto, Shimpei Sugita, Noriaki Aibara, and Tsukasa Yumibayashi for reading this paper.

Appendix A. The time dependence of S(a, t)

In this appendix, the meaning of the time dependence of S(a, t) is discussed. As is well known in classical mechanics [13], for the action

$$S[x] = \int dt L(x, \dot{x}), \tag{A1}$$

the differential of the action is equal to

$$dS(x,t) = pdx - Hdt.$$
(A2)

Then, we obtain

$$p = \frac{\partial S(x,t)}{\partial x},\tag{A3}$$

$$-H = \frac{\partial S(x,t)}{\partial t}.$$
 (A4)

The last equation is the Hamilton–Jacobi equation. The application of the Hamilton–Jacobi equation to fluctuation phenomena is studied by Kitahara [14].

If H takes a constant value E, then we have

$$S(x,t) = -Et + \bar{S}(x), \tag{A5}$$

and Eq. (A2) becomes

$$dS(x,t) = \bar{p}dx - Edt \tag{A6}$$

$$\bar{p} \equiv \frac{\partial \bar{S}(x)}{\partial x}.$$
(A7)

This $\overline{S}(x)$ is the entropy that does not depend on time.

Appendix B. Restriction from the second law of thermodynamics

We want to discuss the restriction from the second law of thermodynamics on our model with a non-linear constitutive equation.

The entropy production is

$$\dot{S} = \frac{\partial S}{\partial a^i} \dot{a}^i + \Phi = \frac{1}{2} L^{ij} X_i X_j + X_i \xi^i$$
(B1)

$$X_i\xi^i = X^2 \left(\partial_l X_k\right)^2 + X^2 X^k \partial^2 X_k - X^2 \partial \cdot \dot{X} + X^i \dot{X}^j \partial_i X_j + X^i X^j \partial_i \dot{X}_j - X^i \ddot{X}_i.$$
(B2)

Since Eq. (A2) can be written as

$$S(a,t) = \Phi t + \bar{S}(a), \tag{B3}$$

we get

$$dS(a,t) = \bar{X}_i(a)da^i + \Phi dt, \tag{B4}$$

$$\bar{X}_i(a) \equiv \frac{\partial \bar{S}(a)}{\partial a^i}.$$
(B5)

Then, under Eq. (B3), Eq. (B2) becomes

$$\bar{X}_i \xi^i = \bar{X}^2 \left(\partial_l \bar{X}_k\right)^2 + \bar{X}^2 \bar{X}^i \partial^2 \bar{X}_i. \tag{B6}$$

Here we note that $\dot{E}_i(\bar{X}) = B_{ij}(\bar{X}) = 0$.

If it is assumed that the second law of thermodynamics cannot be broken, this requires the condition of

$$\frac{1}{2} + \left(\partial_l \bar{X}_k\right)^2 + \bar{X}^i \partial^2 \bar{X}_i \ge 0.$$
(B7)

If we use the Ruppeiner metric [15]:

$$g_{ij}^{R} \equiv \frac{\partial^{2}\bar{S}}{\partial a^{i}\partial a^{j}} = \frac{\partial\bar{X}_{i}}{\partial a^{j}},$$
(B8)

this condition gives

$$2\partial^k \left(\bar{X}^i g^R_{ik} \right) \ge -1 \tag{B9}$$

if g_{ij}^R is constant, then

$$2g^{Rik}g^R_{ik} \ge -1. \tag{B10}$$

Because L^{ij} is positive definite,

$$g^{Rik}g^R_{ik} \ge 0 \tag{B11}$$

always holds.

More generally, in term of the thermodynamic electric field and thermodynamic magnetic field, the non-linear constitutive equation is

$$\dot{a}^{i} = \left(1 + \partial_{k}E^{k}\right)L^{ij}X_{j} + \dot{E}^{i} + \partial_{j}B^{ji}.$$
(B12)

Then, the entropy production can be given by

$$\dot{S} = (\frac{1}{2} + \partial_k E^k) L^{ij} X_i X_j + X_i (\dot{E}^i + \partial_j B^{ji}).$$
(B13)

Therefore, the restriction from the second law of thermodynamics on our model with a non-linear constitutive equation yields

$$\dot{E}^{i} + \partial_{j}B^{ji} \ge -\left(\frac{1}{2} + \partial_{k}E^{k}\right)L^{ij}X_{j}.$$
(B14)

Appendix C. Dimensional analysis

To know the physics, the dimensional analysis is important. From this analysis, we obtain the relations

$$[S] = [k_B], \tag{C1}$$

$$[\Phi] = \frac{[S]}{[t]} = \frac{[k_B]}{[t]},$$
(C2)

$$\left[\frac{\partial}{\partial a^i}\right] = \frac{1}{\left[a^i\right]},\tag{C3}$$

$$[X_i] = \frac{[S]}{[a^i]} = \frac{[k_B]}{[a^i]},\tag{C4}$$

$$\frac{[a^i]}{[t]} = [L^{ij}][X_j] = [L^{ij}]\frac{[k_B]}{[a^i]},$$
(C5)

$$[L^{ij}] = \frac{[a^i][a^j]}{[t][k_B]},$$
(C6)

$$[R_{ij}] = \frac{[t][k_B]}{[a^i][a^j]},\tag{C7}$$

$$[F_{\mu\nu}] = \frac{[k_B]}{[a^{\mu}][a^{\nu}]},\tag{C8}$$

$$[F^{\mu\nu}] = \frac{[a^{\mu}]}{[k_B]} \frac{[a^{\nu}]}{[t^2]},\tag{C9}$$

$$[E_i] = [F_{i0}] = \frac{[k_B]}{[a^i][t]},$$
(C10)

$$[E^{i}] = \frac{[a^{i}]}{[k_{B}][t]},$$
(C11)

$$[B_{ij}] = \frac{[k_B]}{[a^i][a^j]},$$
(C12)

$$[\xi^{i}] = [\dot{a}] = \frac{[a]}{[t]}.$$
(C13)

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$$\lambda = \frac{da^0}{d\tau} - k_B \tilde{t} \partial_i E^i \tag{C14}$$

$$\dot{a}^{i} = (1 + k_{B}\tilde{t}\partial_{k}E^{k})L^{ij}X_{j} + k_{B}\tilde{t}\left(\dot{E}^{i} + \partial_{j}B^{ji}\right).$$
(C15)

Then, if \tilde{t} is long, the non-linear term dominates and our model with a non-linear constitutive equation becomes effective.

Appendix D. Simple example

As a simple example, we assume

$$X_i = g_{ij}^R a^j. \tag{D1}$$

$$\partial_j X_i = g_{ij}^R \tag{D2}$$

$$\xi^{i} = k_{B}\tilde{t} \left(\left(g_{mk}^{R} \right)^{2} L^{in} g_{np}^{R} a^{p} + L^{jk} L^{il} g_{jp}^{R} \dot{a}^{p} g_{lk}^{R} - L^{ij} g_{jp}^{R} \ddot{a}^{p} \right).$$
(D3)

In this case, we can derive the following second-order linear differential equations:

$$A^i_p \ddot{a}^p + B^i_p \dot{a}^p + C^i_p a^p = 0, \tag{D4}$$

$$A_p^i \equiv k_B \tilde{t} L^{ij} g_{jp}^R, \tag{D5}$$

$$B_p^i \equiv \left(1 - k_B \tilde{t} L^{jk} L^{il} g_{jp}^R g_{lk}^R\right),\tag{D6}$$

$$C_p^i \equiv -\left(1 + k_B \tilde{t} (g_{ml}^R)^2\right) L^{ij} g_{jp}^R.$$
 (D7)

If we change the coefficient of the first term in Eq. (D4) to 1, we get

$$\ddot{a}^i + D^i_p \dot{a}^p + E^i_p a^p = 0, \tag{D8}$$

$$D_{p}^{i} \equiv \left(A^{-1}\right)_{j}^{i} B_{p}^{j} = \left(\frac{1}{k_{B}\tilde{t}} g^{Rik} R_{kp} - L^{ik} g^{R}_{kp}\right), \tag{D9}$$

$$E_p^i \equiv (A^{-1})_j^i C_p^j = \epsilon \delta_p^i, \tag{D10}$$

$$\epsilon \equiv -\left(\frac{1}{k_B \tilde{t}} + (g_{ml}^R)^2\right). \tag{D11}$$

Here we note that

$$[D, E] = 0.$$
 (D12)

If $\tilde{t} \to 0$, we get the well known linear constitutive equation.

We take a linear transformation of *b*:

$$a^p = P^p_o b^o, \tag{D13}$$

$$\ddot{b}^i + \left(P^{-1}DP\right)^i_p \dot{b}^p + \epsilon b^p = 0.$$
(D14)

If we assume that it can be diagonalized,

$$\left(P^{-1}DP\right)_{p}^{i} = \lambda_{(i)}\delta_{p}^{i},\tag{D15}$$

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we get

$$\ddot{b}^i + \lambda_{(i)}\dot{b}^i + \epsilon b^i = 0, \tag{D16}$$

$$\omega^2 + \lambda_{(i)}\omega + \epsilon = 0, \tag{D17}$$

$$\omega = \frac{-\lambda_{(i)} \pm \sqrt{\lambda_{(i)}^2 - 4\epsilon}}{2}.$$
 (D18)

Under $\tilde{t} \to \infty$,

$$D_p^i = -L^{ik} g_{kp}^R, (D19)$$

$$\epsilon = -\left(g_{lm}^R\right)^2. \tag{D20}$$

If we assume for simplicity that

$$L^{ik}g^R_{kp} = \alpha \delta^i_p, \tag{D21}$$

then

$$\alpha = \frac{L^{ik} g_{ki}^R}{N},\tag{D22}$$

$$\epsilon = -L^{lk} L^{ms} g^R_{lm} g^R_{ks} = -N\alpha^2, \qquad (D23)$$

$$P_j^i = \delta_j^i, \tag{D24}$$

and we get

$$\ddot{a}^i - \alpha \dot{a}^i + N \alpha^2 a^i = 0, \tag{D25}$$

$$\omega = \frac{\alpha \pm \sqrt{\alpha^2 - 4N\alpha^2}}{2} = \frac{1 \pm i\sqrt{(4N-1)}}{2}\alpha.$$
 (D26)

Then, we get a solution that decays while oscillating:

$$a(t) = a(0)e^{-\frac{\sqrt{4(N-1)}}{2}\left(\frac{L^{ik}g_{ki}^{R}}{N}\right)t}\cos\frac{1}{2}\left(\frac{L^{ik}g_{ki}^{R}}{N}\right)t.$$
 (D27)

If we take the limit $N \to \infty$, the oscillation of Eq. (D27) disappears:

$$a(t) = a(0)e^{-L^{ik}g_{ki}^{R}t}.$$
 (D28)

Non-equilibrium macroscopic oscillatory phenomena have been discussed in the context of chemical reactions [16].

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