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1     **A non-integral form of the reciprocal relation associated with**  
2             **violation of the fluctuation response relation**

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## Abstract

We extend Onsager's reciprocal relation to systems in a nonequilibrium steady state. While Onsager's reciprocal relation concerns the kinetic (Onsager) coefficient, the extended reciprocal relation concerns violation of the fluctuation response relation (FRR) for mechanical and thermal perturbations. This extended relation holds at each frequency when the extent of the FRR violation is expressed in a frequency domain. This non-integral form distinguishes the extended relation from previous relations expressed by integration over a frequency. To obtain this relation, we consider one-particle one-dimensional systems described by an overdamped Langevin equation with a force driving the system away from equilibrium. We assume a special property of the potential in the system. From this Langevin equation, we obtain the Fokker-Planck (FP) equation describing the time evolution of the distribution function of the particle. Using the FP equation, we calculate the responses of the particle velocity and heat current by applying time-dependent perturbations of the driving force and temperature. We express the extent of the FRR violation in terms of these responses with time correlation functions and expand them in powers of the FP operator. This reciprocal relation is valid far from equilibrium. One can also confirm this reciprocal relation through experiments with systems such as colloidal suspensions because the FRR violation can be experimentally observed.

### 1 I. INTRODUCTION

2 Thermal and mechanical perturbations to an equilibrium or nonequilibrium system has a  
3 cross effect on thermal and mechanical responses of the system. An example of perturbation  
4 to an equilibrium system is a heat engine because thermal perturbations change mechanical  
5 variables such as energy [1–3]. For an equilibrium cross effect, there are many relations  
6 including Onsager's reciprocal relation [4] and the fluctuation response relation (FRR) [5, 6],  
7 while nonequilibrium effects are less well studied [7–9]. These studies contrast with those  
8 on mechanical perturbations because they provide many nonequilibrium relations such as  
9 the glassy system FRR [10–19], extended FRR [20–28], and reciprocal relation [29, 30].  
10 Some nonequilibrium studies have also dealt with perturbations other than mechanical ones  
11 [31–36], although the cross effect is out of the scope of these studies.

12 Yamada and Yoshimori have derived a reciprocal relation between thermal and mechani-

1 cal responses by considering the nonequilibrium cross effect of perturbations [7, 8]. When the  
 2 perturbations are applied to a nonequilibrium steady state (NESS) [7, 8, 20–23, 25–32, 37–  
 3 43], neither Onsager’s reciprocal relation nor the FRR is valid. Yamada and Yoshimori  
 4 showed that a reciprocal relation is valid for the extent of the FRR violation in nonequilib-  
 5 rium Brownian systems. Their reciprocal relation is valid for any type of system potential  
 6 and for any driving force strength, which causes the system to deviate from an equilibrium  
 7 state. In addition, their relation can be experimentally confirmed because it consists of  
 8 measurable quantities.

9 Yamada and Yoshimori expressed their reciprocal relation by integrating the extent of the  
 10 FRR violation over a frequency [7, 8]. The integral over a frequency shows that the reciprocal  
 11 relation in the time domain does not hold for all time, but only at zero time. Thus, their  
 12 reciprocal relation contrasts with Onsager’s reciprocal relation, which has a non-integral  
 13 form holding at each frequency and for all time. In a special case, they numerically found  
 14 that their relation has a non-integral form when the potential of the system is proportional  
 15 to a cosine [8]. This result, however, has not exactly been proved.

16 In this study, we exactly derive a non-integral form of a reciprocal relation valid for the  
 17 extent of the FRR violation by assuming a condition of the potential  $U(x)$  of the system.  
 18 This condition is given by  $U''(x) \propto U(x)$ , where  $U''(x)$  represents the second derivative of  
 19  $U(x)$ . Using the potential, we calculate responses to force and temperature perturbations on  
 20 the basis of the one-dimensional one-particle overdamped Langevin equation with a driving  
 21 force. We do not assume the strength of the force driving the system out of equilibrium; thus,  
 22 our reciprocal relation holds even far from an equilibrium state. In addition, we confirm  
 23 our reciprocal relation for various values of the driving force by numerically calculating the  
 24 extent of the FRR violation.

## 25 II. MAIN RESULTS

26 We study a one-particle one-dimensional system described by the overdamped Langevin  
 27 equation

$$\dot{x}(t) = \gamma^{-1} [F(x(t)) + \epsilon_1 f_p(t) + \xi(t)], \quad (1)$$

28 where  $x(t)$  and  $\dot{x}(t)$  are the position and velocity of the particle,  $\gamma$  is the coefficient of friction,  
 29  $\epsilon_1 f_p(t)$  represents the time-dependent mechanical perturbation, and we assume a periodic

1 boundary condition of length  $l$ . We write the force term  $F(x)$  as

$$F(x) = f - \frac{dU(x)}{dx} \quad (2)$$

2 with the periodic potential

$$U(x + l) = U(x), \quad (3)$$

3 where  $f$  is the time-independent driving force shifting the system out of equilibrium. In  
4 Eq. (1), the Gaussian noise  $\xi(t)$  satisfies

$$\langle \xi(t)\xi(s) \rangle_\epsilon = 2\gamma(T + \epsilon_2 T_p(t))\delta(t - s), \quad (4)$$

5 where  $T$  is the time-independent temperature,  $\epsilon_2 T_p(t)$  represents the time-dependent thermal  
6 perturbation, and  $\langle \cdot \cdot \rangle_\epsilon$  is the average in the presence of  $\epsilon_1 f_p(t)$  and  $\epsilon_2 T_p(t)$ . In this paper,  
7 we set the Boltzmann constant to unity. The perturbations  $\epsilon_1 f_p(t)$  and  $\epsilon_2 T_p(t)$  are applied  
8 to the steady-state system at  $t = t_{ini} \rightarrow -\infty$ .

9 In the system described by the Langevin equation, we express the entropy production  
10 using the currents and the affinities. We define entropy production as [1]

$$\Delta S \equiv \int_{-\infty}^t ds \beta(s) \langle \dot{Q}(s) \rangle_\epsilon, \quad (5)$$

11 where  $\beta(s) \equiv 1/[T + \epsilon_2 T_p(s)]$  and

$$\dot{Q}(t) \equiv (\gamma \dot{x}(t) - \xi(t)) \circ \dot{x}(t) \quad (6)$$

12 with the Stratonovich product  $\circ$  [48]. We rewrite Eq. (5) in the form [7, 8]

$$\Delta S = \sum_{i=1}^2 \int_{-\infty}^t ds A_i(s) \langle J_i(t) \rangle_\epsilon \quad (7)$$

$$= \int_{-\infty}^t ds A_1(s) \langle \dot{x}(s) \rangle_\epsilon + \int_{-\infty}^t ds A_2(s) \langle \dot{Q}(s) \rangle_\epsilon, \quad (8)$$

13 where  $J_1(t) \equiv \dot{x}(t)$  and  $J_2(t) \equiv \dot{Q}(t)$  (currents), and  $A_1(t) \equiv (f + \epsilon_1 f_p(t))/T$  and  $A_2(t) \equiv$   
14  $1/(T + \epsilon_2 T_p(t)) - 1/T$  (affinities). To derive Eq. (7) from Eq. (5), we have used  $\langle U(x(t)) \rangle_\epsilon =$   
15  $\langle U(x(-\infty)) \rangle_\epsilon$ , which is obtained from the assumptions of  $f_p(s) = T_p(s) = 0$  for  $s > t_f$  and  
16 of  $t_f \ll t$  [7, 8]. After enough time from the time  $t_f$  when the perturbations are turned off,  
17 the system reaches the same steady state as that at  $t = -\infty$ .

18 By expanding the currents using the affinities, we define the nonequilibrium kinetic coeffi-

1 cients (Onsager's coefficients). By expanding  $\langle \dot{x}(s) \rangle_\epsilon$  and  $\langle \dot{Q}(s) \rangle_\epsilon$  in powers of the perturbed  
 2 parts of the affinities  $\delta A_1(t) \equiv \epsilon_1 f_p(t)/T$  and  $\delta A_2(t) \equiv -\epsilon_2 T_p(t)/T^2$ , we define the nonequi-  
 3 librium kinetic coefficients  $L_{ij}(t)$  as

$$\langle \dot{x}(t) \rangle_\epsilon = J_1^{st} + \int_{-\infty}^t ds L_{11}(t-s) \frac{\epsilon_1 f_p(s)}{T} - \int_{-\infty}^t ds L_{12}(t-s) \frac{\epsilon_2 T_p(s)}{T^2} \dots, \quad (9)$$

$$\langle \dot{Q}(t) \rangle_\epsilon = J_2^{st} + \int_{-\infty}^t ds L_{21}(t-s) \frac{\epsilon_1 f_p(s)}{T} - \int_{-\infty}^t ds L_{22}(t-s) \frac{\epsilon_2 T_p(s)}{T^2} \dots. \quad (10)$$

4 Here,  $J_1^{st}$  and  $J_2^{st}$  are the particle velocity and the heat current in the steady state with  
 5  $\epsilon_1 = \epsilon_2 = 0$ , respectively. We assume  $L_{ij}(t) = 0$  for  $t < 0$ .

6 Using the kinetic coefficients, we express the FRR and Onsager's reciprocal relation in an  
 7 equilibrium state while defining their violation in a nonequilibrium state. To express these  
 8 relations, the time correlation function  $C_{ij}(t)$  is defined as

$$C_{ij}(t) \equiv \langle J_i(t) J_j(0) \rangle_0, \quad (11)$$

9 where  $\langle \dots \rangle_0$  is the average in the absence of perturbations. Using  $C_{ij}(t)$ , the FRR is given  
 10 by [5, 8]

$$C_{ij}(t) = L_{ij}(t) \quad (t > 0), \quad (12)$$

11 and Onsager's reciprocal relation is given by [4, 49]

$$L_{12}(t) = L_{21}(t). \quad (13)$$

12 While Eqs. (12) and (13) are valid for the perturbations applied to the equilibrium state  
 13 ( $f = 0$ ), they are violated for perturbations applied to the NESS ( $f \neq 0$ ). For the NESS, if  
 14 we define the extent of the FRR violation  $\Delta_{ij}(t)$  as

$$\Delta_{ij}(t) \equiv \begin{cases} C_{ij}(t) - L_{ij}(t) & (t > 0) \\ 0 & (t \leq 0), \end{cases} \quad (14)$$

15 then the following reciprocal relation holds [7, 8]:

$$\int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \tilde{\Delta}_{12}(\omega) = \int_{-\infty}^{\infty} \frac{d\omega}{2\pi} \tilde{\Delta}_{21}(\omega) \quad (15)$$

16 with  $\tilde{\Delta}_{ij}(\omega) \equiv \int_{-\infty}^{\infty} dt \Delta_{ij}(t) \exp(-i\omega t)$ .

17 In Sec. IV, we will use the extent of the FRR violation in the NESS to prove the following  
 18 reciprocal relation expressed in a non-integral form:

$$\tilde{\Delta}_{12}(\omega) = \tilde{\Delta}_{21}(\omega). \quad (16)$$

1 To prove Eq. (16), we assume

$$\frac{d^2U(x)}{dx^2} \propto U(x). \quad (17)$$

2 Equation (16) can be expressed in the time domain as

$$\Delta_{12}(t) = \Delta_{21}(t), \quad (18)$$

3 which holds for all time. Equations (16) and (18) are independent of the strength of the  
 4 driving force  $f$ , which shows the extent of deviation from the equilibrium state. In Sec. IV,  
 5 we will prove Eq. (18), which is equivalent to Eq. (16).

6 The reciprocal relation Eq. (16) expressed in the non-integral form is equivalent to  
 7 Eq. (18) holding for all time in the time domain. Equation (18) holds for a wider time  
 8 range than previous nonequilibrium relations, which hold only at zero time [7, 8, 20, 21, 30]  
 9 (see the next paragraph). **For the property, we need to assume Eq. (17), which is satisfied**  
 10 **by an experimentally constructible potential used in many studies, as explained later.** In  
 11 addition, because we do not need the frequency integration, our relation is less difficult to  
 12 confirm experimentally than those in the integral form.

13 Equation (16) contrasts with nonequilibrium relations previously expressed in integral  
 14 forms [7, 8, 20, 21, 30]. Harada and Sasa have expressed the relationship between the heat  
 15 current and FRR violation through an integral identity [20, 21]. Shimizu and Yuge also  
 16 used an integral form to obtain a reciprocal relation between two mechanical perturbations  
 17 [30]. In addition, Yamada and Yoshimori have obtained an integral form of the reciprocal  
 18 relation between the same thermal and mechanical perturbations as those used by this study  
 19 (Eq. (15)) [7, 8]. Onsager's reciprocal relation differs from these nonequilibrium relations in  
 20 that it can be expressed in a non-integral form.

21 To prove that the reciprocal relation holds for all time, we need to assume Eq. (17). **The**  
 22 **potential satisfying Eq. (17) has often been used in theoretical and experimental studies**  
 23 **[20, 44–47].** The potential can be expressed in the form  $U(x) = A \sin(kx + c)$  or  $U(x) =$   
 24  $A \cos(kx + c)$ , where  $A$ ,  $k$ , and  $c$  are constants independent of  $x$ . Such a function has only  
 25 one wavelength; thus, we can consider it to be simplest of the periodic functions assumed in  
 26 Eq. (3). The potential satisfying Eq. (17) can experimentally be constructed and, in fact,  
 27 has been constructed by some experimental studies [44, 45].

1 Equations (16) and (18) can be confirmed by performing experiments on cross effects be-  
 2 tween thermal and mechanical perturbations. In Eq. (16),  $\tilde{\Delta}_{12}(\omega)$  and  $\tilde{\Delta}_{21}(\omega)$  are measurable  
 3 quantities that can be obtained in experimental systems, such as a colloidal suspension. If  
 4 the potential of the experimental system satisfies Eq. (17), one need not integrate the extent  
 5 of the violation over a frequency. In addition, Eqs. (16) and (18) include responses to both  
 6 thermal and mechanical perturbations. Thus, the nonequilibrium cross effect between these  
 7 perturbations can more deeply be understood through our reciprocal relations.

### 8 III. A BRIEF SKETCH OF THE PROOF

9 We will give a brief sketch of the proof before describing its details. We begin the proof  
 10 by expressing the extent of the FRR violation  $\Delta_{ij}(t)$  using the non-perturbed stationary  
 11 distribution function with the Fokker–Planck (FP) operator. To obtain the expression, we  
 12 derive  $L_{ij}(t)$  by expanding the FP equation in powers of  $\epsilon_1$  and  $\epsilon_2$  and derive  $C_{ij}(t)$  using the  
 13 Furutsu–Novikov–Donsker formula. Combining the derived expressions of  $L_{ij}(t)$  and  $C_{ij}(t)$ ,  
 14 we derive the expression of  $\Delta_{ij}(t)$  on the basis of Eq. (14). Details of the derivation have  
 15 been given by Yamada and Yoshimori [7, 8].

16 From this expression of  $\Delta_{ij}(t)$ , we obtain Eq. (18) or  $\Delta_{21}(t) - \Delta_{12}(t) = 0$  by introducing  
 17 the new operator  $\hat{L}_1^\dagger$ . To define  $\hat{L}_1^\dagger$ , we divide the conjugate FP operator into two operators:  
 18 one that includes  $F(x)$  and one that does not include  $F(x)$ . The operator  $\hat{L}_1^\dagger$  is defined by  
 19 one including  $F(x)$ . The exponential operators including  $\hat{L}_1^\dagger$  give the time dependence of  
 20  $\Delta_{21}(t) - \Delta_{12}(t)$ .

21 Using the operator  $\hat{L}_1^\dagger$ , we divide  $\Delta_{21}(t) - \Delta_{12}(t)$  into two parts. This is an important step  
 22 of the proof and will be explained as follows. First, we expand the exponential operators  
 23 in powers of the conjugate FP operator and count the number of  $\hat{L}_1^\dagger$  operators included in  
 24 the expanded term. Next, using this number, we divide the exponential operators into a  
 25 term including an odd number of  $\hat{L}_1^\dagger$  operators and a term including an even number. This  
 26 division of the exponential operators allows us to divide  $\Delta_{21}(t) - \Delta_{12}(t)$  into two parts.

27 Finally, we show that the two divided parts of  $\Delta_{21}(t) - \Delta_{12}(t)$  vanish respectively. We  
 28 can show that one of the parts vanishes without assuming Eq. (17). In contrast, the other  
 29 part vanishes only when Eq. (17) is satisfied. To prove this, the second part is given by the  
 30  $x$ -integration, whose integrand is expressed using the product of  $F(x)$  and  $dF(x)/dx$ . By



1 integrating the expression by parts, we show that the second part vanishes.

## 2 IV. PROOF

3 We express the extent of the FRR violation  $\Delta_{ij}(t)$  in terms of the Fokker–Planck (FP)  
4 operator  $\hat{L}$ ,

$$\hat{L} \equiv -\gamma^{-1} \frac{\partial}{\partial x} \left( F(x) - T \frac{\partial}{\partial x} \right), \quad (19)$$

5 with the stationary distribution function in a non-perturbed system. Using  $\hat{L}$ , we describe  
6 the time development of the distribution function for the particle  $P_\epsilon(x, t)$  using the FP  
7 equation

$$\frac{\partial P_\epsilon(x, t)}{\partial t} = \hat{L} P_\epsilon(x, t) - \gamma^{-1} \frac{\partial}{\partial x} \left( \epsilon_1 f_p(t) - \epsilon_2 T_p(t) \frac{\partial}{\partial x} \right) P_\epsilon(x, t). \quad (20)$$

8 The FP equation (20) is equivalent to the overdamped Langevin equation (1). The non-  
9 perturbed stationary distribution function  $P_{st}(x)$  is defined as the steady-state equation with  
10  $\epsilon_1 = \epsilon_2 = 0$ :

$$\frac{\partial P_{st}(x)}{\partial t} = \hat{L} P_{st}(x) = 0. \quad (21)$$

11 Using the FP operator  $\hat{L}$  with the stationary distribution function  $P_{st}(x)$ , we can express  
12  $\Delta_{12}(t)$  and  $\Delta_{21}(t)$  as follows [7, 8]:

$$\Delta_{12}(t) = \gamma^{-2} \int_0^l dx F(x) e^{t\hat{L}} \hat{J}^2 P_{st}(x), \quad (22)$$

$$\Delta_{21}(t) = \gamma^{-2} \int_0^l dx F(x) \hat{J} e^{t\hat{L}} \hat{J} P_{st}(x), \quad (23)$$

13 where  $\hat{J}$  is the operator defined as

$$\hat{J} \equiv F(x) - T \frac{d}{dx}. \quad (24)$$

14 Using Eqs. (22) and (23), which express  $\Delta_{12}(t)$  and  $\Delta_{21}(t)$ , respectively, we calculate  
15  $\Delta_{21}(t) - \Delta_{12}(t)$ . From Eqs. (22) and (23), we obtain

$$\Delta_{21}(t) - \Delta_{12}(t) = \gamma^{-2} \int_0^l dx F(x) \left[ \hat{J}, e^{t\hat{L}} \right] \hat{J} P_{st}(x), \quad (25)$$

16 where  $[\hat{A}, \hat{B}] \equiv \hat{A}\hat{B} - \hat{B}\hat{A}$  with the operators  $\hat{A}$  and  $\hat{B}$ . Because the differential equation

$$\frac{\partial}{\partial t} \left[ \hat{J}, e^{t\hat{L}} \right] = \hat{L} \left[ \hat{J}, e^{t\hat{L}} \right] + \left[ \hat{J}, \hat{L} \right] e^{t\hat{L}} \quad (26)$$

1 provides

$$\left[ \hat{J}, e^{t\hat{L}} \right] = \gamma^{-1} \int_0^t ds e^{(t-s)\hat{L}} F'(x) \hat{J} e^{s\hat{L}}, \quad (27)$$

2 substituting into Eq. (25) yields

$$\Delta_{21}(t) - \Delta_{12}(t) = \gamma^{-3} \int_0^l dx \int_0^t ds F(x) e^{(t-s)\hat{L}} F'(x) \hat{J} e^{s\hat{L}} \hat{J} P_{st}(x), \quad (28)$$

3 where  $F'(x) = dF(x)/dx$ . From integrating Eq. (28) by parts with respect to  $x$ , we obtain

$$\Delta_{21}(t) - \Delta_{12}(t) = \gamma^{-3} \int_0^l dx \int_0^t ds \left[ e^{(t-s)\hat{L}^\dagger} F(x) \right] F'(x) \hat{J} e^{s\hat{L}} \hat{J} P_{st}(x), \quad (29)$$

4 where the conjugate operator of  $\hat{L}$  is defined as

$$\hat{L}^\dagger \equiv \gamma^{-1} \left( T \frac{d}{dx} + F(x) \right) \frac{d}{dx}. \quad (30)$$

5 In Eq. (29),  $[\hat{O}g(x)]$  indicates that an operator  $\hat{O}$  operates only on a function  $g(x)$  and not  
6 on functions outside of  $[\dots]$ .

7 By expanding  $\Delta_{21}(t) - \Delta_{12}(t)$  in powers of conjugate operators, we divide  $\Delta_{21}(t) - \Delta_{12}(t)$   
8 into two parts. To expand  $\Delta_{21}(t) - \Delta_{12}(t)$ , we rewrite Eq. (29) in the form

$$\Delta_{21}(t) - \Delta_{12}(t) = \gamma^{-3} \int_0^l dx \int_0^t ds \left[ e^{(t-s)\hat{L}^\dagger} F(x) \right] F'(x) \left[ e^{s\hat{L}^{\dagger*}} F(x) \right] \hat{J} P_{st}(x) \quad (31)$$

9 using the formula (Appendix A)

$$\hat{J} e^{s\hat{L}} \hat{J} P_{st}(x) = \left[ e^{s\hat{L}^{\dagger*}} F(x) \right] \hat{J} P_{st}(x), \quad (32)$$

10 where  $\hat{L}^{\dagger*} = \hat{L}_0^\dagger - \hat{L}_1^\dagger$  with

$$\hat{L}_0^\dagger \equiv \gamma^{-1} T \frac{d^2}{dx^2}, \quad (33)$$

$$\hat{L}_1^\dagger \equiv \gamma^{-1} F(x) \frac{d}{dx}. \quad (34)$$

11 In Eq. (31), we expand  $e^{t\hat{L}^\dagger} F(x)$  and  $e^{t\hat{L}^{\dagger*}} F(x)$  in powers of  $\hat{L}^\dagger$  and  $\hat{L}^{\dagger*}$ , respectively, via

$$e^{t\hat{L}^\dagger} F(x) = \sum_{n=0}^{\infty} \frac{t^n}{n!} (\hat{L}^\dagger)^n F(x), \quad (35)$$

$$e^{t\hat{L}^{\dagger*}} F(x) = \sum_{n=0}^{\infty} \frac{t^n}{n!} (\hat{L}^{\dagger*})^n F(x). \quad (36)$$

12 Using  $\hat{L}^{\dagger*} = \hat{L}_0^\dagger - \hat{L}_1^\dagger$  and  $\hat{L}^\dagger = \hat{L}_0^\dagger + \hat{L}_1^\dagger$  obtained from Eq. (30) with Eqs. (33) and (34), we

1 obtain

$$e^{t\hat{L}^\dagger} F(x) = \sum_{n=0}^{\infty} \frac{t^n}{n!} (\hat{L}^\dagger)^n F(x) = g_o(x, t) + g_e(x, t), \quad (37)$$

$$e^{t\hat{L}^{\dagger*}} F(x) = \sum_{n=0}^{\infty} \frac{t^n}{n!} (\hat{L}^{\dagger*})^n F(x) = -g_o(x, t) + g_e(x, t), \quad (38)$$

2 with

$$g_o(x, t) \equiv \frac{1}{2} \left[ e^{t\hat{L}^\dagger} F(x) - e^{t\hat{L}^{\dagger*}} F(x) \right] = \frac{1}{2} \sum_{n=0}^{\infty} \frac{t^n}{n!} \left[ (\hat{L}_0^\dagger + \hat{L}_1^\dagger)^n F(x) - (\hat{L}_0^\dagger - \hat{L}_1^\dagger)^n F(x) \right], \quad (39)$$

$$g_e(x, t) \equiv \frac{1}{2} \left[ e^{t\hat{L}^\dagger} F(x) + e^{t\hat{L}^{\dagger*}} F(x) \right] = \frac{1}{2} \sum_{n=0}^{\infty} \frac{t^n}{n!} \left[ (\hat{L}_0^\dagger + \hat{L}_1^\dagger)^n F(x) + (\hat{L}_0^\dagger - \hat{L}_1^\dagger)^n F(x) \right], \quad (40)$$

3 where  $\hat{L}_1^\dagger$  operates on  $F(x)$  an odd and even number of times, respectively. By substituting  
4 Eqs. (37) and (38) into Eq. (31), we can divide Eq. (31) into two parts,

$$\Delta_{21}(t) - \Delta_{12}(t) = \Delta_o(t) + \Delta_e(t), \quad (41)$$

5 where

$$\begin{aligned} \Delta_o(t) \equiv & -\gamma^{-3} \int_0^l dx \int_0^t ds g_e(x, t-s) F'(x) g_o(x, s) \hat{J}P_{st}(x) \\ & + \gamma^{-3} \int_0^l dx \int_0^t ds g_o(x, t-s) F'(x) g_e(x, s) \hat{J}P_{st}(x), \end{aligned} \quad (42)$$

$$\begin{aligned} \Delta_e(t) \equiv & \gamma^{-3} \int_0^l dx \int_0^t ds g_e(x, t-s) F'(x) g_e(x, s) \hat{J}P_{st}(x) \\ & - \gamma^{-3} \int_0^l dx \int_0^t ds g_o(x, t-s) F'(x) g_o(x, s) \hat{J}P_{st}(x). \end{aligned} \quad (43)$$

6 One of the two divided parts  $\Delta_o(t)$  vanishes. To show this, we transform the variable  $s$   
7 into  $\tau = t - s$  in the first term of Eq. (42) to obtain

$$\begin{aligned} \Delta_o(t) = & -\gamma^{-3} \int_0^l dx \int_0^t d\tau g_e(x, \tau) F'(x) g_o(x, t-\tau) \hat{J}P_{st}(x) \\ & + \gamma^{-3} \int_0^l dx \int_0^t ds g_o(x, t-s) F'(x) g_e(x, s) \hat{J}P_{st}(x). \end{aligned} \quad (44)$$

8 On the right side of Eq. (44), the absolute value of the first term is equivalent to that of the  
9 second term if  $\tau = s$ . These terms cancel out, so we obtain

$$\Delta_o(t) = 0. \quad (45)$$

1 We have not assumed Eq. (17) to derive Eq. (45).

2 The other of the two divided parts,  $\Delta_e(t)$ , can be expressed as a product of  $F(x)$  and  
 3  $F'(x)$  assuming Eq. (17). Using

$$\frac{d^2 F(x)}{dx^2} = \alpha [F(x) - f] \quad (46)$$

4 derived from Eq. (17) with  $U'(x) = \alpha U(x)$ , we rewrite Eqs. (39) and (40) in the forms

$$g_o(x, t) = \sum_{p=0}^{\infty} \sum_{q=0}^{\infty} h_o^{pq}(t) [F(x)]^p [F'(x)]^{2q+1}, \quad (47)$$

$$g_e(x, t) = \sum_{p=0}^{\infty} \sum_{q=0}^{\infty} h_e^{pq}(t) [F(x)]^p [F'(x)]^{2q}, \quad (48)$$

5 where  $h_o^{pq}(t)$  and  $h_e^{pq}(t)$  are functions of  $t$  independent of  $x$  (Appendix B). By substituting  
 6 Eqs. (47) and (48) into Eq. (43), we obtain

$$\Delta_e(t) = \gamma^{-3} \sum_{p=0}^{\infty} \sum_{q=0}^{\infty} \int_0^l dx \int_0^t ds h^{pq}(t, s) [F(x)]^p [F'(x)]^{2q+1} \hat{J}P_{st}(x), \quad (49)$$

7 where  $h^{pq}(t, s)$  is a function of  $t$  and  $s$  independent of  $x$ .

8 Using Eq. (49) expressed in terms of  $F(x)$  and  $F'(x)$ , we show that  $\Delta_e(t)$  vanishes. We  
 9 rewrite the integral part of Eq. (49), using  $[F(x)]^p F'(x) = (p+1)^{-1} d([F(x)]^{p+1})/dx$ , in the  
 10 form

$$\int_0^l dx [F(x)]^p [F'(x)]^{2q+1} \hat{J}P_{st}(x) = \frac{1}{p+1} \int_0^l dx \frac{d[F(x)]^{p+1}}{dx} \left[ \frac{dF(x)}{dx} \right]^{2q} \hat{J}P_{st}(x). \quad (50)$$

11 Using Eqs. (21) and (46), integrating Eq. (50) by parts yields

$$\begin{aligned} \frac{1}{p+1} \int_0^l dx \frac{d[F(x)]^{p+1}}{dx} \left[ \frac{dF(x)}{dx} \right]^{2q} \hat{J}P_{st}(x) &= a_0 \int_0^l dx [F(x)]^{p+2} \left[ \frac{dF(x)}{dx} \right]^{2q-1} \hat{J}P_{st}(x) \\ &+ a_1 \int_0^l dx [F(x)]^{p+1} \left[ \frac{dF(x)}{dx} \right]^{2q-1} \hat{J}P_{st}(x), \end{aligned} \quad (51)$$

12 where  $a_0$  and  $a_1$  are  $x$ -independent constants determined by  $p$ ,  $q$ , and  $\alpha$ . By integrating  
 13 Eq. (51)  $p$  times by parts, we obtain

$$\begin{aligned}
\int_0^l dx [F(x)]^p \left[ \frac{dF(x)}{dx} \right]^{2q+1} \hat{J}P_{st}(x) &= \sum_{i=0}^q C_i \int_0^l dx [F(x)]^{p+q+i} \left[ \frac{dF(x)}{dx} \right] \hat{J}P_{st}(x) \\
&= \sum_{i=0}^q \frac{C_i}{p+q+i+1} \int_0^l dx \frac{d[F(x)]^{p+q+i+1}}{dx} \hat{J}P_{st}(x) \\
&= - \sum_{i=0}^q \frac{C_i}{p+q+i+1} \int_0^l dx [F(x)]^{p+q+i+1} \frac{d}{dx} \hat{J}P_{st}(x) \\
&= 0,
\end{aligned} \tag{52}$$

1 where  $C_i$  is a constant expressed in terms of  $a_0$  and  $a_1$ . Because Eq. (52) shows

$$\Delta_e(t) = 0, \tag{53}$$

2 we finally obtain Eq. (18) from Eqs. (45) and (53) with Eq. (41).

### 3 V. NUMERICAL CALCULATIONS

4 We demonstrate the reciprocal relation derived in the previous section by numerically  
5 calculating  $\Delta_{12}(t)$  and  $\Delta_{21}(t)$  using the following form of the potential  $U(x)$ :

$$\frac{U(x)}{T} = \cos \frac{2\pi x}{l} + a \cos \frac{4\pi x}{l}, \tag{54}$$

6 where  $a$  is a parameter independent of  $x$ . The potential given by Eq. (54) does not satisfy  
7 the condition of Eq. (17) for  $a \neq 0$ , but satisfies the condition for  $a = 0$ . To calculate  $\Delta_{ij}(t)$ ,  
8 we convert all quantities to dimensionless forms using the time unit  $\gamma T^{-1} l^2$ , energy  $T$ , and  
9 length  $l$ .

10 We numerically calculate  $\Delta_{12}(t)$  and  $\Delta_{21}(t)$  on the basis of Eqs. (22) and (23) using the  
11 FP equation [7, 8]. Because Eqs. (22) and (23) do not include the perturbations  $\epsilon_1 f_p(t)$  and  
12  $\epsilon_2 T_p(t)$ , the calculations do not need the explicit forms of the perturbations. Equations (22)  
13 and (23) are represented by

$$\Delta_{12}(t) = \gamma^{-2} \int_0^l dx F(x) P_{12}(x, t), \tag{55}$$

$$\Delta_{21}(t) = \gamma^{-2} \int_0^l dx F(x) \hat{J}P_{21}(x, t), \tag{56}$$

14 where  $P_{12}(x, t)$  and  $P_{21}(x, t)$  are given by  $P_{12}(x, t) = e^{t\hat{L}} \hat{J}^2 P_{st}(x)$  and  $P_{21}(x, t) = e^{t\hat{L}} \hat{J} P_{st}(x)$ .

15 We obtain the distribution functions  $P_{12}(x, t)$  and  $P_{21}(x, t)$  by solving Eq. (20) with

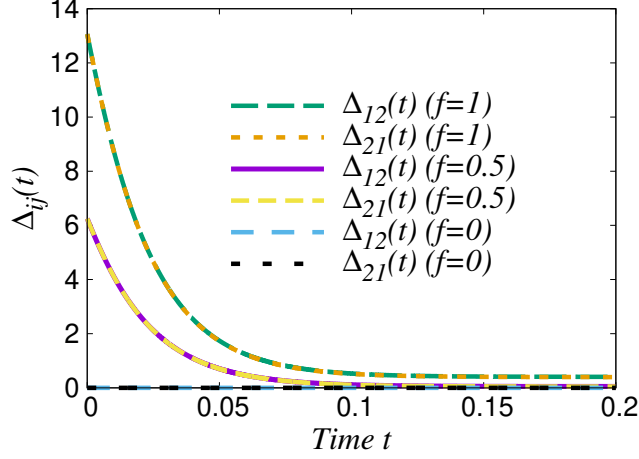


FIG. 1. Time dependence of the extent of the FRR violation  $\Delta_{12}(t)$  or  $\Delta_{21}(t)$  (Eq. (14)) calculated using the potential in Eq. (54) and the driving forces  $f = 0, 0.5$ , and  $1$  with  $a = 0$ . We use a one-particle one-dimensional model described by a driving overdamped Langevin equation. We convert all quantities into dimensionless forms using the time unit  $\gamma T^{-1} l^2$ , energy  $T$ , and length  $l$ , where  $\gamma$  is the friction coefficient.

1  $\epsilon_1 = \epsilon_2 = 0$  under the initial conditions  $P_{12}(x, 0) = \hat{J}^2 P_{st}(x)$  and  $P_{21}(x, 0) = \hat{J} P_{st}(x)$ .  
 2 We numerically solve the FP equation with the Euler method and spatial finite difference  
 3 method, setting the time and length steps at  $\Delta t = 6.25 \times 10^{-7}$  and  $\Delta x = 1.25 \times 10^{-3}$ ,  
 4 respectively.

5 First, we numerically confirm that Eq. (18) is valid using a range of values of the driving  
 6 force  $f$ , which shows the extent of deviation from an equilibrium state (Fig. 1). Because  
 7 Eq. (18) is valid at  $a = 0$  in Eq. (54), we calculate  $\Delta_{ij}(t)$  for the potential at  $a = 0$ . For  
 8  $f = 0$ ,  $\Delta_{12}(t) = \Delta_{21}(t) = 0$  because the FRR is valid in the equilibrium state. We confirm  
 9  $\Delta_{12}(t) = \Delta_{21}(t)$  for **all the calculated values**. This result shows that our reciprocal relation  
 10 is valid in some nonequilibrium states.

11 Second, we calculate  $\Delta_{12}(t) - \Delta_{21}(t)$  when the potential  $U(x)$  does not satisfy Eq. (17)  
 12 (Fig. 2). At  $t = 0$ ,  $\Delta_{12}(t) - \Delta_{21}(t) = 0$  for all values of  $a$ , as Yamada and Yoshimori  
 13 showed [7, 8]. When  $t$  increases from 0,  $\Delta_{12}(t) - \Delta_{21}(t)$  increases from 0 to a positive value  
 14 and reaches a peak between  $t = 0.02$  and  $0.03$ . The peak value increases with  $a$  except  
 15 for  $a = 1.0$ , where the peak is lower than at  $a = 0.75$ . In contrast, for a longer time,  
 16  $\Delta_{12}(t) - \Delta_{21}(t)$  is larger at  $a = 1.0$  than at  $a = 0.75$ .

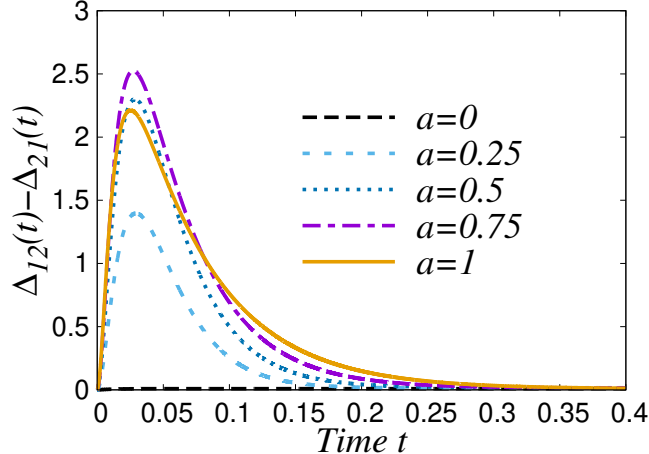


FIG. 2. Time dependence of the difference in extent of the FRR violations  $\Delta_{12}(t)$  and  $\Delta_{21}(t)$  (Eq. (14)) calculated using the potential in Eq. (54) for five values of the parameter  $a$ . We use a one-particle one-dimensional model described by an overdamped Langevin equation with the driving force  $f = 1.0$ . We convert all quantities into dimensionless forms using the time unit  $\gamma T^{-1} l^2$ , energy  $T$ , and length  $l$ , where  $\gamma$  is the friction coefficient.

## 1 VI. DISCUSSION

2 We have exactly proved the reciprocal relations of Eqs. (16) and (18) assuming Eq. (17).  
 3 We now discuss why Eq. (17) was necessary for deriving Eqs. (16) and (18). Equation (17)  
 4 has been used to show  $\Delta_e(t) = 0$ , where  $\Delta_e(t)$  is given by the division of  $\Delta_{21}(t) - \Delta_{12}(t)$   
 5 into  $\Delta_o(t)$  and  $\Delta_e(t)$ . To show  $\Delta_e(t) = 0$ , we have to express  $g_o(x, t)$  and  $g_e(x, t)$  in the  
 6 forms of Eqs. (47) and (48), where any higher derivative of  $F(x)$  is expressed by  $F(x)$  and  
 7  $F'(x)$ . The expressions of the higher derivative can be obtained using Eq. (46) derived from  
 8 Eq. (17) and have also been applied to Eq. (51).

9 In the following, we discuss whether our result can be transferred to non-equilibrium  
 10 systems other than the systems considered in this study. First, we discuss the transferability  
 11 to systems where the potential does not satisfy Eq. (17). Because we can prove  $\Delta_o(t) = 0$   
 12 without Eq. (17), Eqs. (16) and (18) are valid for systems with  $\Delta_e(t) = 0$ . Thus, even if  
 13 Eq. (17) is not satisfied, we can obtain  $\Delta_{21}(t) - \Delta_{12}(t) = 0$ , for instance, in the case of  
 14  $g_e(x, t) = g_o(x, t)$  in Eq. (43). Because it is not clear whether such a system exists, we have  
 15 to study the possibility in future work.

16 Next, we discuss the transferability to the underdamped Langevin case, where we have

1 to consider the particle momentum  $p$  as well as the position  $x$ . Because of the consideration  
 2 of  $p$ , we cannot divide  $\Delta_{21}(t) - \Delta_{12}(t)$  in the same way as in the overdamped case. Even if  
 3 we can divide it in another way, we cannot show that the two divided parts vanish. This is  
 4 because considering  $p$  does not allow us to obtain equations valid in the overdamped case.  
 5 We obtain  $\Delta_o(t) = 0$  from Eq. (31) and  $\Delta_e(t) = 0$  from Eqs. (47) and (48), but we cannot  
 6 obtain such equations in the underdamped case.

7 Finally, we discuss the transferability to a many-particle three-dimensional system de-  
 8 scribed by the overdamped Langevin equation [8]. In this case, we assume

$$\frac{d^2 U(\{\mathbf{x}_i\})}{d\mathbf{x}_i^2} \propto U(\{\mathbf{x}_i\}), \quad (57)$$

9 where  $U(\{\mathbf{x}_i\})$  is the potential including particle interaction terms,  $\mathbf{x}_i$  is the position of  
 10 particle  $i$ , and  $\{\mathbf{x}_i\} = \mathbf{x}_1, \mathbf{x}_2, \dots$ . In this system, we can divide  $\Delta_{21}(t) - \Delta_{12}(t)$  in the  
 11 same way as in the one-particle one-dimensional system, so we obtain  $\Delta_o(t)$  and  $\Delta_e(t)$ .  
 12 Nevertheless, we cannot show  $\Delta_o(t) = 0$  because Eq. (31) is not valid in this system. In  
 13 addition, we cannot show  $\Delta_e(t) = 0$  even using Eq. (57) because it is not possible to obtain  
 14 equations similar to Eqs. (47) and (48).

## 15 VII. CONCLUSION

16 In this work, we have exactly derived the reciprocal relation (16), which is valid in the  
 17 NESS, from an overdamped Langevin equation assuming Eq. (17). Our reciprocal relation  
 18 can be expressed in a non-integral form with respect to the frequency, in contrast to other  
 19 relations derived by previous studies. This relation is valid far from an equilibrium state  
 20 because the derivation of the relation is independent of the driving force  $f$  representing the  
 21 extent of the nonequilibrium state. Because our reciprocal relation is expressed only with  
 22 measurable quantities, one can verify its validity through experiments on systems such as a  
 23 colloidal suspension. Our reciprocal relation gives deeper understanding of the cross effect  
 24 between thermal and mechanical perturbations to the NESS.

### 25 Appendix A: Derivation of Eq. (32)

26 In this appendix, we derive Eq. (32), by expanding  $e^{s\hat{L}}$  in powers of  $\hat{L}$ . We expand  $e^{s\hat{L}}$



1 on the left side of Eq. (32) to obtain

$$\hat{J}e^{s\hat{L}}\hat{J}P_{st}(x) = \hat{J}\sum_{n=0}^{\infty}\frac{s^n}{n!}\hat{L}^n\hat{J}P_{st}(x). \quad (\text{A1})$$

2 Substituting Eqs. (19) and (24) into Eq. (A1), we obtain

$$\hat{J}\sum_{n=0}^{\infty}\frac{s^n}{n!}\hat{L}^n\hat{J}P_{st}(x) = \hat{J}\sum_{n=0}^{\infty}\frac{s^n}{n!}\left(-\gamma^{-1}\frac{d}{dx}\hat{J}\right)^n\hat{J}P_{st}(x). \quad (\text{A2})$$

3 We rewrite the right side of Eq. (A2) in the form

$$\hat{J}\sum_{n=0}^{\infty}\frac{s^n}{n!}\left(-\gamma^{-1}\frac{d}{dx}\hat{J}\right)^n\hat{J}P_{st}(x) = \sum_{n=0}^{\infty}\frac{s^n}{n!}\left(-\gamma^{-1}\hat{J}\frac{d}{dx}\right)^n\hat{J}^2P_{st}(x). \quad (\text{A3})$$

4 From Eq. (A3) with  $\hat{L}^{\dagger*} = -\gamma^{-1}\hat{J}d/dx$  and

$$e^{s\hat{L}^{\dagger*}} = \sum_{n=0}^{\infty}\frac{s^n}{n!}\left(\hat{L}^{\dagger*}\right)^n, \quad (\text{A4})$$

5 we obtain

$$\hat{J}e^{s\hat{L}}\hat{J}P_{st}(x) = e^{s\hat{L}^{\dagger*}}\hat{J}^2P_{st}(x). \quad (\text{A5})$$

6 We can derive Eq. (32) from Eq. (A5), obtained by expanding  $e^{s\hat{L}}$ , using the property of  
7 the stationary distribution function  $P_{st}(x)$ . Because Eq. (24) leads to the property

$$\frac{d}{dx}\hat{J}P_{st}(x) = 0, \quad (\text{A6})$$

8 we obtain

$$e^{s\hat{L}^{\dagger*}}\hat{J}^2P_{st}(x) = e^{s\hat{L}^{\dagger*}}F(x)\hat{J}P_{st}(x). \quad (\text{A7})$$

9 By applying the operator  $e^{s\hat{L}^{\dagger*}}$  to  $F(x)\hat{J}P_{st}(x)$  in Eq. (A7) and using  $\hat{L}^{\dagger*} = -\gamma^{-1}\hat{J}d/dx$  and  
10 Eq. (A6), we rewrite the right side of Eq. (A7) in the form

$$e^{s\hat{L}^{\dagger*}}F(x)\hat{J}P_{st}(x) = \left[e^{s\hat{L}^{\dagger*}}F(x)\right]\hat{J}P_{st}(x). \quad (\text{A8})$$

11 From Eqs. (A5), (A7), and (A8), we finally obtain Eq. (32).

## 12 **Appendix B: Derivation of Eqs. (47) and (48)**

13 Using Eq. (46), we obtain

$$\begin{aligned} \hat{L}_1^\dagger[F(x)]^m[F'(x)]^n &= c_1[F(x)]^m[F'(x)]^{n+1} + c_2[F(x)]^{m+2}[F'(x)]^{n-1} + c_3[F(x)]^{m+1}[F'(x)]^{n-1} \quad (m \geq 0, n \geq 1), \\ & \end{aligned} \tag{B1}$$

$$\begin{aligned} \hat{L}_0^\dagger[F(x)]^m[F'(x)]^n &= c'_1[F(x)]^{m+2}[F'(x)]^{n-2} + c'_2[F(x)]^m[F'(x)]^n + c'_3[F(x)]^{m-2}[F'(x)]^{n+2} \\ &+ c'_4[F(x)]^{m+1}[F'(x)]^{n-2} + c'_5[F(x)]^{m-1}[F'(x)]^n + c'_6[F(x)]^m[F'(x)]^{n-2} \quad (m \geq 2, n \geq 2), \\ & \end{aligned} \tag{B2}$$

1 where  $m$  and  $n$  are integers, and  $c_i$  and  $c'_i$  are constants independent of  $x$ . Equation (B1)  
2 shows that  $\hat{L}_1^\dagger$  changes the exponent of  $F'(x)$  into an odd number when  $n$  is even. When  $n$   
3 is odd,  $\hat{L}_1^\dagger$  changes the exponent into an even number. In contrast, we find from Eq. (B2)  
4 that  $\hat{L}_0^\dagger$  does not change the parity of the exponent of  $F'(x)$ . Because the same situations  
5 are valid for  $n < 2$  or  $m < 2$ , we can rewrite Eqs. (39) and (40) in the forms of Eqs. (47)  
6 and (48).

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