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Reconstruction Lemma and Fluctuation-Dissipation Theorem

Alexander N. Gorban
Institute of Computational Modeling, Russian Academy of Sciences
660036 Krasnoyarsk, Russia

Iliya V. Karlin
ETH Zürich, Institute of Polymers
CH-8092 Zürich, Switzerland
e-mail: ikarlin@ifp.mat.ethz.ch

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We discuss a new approach to nonequilibrium statistical thermodynamics based on mappings of the microscopic dynamics into the macroscopic dynamics. Near stationary solutions, this mapping results in a compact formula for the macroscopic vector field without a hypothesis of a separation of time scales. Relations of this formula to short-memory approximation, the Green-Kubo formula, and expressions of transport coefficients in terms of Lyapunov exponents are discussed.

Keywords: Nonequilibrium statical mechanics, coarse-graining, exact fluctuation-dissipation relation

Se discute una nueva aproximación a la termodinámica estadística fuera de equilibrio basándose en mapeos de la dinámica microscópica dentro de la dinámica macroscópica. En soluciones casi estacionarias, este mapeo da lugar a una fórmula compacta para el campo vectorial macroscópico, sin una hipótesis de separación de escalas de tiempo. Se discuten las relaciones de esta fórmula con la aproximación de memoria corta, la fórmula de Green-Kubo y las expresiones de los coeficientes de transporte en términos de los exponentes de Lyapunov.

Descriptores: Mecánica estadística fuera de equilibrio; relación de fluctuación-disipación exacta.

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Dedicated to Leo García-Colín on the occasion of his 70th birthday.

1. Introduction

One of the most important issues of the mathematical modeling in natural sciences is the description of a projection of the detailed microscopic dynamics onto a coarser macroscopic dynamics. This issue becomes of increasing importance, in particular, in molecular biology where even the terms in which the "micro" and the "macro" are accessible for a sensible formalization are not completely settled [1].

Leaving aside the most general questions of mappings between various dynamics, in this paper we intend to discuss some aspects of this problem as it arises in a much more studied discipline, the classical statistical mechanics of nonequilibrium systems. Seminal works of Ehrenfest [2], Onsager [3], Green [4], Kubo [5], have posed a question of derivation of macroscopic dynamics from the microscopic dynamics. An answer of remarkable generality has been first suggested by Onsager [3] by imposing his famous condition that a regression towards the equilibrium is the same as the dynamics of fluctuations in the equilibrium. A technical implementation of Onsager's condition for the Liouville equation has been first achieved by Green [4] in the case of a wide separation of time scales of the microscopic and the macroscopic motions, and is by now the textbook material [6].

Recently [7, 8], the geometric feature of the coarse-graining has been formalized to the following observation: given a finite coarse-graining time interval τ, it is possible to reconstruct uniquely the macroscopic dynamics from a single condition. In order to formulate this condition for the purpose of what will follow, let us consider a microscopic dynamics given by the linear equation for the distribution function $f(x,t)$,

$$\dot{f} = Lf, \tag{1}$$

where L is the linear operator. Furthermore, let $\mathbf{m}[f]$ be a set of linear functionals whose values, $\mathbf{M} = \mathbf{m}[f]$ representing the macroscopic variables, and $f(\mathbf{M})$ a set of distribution functions satisfying the consistency condition,

$$\mathbf{m}[f(\mathbf{M})] = \mathbf{M}. \tag{2}$$

Let us denote as $\mathbf{M}(t)$ the initial condition at the time t to the (*unknown*!) equations of the macroscopic motion, and let us take $f[\mathbf{m}(t)]$ for the initial condition of the microscopic equation (1) at the time t. Then the condition for the reconstruction of the macroscopic dynamics [7, 8] reads as follows:

For every initial condition $\{\mathbf{M}(t), t\}$*, solutions to the macroscopic dynamic equations at the time* $t+\tau$ *are equal to the values of the macroscopic variables on the solution to Eq. (1) with the initial condition* $\{f(\mathbf{M}(t)), t\}$

$$\mathbf{M}(t+\tau) = \mathbf{m}\Big\{\mathrm{e}^{L\tau} f[\mathbf{M}(t)]\Big\}. \tag{3}$$

The right hand side of Eq. (3) represents an operation on trajectories of the microscopic equation (1), introduced in a particular form by Ehrenfest [2] (the coarse-graining): the solution at the time $t+\tau$ is replaced by the state on the manifold $f(\mathbf{M})$. Notice that the coarse-graining time τ in Eq. (3)

is finite, and we stress the required independence of the condition (3) from the initial time t and the initial condition at t.

The major observation of Ref. 7 and 8 is that the condition just formulated is sufficient for the unique reconstruction of the macroscopic dynamics which carries on the macroscopic variables in the left-hand side of Eq. (3). The essence of the derivation is as follows: seeking the macroscopic equations in the form

$$\dot{\mathbf{M}} = \mathbf{R}(\mathbf{M}, \tau), \tag{4}$$

we proceed with Taylor expansion of the unknown functions $\mathbf{R}$ in terms of powers τ^n, where $n = 0, 1, \ldots$, and require that approximation, $\mathbf{R}^{(n)}$, of the order n, is such that resulting macroscopic solutions verify the condition (3) to the order τ^{n+1}. This process of successive approximation is solvable. Thus, the unknown macroscopic equation (4) is reconstructed to any given accuracy from the condition (3).

The result [7, 8] is sufficiently general, it applies even if the microscopic equation is nonlinear (for example, to the Liouville equation in the presence of mean field interactions). In the sequel, we refer only to a particularly interesting case of the microscopic dynamics which conserves a concave functional $S(f)$ along any trajectory (the entropy). This is the case of the standard Liouville equation. Among possible sets $f(\mathbf{M})$, distinguished rôle is played by quasi-equilibrium approximations $f^*(\mathbf{M})$ which are maximizers of the functional $S(f)$ for fixed $\mathbf{M}$, and which are not invariant under the microscopic dynamics [Eq. (1)]. Then the first and higher order approximations, $\mathbf{R}^{(1)}$, and so on, to the macroscopic dynamics (4) are dissipative with the Lyapunov function $S^*(\mathbf{M}) = S[f^*(\mathbf{M})]$, and explicit entropy production formula has been demonstrated for the vector field $\mathbf{R}^{(1)}$. We notice it in passing that, since the work of Jaynes [9], the usefulness of quasi-equilibrium approximations is well understood in various versions of projection operator formalism [10–13], (see also Remarks 3 and 4) below), as well as for the dissipative dynamics [14–16]. Relatively less studied remains the case of open or externally driven systems, where invariant quasi-equilibrium manifolds may become unstable [17].

Examples have been provided [8], demonstrating that several well known dissipative macroscopic equations, such as the Navier-Stokes equation and the diffusion equation for the one-body distribution function, are derived as the lowest order approximations of this construction. Thus, the construction [7, 8] outlined above has formalized the original Ehrenfest's idea [2] by bridging the apparent gap between the discrete time coarse-graining and the continuous time macroscopic dynamics. In the sequel, we term it as the *reconstruction lemma*.

Remark 1. One instance where the condition (3) has been used in the past is the work of Lewis [18], who suggested a formal replacement

$$\mathbf{M}(t+\tau) \to \mathbf{M}(t) + \tau\dot{\mathbf{M}},$$

on the left hand side of Eq. (3), and

$$\dot{\mathbf{M}} = \frac{1}{\tau}\Big\{\mathbf{m}\big[\mathrm{e}^{L\tau} f^*(\mathbf{M})\big] - \mathbf{M}\Big\} \tag{5}$$

This is very different from Refs. 7 and 8: Eq. (5) does not establish a mapping of the microscopic trajectories into the macroscopic, and is identified as a well known differential pursuit. In particular, Eq. (5) does not pass a consistency test since it gives a non-trivial entropy production in the case when the quasi-equilibrium solves the Liouville equation.

Remark 2. The Ehrenfest's argument has been used in a different way by del Río-Correa and García-Colín [19]: if the coarse-graining time τ is considered as the *dynamically unresolved scale* then the macroscopic dynamics on a much larger scale is seen as a (stationary) Markov process.

Remark 3. It is instructive to compare the present approach with the projection operator formalism mentioned above. These approaches can be represented as the following two steps

i) Introducing a projection operator P, the Liouville equation is rewritten into equivalent system describing the motion along and transverse the manifold $f(\mathbf{M})$. The choice of projector is a priori not unique. For quasi-equilibrium manifolds $f^*(\mathbf{M})$, it is prompted by the fact that the projection of the vector field Lf^* attached to each point of the quasi-equilibrium manifold being projected by the quasi-equilibrium projector, $P^* = D_{\mathbf{M}} f^* \cdot \mathbf{m}$, preserves the entropic property of the Liouville equation: if $D_f S \cdot Lf = 0$ for any f, then $D_{\mathbf{M}} S^*(\mathbf{M}) \cdot P^* L f^*(\mathbf{M}) = 0$ for any $\mathbf{M}$ (if entropy is conserved by the microscopic dynamics, the quasi-equilibrium entropy is also conserved by the quasi-equilibrium approximation to the macroscopic dynamics, or, the zero-order approximation $\mathbf{R}^{(0)}$ [Eq. (3)] is non-dissipative, and is purely kinematics [13]). This way of representing the Liouville equation has been introduced by Robertson [10], and has been used with some modifications by many authors. Modifications, in the first place, concern the choice of the representation of Robertson's projection operator (notice that P^* acts on vector fields, and maps vector fields onto tangent space of the quasi-equilibrium manifold), such as the affine projector (acting on the distribution functions) and the adjoint projector (acting on the adjoint space, or "observables", in the language of quantum mechanics), see, *e.g.* [11, 12], for relations between various representations of the projection operator.

ii) Any system equivalent to the Liouville equation is not dissipative (we do not consider here the case of *infinite* systems which can demonstrate behaviour atypical to their finite and arbitrary large counterparts. For instance, the infinite ideal gas without collisions is a K-system [20]). Therefore, an analog of coarse-graining is introduced on the later stages of the projector operator formalism. To the best of our knowledge, a general discussion of the latter step is still missing, in spite of some efforts (see, *e.g.* Ref. 21). Notice that, in the reconstruction lemma, the projection operator is not introduced a priori, although it arises quite naturally in the result [7, 8].

Remark 4. Let us comment on how the coarse-graining is implemented in the method of nonequilibrium statistical operator introduced by Zubarev [12]. Instead of considering individual solutions to the Liouville equation (1), $f(t) = U(t,t')f^*(t')$, where $U(t,t') = \exp[(t-t')L]$, and $f^*(t')$ is the initial quasi-equilibrium condition, one averages over a set of initial conditions, and the "true nonequilibrium state" $\overline{f}(t)$ is considered to be

$$\overline{f}(t) = \frac{1}{t-t_0}\int_{t_0}^{t} U(t,t')f^*[\,\overline{f}(t')]\,dt'. \qquad (6)$$

Here t_0 is some initial time taken far enough in the past relative to t. In order to make it explicit the way the coarse-graining is implemented by Eq. (6), let us do the following: solving Eq. (6) by iterations, we obtain a sequence of approximations $\overline{f}_n$, where

$$\overline{f}_{n+1}(t) = \frac{1}{t-t_0}\int_{t_0}^{t} U(t,t')f^*[\,\overline{f}_n(t')]\,dt'. \qquad (7)$$

On the first iteration we have,

$$\overline{f}_1(t) = \frac{1}{t-t_0}\int_{t_0}^{t} U(t,t')f^*[U(t',t_0)f^*(t_0)]\,dt'. \qquad (8)$$

Let us now approximate the time integral in Eq. (8) by the integral sum: partitioning the interval $t-t_0$ into N segments of length τ, we may write

$$\overline{f}_1(t) \approx \frac{1}{N+1}\sum_{k=0}^{N} U(t,t_k)f_k^*, \qquad (9)$$

where $t_k = t_0 + k\tau$, and

$$f_k^* = f^*[U(t_k,t_0)f^*(t_0)], \qquad (10)$$

are the initial conditions on the quasi-equilibrium manifold obtained by coarse-graining the solution $U(t,t_0)f^*(t_0)$ at the times t_k. Thus, the sense of the averaged nonequilibrium state $\overline{f}_1$ is as follows:

a) By coarse-graining the solution $U(t,t_0)f^*(t_0)$, we create a set of quasi-equilibrium states f_k^*.

b) Taking these states as initial conditions, we find corresponding solutions to the Liouville equation at the time t.

c) The nonequilibrium state $\overline{f}_1(t)$ is taken as the average of these solutions.

This process is visualized in Fig. 1. Once the first iteration $\overline{f}_1(t)$ is obtained, the entire process is iterated according to Eq. (7). Though the coarse-graining behind Eq. (6) is rather involved, the differential equation for the function $\overline{f}$ has a compact form,

$$\dot{\overline{f}}(t) = L\overline{f}(t) - \frac{1}{t-t_0}\{\overline{f}(t) - f^*[\overline{f}(t)]\}. \qquad (11)$$

This is the Liouville equation with the source term which implements the coarse-graining. The time segment $t-t_0$ is regarded as the time necessary for the system to develop nonequilibrium correlations. By sending the initial time t_0 to $-\infty$ one expects that all the relevant correlations will have enough time to develop. With this, one comes up with the Liouville equation *with broken time-symmetry*:

$$\dot{\overline{f}}(t) = L\overline{f}(t) - \epsilon\{\,\overline{f}(t) - f^*[\overline{f}(t)]\}, \qquad (12)$$

where $\epsilon \to +0$ (the latter limit should be taken after finding solutions for finite ϵ, and after taking the thermodynamic limit).

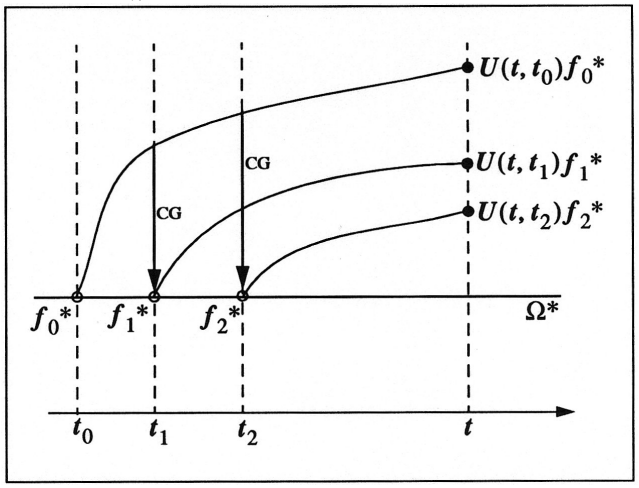

FIGURE 1. Coarse-graining in the method of nonequillibrium statistical operator. The manifold of quasi-equilibrium states is denoted as Ω^*. Thin curves represent solutions to the Liouville equation. Arrows represent coarse-graining (CG) which replaces the solution at discrete times t_k by the quasi-equilibrium states f_k^* (empty circles). The averaged nonequilibrium state $\overline{f}(t)$ (9) is the mean of filled circles $U(t,t_k)f_k^*$.

It can be said that the limit $\epsilon \to +0$ in Zubarev's formalism implements a "weak" coarse-graining (as opposed to the "hard" coarse-graining in the reconstruction lemma). Indeed, the sequence of quasi-equilibrium states can be obtained by solving Eq. (11) with finite $\epsilon = 1/\tau$ with a splitting algorithm[a]. However, this analogy is incomplete. A single sequence of quasi-equilibrium states does not form the condition (3), missing are two ingredients

1) The macroscopic motion [the left hand side of Eq. (3)], and, most important.

2) Condition (3) should hold for *every* such sequence, regardless of the initial conditions [22].

The advantage of the reconstruction lemma is the locality of construction since only Taylor series expansion in the vicinity of the manifold is involved. This is also its limitation. From the physical standpoint, finite and fixed coarse-graining time τ is a phenomenological device which makes it possible to infer the form of the macroscopic equations by a non-complicated computation rather than to derive a full form thereof. While using the fixed coarse-graining time we, in some cases, just mimic a true physical process of coarse-graining by a generalized thermostat, resulting in the short

memory approximation. For instance, the form of the Navier-Stokes equations can be derived from the simplest model of free motion of particles without collisions, in which case the coarse-graining is a substitution for the collisions. Going away from the limitations imposed by the reconstruction lemma [7, 8] is the major problem of the mathematically consistent formulation of the nonequilibrium statistical mechanics based on mappings between the microscopic and the macroscopic dynamics. In this paper, we demonstrate it that taking the limit of the infinite coarse-graining time τ in the above construction, *and without invoking any additional assumptions*, results in a compact formula for the macroscopic dynamics near equilibrium microscopic solutions. This result has been only briefly mentioned already [8] but here we argue that it covers all possible outcomes of the macroscopic dynamics near the equilibrium. In particular, we demonstrate that the Green-Kubo formula is contained in our result if one restores to the argument about separation of time scales. We shall also comment on the extensions of this result to the nonlinear domain, as well as relations to dynamic-theoretic approach to transport coefficients.

We consider here the case of a dynamics close to the equilibrium state, $f^{\rm eq}$, such that $Lf^{\rm eq}=0$, and $f^{\rm eq}$ is normalized. We also assume that the macroscopic variables $\mathbf{M}$ vanish at equilibrium, and are normalized in such a way that

$$\mathbf{m}[f^{\rm eq}\mathbf{m}^\dagger]=\mathbf{1},$$

where $(\dagger)$ denotes transposition, and where $\mathbf{1}$ is an appropriate identity operator. To the first order in $\mathbf{M}$, the quasi-equilibrium is a linear manifold,

$$f^*(\mathbf{M})=f^{\rm eq}(1+\mathbf{m}^\dagger\cdot\mathbf{M}).$$

The linear dynamics of the macroscopic variables M is thought in the form,

$$\dot{\mathbf{M}}=\mathbf{A}\mathbf{M}, \tag{13}$$

where $\mathbf{A}$ is a linear operator to be determined from the coarse-graining condition (3). In order to do this, we use exponential representation of solutions to Eq. (13), and write the condition (3) as follows:

$$\mathrm{e}^{\tau\mathbf{A}}\mathbf{M}=\mathbf{m}\big[\mathrm{e}^{L\tau}f^*(\mathbf{M})\big]=\mathbf{m}\big[\mathrm{e}^{L\tau}f^{\rm eq}\mathbf{m}^\dagger\big]\cdot\mathbf{M}. \tag{14}$$

Next we require that this condition should be valid for *every* $\mathbf{M}$ in the (formal) limit of infinitely large coarse-graining time. Thus,

$$\mathbf{A}=\lim_{\tau\to\infty}\frac{1}{\tau}\ln\big[\mathbf{m}\big(\mathrm{e}^{L\tau}f^{\rm eq}\mathbf{m}^\dagger\big)\big]. \tag{15}$$

Equation (15) is compact but, in fact, a rather complicated expression involving logarithm of non-commuting operators $\mathbf{m}$ and L. It is the general expression for the near-equilibrium macroscopic dynamics, as it follows from the condition (3) in the limit of large coarse-graining time.

Remark 5. In general, Eq. (15) does not require a separation of the time scales of the microscopic and the macroscopic evolution. On the other hand, intuitively, if there is such a separation, then we expect a "two-time coarse-graining condition" instead of the "one-time coarse-graining condition" (3),

$$\mathbf{M}(t+\tau_1)=\mathbf{m}\big\{\mathrm{e}^{L\tau_2}f^*[\mathbf{m}(t)]\big\}, \tag{16}$$

for $\tau_1\ll\tau_2$. However, the reconstruction lemma cannot be implemented rigorously starting with Eq. (16).

Let us now demonstrate how the result (15) reduces to the well known Green-Kubo formula in the limit of a large separation of the macroscopic and the microscopic motions. In order to do so, for simplicity, we shall assume that the kinematic contribution,

$$\mathbf{A}^{(0)}=\mathbf{m}[Lf^{\rm eq}\mathbf{m}^\dagger], \tag{17}$$

is equal to zero. Next, using an operator identity (which can be checked by differentiation),

$$\mathrm{e}^{L\tau}=1+L\tau+L\tau\underline{\left[\frac{1}{\tau}\int_0^\tau(\tau-t)\mathrm{e}^{tL}\,dt\right]}L, \tag{18}$$

in the Eq. (15), we further *assume* that

i) The microscopic dynamics is such that the underlined term in Eq. (18) for large enough τ is well approximated by the following expression

$$\int_0^\infty \mathrm{e}^{tL}\,dt.$$

ii) The expansion of the logarithm in terms of τ to first order is a valid approximation *before* taking the limit.

With these two crucial assumptions, and taking into account the simplifying assumption (17), we obtain in Eq. (15),

$$\mathbf{A}=\mathbf{m}\left[L\left(\int_0^\infty\mathrm{e}^{tL}\,dt\right)Lf^{\rm eq}\mathbf{m}^\dagger\right]. \tag{19}$$

Finally, using the two properties of the Liouville operator L, $L^\dagger=-L$ (anti-symmetry), and $L(fg)=fLg+gLf$ (operator L is differentiation), and introducing the usual notation, $\dot{\mathbf{m}}=L^\dagger\mathbf{m}$, and $\langle\ldots\rangle=\int f^{\rm eq}(x)\ldots dx$ (averaging over the equilibrium state), we arrive in Eq. (19) at

$$\mathbf{A}=\int_0^\infty\langle\dot{\mathbf{m}}(0)\dot{\mathbf{m}}(t)\rangle\,dt. \tag{20}$$

This is the most standard case of the Green-Kubo formula, or the *fluctuation-dissipation theorem*, which expresses the matrix of transport coefficients $\mathbf{A}$ in terms of the autocorrelation function of the corresponding fluxes.

The above analysis demonstrates that though the Eq. (15) reduces to the Green-Kubo formula, this derivation uses a set of sensitive assumptions. The necessity of using these assumptions stems from the fact already mentioned above that the derivation of the Eq. (15) does not uses a two time scales picture. It would be interesting to test equation (15) in those cases where the Green-Kubo Eq. (20) is known to be inapplicable [23]. Extension of the present approach to the general quasi-equilibrium manifolds is a challenge for our further study.

Final remarks. The *existence* of the non-trivial limit (15) is a difficult problem which requires a separate investigation. Strictly speaking, the limit is expected to be non-trivial only in the thermodynamic limit. On the other hand, if dissipativity is brought into the Liouville equation, for example, in the form of a thermostatting mechanism of NEMD [24], then the non-trivial limit (15) will exist also for finite systems. It is not surprising, therefore, that the form of the generalized fluctuation-dissipation theorem (15) is quite similar to expressions of transport coefficients in terms of Lyapunov exponents (see, *e.g.* [25], and references therein).

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