GENERAL LINEAR RESPONSE FORMULA IN STATISTICAL MECHANICS, AND THE FLUCTUATION-DISSIPATION THEOREM FAR FROM EQUILIBRIUM.

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Abstract. *Given a nonequilibrium steady state* ρ *we derive formally the linear response formula given by equation (6) in the text for the variation of an expectation value at time* t *under a time-dependent infinitesimal perturbation* $\delta_\tau F$ *of the acting forces. This leads to a form of the fluctuation-dissipation theorem valid far from equilibrium: the complex singularities of the susceptibility are in part those of the spectral density, and in part of a different nature to be discussed.*

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Serious attention is now starting to be paid to the chaotic dynamics underlying nonequilibrium statistical mechanics. (See for instance Chernov et al. [1]). A statistical mechanical system is kept far away from equilibrium by nonhamiltonian forces, and "cooled" by a "Gaussian thermostat" (Hoover [2], Evans and Morriss [3]). What this amounts to is that the phase space M of the system is taken to be compact, and its time evolution given by

$$\frac{dx}{dt} = F(x,t) \tag{1}$$

where no particular assumption is made on F (except smoothness, and often t-independence).

It would at first appear that the framework provided by

$$\frac{dx}{dt} = F(x) \tag{2}$$

is much too general to provide results of interest for nonequilibrium statistical mechanics. It is reasonable however to assume that (2) defines a chaotic time evolution, and that we may exclude a set of Lebesgue measure 0 of initial conditions in the distant past. These assumptions have surprisingly strong consequences. If we translate *chaos* mathematically by *uniform hyperbolicity*, then time averages are uniquely determined and given by a so-called SRB measure (see below). Physical time evolutions are often hyperbolic in the weaker sense that most Lyapunov exponents are different from 0. A natural idea is thus to proceed as if physical systems were uniformly hyperbolic (and then compare the results with experiments). This has been called the *chaotic hypothesis* by Gallavotti and Cohen (who also assume microscopic reversibility). The approach just outlined has been vindicated in the case of the Gallavotti-Cohen fluctuation theorem [4], which agrees with numerical experiments far from equilibrium [5], [6]. From the chaotic principle one also recovers near equilibrium the Onsager reciprocity relations [7], [8]. The present letter follows the same philosophy for the study of linear response far from equilibrium. Our calculations will be formal and easy. They can be made rigorous if uniform hyperbolicity holds [9], but this is harder. A rigorous analysis in a more general setup seems difficult and one may have to untroduce new ideas like the limit of a large system (thermodynamic limit). Such a limit will be used anyway to compute transport coefficients like the viscosity. What may be said now is that if linear response for physical systems has a simple expression, it must be the one given by our formal calculation. The formulae that we obtain appear thus unavoidable, and should be fundamental for nonequilibrium statistical mechanics far from equilibrium.

Integrating (1) with initial condition x at time s gives at time t a point $x(s,t)$. Suppose now that we are in the time independent situation of (2). We may thus write

$$x(s,t) = x(t-s) = f^{t-s}x \tag{3}$$

If m is a probability measure absolutely continuous with respect to the volume element dx on the phase space M, and if m under the time evolution f^t tends weakly to a limit ρ, then ρ is a good candidate to describe a nonequilibrium steady state. Using the notation

$$m(\Phi) = \int m(dx)\Phi(x) \qquad , \qquad \rho(\Phi) = \int \rho(dx)\Phi(x)$$

we have by assumption, for every continuous Φ,

$$\lim_{s\to-\infty} \int m(dx)\Phi(f^{-s}x) = \int \rho(dx)\Phi(x) \tag{4}$$

We call $\rho = \rho_F$ an SRB measure, it is usually not absolutely continuous with respect to dx. Such measures were introduced by Sinai, Ruelle, and Bowen [10], [11], [12] for uniformly hyperbolic dynamical systems, where it was shown that there is a unique SRB measure on each mixing attractor. A much more general study of SRB measures was then made by Ledrappier, Strelcyn, and Young [13], [14] (see Young [15] for recent results). The use of SRB measures to describe nonequilibrium steady states in statistical mechanics was advocated early by Ruelle [16], but only recently did it lead to useful results with the *Fluctuation Theorem* of Gallavotti and Cohen [4].

One problem with SRB measures is that their characterizations [10], [11], [12], [13], [14] are difficult to use. It is however relatively easy to expand $\rho_{F+\delta F}$ with respect to δF [17]. This has been done rigorously in a special case [9]. Here we proceed instead formally, and to first order with respect to a *time dependent* perturbation $\delta_t F$ (we keep F *time independent*). We shall thus recover in a new manner some classical results of nonequilibrium statistical mechanics, for which see for instance [18], and also obtain new results.

Using (4), we have formally

$$\delta_t \rho(\Phi) = \delta \lim_{s\to-\infty} \int m(dx)\, \Phi(x(s,t)) = \lim_{s\to-\infty} \int m(dx)\, \delta x(s,t) \cdot \nabla_{x(s,t)} \Phi$$

and we may assume $s < t$. Writing also $T_x f^\sigma$ for the tangent map at x to f^σ (in coordinates this is the matrix of partial derivatives), we have

$$\delta x(s,t) = \int_s^t d\tau\, (T_{x(s,\tau)} f^{t-\tau}) \delta_\tau F(x(s,\tau))$$

where we may replace $x(s,\tau)$ by $f^{\tau-s}x$. Using also the notation $(f^* m)(\Phi) = m(\Phi \circ f)$, so that $\lim_{\sigma\to\infty}(f^{\sigma *} m)(\Phi) = \rho_F(\Phi)$, we obtain

$$\delta_t \rho(\Phi) = \lim_{s\to-\infty} \int_s^t d\tau \int ((f^{\tau-s})^* m)(dy)\, \big((T_y f^{t-\tau}) \delta_\tau F(y)\big) \cdot \nabla_{y(\tau,t)} \Phi$$

With the notation of (3), our formal calculation gives thus

$$\delta_t\rho(\Phi) = \int_{-\infty}^{t} d\tau \int \rho_F(dy)\,\big((T_y f^{t-\tau})\delta_\tau F(y)\big)\cdot\nabla_{y(t-\tau)}\Phi$$

which can be rewritten in the equivalent forms

$$\delta_t\rho(\Phi) = \int_{-\infty}^{t} d\tau \int \rho_F(dx)\,\big((T_{x(\tau-t)} f^{t-\tau})\delta_\tau F(x(\tau-t))\big)\cdot\nabla_x\Phi \tag{5}$$

or

$$\delta_t\rho(\Phi) = \int_{-\infty}^{t} d\tau \int \rho_F(dy)\,\delta_\tau F(y)\cdot\nabla_y(\Phi\circ f^{t-\tau}) \tag{6}$$

Assuming that Φ is in a suitable space $\mathcal{B}$ of functions on M, and δF in a suitable space $\mathcal{X}$ of vector fields, we can rewrite (5) and (6) as

$$\delta_t\rho = \int d\tau\kappa_{t-\tau}\delta_\tau F = \int d\sigma\kappa_\sigma\delta_{t-\sigma}F$$

where the linear operator κ_σ maps $\mathcal{X}$ to the dual $\mathcal{B}^*$ of $\mathcal{B}$ ($\mathcal{B}^*$ consists of linear functionals on $\mathcal{B}$), and

$$(\kappa_\sigma X)\Phi = 0 \qquad \text{for } \sigma < 0$$

$$(\kappa_\sigma X)\Phi = \int \rho_F(dx)\big((T_{x(-\sigma)}f^\sigma)X(f^{-\sigma}x)\big)\cdot\nabla_x\Phi = \int \rho_F(dy)X(y)\cdot\nabla_y(\Phi\circ f^\sigma)$$

$$\text{for } \sigma \geq 0$$

The (operator-valued) *response function* $\sigma \to \kappa_\sigma$ vanishes for $\sigma < 0$: this is called *causality.*

As we have said, formulas like (5), (6) can be proved rigorously [9] under *uniform hyperbolicity* assumptions. They can be extended to time dependent F and ρ_F, and to random forces [9]. Near equilibrium and assuming reversibility one can use (6) to prove the Onsager reciprocity relation [7], [8] and compute higher order corrections [19].

To study the convergence of the right hand side of (6), assume that all Lyapunov exponents with respect to ρ_F are $\neq 0$, except one corresponding to the direction of the flow, and write $\delta_\tau F = X^s_\tau + \phi_\tau F + X^u_\tau$, where X^s_τ is in the stable (contracting) and X^u_τ is in the unstable (expanding) direction. We have then

$$\delta_t\rho(\Phi) = \int_0^\infty d\sigma \int \rho_F(dy)[\big((T_y f^\sigma)X^s_{t-\sigma}(y)\big)\cdot\nabla_{y(\sigma)}\Phi - (F\cdot\nabla\phi_{t-\sigma} + \mathrm{div}^u X^u_{t-\sigma})(y).\Phi(y(\sigma))]$$

where div^u is the divergence in the unstable direction. [The SRB measure ρ_F is smooth in the unstable direction, so that an integration by part in this direction can be performed, see [9]]. The convergence of the right hand side depends on the exponential decrease of $\sigma \mapsto (T_y f^\sigma)X^s$ and on the decay of the correlation function $\sigma \mapsto \rho_F((F\cdot\nabla\phi + \mathrm{div}^u X^u)(\Phi\circ f^\sigma))$.

In the same manner one obtains for the *susceptibility*, *i.e.*, the Fourier transform

$$\hat\kappa_\omega = \int \kappa_\sigma e^{i\omega\sigma} d\sigma$$

the formula

$$(\hat\kappa_\omega X)\Phi$$

$$= \int_0^\infty e^{i\omega\sigma} d\sigma \int \rho_F(dy)[\big((T_y f^\sigma)X^s(y)\big)\cdot\nabla_{y(\sigma)}\Phi - (F\cdot\nabla\phi + \mathrm{div}^u X^u)(y).\Phi(y(\sigma))]$$

where $X = X^s + \phi F + X^u$, or

$$\hat\kappa_\omega = \hat\kappa^s_\omega + \hat\kappa^u_\omega$$

where

$$(\hat\kappa^s_\omega X)\Phi = \rho_F[\Big(\int_0^\infty e^{i\omega\sigma} d\sigma((Tf^\sigma)X^s)\circ f^{-\sigma}\Big)\cdot\nabla\Phi] \tag{7}$$

$$(\hat\kappa^u_\omega X)\Phi = -\int_0^\infty e^{i\omega\sigma} d\sigma\, \rho_F\big((F\cdot\nabla\phi + \mathrm{div}^u X^u)\circ f^{-\sigma}).\Phi\big) \tag{8}$$

At equilibrium, an important property of the susceptibility function is given by the *fluctuation-dissipation* theorem [20]. We shall now show that part of this property survives away from equilibrium, as a statement about singularities of the susceptibility function. First notice that, by causality, $\hat{\kappa}_\omega$ extends to an analytic function for $\mathrm{Im}\,\omega > 0$. ¿From this one can deduce the Kramers-Kronig *dispersion relations* [21]. The dispersion relations therefore also hold far from equilibrium. We discuss now the singularities of $\hat{\kappa}_\omega$ for $\mathrm{Im}\,\omega < 0$. For the sake of clarity, we consider the situation first at equilibrium, then away from equilibrium. Our discussion will not make use of microscopic reversibility.

At equilibrium, the SRB measure ρ_F is absolutely continuous with respect to the volume element of M, and we simply write $\rho_F(dx) = dx$. From (7) and (8), it follows that the singularities of $(\hat{\kappa}^{s,u}_\omega X)\Phi$ for $\mathrm{Im}\,\omega < 0$ are the same as those of

$$\int_{-\infty}^{\infty} e^{i\omega\sigma} d\sigma \rho_F((\Phi \circ f^\sigma).\Psi) = S_\omega(\Phi, \Psi) \tag{9}$$

for $\Psi = \mathrm{div}^s X^s$, or $F \cdot \nabla\phi + \mathrm{div}^u X^u$. The right-hand side of (9) is the Fourier transform of the correlation function $\sigma \mapsto \rho_F((\Phi \circ f^\sigma).\Psi)$, and S_ω is called the *spectral density.* For sufficiently regular Φ, Ψ, one can extend $S_\omega(\Phi, \Psi)$ to complex ω. The singularities of $\hat{\kappa}^s_\omega$ and $\hat{\kappa}^u_\omega$ (and $\hat{\kappa}_\omega$) are thus expected to be the same as those of S_ω for $\mathrm{Im}\,\omega < 0$. This connection between $\hat{\kappa}_\omega$ and S_ω is basically the fluctation-dissipation theorem.

Far away from equilibrium, the singularities of $\hat{\kappa}^u_\omega$ are (in view of (8)) again the same as the singularities with $\mathrm{Im}\,\omega < 0$ of the spectral density S_ω. For instance a simple pole of S_ω at ω_0 (with $\mathrm{Im}\,\omega_0 < 0$) corresponds to a simple pole $a(\omega - \omega_0)^{-1}$ of $\hat{\kappa}^u_\omega$ (even if it is not clear how to determine the residue a in practice). This is what remains here of the fluctuation-dissipation theorem. The singularities of $\hat{\kappa}^s_\omega$, however, become different from those of $\hat{\kappa}^u_\omega$. Define $\mathcal{T}^\sigma$ on the vector fields X^s (in the stable direction) by

$$(\mathcal{T}^\sigma X^s)(x) = (T_{f^{-\sigma}x} f^\sigma) X^s(f^{-\sigma}x) \tag{10}$$

Then, $(\mathcal{T}^\sigma)_{\sigma\geq 0}$ is a contraction semigroup and, if $-H$ is its infinitesimal generator, we have by the Hille-Yosida theorem [22]

$$\int_0^\infty e^{i\omega\sigma} d\sigma ((Tf^\sigma)X^s) \circ f^{-\sigma} = (H - i\omega)^{-1}$$

so that $(\hat{\kappa}^s_\omega X)(\Phi) = \rho_F[(H - i\omega)^{-1} X^s \cdot \nabla\Phi]$. The singularities of $\hat{\kappa}^s_\omega$ are thus related to the spectrum of H.

To summarize, we see that at equilibrium the singularities of the susceptibility $\hat{\kappa}_\omega$ are the singularities of the spectral density S_ω with $\mathrm{Im}\,\omega < 0$. Outside of equilibrium, the singularities of the susceptibility $\hat{\kappa}_\omega$ bifurcate into those of $\hat{\kappa}^u_\omega$ which are again the singularities of the spectral density S_ω with $\mathrm{Im}\,\omega < 0$, and those of $\hat{\kappa}^s_\omega$ which are different (and related to the spectrum of the infinitesimal generator of the semigroup $(\mathcal{T}^\sigma)_{\sigma\geq 0}$ defined by (10)). It is thus in principle possible to distinguish the singularities of $\hat{\kappa}^u_\omega$ and of $\hat{\kappa}^s_\omega$, and it would be interesting to see them in an experimental study bifurcating from each other as one moves away from equilibrium [23].

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