Generalized Langevin Equations: A Useful Tool for the Perplexed Modeller of Nonequilibrium Fluctuations?

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Abstract. The author identifies some subtle difficulties that one encounters within the framework of nonlinear generalized Langevin equations. These difficulties become even more pronounced when describing an open system that is in contact with more than one heat bath.

A well known technique to describe the dynamics of a system which is governed by fluctuations is the method of generalized master equations and the methodology of generalized Langevin equations. This strategy is by now well developed for thermal equilibrium systems. Here, the projector operator methodology (Mori 1965, Kawasaki 1973, Nordholm and Zwanzig 1975, Grabert et al. 1980, Grabert 1982) yields a clear-cut way to obtain the formal equations, either for the rate of change of the probability, i.e., the Generalized Master Equation (GME), or the generally nonlinear Generalized Langevin Equation (GLE). Take the case of relaxation of a system that is coupled to a thermal bath towards its unique equilibrium as specified by a single temperature T. Then, the equivalence between the two approaches is expected, but it is by no means transparent. Clearly, it are the statistical properties of the fluctuational force that determine this equivalence, such as its cumulant averages to an arbitrary high order, see in (Grabert et al. 1980). This fact is not widely appreciated because one often restricts the discussion of the statistical properties of the fluctuating force to the first two cumulants only; namely its average and its autocorrelation. Fact is that little is known between the connection of the GME and the corresponding GLE. One such relation which relates the generalized, generally nonlinear memory-diffusion matrix in the GME with the generally nonlinear memory-friction kernel in the GLE has been put forward with eqs. (55) and (56) in (Grabert et al. 1980).

A popular model consists of coupling a nonlinear system S bilinearly to a bath of harmonic oscillators. Then, the total Hamiltonian of a particle with mass M moving in a potential $U(x)$ in presence of a bath of harmonic oscillators reads (Zwanzig 1973, Ford and Kac 1987, Pollak 1986):

$$H = \frac{p^2}{2M} + U(x) + \sum_\alpha \left[\frac{p_\alpha^2}{2m_\alpha} + \frac{m_\alpha \omega_\alpha^2}{2} \left(q_\alpha - \frac{c_\alpha}{m_\alpha \omega_\alpha^2}\, x \right)^2 \right] . \tag{1}$$

Let us elucidate the reasoning which leads to the GLE. With (1) the equations of motion for the system degrees of freedom read

$$\begin{aligned} M\dot{x} &= p \\ \dot{p} &= -\frac{\partial U}{\partial x} + \sum_\alpha c_\alpha (q_\alpha - \frac{c_\alpha}{m_\alpha \omega_\alpha^2} x) \ , \end{aligned} \tag{2}$$

and for the bath degrees of freedom, respectively

$$\begin{aligned} m_\alpha \dot{q}_\alpha &= p_\alpha \\ \dot{p}_\alpha &= -m_\alpha \omega_\alpha^2 q_\alpha + c_\alpha x \ . \end{aligned} \tag{3}$$

Next we integrate the bath degrees of freedom, which obey first order ordinary, linear differential equations with an inhomogeneity $c_\alpha x(t)$. Considering the solution $x(t)$ as given, we formally solve (3) in terms of the Green's function for the bath oscillator, $\theta(t-s)\sin(\omega_\alpha(t-s))/\omega_\alpha$, i.e.,

$$\begin{aligned} q_\alpha(t) = q_\alpha(t_0)\cos(\omega_\alpha(t-t_0)) &+ \frac{p_\alpha(t_0)}{m_\alpha \omega_\alpha}\sin(\omega_\alpha(t-t_0)) \\ &+ \frac{c_\alpha}{m_\alpha \omega_\alpha}\int_{t_0}^t \mathrm{ds}\ \sin(\omega_\alpha(t-s))x(s) \ . \end{aligned} \tag{4}$$

Upon inserting (4) into (2) we find

$$\begin{aligned} \dot{p} = M\ddot{x} = -\frac{\partial U}{\partial x} &- \sum_\alpha \frac{c_\alpha^2}{m_\alpha \omega_\alpha^2} x(t) \\ &+ \sum_\alpha \frac{c_\alpha^2}{m_\alpha \omega_\alpha^2}\int_{t_0}^t \mathrm{ds}\ \omega_\alpha \sin(\omega_\alpha(t-s))x(s) \\ &+ \sum_\alpha c_\alpha \left[q_\alpha(t_0)\cos(\omega_\alpha(t-t_0)) + \frac{p_\alpha(t_0)}{m_\alpha \omega_\alpha}\sin(\omega_\alpha(t-t_0)) \right] \ . \end{aligned} \tag{5}$$

The last term will be denoted by $F(t)$. It does determine a fluctuating force. The second and the third term can be combined to read

$$\dot{p} = -\frac{\partial U}{\partial x} + \sum_\alpha \frac{c_\alpha^2}{m_\alpha \omega_\alpha^2}\left[\int_{t_0}^t \mathrm{ds}\ x(s)\frac{\partial}{\partial s}\cos(\omega_\alpha(t-s)) \ - \ x(t) \right] + F(t) \ . \tag{6}$$

With a partial integration we thus arrive at the GLE structure, i.e.,

$$\begin{aligned} \dot{p} = -\frac{\partial U}{\partial x} - \sum_\alpha \frac{c_\alpha^2}{m_\alpha \omega_\alpha^2} \Big[&\int_{t_0}^t \mathrm{ds}\ \dot{x}(s)\cos(\omega_\alpha(t-s)) \\ &+ x(t_0)\cos(\omega_\alpha(t-t_0)) \Big] + F(t) \ . \end{aligned} \tag{7}$$

Note the dependence on the initial value $x(t_0)$ in the second contribution. Hence, we have found already the exact result for the GLE, (Zwanzig 1973, Ford and Kac 1987):

$$M\ddot{x} + M\int_{t_0}^{t} \mathrm{ds}\ \gamma(t-s)\dot{x}(s) + \frac{\partial U}{\partial x} = -M\gamma(t-t_0)x(t_0) + F(t), \tag{8}$$

where the memory friction reads

$$\gamma(t-s) = \frac{1}{M}\sum_{\alpha}\frac{c_\alpha^2}{m_\alpha\omega_\alpha^2}\ \cos(\omega_\alpha(t-s)), \tag{9}$$

and

$$F(t) = \sum_{\alpha} c_\alpha \left[q_\alpha(t_0)\ \cos(\omega_\alpha(t-t_0)) + \frac{p_\alpha(t_0)}{m_\alpha\omega_\alpha}\sin(\omega_\alpha(t-t_0))\right] \tag{10}$$

is a colored (i.e. the noise has a finite correlation time) Gaussian fluctuating force, which obeys the fluctuation-dissipation theorem of the second kind, i.e.,

$$\begin{aligned}\langle F(t)\rangle_{\rho_B} &= 0\\ \langle F(t)\ F(s)\rangle_{\rho_B} &= MkT\gamma(t-s)\end{aligned} \tag{11}$$

where the average is taken with respect to the unperturbed bath, i.e.,

$$\rho_B = Z^{-1}\exp\left\{-\beta\left[\sum_{\alpha}\left(\frac{p_\alpha^2}{2m_\alpha} + \frac{m_\alpha\omega_\alpha^2}{2}\ q_\alpha^2\right)\right]\right\}, \tag{12}$$

and $\beta = (kT)^{-1}$.

Here, we already notice the role of the initial slip, $\gamma(t-t_0)x(t_0)$, see also in (Bez 1980, Canizares and Sols 1994), which usually is simply brushed under the rug, i.e. simply 'dropped'. For strict ohmic dissipation it reduces to a δ-function contribution, $2\gamma\delta(t-t_0)$: Although being zero for finite times t it still affects, of course, the time evolution of the realization, as well as the relaxation of time-dependent averages of the coordinate $x(t)$ and momentum $\dot{x}(t)$ processes, which indeed depend on all previous times $t \geq t_0$. The result in (8) is not yet the usual GLE, although it begins to look close. Next, we absorb the initial slip term into the stochastic force. Then, the noise

$$\begin{aligned}\zeta(t) &\equiv F(t) - M\ \gamma(t-t_0)x(t_0)\\ &= \sum_{\alpha} c_\alpha\left[\ \left(q_\alpha(t_0) - \frac{c_\alpha}{m_\alpha\omega_\alpha^2}x(t_0)\right)\cos(\omega_\alpha(t-t_0))\right.\\ &\qquad\left. + \frac{p_\alpha(t_0)}{m_\alpha\omega_\alpha}\sin(\omega_\alpha(t-t_0))\ \right]\end{aligned} \tag{13}$$

no longer has a stationary autocorrelation when averaged with the 'sudden' bath ensemble ρ_B. The combined stochastic force $\zeta(t)$ is again stationary and colored Gaussian, however, if conditionally averaged with respect to the Gaussian equilibrium ensemble

$$\begin{aligned} &\hat{\rho}(\{p_\alpha, q_\alpha\} \mid x(t_0) = x) \\ &= Z^{-1} \exp\left\{-\beta\left[\sum_\alpha \frac{p_\alpha^2}{2m_\alpha} + \frac{m_\alpha \omega_\alpha^2}{2}\left(q_\alpha - \frac{c_\alpha}{m_\alpha \omega_\alpha^2}\, x\right)^2\right]\right\}. \end{aligned} \tag{14}$$

With this conditional probability for the bath variables the statistical force $\zeta(t)$ obeys the identical fluctuation-dissipation relations as in eq. (11), i.e.,

$$\begin{aligned} \langle \zeta(t) \rangle_{\hat{\rho}} &= 0 \\ \langle \zeta(t)\, \zeta(s) \rangle_{\hat{\rho}} = \langle \zeta(t-s+t_0)\, \zeta(t_0) \rangle_{\hat{\rho}} &= MkT\gamma(t-s)\;. \end{aligned} \tag{15}$$

Hence, care must be applied if one refers to the statistical properties of the two different stochastic forces $F(t)$ and $\zeta(t)$, respectively! Our exercise elucidates that this exact GLE does not pop out of Pandoras box, but can be obtained in a straightforward manner. Nevertheless, it needed a master like Zwanzig (1973), who paved the way for us. The above derivation is worth to be commented on further: Note that the steps in (2) - (7) carry through for the corresponding Heisenberg equations of motion as well. This procedure yields the exact (operator)-GLE for the quantum case, see in (Ford and Kac 1987). Interestingly enough, with $U(x)$ a harmonic oscillator potential and without the 'counter-term', i.e. without the second term in (5) so that $U(x)$ undergoes a potential renormalization in the GLE, the quantum-GLE together with a discussion of the corresponding quantum Nyquist formula has already been presented clearly – on one and a quarter page – by Magalinskij as early as (1959). In the quantum case the stochastic forces are no longer c-numbers, but become operators which act on the Hilbert space of all bath degrees of freedom (and composite Hilbert space of system plus bath for the combined stochastic force $\zeta(t)$). The symmetrized autocorrelation of the stochastic force(s) obey a quantum generalization of the fluctuation-dissipation theorem of the second kind (quantum Nyquist theorem); i.e., the symmetric quantum noise autocorrelation can be expressed solely in terms of the macroscopic memory friction kernel $\gamma(t-s)$; not withstanding recent incorrect claims to the contrary (Cortes et al. 1985).

Next, let me return to classical statistical mechanics. As already demonstrated by Zwanzig (1973), the above GLE can be generalized to a nonlinear system-bath coupling, i.e. we set for the coupling, $-c_\alpha q_\alpha x \longrightarrow -c_\alpha q_\alpha G(x)$, in (1). The bath motions are then still linear; thus we again can formally integrate the bath degrees of freedom, thereby generalizing the result in (4). With $\frac{dG(x)}{dx} \equiv g(x)$ the exact GLE for this nonlinear system-bath interaction is then evaluated to give the following GLE

$$M\ddot{x} + Mg(x(t)) \int_{t_0}^{t} \mathrm{ds}\, \gamma(t-s) g(x(s))\, \dot{x}(s) \; + \; \frac{\partial U}{\partial x} = \\ -M\gamma(t-t_0)\, g(x(t_0))\, G(x(t_0)) + g(x(t))F(t) \; . \qquad (16)$$

With $G(x) = x$ it reduces to the previous result in (8). This nonlinear GLE exhibits multiplicative noise with a generalized, state-dependent memory friction. The noise $F(t)$ and the combined fluctuating force, $F(t) - M\gamma(t - t_0)G(x(t_0))$, then obey the corresponding relations in (11) and (15), respectively. An unsolved problem, to the best of my knowledge, is the detailed connection between the GLE in (8,16), with the stochastic force given either by (11), or by (15), and the corresponding non-Markovian GME for the joint probability density of coordinate and momentum variable; i.e.

$$\dot{p}(x, \dot{x}; t) = \ldots ? \qquad (17)$$

Although the noise is Gaussian, the joint-process $[(x(t), \dot{x}(t)]$ is with a (non-quadratic) nonlinear potential no longer Gaussian. Then the difficulty in obtaining the GME is rooted in the fact that its derivation requires the knowledge of corresponding functional derivatives, (Hänggi 1978,1989), such as $(\delta\dot{x}(t)/\delta\, \zeta(s))$ at times $s \leq t$; hence, the GME is at best known formally only. With $U(x)$ being at most a quadratic function of x, the process pair $[x(t), \dot{x}(t)]$, given $x(t_0) = x_0, \dot{x}(t_0) = v_0$ can be shown to be Gaussian, but non-Markovian. Then, a master equation for $\dot{p}(x, \dot{x}; t)$ for a stable process, i.e. $U''(x) \geq 0$, and an initial equilibrium preparation (Adelman 1976) or, more generally, with an initial *non*-equilibrium preparation (Hänggi 1978,1989), as well as for an unstable process with $U''(x) \leq 0$, see in (Hänggi and Mojtabai 1982), can readily be constructed. It exhibits a *time-convolutionless* structure. Thus, it clearly does not have the retarded (memory) structure predicted by the (formal) projection operator methodology (Grabert et al. 1980, Grabert 1982). What is the corresponding memory-GME in this linear case? – I simply do not know. –

Sailing becomes much smoother in the Markovian limit, with $\zeta(t)$ approaching stationary Gaussian white noise. Given an arbitrary potential $U(x)$, the corresponding connection is the well-known relationship between the Langevin equation and the Fokker-Planck equation, i.e. the Klein-Kramers equation, (Klein 1922, Kramers 1940), for which, even for the case that a spatially-dependent friction coefficient governs the inertia motion, no Ito-Stratonovich dilemma occurs. Another challenge presents the task of a microscopic modelling of fluctuations in a general nonequilibrium system such as e.g. a thermal ratchet system (for a recent review see Hänggi and Bartussek 1996). Now, matters become even worse. For example, given the fact that the detailed balance symmetry of the total system (i.e. system plus all degrees of freedom of all baths) survives a coarse graining operation, see eq. (4.3.8) in (Hänggi and Thomas 1982), it is, *prima facie*, not obvious how the violation of the detailed balance symmetry for a stationary nonequilibrium system does occur. For an elucidating discussion of this point the reader is referred to page 267 in (Hänggi and Thomas 1982).

To gain further insight, we consider a single particle S which is coupled simultaneously (!) in a bilinear way, cf. (1), to two harmonic baths at different temperatures, with the baths composed of a large number of oscillators. This situation distinctly differs from the Carnot machine case, where the two baths never influence the system at the same time. Related in this spirit are the theoretical models of heat conduction in harmonic chains whose ends are brought into contact with independent heat baths, which have been modelled phenomelogically by two independent Gaussian noise forces together with ohmic friction for the end points, as studied classically by Rieder, Lebowitz and Lieb (1967), and quantum mechanically by Zürcher and Talkner (1990). We next assume that two baths are initially prepared in a thermal product state at the individual temperature T_1 and T_2, respectively. Then, the resulting exact GLE for this situation has the same form as in (8), but with two memory kernels, and two stochastic forces of the type in (10), obeying corresponding statistical properties as given in (11). Most important is the observation that two different initial slip terms of the form in (8) appear. Likewise, one can introduce two different stochastic forces of the type in (13).

By forming cross-correlations, or by combining them into a single stochastic force we encounter, however, difficulties: What β-value should be chosen for the conditional, longtime equilibrium ensemble in (14)? Although the two baths are not coupled between each other, they are in fact in contact via their finite coupling to the system S. Thus, it is the (relative) heat capacities of the two baths that ultimately determine the overall equilibrium temperature. Moreover, the two initial slip terms couple, via the dependence on $x(t_0)$, the two stochastic forces $\zeta_1(t)$ and $\zeta_2(t)$. I am not aware that these subtle aspects have formerly been discussed in the literature. Previous attempts, a most recent can be found in (Millonas 1995), can be shown to be inconsistent (neglect of initial slips, and mix up with different ensemble averages, etc.).

Playing naive, let us discuss the simplest situation, namely we use the Markovian limits, i.e. $\gamma(t-s) \longrightarrow 2\gamma\delta(t-s)$, for the stochastic forces $F_1(t)$ and $F_2(t)$ of the two baths at temperature T_1 and T_2, respectively. Further, we simply neglect corresponding initial slips such that $F_1(t) = \zeta_1(t), F_2(t) = \zeta_2(t)$. Note that the equality does not hold when $t = t_0$. Then, the autocorrelation for the force $\zeta_1(t)$ reads

$$\langle \zeta_1(t)\, \zeta_1(s) \rangle = 2M\gamma\, T_1\, \delta(t-s). \tag{18}$$

With $\gamma_2 = \gamma_1 = \gamma$, the stochastic force corresponding to the bath at temperature T_2 obeys

$$\langle \zeta_2(t)\, \zeta_2(s) \rangle = 2M\gamma\, T_2\, \delta(t-s)\ . \tag{19}$$

Next, we intuitively set

$$\langle \zeta_1(t)\, \zeta_2(s) \rangle = 0\ . \tag{20}$$

Moreover, we use a vanishing potential field $U(x) = 0$. With $v = \dot{x}$, the Langevin equation which governs the relaxation dynamics is then expected to be of the form,

$$M\dot{v} = -2\gamma\, v + \zeta_1(t) + \zeta_2(t). \tag{21}$$

With the equilibrium temperature being $T \equiv \frac{1}{2}(T_1 + T_2)$, the corresponding Fokker-Planck equation for the nonequilibrium situation, with the two baths at different temperatures $T_1 \neq T_2$ can then formally be recast by the stochastically equivalent Langevin equation of a single bath at temperature T; namely

$$M\dot{v} = -2\gamma\, v + \zeta(t) \tag{22}$$

with $\zeta(t)$ being a single white Gaussian noise, obeying

$$\langle \zeta(t)\ \zeta(s)\rangle = 2M\,(2\gamma)T\delta(t-s). \tag{23}$$

Both equations (21), and (22) with (23), indeed yield the identical Fokker-Planck equation

$$\dot{p}(v,t) = -\frac{\partial}{\partial\, v}\left[-2\gamma v p(v,t)\right] + 2M\gamma\, T\frac{\partial^2}{\partial\, v^2}\, p(v,t). \tag{24}$$

What does this result imply: Is the nonequilibrium relaxation dynamics of $v(t)$, given two harmonic baths at temperature T_1 and T_2, at all later times t identical to the equilibrium relaxation dynamics of $v(t)$ in contact with a single bath at $T = \frac{1}{2}(T_1 + T_2)$? Clearly, the answer must be 'no'. Thus, we must have made mistakes for this most simple situation already, such as e.g. mixing different averages in eqs. $(18 - 23)$.

The Langevin equation in (21) and (22) should describe different physical situations at finite times $t > t_0$; in the longtime limit, $t_0 \longrightarrow -\infty$, however, these differences vanish and both equations describe the same equilibrium Boltzmann distribution for the momentum variable v. In this sense, the two Langevin equations become equivalent in the asymptotic longtime limit; however, (21) no longer, of course, can be used to describe the nonequilibrium relaxation of averages towards asymptotic equilibrium.

The generalization with quantum mechanics taken fully into account is even more subtle. Then, as has been indicated above, an exact (operator) GLE – which solely acts on the Hilbert space of the system S alone – does simply not exist.

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