

# Generalized Langevin Equation and fluctuation-dissipation theorem for particle-bath systems in external oscillating fields

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The Generalized Langevin Equation (GLE) can be derived from a particle-bath Hamiltonian, in both classical and quantum dynamics, and provides a route to the (both Markovian and non-Markovian) fluctuation-dissipation theorem (FDT). All previous studies have focused either on particle-bath systems with time-independent external forces only, or on the simplified case where only the tagged particle is subject to the external time-dependent oscillatory field. Here we extend the GLE and the corresponding FDT for the more general case where both the tagged particle and the bath oscillators respond to an external oscillatory field. This is the example of a charged or polarisable particle immersed in a bath of other particles that are also charged or polarizable, under an external AC electric field. For this Hamiltonian, we find that the ensemble average of the stochastic force is not zero, but proportional to the AC field. The associated FDT reads as  $\langle F_P(t)F_P(t') \rangle = mk_B T \nu(t-t') + (\gamma e)^2 E(t)E(t')$ , where  $F_P$  is the random force,  $\nu(t-t')$  is the friction memory function, and  $\gamma$  is a numerical prefactor.

The theory of Brownian motion is so far the simplest approximation to the dynamics of nonequilibrium systems. The Langevin equation, and its equivalent counterpart, the Smoluchowski diffusion equation, describe the motion of a Brownian particle in an external force field and under the action of thermal agitation from the bath of solvent molecules, in which the Brownian particle is immersed. At steady-state, the thermal fluctuations that cause rapid changes in the particle velocity are dissipated by viscous drag.

This is a manifestation of the fluctuation-dissipation theorem (FDT), as was originally formulated in different contexts by Einstein and by Nyquist, and generalized by Onsager [1] and by Callen and Welton [2]. Later, the theorem has been further elaborated in many different contexts [3–12]. FDT stipulates that the response of a system in thermodynamic equilibrium to a small applied force is the same as its response to a spontaneous fluctuation, thus connecting the linear response relaxation of a system to equilibrium, from a prepared nonequilibrium state, with its statistical fluctuation properties in equilibrium. FDT applies to both classical and quantum mechanical systems [13, 14] and has been generalized to non-Markovian processes for classical systems by Zwanzig [15]. In the latter case, the noise is no longer uncorrelated in time, and the time correlation of the stochastic force is dictated by the time correlation of the friction which plays the role of the memory function in the Generalized Langevin Equation (GLE). The non-Markovianity arises from the dynamical coupling of the tagged Brownian particle with many particles (harmonic oscillators) forming the heat bath. This coupling, which in physical systems may be provided by long-range molecular interactions, is thus responsible for both the thermal agitation and

the damping experienced by the tagged particle.

All versions of the GLE, and of the associated FDT which can be derived from it, that have been considered in the past are limited to either systems in the absence of external time-dependent forces or, if an external time-dependent force is present, its action is restricted to the tagged Brownian particle, leaving the bath oscillators unaffected by the external field [5, 16]. Harada and Sasa considered time-dependent driving forces being exerted on the tagged particle and they found a violation of the fluctuation-response relation in driven nonequilibrium systems [17, 18]. More recently, Maes and co-workers derived FDT for nonequilibrium systems by implementing mutual interactions between bath particles and also the effect of stochastic white-noise force on the bath particles dynamics [19, 20].

This limitation is obviously artificial and not realistic, because in many systems there is no reason to justify why the tagged particle is subjected to the external AC field whereas the bath oscillators remain unaffected by the field. This limitation clearly leaves out a number of important physical problems, where not only the tagged particle is subjected to the AC field, but also the particles that constitute the heat bath are subjected to it. This situation is clearly encountered in dielectric matter under a uniform AC electric field  $E(t)$ . In this case, every building block (atom, molecule, ion) is polarisable or charged such that it is subjected to a force from the electric field. Hence, if the bath is constituted by polarisable or charged particles, these will also respond to the AC field and it is unphysical to neglect the effect of the AC field on the dynamics of the bath oscillators. This situation is schematically depicted in Fig. 1.

A similar situation arises in the field of microrheology [21] where not only the probe Brownian particle, but

also the building-blocks of the medium do respond to the oscillatory mechanical field.

In this contribution, we provide a solution to this problem by formulating a Caldeira-Leggett particle-bath Hamiltonian where both the particle and the bath oscillators are subjected to the external AC electric field, which is explicitly accounted for in both the Hamiltonian of the particle and the Hamiltonian of the bath. The two Hamiltonians are then connected via a bi-linear coupling as is standard in this type of models. We analytically solve the coupled Hamiltonian for the tagged particle to find a new Generalized Langevin Equation, which, for the first time, accounts for the effect of the polarization of the bath under the AC field on the dynamics of the tagged particle.

We also derive the associated FDT and find a surprising result: the time-correlation of the stochastic force is not just equal to the memory function for the friction, but there is an additional term which is proportional to the time-correlation of the AC field, and hence to the amplitude of the AC field squared. This term is absent in all previously studied versions of the FDT. This result shows that the strength of the fluctuating force can be controlled by the external force field in the limit of sufficiently strong external driving. This framework opens new avenues to understanding the macroscopic response of complex liquids, plasmas, colloids and other condensed matter under oscillatory fields.

We study the classical version of the Caldeira-Leggett coupling between the tagged particle and a bath of harmonic oscillators, which was actually proposed already in 1973 by Zwanzig [15], and add a new term,  $H_E$ , which contains the force due to the applied AC electric field acting on both the tagged particle and the harmonic oscillators:

$$H = H_S + H_B + H_E \quad (1)$$

where  $H_S = P^2/2m + V(Q)$  is the Hamiltonian of the tagged particle without external field. The second term is the standard Hamiltonian of the bath of harmonic oscillators that are coupled to the tagged particle,  $H_B = \frac{1}{2} \sum_{\alpha=1}^N \left[ \frac{P_{\alpha}^2}{m_{\alpha}} + m_{\alpha} \omega_{\alpha}^2 \left( X_{\alpha} - \frac{F_{\alpha}(Q)}{\omega_{\alpha}^2} \right)^2 \right]$ , consisting of the standard harmonic oscillator expression for each bath oscillator  $\alpha$ , and of the coupling term between the tagged particle and the  $\alpha$ -th bath oscillator, which contains the coupling function  $F_{\alpha}(Q)$ .

The new term

$$\begin{aligned} H_E &= -ze\Phi(Q, t) - \sum_{\alpha} e_{\alpha}\Phi(X_{\alpha}, t) \\ &= -E_0 \sin(\omega t) (zeQ + \sum_{\alpha} e_{\alpha}X_{\alpha}) \end{aligned} \quad (2)$$

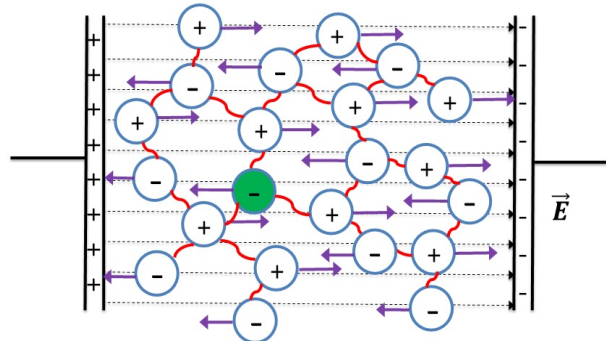


FIG. 1. Schematic example of system of charged (solid circles) or polarisable molecules. In the former case the particles could be ions in a plasma or ions and electrons in a liquid metal. In the latter case, pairs of negatively and positively charged particles represent the electron cloud and the molecular ion of a polarised neutral molecule as in e.g. dielectric relaxation of molecular liquids. A particle-bath Hamiltonian like the one of Eqs.(1-2) can be applied to these systems where a tagged particle (coloured, green) interacts with the local environment via an interaction potential  $V(Q)$ , which may represent the interaction potential with neighbours. The tagged particle interacts also with all other degrees of freedom in the system which can be effectively represented as a bath of harmonic oscillators to which the tagged particle is coupled via a set of coupling constants  $c_{\alpha}$ , where  $\alpha$  runs over all other bath oscillators in the system. In traditional models of bath-oscillator dynamics, only the tagged particle is subjected to the external AC field, see e.g. [5], whereas the other particles are not. In the proposed model, both the tagged particle and also all the other particles (forming the bath) are responding to the AC electric field, which leads to new physics.

represents the influence of electric field on both the tagged particle (first term on the RHS) and the bath oscillators (second term on the RHS). Here,  $e$  is the unit charge and hence  $ze$  is the total charge of tagged particle, where e.g.  $z = \pm 1$  for monovalent ions/electrons in a plasma or  $z = -1$  for the case of molecular dielectrics where a molecule polarizes into a negatively charged electron cloud which oscillates about a positively charged molecular ion;  $e_{\alpha}$  is the net charge of bath particle  $\alpha$  which is subjected to the same polarization. Here, we only consider motion along the direction of electric field.

In the Caldeira-Leggett Hamiltonian, the coupling function is taken to be linear in the particle coordinate  $Q$ ,  $F_{\alpha}(Q) = c_{\alpha}Q$ , where  $c_{\alpha}$  is known as the strength of coupling between the tagged atom and the  $\alpha$ -th bath oscillator.

This Hamiltonian leads in a straightforward manner to the following system of differential equations:

$$\begin{aligned} \frac{dQ}{dt} &= \frac{P}{m}; \quad \frac{dP}{dt} = -V'(Q) + \sum_{\alpha} m_{\alpha} c_{\alpha} (X_{\alpha} - \frac{c_{\alpha} Q}{\omega_{\alpha}^2}) + zeE_0 \sin(\omega t) \\ \frac{dX_{\alpha}}{dt} &= \frac{P_{\alpha}}{m_{\alpha}}; \quad \frac{dP_{\alpha}}{dt} = -m_{\alpha} \omega_{\alpha}^2 X_{\alpha} + m_{\alpha} c_{\alpha} Q + e_{\alpha} E_0 \sin(\omega t) \end{aligned} \quad (3)$$

From the second line, upon solving the second-order inhomogeneous ODE with the Green's function method (or alternatively with the Wronskian method), we get

$$\begin{aligned} X_{\alpha}(t) &= X_{\alpha}(0) \cos(\omega_{\alpha} t) + \frac{P_{\alpha}(0) \sin(\omega_{\alpha} t)}{m_{\alpha} \omega_{\alpha}} \\ &+ \int_0^t [c_{\alpha} Q(s) + \frac{e_{\alpha}}{m_{\alpha}} E_0 \sin(\omega s)] \frac{\sin(\omega_{\alpha}(t-s))}{\omega_{\alpha}} ds \end{aligned} \quad (4)$$

The integral  $\int_0^t c_{\alpha} Q(s) \frac{\sin(\omega_{\alpha}(t-s))}{\omega_{\alpha}} ds$  can be evaluated via integration by parts. Upon further denoting  $E_{\alpha}(t) =$

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$\int_0^t e_{\alpha} E_0 \sin(\omega s) \frac{\sin(\omega_{\alpha}(t-s))}{\omega_{\alpha}} ds$ , we obtain

$$\begin{aligned} X_{\alpha}(t) - \frac{c_{\alpha} Q(t)}{\omega_{\alpha}^2} &= \left( X_{\alpha}(0) - \frac{c_{\alpha} Q(0)}{\omega_{\alpha}^2} \right) \cos(\omega_{\alpha} t) \\ &+ P_{\alpha}(0) \frac{\sin(\omega_{\alpha} t)}{m_{\alpha} \omega_{\alpha}} - \int_0^t \frac{c_{\alpha} P(s) \cos(\omega_{\alpha}(t-s))}{m \omega_{\alpha}^2} ds + \frac{E_{\alpha}(t)}{m_{\alpha}} \end{aligned} \quad (5)$$

Substituting Eq. (5) into the equation for  $P(t)$  in Eq. (3), we derive the following Generalized Langevin Equation for the tagged particle motion, which accounts for the AC electric field acting on both the tagged particle and the bath oscillators:

$$\begin{aligned} \frac{dP}{dt} &= -V'(Q(t)) - \sum_{\alpha} \int_0^t \frac{m_{\alpha} \cos(\omega_{\alpha}(t-s))}{m \omega_{\alpha}^2} c_{\alpha}^2 P(s) ds + zeE_0 \sin(\omega t) \\ &+ \sum_{\alpha} \left\{ m_{\alpha} c_{\alpha} \left[ X_{\alpha}(0) - \frac{c_{\alpha} Q(0)}{\omega_{\alpha}^2} \right] \cos(\omega_{\alpha} t) + c_{\alpha} P_{\alpha}(0) \frac{\sin(\omega_{\alpha} t)}{\omega_{\alpha}} + c_{\alpha} E_{\alpha}(t) \right\} \\ &= -V'(Q(t)) - \int_0^t \nu(s) \frac{m_{\alpha} P(t-s)}{m} ds + zeE_0 \sin(\omega t) + F_P(t). \end{aligned} \quad (6)$$

We have introduced the noise or stochastic force  $F_P(t)$  which is equal to the second line after the first equality,

$$\begin{aligned} F_P(t) &= \sum_{\alpha} \left\{ m_{\alpha} c_{\alpha} \left[ X_{\alpha}(0) - \frac{c_{\alpha} Q(0)}{\omega_{\alpha}^2} \right] \cos(\omega_{\alpha} t) \right. \\ &\left. + c_{\alpha} P_{\alpha}(0) \frac{\sin(\omega_{\alpha} t)}{\omega_{\alpha}} + c_{\alpha} E_{\alpha}(t) \right\}. \end{aligned} \quad (7)$$

One should note that this expression for the stochastic force is identical with the one derived by Zwanzig for a particle-bath system without external field, except for the important term  $c_{\alpha} E_{\alpha}(t)$ , which is new and contains the effect of the external AC field on the bath oscillators dynamics. This is a crucial point because the dynamical response of the bath oscillators to the external AC field in turn produces a modification of spectral properties of the bath fluctuations, and thus leads to a new form of the stochastic force which is different from those studied in previous works.

The memory function for the friction  $\nu(t) =$

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$\sum_{\alpha} \frac{m_{\alpha} c_{\alpha}^2}{m \omega_{\alpha}^2} \cos(\omega_{\alpha} t)$  is identical to the memory function of systems without external time-dependent forces such as the one derived by Zwanzig [15].

The integral in the function  $E_{\alpha}(t) = \int_0^t e_{\alpha} E_0 \sin(\omega s) \frac{\sin(\omega_{\alpha}(t-s))}{\omega_{\alpha}} ds$ , can be evaluated using trigonometric identities which leads to

$$E_{\alpha}(t) = \frac{e_{\alpha} E_0 (\omega \sin(\omega_{\alpha} t) - \omega_{\alpha} \sin(\omega t))}{\omega_{\alpha} (\omega^2 - \omega_{\alpha}^2)}. \quad (8)$$

As for the case without external time-dependent fields, we find that our  $F_P(t)$  is defined in terms of initial positions and momenta of bath oscillators, but in our case it now also depends on the sinusoidal electric field at time  $t$ . Note that, by shifting the time origin, it can be easily verified that the statistical average is stationary. Following Zwanzig [15], we assume the initial conditions for the bath oscillators can be taken to be Boltzmann-distributed  $\sim \exp(-H_B/k_B T)$ , where the bath is in thermal equilibrium with respect to a frozen or constrained system coordinate  $X(0)$ .

Then for the average of  $X$  and  $P$  we find:

$$\langle X_\alpha(0) - \frac{c_\alpha Q(0)}{\omega_\alpha^2} \rangle = 0, \quad \langle P_\alpha(0) \rangle = 0. \quad (9)$$

The second moments are

$$\left\langle \left( X_\alpha(0) - \frac{c_\alpha Q(0)}{\omega_\alpha^2} \right)^2 \right\rangle = \frac{k_B T}{m_\alpha \omega_\alpha^2}, \quad \langle P_\alpha(0)^2 \rangle = m_\alpha k_B T. \quad (10)$$

Both these results are consistent with what one finds for systems without external time-dependent fields, since obviously they descend directly from the assumption of Boltzmann-distributed degrees of freedom at the initial time.

As for  $\sum_\alpha c_\alpha E_\alpha(t)$ , we first note that there is no singularity when  $\omega_\alpha \rightarrow \omega$  and  $\omega_\alpha \rightarrow 0$ :

$$\begin{aligned} \lim_{\omega_\alpha \rightarrow \omega} \frac{\omega \sin(\omega_\alpha t) - \omega_\alpha \sin(\omega t)}{\omega_\alpha(\omega^2 - \omega_\alpha^2)} &= -\frac{\omega t \cos(\omega t) - \sin(\omega t)}{2\omega^2} \\ \lim_{\omega_\alpha \rightarrow 0} \frac{\omega \sin(\omega_\alpha t) - \omega_\alpha \sin(\omega t)}{\omega_\alpha(\omega^2 - \omega_\alpha^2)} &= \frac{\omega t - \sin(\omega t)}{\omega^2}. \end{aligned} \quad (11)$$

Focusing on ions, atoms or molecules,  $\omega_\alpha$  is at least in the THz regime or higher, which is orders of mag-

nitude larger than the frequency of the applied field  $\omega$  (this assumption of course may not hold for THz spectroscopy, which deserves a separate treatment in future work). Hence, the first term in the numerator in the RHS of Eq. (8) can be neglected along with the first term in the bracket in the denominator, and we can approximate as follows:

$$\sum_\alpha c_\alpha E_\alpha(t) \approx \sum_\alpha \frac{c_\alpha e_\alpha}{\omega_\alpha^2} E_0 \sin(\omega t) \propto eE(t). \quad (12)$$

We now take the Boltzmann average of the stochastic force, Eq. (7), and we find:

$$\langle F_P(t) \rangle = \gamma e E(t), \quad (13)$$

for some constant  $\gamma$ . That is, the average of the stochastic force  $F_P$  is not zero, unlike in all previously studied GLEs of particle-bath systems and, remarkably, is directly proportional to the AC field.

Now, by direct calculation, using Eqs. (9) and (10) and standard trigonometric identities, we can get the fluctuation-dissipation theorem (FDT) for our particle-bath Hamiltonian under a uniform AC field:

$$\begin{aligned} \langle F_P(t) F_P(t') \rangle &= \frac{1}{Z_N} \int F_P(t) F_P(t') \exp\left(-\frac{H_B}{k_B T}\right) d\mathbf{X}(0) d\mathbf{P}(0) \\ &= \sum_\alpha \left( m_\alpha c_\alpha^2 \frac{k_B T}{\omega_\alpha^2} \cos(\omega_\alpha t) \cos(\omega_\alpha t') + m_\alpha c_\alpha^2 \frac{k_B T}{\omega_\alpha^2} \sin(\omega_\alpha t) \sin(\omega_\alpha t') \right) + (\gamma e)^2 E(t) E(t') \\ &= k_B T \sum_\alpha \frac{m_\alpha c_\alpha^2}{\omega_\alpha^2} \cos(\omega_\alpha(t-t')) + (\gamma e)^2 E(t) E(t') \\ &= m k_B T \nu(t-t') + (\gamma e)^2 E(t) E(t') \end{aligned} \quad (14)$$

where  $Z_N$  is the canonical partition function

$$Z_N = \int \exp\left(-\frac{H_B}{k_B T}\right) d\mathbf{X}(0) d\mathbf{P}(0) \quad (15)$$

and  $\mathbf{X}(0) = \{X_1(0), X_2(0), \dots\}$ ,  $\mathbf{P}(0) = \{P_1(0), P_2(0), \dots\}$ .

Equation (14) is a key result of this paper and is the FDT associated with the GLE given by Eq. (6). This is a remarkable result which shows that in the presence of an external AC field which affects the microscopic dynamics of both the tagged particle and the bath oscillators, the strength of the noise is no longer proportional to thermal energy only, but also has an important deterministic contribution proportional to the AC field amplitude squared.

The implications of this result for statistical mechanics and its applications in condensed matter are vast and

profound and will be explored in future works, including experimental verifications and extension to nonequilibrium environments [17–20]. We can however notice that the presence of the external AC field makes it impossible for the noise to be white noise. This is true even if the memory function is delta-correlated, because of the second term controlled by the AC field in Eq. (14), which inevitably introduces a deterministic temperature-independent correlation into the noise.

In conclusion, we have introduced a more general version of the classical particle-bath Hamiltonian, which is used as a starting point to derive Generalized Langevin Equations, for systems subjected to an external time-dependent (oscillating) field. Unlike in previous models where the bath oscillators were always taken to be unaffected by the external field, here we added the time-

dependent force due to the field to both the Hamiltonian of the particle and the Hamiltonian of the bath oscillators. The resulting Hamiltonian has been solved analytically and the resulting GLE and fluctuation-dissipation theorem have been found. The formal structure of the GLE is still formally identical to that of standard GLE with external field acting on the particle only [5] (and the memory function for the friction is the same), but the stochastic force is very different. Its ensemble average is non-zero and directly proportional to the AC field. The associated fluctuation-dissipation theorem, remarkably, has an additional term given by the time-correlation of the AC field, and is thus quadratic in the field amplitude.

An immediate application of this result is to dielectric relaxation and dielectric spectroscopy of liquids and glasses. The Debye model treats each molecule as fully independent from all other molecules in the material and describes it with a Langevin equation for the orientation of the molecule in the field [22, 23]. More refined models, e.g. Mode-Coupling Theory, are able to account for the coupling of each molecule to collective degrees of freedom [24, 25], but do not resolve the combined effect of the AC field on the dynamics of both the single molecule and the other molecules which provide the memory effect in dielectric relaxation. The GLE derived here will open the possibility of describing both these effects at the same time, within the GLE picture that has been proposed more recently for the dielectric response of liquids and glasses [26–28]. Furthermore, the GLE derived here can be used as the starting point for a more microscopic description of nonlinear effects in dielectric relaxation of supercooled liquids under strong fields [29, 30].

In the context of microrheology of soft and biological matter, the FDT is the starting point to derive the key relationship (generalized Stokes-Einstein relation, GSE) that is used to infer the viscoelastic response of the medium to an oscillatory mechanical field from the optically monitored dynamics of a probe particle [21, 31–34]. This method based on FDT has found widespread application in biophysics, to measure the viscoelasticity of cytoskeleton and other biological matters. However, all these works, including the most recent ones (see e.g. Ref. [35]) always neglect the effect of the oscillatory field on the dynamics of the medium (the bath). Clearly, the system as a whole is composed of many building blocks (molecules, filaments) which also respond dynamically to the oscillatory shear field and behave as the bath for the probe particle. Therefore, the same effect described in our paper is certainly present also in all those systems, and yet it has always been neglected in all the literature so far.

Further applications of the proposed framework include quantum dissipative transport and Josephson tunnelling with dissipation [16, 36, 37], driven dynamics of colloids in soft matter systems [38–40], molecular dynam-

ics simulations of liquids [41] and of amorphous solids in oscillatory shear [42].

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