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ABSTRACT

Five requirements are proposed which should be satisfied by any formalism attempting to calculate approximately a nonlinear response solely from a knowledge of the external field and the equilibrium fluctuations of the degree of freedom of interest. One possible approximate nonlinear response theory is then derived and discussed; it satisfies four of the five requirements while a fifth cannot be checked.

Linear response theory [1-7] was derived many years ago and is today a standard tool used in many parts of physics. The nonlinear generalization [6,8,9] is complex and not very useful, however. The problem is that, in the expansion of the response in terms of the external field E , the coefficients cannot be expressed in terms of equilibrium fluctuations of the degree of freedom, Q , that couples to the field [10]. The best one can hope for is an approximate theory that estimates the nonlinear response from equilibrium fluctuations. There is another problem which must be faced. In a non-infinitesimal field the dissipated heat cannot be ignored and the response depends on the way this heat is removed. A general formalism that estimates the response from equilibrium fluctuations does not take this into account and should be applied only at fields small enough to ensure that the temperature is almost constant throughout the sample. In many cases this condition is fulfilled for sufficiently small samples. Note that some important nonlinear phenomena actually do take place at a very small dissipation rate. An example is the flow of a polymeric liquid. It is an empirical fact [11,12] that this flow becomes nonlinear at a shear rate of order τ_M^{-1} where τ_M is the (linear) Maxwell relaxation time. Thus, if G_∞ denotes the infinite frequency shear modulus and η is the linear viscosity, the dissipation rate per unit volume is of the order G_∞^2/η at the onset of nonlinearity. For large viscosity this quantity is very small and the temperature is almost constant throughout even a relatively large sample.

In this paper five requirements are proposed which an approximate nonlinear response theory should reasonably satisfy.

Then a maximum entropy based response theory is briefly discussed, followed by the development of a more general formalism. To avoid the complications of quantum mechanics, the underlying dynamics is assumed to be classical throughout. For simplicity, it is also assumed that Q does not change sign under time-reversal.

An approximate nonlinear response theory should, by the above definition, (1), calculate the response solely from a knowledge of the external field and of the equilibrium fluctuations of $Q(t)$. The theory should, (2), satisfy causality and, (3), reflect the time-reversal invariance of the underlying equation of motion. Points (2) and (3) ensure any approximate nonlinear response theory is an extension of linear response theory. As shown by Bochkov and Kuzovlev [13,14], time-reversal invariance is conveniently expressed in terms of the "path probability" $P_{E(\tau)}[\dot{Q}(t)]$ (the probability of a $\dot{Q}(t)$ -fluctuation in the external field $E(\tau)$), as

$$\frac{P_{E(\tau)}[\dot{Q}(t)]}{P_{E(-\tau)}[-\dot{Q}(-t)]} = \exp \left\{ \beta \int_{-\infty}^{\infty} E(t) \dot{Q}(t) dt \right\} \quad (1)$$

where β is the inverse temperature. Equation (1) is derived by assuming thermal equilibrium at $t = -\infty$ followed by a decoupling of the heat bath and subsequent evolution in time according to the canonical equations of motion for the system interacting with the external field. The requirements (1)-(3) are sine qua non for any approximate nonlinear response theory, but there are further requirements which such a theory should reasonably satisfy. In statistical mechanics the occurrence of nonlinearities always correlates to the existence of non-Gaussian equilibrium fluctua-

tions. For instance, a magnetic system has a field-independent susceptibility if and only if the magnetization fluctuations in equilibrium are Gaussian. In analogy to statistical mechanics it is reasonable to require that, (4), any approximate nonlinear response theory should predict an exactly linear response whenever the equilibrium $Q(t)$ -fluctuations are Gaussian. Finally, there is a requirement which is valid also for linear response theory, (5): If Q_1 and Q_2 fluctuate independently in equilibrium, their response should be uncorrelated. Thus, if Q_1 and Q_2 are coupled to two different fields, the field E_1 should not influence the Q_2 -response and vice versa. The case when the same field couples to Q_1 and Q_2 is important. Then requirement (5) ensures consistency for the response of a bulk degree of freedom, Q , since Q may be written $Q=Q_1+Q_2$ where Q_1 and Q_2 refer to different volumes of the system and therefore fluctuate independently.

The simplest solution of Eq. (1) is the "maximum entropy ansatz" [15]

$$P_{E(\tau)}[\dot{Q}(t)] = N^{-1} P_0[\dot{Q}(t)] \exp\left\{\beta/2 \int_{-\infty}^{\infty} E(t) \dot{Q}(t) dt\right\} \quad (2)$$

where $P_0[\dot{Q}(t)]$ is the equilibrium path probability and N is a normalization constant. Equation (2) obviously violates causality. Therefore Eq. (2) makes sense only for constant fields. In this case the response is given by [15]

$$\langle \dot{Q} \rangle_E = \sum_{n=1}^{\infty} \frac{1}{n!} \left(\frac{\beta E}{2}\right)^n \int_{-\infty}^{\infty} dt_1 \cdots dt_n \langle \dot{Q}(0), \dot{Q}(t_1), \cdots, \dot{Q}(t_n) \rangle_0 \quad (3)$$

where the sharp brackets on the right hand side denote equilibrium cumulant averages. An equivalent of Eq. (3) was first derived by Stratonovich for stochastic systems [16]. In this case Eq. (3) results from the assumption that the energy maximum to be overcome in a transition between two minima is placed midway between the two minima (in the direction of Q). To check the five requirements against Eq. (3), we note that requirements (1) and (3) are satisfied, while (2) is not. Requirement (4) is also satisfied since a system is Gaussian whenever all higher than second order cumulant averages are zero. It is easy to show that the generalization of Eq. (3) to N external fields is

$$\langle \dot{Q}_i \rangle_{\{E_j\}} = \sum_{n=1}^{\infty} \frac{1}{n!} \left(\frac{\beta}{2}\right)^n \sum_{i_1, \dots, i_n} E_{i_1} \dots E_{i_n} \times \int_{-\infty}^{\infty} dt_1 \dots dt_n \langle \dot{Q}_i(0), \dot{Q}_{i_1}(t_1), \dots, \dot{Q}_{i_n}(t_n) \rangle_0. \quad (4)$$

From this it follows that if, e. g. , the Q_1 and Q_2 equilibrium fluctuations are uncorrelated, there is no coupling from the field E_1 to Q_2 . Thus, Eq. (3) satisfies all 5 requirements for a nonlinear response theory except causality. Note that, despite violating causality, Eq. (3) for time-independent external fields is an extension of linear response theory. It should also be noted that Eq. (2) for constant fields does have non-trivial applications, e. g. , to the calculation of excess current noise in random walk models where Eq. (2) leads to the correct expression [15,17].

We now turn to the problem of deriving an approximate nonlinear response theory applicable to the case with a time-dependent

external field. This is achieved by approximating the exact density matrix by an expression referring to the equilibrium dynamics. For simplicity, we consider first the case of only one external field $E(t)$. The Hamiltonian is $H(t) = H_0 - E(t)Q$. Suppose that, for a system initially in thermal equilibrium, the field is turned on at $t=0$. For $t>0$ the "internal energy" $H_0(t)$ is determined from the Poisson bracket expression

$$\begin{aligned} \frac{dH_0}{dt} &= \{H, H_0\} \\ &= -E(t)\{Q, H_0\} = E(t)\{H, Q\} = E(t)\dot{Q}. \end{aligned} \quad (5)$$

If $X(0)$ and $X(t)$ denote, respectively, the starting and ending point of a path in phase space, an integration of Eq. (5) leads to

$$H_0(X(t)) - H_0(X(0)) = \int_0^t E(\tau) \dot{Q}(X(\tau)) d\tau. \quad (6)$$

Equation (6) just expresses the fact that the change in internal energy is equal to the work performed by the external field. Now the Liouville equation for the density matrix ρ , $\frac{d\rho}{dt} = 0$, implies $\rho(X(t)) = \rho(X(0))$. Initially, the density matrix is $\rho(X(0)) = \exp[-\beta H_0(X(0)) + \beta F_0]$ where F_0 is the equilibrium free energy. Thus, from Eq. (6) we get

$$\rho(X(t)) = \exp\left[-\beta H_0(X(t)) + \beta \int_0^t E(\tau) \dot{Q}(X(\tau)) d\tau + \beta F_0\right]. \quad (7)$$

Define now the functional $Z_E[E(\tau)]$ by

$$Z_\varepsilon [E(\tau)] = \text{Tr}_{\text{cl}, X(t)} \exp \left[-\beta H_0(X(t)) + \beta \int_0^{t+\varepsilon} E(\tau) \dot{Q}(X(\tau)) d\tau + \beta F_0 \right] \quad (8)$$

where $\text{Tr}_{\text{cl}, X(t)}$ denotes the classical trace, i. e. , an integration over all phase space points $X(t)$. We obviously have

$$\langle \dot{Q}(t) \rangle_{E(t)} = \beta^{-1} \lim_{\varepsilon \rightarrow 0} \frac{\delta}{\delta E(t)} \ln Z_\varepsilon [E(\tau)] . \quad (9)$$

This result is exact. To arrive at an approximation that allows a calculation of the response in terms of equilibrium averages, we replace the exact path in phase space from $X(0)$ to $X(t)$ by the unperturbed path that ends in $X(t)$. Then Eq. (8) reduces to an equilibrium average:

$$Z_\varepsilon [E(\tau)] = \left\langle e^{\beta \int_0^{t+\varepsilon} E(\tau) \dot{Q}(\tau) d\tau} \right\rangle = \exp \left[\sum_{n=2}^{\infty} \frac{\beta^n}{n!} \int_0^{t+\varepsilon} dt_1 \dots dt_n E(t_1) \dots E(t_n) \langle \dot{Q}(t_1), \dots, \dot{Q}(t_n) \rangle_0 \right] . \quad (10)$$

In this approximation the predicted response is easily found from Eqs. (9) and (10),

$$\langle \dot{Q}(t) \rangle_{E(t)} = \sum_{n=1}^{\infty} \frac{\beta^n}{n!} \int_{-\infty}^t dt_1 \dots dt_n E(t_1) \dots E(t_n) \langle \dot{Q}(t), \dot{Q}(t_1), \dots, \dot{Q}(t_n) \rangle_0 . \quad (11)$$

The generalization of this result to the case of several external fields is straightforward. One finds

$$\langle \dot{Q}_i(t) \rangle_{\{E_j(\tau)\}} = \sum_{n=1}^{\infty} \frac{\beta^n}{n!} \sum_{i_1, \dots, i_n} \int_{-\infty}^t dt_1 \dots dt_n E_{i_1}(t_1) \dots E_{i_n}(t_n) \times \langle \dot{Q}_i(t), \dot{Q}_{i_1}(t_1), \dots, \dot{Q}_{i_n}(t_n) \rangle_0 . \quad (12)$$

It is also easy to show that, in the case of one external field the response of an arbitrary degree of freedom A is given by

$$\langle A(t) \rangle_{E(t)} = \sum_{n=1}^{\infty} \frac{\beta^n}{n!} \int_{-\infty}^t dt_1 \cdots dt_n E(t_1) \cdots E(t_n) \langle A(t), \dot{Q}(t_1), \dots, \dot{Q}(t_n) \rangle_0 \quad (13)$$

Equation (12) satisfies all 5 requirements for an approximate nonlinear response theory except, perhaps, time-reversal invariance. This requirement cannot be checked because Eq. (12) is not based on an expression for the path probability. On the other hand, there is no indication that Eq. (12) violates time-reversal invariance. Thus, Eq. (13) reduces to the correct expression in the linear limit which is not obvious since linear response theory is derived from time-reversal invariance. Furthermore, the second order term of Eq. (13) is also correct [8,13]. In many cases, of course, this term is zero.

As an application of Eq. (11) we now evaluate the nonlinear creep function. This quantity is defined as the average increase of Q in time t after a constant field E is introduced at $t=0$. Denoting the creep function by $\langle \Delta Q(t) \rangle_{E \theta(t)}$, we first note that Eq. (11) may be rewritten as

$$e^{\beta \int_{-\infty}^t E(\tau) \langle \dot{Q}(\tau) \rangle_{E(\tau')} d\tau} = \left\langle e^{\beta \int_{-\infty}^t E(\tau) \dot{Q}(\tau) d\tau} \right\rangle_0 \quad (14)$$

Since $E(t)=E \theta(t)$ Eq. (14) implies for $t>0$

$$e^{\beta E} \langle \Delta Q(t) \rangle_{E\theta(t)} = \left\langle e^{\beta E \Delta Q(t)} \right\rangle_0 \quad (15)$$

In the $E \rightarrow 0$ limit Eq. (15) reduces to the well-known expression for the linear creep function

$$\langle \Delta Q(t) \rangle_{E\theta(t)} = \frac{1}{2} \beta E \langle \Delta Q^2(t) \rangle_0 \quad (16)$$

Equations (11)-(15) were derived for a Hamiltonian system but make sense also for a stochastic system. An example is random walk in one dimension. To calculate the right hand side of Eq. (15) we note that the number of jumps in time t is Poisson distributed around $N = \gamma_0 t$ where γ_0 is the zero field jump frequency. From this it is easy to see that Eq. (15) implies

$$e^{\beta E} \langle \Delta Q(t) \rangle_{E\theta(t)} = e^{\gamma_0 t [\cosh(\beta E a) - 1]} \quad (17)$$

In this simple example there are no transient effects and the predicted average velocity is given by

$$\langle \dot{Q} \rangle_E = \gamma_0 a \frac{\cosh(\beta E a) - 1}{\beta E a} \quad (18)$$

The theory predicts an exponential nonlinearity which is typical for random walks. For large fields the velocity varies as $\exp(\beta E a)$ whereas for the commonly used "symmetric" jump rate assignment one has asymptotically $\dot{Q} \propto \exp(\beta E a / 2)$. On the other hand, for the "asymmetric" jump rate assignment $\gamma = \gamma_0 \exp(\beta E a)$ in the direction of the field and $\gamma = \gamma_0$ in the opposite direction, the predicted velocity is asymptotically correct. There are, however,

many other possible jump rate assignments in an external field, and this illustrates well the impossibility of constructing a generally applicable approximate nonlinear response theory.

Another simple example of Eq. (15) is the case of a free particle in a box. It is a simple matter to evaluate $\langle e^{\beta E \Delta Q(t)} \rangle_0$ in the $t \rightarrow \infty$ limit where Q is the box length coordinate. If L denotes the length of the box, one finds

$$\lim_{t \rightarrow \infty} \langle e^{\beta E \Delta Q(t)} \rangle_0 = \langle e^{\beta E Q} \rangle \langle e^{-\beta E Q} \rangle = \frac{2 [\cosh(\beta E L) - 1]}{(\beta E L)^2} \quad (19)$$

where it has been assumed that $Q(t)$ and $Q(0)$ are uncorrelated for $t \rightarrow \infty$. For large fields Eq. (19) predicts an average displacement equal to L . The correct result is $L/2$, of course. The theory gives a qualitatively correct result, for instance there is no drift of the particle after some time, but the result is not exact.

To summarize, the nonexistence of a useful exact nonlinear response theory raises the question whether any approximate nonlinear response theory exists. Five requirements have been proposed which such a theory should satisfy. It should, (1), calculate the response solely from a knowledge of the external field and of the equilibrium fluctuations of $Q(t)$. It should, (2), satisfy causality and, (3), time-reversal invariance, and also, (4), predict a linear response whenever the equilibrium fluctuations of $Q(t)$ are gaussian. Finally, (5), if Q_1 and Q_2 fluctuate independently in equilibrium, a field coupling to Q_1 should not influence the Q_2 -response and vice versa. It is not clear whether any theory exists which satisfies these five require-

ments. A candidate for such a theory has been proposed (Eq. (11), and more generally Eq. (12)). In deriving Eqs. (11) and (12) the external field density matrix was estimated by replacing the exact phase space paths by the unperturbed paths.

Whenever the cumulant averages of $Q(t)$ are smooth functions of time, the theory predicts the nonlinear effects to appear only some time after the field is introduced. This is a feature which is often seen in experiment where a system for a given external field may be almost linear on a short time scale while the DC response is strongly nonlinear [12].

The proposed approximate nonlinear response theory satisfies all five requirements except, perhaps, time-reversal invariance which cannot be checked. On the other hand, there are no indications of a violation of time-reversal invariance. Thus, the theory is exact to second order, which is as far as there are general results calculating the response from equilibrium fluctuations [14]. To properly check time-reversal invariance, the full path probability must be known. This is equivalent to knowing all cumulant averages of $\dot{Q}(t)$ in the external field. A possible generalization of Eq. (11) to the calculation of these averages is

$$\begin{aligned} \langle \dot{Q}(t_1), \dots, \dot{Q}(t_n) \rangle_{E(t)} &= \sum_{m=0}^{\infty} \frac{\beta^m}{m!} \int_{-\infty}^{t_n} dt'_1 \dots dt'_m E(t'_1) \dots E(t'_m) \\ &\times \langle \dot{Q}(t_1), \dots, \dot{Q}(t_n), \dot{Q}(t'_1), \dots, \dot{Q}(t'_m) \rangle_0 \quad (t_1 < \dots < t_n) \end{aligned} \quad (20)$$

with an obvious generalization to the case of several external fields. However, Eq. (20) explicitly violates causality which, as shown by Bochkov and Kuzovlev [14], implies the following exact

relation between the various cumulant averages

$$\langle \dot{Q}(t) \rangle_{E(t)} = \sum_{n=1}^{\infty} (-1)^{n-1} \frac{\beta^n}{n!} \int_{-\infty}^t dt_1 \cdots dt_n E(t_1) \cdots E(t_n) \quad (21)$$

$$\times \langle \dot{Q}(t), \dot{Q}(t_1), \dots, \dot{Q}(t_n) \rangle_{E(t)}$$

It is an open problem whether it is possible to derive Eqs. (11) and (12) from a path probability that satisfies time-reversal invariance (Eq. (1)). Another open problem is to derive a criterion for the applicability of Eqs. (11) and (12). As a matter of fact it is not clear whether any system exists for which the proposed response theory is satisfactory. On the other hand it is likely that no other reasonable approximate nonlinear response theory exists. Thus, accepting requirement (4), the occurrence of a nonlinearity must be coupled to the existence of nonzero higher than second order equilibrium cumulant averages of $\dot{Q}(t)$. Similarly, requirement (5) more or less dictates the occurrence of cumulant averages in the expression for the response, since cumulants are additive for independently fluctuating degrees of freedom. Finally, in the case where $\{\dot{Q}_i\}$ is a vector, the transformation properties of the average $\{\langle \dot{Q}_i \rangle_E\}$ almost dictates the n'th order term to involve n+1 \dot{Q} 's.

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