Classical Langevin Dynamics for Model Hamiltonians

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We propose a scheme for extending the model Hamiltonian method developed originally for studying the equilibrium properties of complex perovskite systems to include Langevin dynamics. The extension is based on Zwanzig’s treatment of nonlinear generalized Langevin’s equations. The parameters entering the equations of motion are to be determined by mapping from first-principles calculations, as in the original model Hamiltonian method. The scheme makes possible, in principle, the study of the dynamics and kinetics of structural transformations inaccessible to the original model Hamiltonian method. Moreover, we show that the equilibrium properties are governed by an effective Hamiltonian which differs from that used in previous work by a term which captures the coherent part of the previously ignored dynamical interaction with the omitted degrees of freedom. We describe how the additional information required for the Langevin equations can be obtained by a minor extension of the previous mapping.

This paper is dedicated to Professor Josef Devreese on the occasion of his 65th birthday and his formal retirement. May he continue to have many more productive years in theoretical physics.

I. INTRODUCTION

Our understanding of the properties of relativity simple materials has advanced markedly over the last half century. A wealth of experimental data exists with which theory, both as formal analysis and as first-principles computation, is in quantitative agreement. More recently, attention has turned to such complex materials as multication oxides which, even at this advanced stage of condensed matter physics and materials science, still pose a compelling challenge. Those materials possess an immensely rich phenomenology - antiferromagnetism and high-temperature superconductivity in cuprates; colossal magnetoresistance, charge, spin, and orbital ordering in manganates; ferroelectricity, antiferroelectricity, ferrodistortion, and relaxors in perovskites and related materials; and, more recently, a colossal temperature-independent dielectric constant in single crystal CaCu$_3$Ti$_4$O$_{12}$ (CCTO). First-principles studies of these complex oxides, however, have been limited to equilibrium properties or responses of perfect crystals and to relatively simple multilayer structures. More difficult equilibrium questions have been addressed for the perovskite ferroelectrics by constructing a simplified model Hamiltonian the parameters of which are obtained from first principles computations. The construction of the model Hamiltonians for the perovskite ferroelectrics proceeds as follows. Those optical branches which are unstable in the high symmetry cubic structure are identified$^1$. From them, local modes are constructed which approximate the optimally localized lattice Wannier functions$^2$ and which capture the dominant anharmonicities in the total energy. These local modes form the basis for the construction of an “effective” or “model” Hamiltonian$^2$ simple enough to use for Monte Carlo simulations of equilibrium structures and properties.$^3$–$^5$ The parameters of the model Hamiltonian are few enough in number to admit computation by first-principles methods via a mapping to the model Hamiltonian.

There are many dynamic or kinetic phenomena of great interest which cannot be addressed via the model Hamiltonian scheme at its present level of development because the model Hamiltonians are too incomplete to use for molecular dynamics. Among these are nucleation and growth in first-order phase transitions, nonequilibrium kinetics associated with phase transitions, domain wall movement in ferroelectrics, dielectric dissipation in insulators and many others. One such problem is presented by the phenomenon of pressure amorphization. This has been successfully addressed at the conceptual level by augmenting a highly simplified model Hamiltonian with Langivan dynamics,$^6$,$^7$ showing how pressure amorphization can occur by the emergence of metastable displacive disorder. In the present paper, Zwanzig’s theory of nonlinear generalized Langevin equations$^8$ is used to create a classical dynamic extension of the model Hamiltonian method suitable for addressing such problems beyond the present scope of that method.

In Langevin dynamics, a system evolves under the influence of its internal dynamics; of a relaxation term or, more generally, of a memory kernel which captures the coherent part of its interaction with a thermal bath, and of the remaining stochastic forces from the bath. In Section 2, we show how to decompose the Born-Oppenheimer Hamiltonian of a material into a model system, a bath, and an interaction between them in the form with which Zwanzig starts. In Section 3, we recapitulate Zwanzig’s development to add clarity and to make the present paper more nearly selfcontained. In Section 4, we transform Zwanzig’s abstract equations into explicit Langevin equations of motion for the model coordinates.
and momenta. We then comment on implementation in Section 5 and conclude there that exploring the computational feasibility of the scheme would be worthwhile. We emphasize in Section 6 that the equilibrium properties of the system are governed by a model Hamiltonian which differs from that previously obtained by a term which captures the coherent consequences of the interaction of the model degrees of freedom with those omitted from the model Hamiltonians and comment on generalizing the formalism.

2. THE SYSTEM PLUS BATH DECOMPOSITION

We make the Born-Oppenheimer approximation and confine ourselves to classical dynamics for the nuclear coordinates. The resulting Hamiltonian \( \mathcal{H} \) contains the nuclear coordinates and momenta. We divide the nuclear coordinates and momenta into two groups, the system variables \( x \) which contribute all the anharmonicity in \( \mathcal{H} \) and the bath variables \( y \) the dependence of \( \mathcal{H} \) on which is harmonic. We are thus supposing that the \( x \) are the important structural variables in analogy with the model Hamiltonian method, but are retaining the remaining dynamic variables \( y \) of the material viewed as providing a thermal reservoir to the system variables. \( x \) and \( y \) are column vectors with coordinate subvectors \( Q \) and \( q \), respectively, and momentum subvectors \( P \) and \( p \), respectively:

\[
\begin{align*}
  x &= \left( \begin{array}{c} Q \\ P \end{array} \right), & y &= \left( \begin{array}{c} q \\ p \end{array} \right)
\end{align*}
\]

\( \mathcal{H} \) can now be decomposed into a system part \( \mathcal{H}'_s(x) \), a bath part \( \mathcal{H}_b(y) \), and a system-bath interaction \( \mathcal{H}_{SB}(x,y) \),

\[
\mathcal{H} = \mathcal{H}'_s(x) + \mathcal{H}'_{sb}(x,y) + \mathcal{H}_b(y)
\]

(2)

The \( Q \) correspond to the local modes of the model Hamiltonian method, and the potential energy part of \( \mathcal{H}'_s(x) \) corresponds to the entire model Hamiltonian of the prior schemes\(^3\)\(^-\)\(^5\).

The bath Hamiltonian \( \mathcal{H}'_b(y) \) is harmonic,

\[
\mathcal{H}_b(y) = \frac{1}{2} y^T \cdot K \cdot y,
\]

(3)

where \( y^T \) is the row vector \((q^T, p^T)\). \( K \) is a real, symmetric, positive-definite matrix in the \( y \) space with the submatrices

\[
K = \begin{pmatrix}
K_{qq} & K_{qp} \\
K_{pq} & K_{pp}
\end{pmatrix}
\]

(4)

In all cases contemplated here, there are no velocity-dependent forces so that \( K_{qp} = K_{pq} = 0 \). The system-bath interaction is linear in \( y \) but nonlinear in \( x \) in general,

\[
\mathcal{H}'_{SB}(x,y) = -\frac{1}{2} (a^T(x) \cdot y + y^T \cdot a(x)),
\]

(5)

where \( a(x) \) is a column vector in the \( y \) space which is in general a nonlinear function of \( x \).

To make contact with Zwanzig’s development, \( \mathcal{H} \) must be rewritten. \( \mathcal{H}'_s(x) \) contains both kinetic, \( T_s(P) \), and potential, \( V'_s(Q) \), energies,

\[
\mathcal{H}'_s(x) = T_s(P) + V'_s(Q),
\]

(6)

with \( V'_s(Q) \) anharmonic. We now rewrite \( \mathcal{H} \) in the form used by Zwanzig

\[
\mathcal{H}(x, y) = \mathcal{H}_s(x) + \mathcal{H}_b(x, y)
\]

(7)

so that

\[
\mathcal{H}_b(x, y) = \frac{1}{2} (y - a(x))^T \cdot K \cdot (y - a(x)),
\]

(8)

absorbing \( \mathcal{H}'_{SB} \) into \( \mathcal{H}_b \). This introduces an additional potential energy term into \( \mathcal{H}'_s \) so that

\[
\mathcal{H}_s(X) = T_s(P) + V_s(Q),
\]

(9)

\[
V_s(Q) = V'_s(Q) - \frac{1}{2} a^T(x) \cdot K \cdot a(x).
\]

(10)

Thus \( a(x) \) becomes the instantaneous equilibrium value of \( y \), and the non-negative term subtracted from \( V'_s(x) \) in (9b) softens \( V_s(x) \). Thus the potential energy of the model system differs importantly from the original model Hamiltonian and should yield improved equilibrium properties. The Hamiltonian (7) is the starting point of Zwanzig’s development.

3. ZWANZIG’S FORMAL LANGEVIN DYNAMICS

3.1 Kerner Dynamics

One can write Hamilton’s equations of motion for the system variables in vector form

\[
\begin{pmatrix}
\dot{Q} \\
\dot{P}
\end{pmatrix} = \begin{pmatrix}
\partial \mathcal{H}/\partial Q \\
-\partial \mathcal{H}/\partial P
\end{pmatrix}
\]

(11)

Kerner’s procedure\(^9\) collects these into a single equation through the introduction of the antisymmetric matrix \( A \) operating in the \( x \) space

\[
A = \begin{pmatrix}
0 & 1 \\
-1 & 0
\end{pmatrix}.
\]

(12)

The equation of motion then becomes

\[
\dot{x} = A \cdot \nabla_x \mathcal{H},
\]

(13)
\[ \dot{y} = B \cdot \nabla_y \mathcal{H}, \]  

where B is the analog of A in the y space.

### 3.2 Motion in the y Space

Introducing the subscript \( t \) to specify the time at which the dependent variables are evaluated, (14) becomes

\[ \dot{y}_t = B \cdot K \cdot [y_t - a(x_t)]. \]

The formal solution to (15) can be written as

\[ y_t - a(x_t) = e^{tB} [y_o - a(x_o)] - \int_0^t dt' e^{t'K} \cdot \nabla_x a(x_{t-t'}) \cdot \dot{x}_{t-t'} \]

after partial integration subsequent to the direct integration of (15).

### 3.3 Motion in the x-Space

The equation of motion (13) for x is, more explicitly,

\[ \dot{x}_t = A \cdot \nabla_x \mathcal{H}_S - A \cdot \nabla_x a T(x_t) \cdot K \cdot [y_t - a(x_t)]. \]

Substituting (16) into (17) yields a closed equation of motion for x which will turn out to be the desired Langevin equation,

\[ \dot{x}_t = A \cdot \nabla_x \mathcal{H}_S(x_t) + \int_0^t dt' A \cdot \nabla_x a(x_t) T \cdot K \cdot e^{t'K} \cdot \nabla_x a(x_{t-t'}) \cdot \dot{x}_{t-t'} \\
- A \cdot \nabla_x a(x_t) \cdot K \cdot e^{t'K} [y_o - a(x_o)]. \]

The first line on the right side of (18) is a deterministic, conservative, nonlinear force. The second line contains a state-dependent, that is nonlinear, memory function which gives rise to a dissipative force which is nonlocal in time. The third line contains a state-dependent response to a stochastic force. In the next section we show the latter to be Gaussian random colored noise.

### 3.4 The Random Force

Define the vector \( F_t \) in y space as a force

\[ F_t = -K \cdot e^{tB} [y_o - a(x_o)]. \]

The equation (18) for \( \dot{x}_t \) is solved for \( x_t \) given \( x_o \), the initial condition. One could in principle also fix \( y_o \) as an initial condition and solve Eq. (18) deterministically by molecular dynamics. One would then, for each value of \( x_o \) chosen, have to compute the complete trajectories to establish the statistical dynamics of the system over and over for different \( y_o \). In doing so, any external coupling to a reservoir forcing the system to relax towards equilibrium is ignored. Zwanzig instead makes the assumption that the bath is in thermodynamic equilibrium at \( t_o \) given \( x_o \). This implies that the initial state \( y_o \) has a probability distribution

\[ \text{PROB}[y_o|x_o] = \frac{e^{-\beta \mathcal{H}_B(y_o,x_o)}}{\int dy_o e^{-\beta \mathcal{H}_B(y_o,x_o)}} \]

so that \( y_o \) is a Gaussian random variable (GRV). \( F_t \), [19], is a superposition of GRV’s and is therefore a Gaussian random variable itself with

\[ \langle F_t \rangle = 0, \quad \langle F_t F_{t'} \rangle = k_B T K \cdot e^{(t-t')B} K. \]

### 3.5 The Langevin Equation

We can now interpret [16] as a generalized, nonlinear Langevin equation,

\[ \dot{x}_t = A \cdot [\nabla_x \mathcal{H}_S(x_t)] + \int_0^t dt' M(t,t') \cdot \dot{x}'_t + \nabla_x a^T(x_t) \cdot F_t, \]

where

\[ M(t,t') = \nabla_x a(x_t) T \cdot K \cdot e^{(t-t')B} K \cdot \nabla_x a(x_{t'}). \]

### 4. EQUATIONS OF MOTION FOR Q AND P

#### 4.1 Q and P

As asserted earlier, we exclude velocity-dependent forces, which simplifies the formalism. We also absorb the nuclear masses into the definition of the momenta so that

\[ \mathcal{H}_S = \frac{1}{2} P \cdot P + V(Q), \]

\[ \mathcal{H}_B = \frac{1}{2} p \cdot p + (q - a_q(Q))^T \cdot K_{qq} \cdot (q - a_q(Q)), \]

\[ a(x) = \begin{pmatrix} a_q(Q) \\ 0 \end{pmatrix}, \]

\[ \nabla_x a(x) = \begin{pmatrix} \nabla_q a_q(Q) & 0 \\ 0 & 0 \end{pmatrix}. \]

If we define

\[ L(t) = K \cdot e^{tB} K, \]
then \( L_{qq} \) is its only nonzero submatrix so that

\[
M(t, t') = \begin{pmatrix}
(\nabla Q a_q(Q_t))^T \cdot L_{qq}(t - t') \cdot \nabla Q a_q(Q_t) & 0 \\
0 & 0
\end{pmatrix}
\]  
\hspace{1cm}
(30)

All this results in the much simpler, explicit equations of motion

\[
\dot{Q}_t = P_t,  
\hspace{1cm}
(31)
\]

\[
\dot{P}_t = -\nabla Q V(Q_t) - \int_0^t dt' M_{pp}(t, t') \cdot P_{t'},
\hspace{1cm}
(32)
\]

\[
M_{pp}(t, t') = \nabla Q a_q(Q_t)^T \cdot L_{qq}(t - t') \cdot \nabla Q a_q(Q_t),
\hspace{1cm}
(33)
\]

\[
F_{qt} = -L_{qq}(t) \cdot [q_\lambda - a_q(Q_\lambda)],
\hspace{1cm}
(34)
\]

\[
\langle F_{qt} F_{q't'} \rangle = k_BT_L L_{qq}(t - t').
\hspace{1cm}
(35)
\]

The remaining task is to simplify \( L_{qq}(t - t') \).

4.2 Bath Normal Modes

We now transform to the normal modes of \( K \) in the \( y \) space. \( K \) and \( B \) are now comprised entirely of a set of \( 2 \times 2 \) diagonal submatrices \( K_{\lambda\lambda} \) and \( B_{\lambda\lambda} \) along the main diagonal with

\[
K_{\lambda\lambda} = \begin{pmatrix}
\omega_\lambda^2 & 0 \\
0 & 0
\end{pmatrix},
\hspace{1cm}
(36)
\]

and correspondingly

\[
B_{\lambda\lambda} = \begin{pmatrix}
0 & -1 \\
1 & 0
\end{pmatrix}.
\hspace{1cm}
(37)
\]

Inserting these into \( L_{qq}(t - t') \) yields

\[
L_{q_\lambda q_{\lambda'}}(t - t') = \omega_\lambda \cos \omega_\lambda(t - t') \delta_{\lambda\lambda'},
\hspace{1cm}
(38)
\]

greatly simplifying (35) and leading to

\[
M_{PP}(t, t') = \int d\omega g(\omega) \omega^2 \cos \omega(t - t') W_{PP}(\omega; t, t'),
\hspace{1cm}
(39)
\]

\[
W_{PP}(\omega; t, t') = \sum_{\lambda} \delta(\omega_\lambda - \omega) \nabla Q a_{q_\lambda}(Q_t) \nabla Q a_{q_\lambda}(Q_{t'}) / g(\omega),
\hspace{1cm}
(40)
\]

\[
g(\omega) = \sum_{\lambda} \delta(\omega_\lambda - \omega)
\hspace{1cm}
(41)
\]

From (40), \( g(\omega) \) is the total phonon density of states of the bath.

Still more explicitly, let \( \ell \) be the index of the degrees of freedom of \( S \). The equations of motion become

\[
\dot{Q}_{\ell t} = P_{\ell t},
\hspace{1cm}
(42)
\]

\[
\dot{P}_{\ell t} = -\nabla Q V(Q) - \sum_{\ell} \int_0^t dt' M'_{\ell\ell}(t, t') P_{\ell t'},
\hspace{1cm}
(43)
\]

\[
M_{\ell\ell} = \int d\omega g(\omega) \omega^2 \cos \omega(t - t') W_{\ell\ell}(\omega; t, t'),
\hspace{1cm}
(44)
\]

\[
W_{\ell\ell}(\omega; t, t') = \sum_{\lambda} \delta(\omega_\lambda - \omega) \nabla Q a_{q_\lambda}(Q) \nabla Q a_{q_\lambda}(Q') / g(\omega),
\hspace{1cm}
(45)
\]

\[
\langle F_{q_\ell t} F_{q'_{\ell'} t'} \rangle = k_BT \omega_\lambda^2 \cos \omega_\lambda(t - t') \delta_{\lambda\lambda'}.
\hspace{1cm}
(46)
\]

5. COMMENTS ON IMPLEMENTATION

To focus our attention, we discuss as an illustrative example the dynamics of a first-order structural phase transition under pressure from crystalline phase \( C \) to crystalline phase \( C' \). We suppose that \( C \) is the simpler, more symmetric structure. The transition is associated with the development of a soft branch of the phonon spectrum of \( C \). There is a spinodal in the pressure-temperature plane at which \( C \) becomes locally unstable. Implementation of the Langevin dynamics for studying the phase transition proceeds in a series of steps which we sketch in the following.

5.1 Decomposition into System Plus Bath

Consider pressures such that \( C \) is unstable at \( T=0 \). Calculate the harmonic normal mode spectrum of \( C \). Establish which are the unstable modes or branches of \( C \). Construct local modes from the unstable modes and identify their amplitudes with \( Q \). All other normal modes are allocated to the bath, and \( K \) and the normal modes \( q_\lambda \) are determined.

5.2 Mapping from \( \mathcal{H} \) to \( \mathcal{H}_S + \mathcal{H}_B \)

One first assumes simple analytic forms for the \( Q \) dependence of \( V(Q) \) and \( a(Q) \) motivated by the insights underlying the decomposition of the material into system plus bath. Practical considerations limit both the complexity of the \( Q \) dependence of the forms assumed for \( V(Q) \) and \( a(Q) \) and the fineness with which the \( Q \) and \( q \) spaces are sampled. \( V(Q) \) is then obtained from first-principles electronic-structure computations of the total energies for the fixed sample of \( Q \) values which are fully relaxed with regard to the sampled \( q \) variables. The \( a(Q) \) are then simply equal to the relaxed values of the \( q_\lambda \), according to Eqs. (2)-(9). The parameters of the simple analytic forms for \( V(Q) \) and \( a(Q) \) are then determined by a best fit to the numerical data. Thus the computational cost of generating the necessary information on which to base the Langevin simulations is increased beyond that of the previous model-Hamiltonian constructions only by the relaxation.
5.3 Constructing the Langevin Equation

Knowledge of \( V(Q) \) yields the internal forces \( \nabla_Q V(Q) \). Knowledge of \( a(Q) \) yields the state-dependent quantities \( \nabla_Q a(Q) \). Knowledge of \( K \) yields the frequency spectrum \( g(\omega) \), the random force autocorrelation function, and, together with \( \nabla_Q a(Q) \), \( W_{\ell\ell}(t - t') \). Thus everything entering the Langevin equations is known once the parameters of \( H_S + H_B \) are established by the mapping of Section 5.2.

5.4 Solving the Langevin Equation

Apart from the issue of the number of variables to keep track of, there are three essential difficulties in standing in the way of implementing numerically the solution of the Langevin equations: 1) Each of the two terms in the equation for \( P_t \) is nonlinear. 2) There is a memory, and the noise is colored, which require remembering \( Q, P, \) and \( F_{\ell\ell} \), over an interval of time longer than a relevant bath phonon period. 3) There are two quite different time scales in the problem, the bath phonon period and the characteristic relaxation times implicit in the memory function. Issues 1.) and 3.) are encountered in molecular dynamics. Issue 1.) is not serious, and issue 3.) has been confronted with varying degrees of success in MD. Thus the main difficulty is the increase in memory requirement associated with issue 2.). Assuming that issue 2.) can be overcome, one would start the simulation of the first order transition from \( C \) to \( C' \) by drawing an initial value of \( x \) from the truncated canonical ensemble

\[
\rho(x) = \frac{e^{-\beta H_S(x)}}{\int dx' e^{-\beta H_S(x')}}
\]  

(47)

with \( x \) and \( x' \) limited to the basin of attraction of \( C \) in \( V_S(Q) \). One would then choose a time step which permitted adequate sampling of the memory function and the random force autocorrelation function. Forward time integration could proceed by a suitable generalization of the Verlet algorithm adapted for different-integral equations. Updating the random force at each time step need not be onerous. In molecular dynamics only \( P \) and \( Q \) need be remembered at each time step, and only \( \nabla Q V \) needs to be updated. In the current Langevin dynamics, \( P, Q \nabla Q V, \nabla Q a(Q), \) and \( F_{\ell\ell} \) all need to be updated. Thus, the upper bound to the number of degrees of freedom which can feasibly be incorporated into \( H_S \) will turn but to be much less than can be incorporated into a standard molecular dynamics computation with classical pairwise forces. The latter now exceeds \( 10^6 \). Losing as many as three to four orders of magnitude in feasible cell size would still allow quite complex problems to be tackled. We therefore propose that investigating the computational complexity of the proposed Langevin dynamics scheme with the goal of establishing its feasibility and, if feasible, its size limitations would be highly worthwhile.

6. CONCLUSIONS

The current procedure for generating the “model” or “effective” Hamiltonian from first-principles calculations\(^{3-5} \), generates only the potential \( V'_S(Q) \) in \( H'_S(x) \), Eq.(6). By transforming the full Hamiltonian of Eq.(2) into the Zwanzig form Eq.(7), we have added an additional, softening term to \( V'_S(Q) \) to arrive at a system potential \( V_S(Q) \), Eq.(10), which includes all of the coherent effects of the interaction between the reduced model system and the rest of the material. If one constructs the master equation for the probability distribution of \( x \) at \( t \) given \( x_o \) at \( t = 0 \), \( \text{PROB}[x,t — x_o] \), a generalized Fokker-Planck equation, one finds that \( \text{PROB}[x,t — x_o] \) asymptotically approaches a unique stationary solution which is a canonical ensemble for the Hamiltonian \( H_S(x) \), not \( H'_S(x) \),

\[
\text{PROB}[x,t|x_o] \rightarrow \frac{e^{-\beta H_S(x)}}{\int dx' e^{-\beta H_S(x')}} \quad t \rightarrow \infty,
\]  

(48)

which differs from \( \rho(x) \) in Eq.(47) because there is no truncation here. This change is a direct consequence of the form of \( H_B \), Eq.(8), which states that the bath variables relax towards and oscillate around their instantaneous equilibrium positions \( a(x) \). Thus, the use of \( V'_S(Q) \) and \( H'_S(x) \) for the calculation of equilibrium properties is conceptually wrong.

A quantitative analysis of the errors in the current first-principles effective-hamiltonian approach to ferroelectric perovskites has recently been carried out by Tinte et al. They show that not calculating the fully relaxed \( V_S(Q) \) and calculating instead the unrelaxed \( V'^{rs}_S(Q) \) does not, for the materials studied, introduce a major error. Nevertheless, as discussed in Section 5.2, the fully relaxed calculation is necessary to generate the interaction amplitudes \( a(Q) \) essential for implementing the Langevin formalism. Tinte et al. report that the major error arises from ignoring thermal expansion. The import of that conclusion for the present analysis is that anharmonicity in the bath coordinates \( q \) should be included. The only practical way to do so without losing the distinction between system and bath is through the use of a pseudo-harmonic approximation to the time evolution and statistical distribution of the bath variables \( y \). We reserve that generalization of the present formalism for a future publication.

We reiterate the conclusion of Section 5.4 that exploring the feasibility of numerical implementation of the present Langevin dynamics scheme would be highly worthwhile because of the great extension of the reach of model Hamilton methods\(^{3-5,10-14} \) which could result.

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