Nosé-Hoover dynamics for coherent states

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Abstract

The popular method of Nosé and Hoover to create canonically distributed positions and momenta in classical molecular dynamics simulations is generalized to a genuine quantum system of infinite dimensionality. We show that for the quantum harmonic oscillator, the equations of motion in terms of coherent states can easily be modified in an analogous manner to mimic the coupling of the system to a thermal bath and create a quantum canonical ensemble. Possible applications to more complex systems, especially interacting Fermion systems, are proposed.

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1 Introduction and summary

The typical problem in statistical physics is the determination of ensemble averages. The canonical ensemble is characterized by a constant temperature, i.e. the total energy of the system is allowed to fluctuate around its mean value, but the system is kept at a constant temperature by thermal contact with an external heat bath. Besides the direct evaluation of ensemble averages which is impossible in many cases, especially in interacting many-body systems, numerous different approaches have been developed to calculate canonical ensemble properties, among them Monte Carlo approaches and stochastic techniques. In classical molecular dynamics, Nosé has developed a scheme that allows to calculate canonical averages by averaging over a deterministic isothermal time evolution [1,2]. This scheme is called the classical Nosé-thermostat and

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has attracted much interest. In the Nosé-method, a degree of freedom \( s \) and its conjugate momentum \( p_s \) are added to the original system for temperature control. The additional degree of freedom \( s \) acts as a scaling factor for the positions and momenta of the original system. The idea is now that the isoenergetic time evolution of the extended system (that conserves the total energy of the extended system) yields an isothermal time evolution in the subspace of the variables of the original system. This holds for ergodic time evolutions. For a detailed review, see [2].

Although in practice the original formulation of Nosé turned out to be too cumbersome and featured ergodicity problems in many cases, it allowed for a number of improvements that led to very effective and versatile methods [3]. Simply speaking, the resulting schemes exploit the equipartition theorem of classical mechanics to determine the equations of motion of pseudofriction coefficients. The most reliable methods are the so-called Nosé-Hoover chains [4] and the demon method of Kusnezov, Bulgac, and Bauer (KBB) [5] for which even the simple one-dimensional harmonic oscillator is ergodic.

For quantum systems, equivalent methods of comparable power are not yet available. Grilli and Tosatti have found a theorem that provides a basis for a seemingly possible translation of the Nosé-method to quantum mechanics [6]. However, in practice, their method features substantial problems [7,8]. Kusnezov has proposed a method for finite dimensional quantum systems that can be applied if all eigenvectors and eigenvalues of the Hamiltonian are known [9].

For coherent states, a quantum phase space \((r, p)\) can be defined properly and a thermal weight function \(w_{qm}(\beta; r, p)\) exists that permits the calculation of canonical ensemble averages as phase space integrals [10]. In this article, we present a modification of the quantum equations of motion of coherent states in a one-dimensional harmonic oscillator, following closely the ideas of Nosé, Hoover, and KBB for classical systems. In order to calculate ensemble averages by time averaging, the quantum equations of motion of the parameters \((r, p)\) of the coherent states are modified. More precisely, a classical pseudofriction coefficient \(p_\eta\) is added to the system and the equations of motion are designed in such a way that the distribution

\[
f(\beta; r, p, p_\eta) \propto w_{qm}(\beta; r, p) \exp\left(-\beta \frac{p_\eta^2}{2Q}\right)
\]

which is defined on the mixed quantum-classical phase space \((r, p, p_\eta)\) is a stationary solution of a generalized Liouville equation. As a consequence, if the system is ergodic, \(f\) is the stationary probability distribution generated by the modified quantum dynamics, and canonical ensemble averages can be calculated by time averages over the trajectories thus generated. Hence, our
method provides an isothermal quantum time evolution for a quantum system of infinite dimensionality. It is straightforward to generalize it to systems of many distinguishable particles in a three-dimensional harmonic oscillator as well as to free particles. Even non-interacting fermions may be thermalized, since in this case, the quantum distribution function is also known [11].

2 Method and setup

2.1 Coherent states in a harmonic oscillator potential

Given the Hamilton operator $H$ of the one-dimensional harmonic oscillator

$$H = \hbar \omega \left( a^\dagger a + \frac{1}{2} \right),$$

coherent states are defined as eigenstates of the destruction operator $a$

$$a \left| z \right\rangle = z \left| z \right\rangle, \quad z = \sqrt{\frac{m\omega}{2\hbar}} r + \frac{i}{\sqrt{2m\omega}} p. \quad (2)$$

A coherent state is labelled by its complex eigenvalue $z$ which corresponds to a pair of real parameters $(r, p)$. Explicitly, in coordinate representation, coherent states are shifted Gaussian wave packets characterized by the parameters $r$ (mean position) and $p$ (mean momentum):

$$\langle x \left| z \right\rangle = \langle x \left| r, p \right\rangle \propto \exp \left\{ -\frac{(x - r)^2}{2\hbar} \frac{m\omega}{\hbar} + i\frac{p}{\hbar} x \right\}. \quad (3)$$

Coherent states have been extensively investigated [12]. In particular, the following equality is useful for considering the time evolution of coherent states in a harmonic oscillator potential:

$$\exp(-i \omega a^\dagger a t) \left| z \right\rangle = \left| \exp(-i\omega t)z \right\rangle. \quad (4)$$

This implies that the exact quantum time evolution of a coherent state in a harmonic oscillator potential is given by the following equations of motion for the parameters $r$ and $p$

$$\frac{d}{dt} r = \frac{p}{m}, \quad \frac{d}{dt} p = -m\omega^2 r. \quad (5)$$
We stress that in a harmonic oscillator potential the solution of these two coupled ordinary differential equations provides the exact quantum time evolution of coherent states.

Furthermore, the set of all coherent states forms an overcomplete basis of the Hilbert space with \( \int \frac{dr}{(2\pi \hbar)} \mid r, p \rangle \langle r, p \mid = 1 \). As a consequence, given an observable \( B \), its thermodynamic mean value may be evaluated using coherent states:

\[
\langle \langle B \rangle \rangle = \frac{1}{Z(\beta)} \text{tr} \left( B e^{-\beta H} \right) = \frac{1}{Z(\beta)} \int \frac{dr}{(2\pi \hbar)} \int \frac{dp}{(2\pi \hbar)} \langle r, p \mid B e^{-\beta H} \mid r, p \rangle ,
\]

where \( \beta = \frac{1}{k_B T} \) is the inverse temperature, \( Z(\beta) = \text{tr} \left( e^{-\beta H} \right) \) is the usual canonical partition function and \( \langle \langle \cdot \rangle \rangle \) denotes canonical averages.

As shown in [10], one can interpret the space of the continuous parameters \( r \) and \( p \) as a phase space and rewrite (6) as a phase space integral with the thermal weight function \( w_{qm}(\beta; r, p) \)

\[
\langle \langle B \rangle \rangle = \frac{1}{Z(\beta)} \int \frac{dr}{(2\pi \hbar)} \int \frac{dp}{(2\pi \hbar)} w_{qm}(\beta; r, p) B(r, p) ,
\]

\[
Z(\beta) = \int \frac{dr}{(2\pi \hbar)} \int \frac{dp}{(2\pi \hbar)} w_{qm}(\beta; r, p) ,
\]

with

\[
B(r, p) = \langle r, p \mid B \mid r, p \rangle ,
\]

\[
w_{qm}(\beta; r, p) = e^{-|z|^2(e^{\beta \hbar \omega} - 1)} = e^{-\left(\frac{p^2}{2m} + \frac{1}{2} m \omega^2 r^2\right)\left(e^{\beta \hbar \omega} - 1\right)}/(\hbar \omega) .
\]

The function \( w_{qm} \) contains all quantum statistical properties of the system. From (10) it can be inferred that formally, it differs from the classical distribution function of the harmonic oscillator by the factor \( (e^{\beta \hbar \omega} - 1)/\left(\beta \hbar \omega\right) \). Note that this factor tends to 1 in both the classical (\( \hbar \to 0 \)) and the high-temperature (\( \beta \to 0 \)) limit.

2.2 Modification of the equations of motion

The idea of our method is to modify the equations of motion (5) of the coherent states in such a way that the distribution function \( w_{qm} \) is sampled provided
the time evolution is ergodic. To this end, we proceed in a way which is in close analogy to the approaches in classical molecular dynamics [3–5]. The equation of motion of the parameter $p$ is supplemented by a term similar to a frictional force. The time evolution of the pseudofriction coefficient is then determined by the condition that the desired distribution function is a stationary solution of a generalized Liouville equation in the generalized phase space.

2.2.1 Nosé-Hoover thermostat and Nosé-Hoover chain

Adopting the notation of Martyna et al. [4], we investigate the following analogue of the classical Nosé-Hoover dynamics for the quantum dynamics of coherent states:

$$\frac{d}{dt}r = \frac{p}{m}, \quad \frac{d}{dt}p = -m\omega^2 r - p\eta_Q. \tag{11}$$

The key point is the equation of motion of the pseudofriction coefficient $p\eta$. It is determined by the condition that the distribution function

$$f(\beta; r, p, p\eta) \propto w_{qm}(\beta; r, p) \exp(-\beta\frac{p^2}{2Q}) \tag{12}$$

$$\propto \exp\left(-\left(\frac{p^2}{2m} + \frac{1}{2}m\omega^2 r^2\right)e^{\beta\hbar\omega} - \beta\frac{p^2}{2Q}\right)$$

is a stationary solution of the following generalized Liouville equation in the mixed quantum-classical phase space $\Gamma = (r, p, p\eta)$:

$$\frac{d}{dt}f = -f \cdot \left(\frac{\partial}{\partial \Gamma} \cdot \dot{\Gamma}\right) \tag{13}$$

$$= -f \cdot \left(\frac{\partial}{\partial \dot{r}} \ddot{r} + \frac{\partial}{\partial \dot{p}} \ddot{p} + \frac{\partial}{\partial \dot{p}\eta} \ddot{p}\eta\right).$$

We calculate the left-hand side of (13), employing the equations of motion of $r$ and $p$, (11):

$$\frac{d}{dt}f = \frac{\partial f}{\partial \dot{p}} \ddot{p} + \frac{\partial f}{\partial \dot{r}} \ddot{r} + \frac{\partial f}{\partial \dot{p}\eta} \ddot{p}\eta$$

$$= f \cdot \left(-\left(\frac{p}{m} \ddot{p} + m\omega^2 r \ddot{r}\right)e^{\beta\hbar\omega} - \beta\frac{p\eta_Q}{Q}\right)$$

$$= f \cdot \left(\frac{p^2}{m} \frac{e^{\beta\hbar\omega} - 1}{\hbar\omega} - \beta\frac{p\eta_Q}{Q}\right).$$
On the right-hand side of (13), we have the freedom to impose the constraint \( \partial p_\eta / \partial p_\eta = 0 \) that is common in this context [2]. We obtain

\[-f \cdot \left( \frac{\partial}{\partial t} \cdot \dot{\Gamma} \right) = f \frac{p_\eta}{Q} \, .\]  

(15)

Equating (14) and (15) yields the following equation of motion for \( p_\eta \):

\[ \frac{d}{dt} p_\eta = \frac{1}{\beta} \left( \frac{p_\eta^2 e^{\beta \hbar \omega} - 1}{m \hbar \omega} - 1 \right) \, .\]  

(16)

Again, the only difference between this equation and its classical counterpart is given by the factor \((e^{\beta \hbar \omega} - 1)/(\beta \hbar \omega)\). Moreover, (16) retains the property of its classical counterpart that the time evolution of the pseudofriction coefficient is governed by the deviation of the actual value of a quantity related to the kinetic energy from its canonical average value. This can be inferred by evaluating

\[ \left\langle \frac{p_\eta^2 e^{\beta \hbar \omega} - 1}{m \hbar \omega} \right\rangle = 1 \, .\]  

(17)

using (7).

Finally, it is easily confirmed that the set of dynamical equations (11), (16) conserve the quantity

\[ H^* = \left( \frac{p_\eta^2}{2m} + \frac{1}{2} m \omega^2 r^2 \right) e^{\beta \hbar \omega} - \frac{1}{\beta \hbar \omega} + \frac{p_\eta^2}{2Q} + \int^t dt' \frac{p_\eta(t')}{\beta} \, .\]  

(18)

The equations (11) and (16) form a genuine quantum Nosé-Hoover thermostat for coherent states. Since in classical molecular dynamics, these equations of motion frequently feature ergodicity problems, Martyna et al. have developed the idea of a chain thermostat [4]. This method implies to impose another thermostating pseudofriction coefficient on \( p_\eta \) which may be coupled to yet another pseudofriction coefficient, and so on, thereby forming a chain of thermostats. The application of the idea to the quantum case does not infer anything new compared to the classical case, since only the first pseudofriction coefficient of the chain interacts with the quantum phase space variables. Therefore, for further particulars we refer the reader to [4].
2.2.2 KBB-method

Another generalization of the Nosé-Hoover thermostat that is frequently used in classical molecular dynamics is the so-called demon method proposed by Kusnezov, Bulgac, and Bauer [5]. The advantage of this method is that the Hamilton function of the envisaged system does not have to contain a kinetic energy term for temperature control; instead, the time derivative of the temperature control variables is postulated to be proportional to the difference of two arbitrary quantities whose ratio of canonical averages is \(1/\beta\).

At least two pseudofriction coefficients, so-called demons, are introduced for temperature control. Both the equations of motion for positions and momenta are supplemented by additional terms. We introduce the demons into the quantum equations of motion of the parameters of coherent states:

\[
\frac{d}{dt} r = \frac{p}{m} - g'_2(\xi) F(r, p) , \quad \frac{d}{dt} p = -m\omega^2 r - g'_1(\zeta) G(r, p) .
\]  

(19)

\(F(r, p), G(r, p)\) are arbitrary functions of the quantum phase space variables. \(g_1(\zeta), g_2(\xi)\) are functions of the demons which have to be chosen so that the integration of the distribution function \(f\) converges. \(g'_1, g'_2\) are the respective derivatives. The distribution function on the phase space \(\Gamma = (r, p, \zeta, \xi)\) reads

\[
f(r, p, \xi, \zeta) = \exp\left(-\frac{p^2}{2m} + \frac{1}{2}m\omega^2 r^2 \frac{e^{\beta \hbar \omega} - 1}{\hbar \omega} - \beta \frac{(g_2(\xi)}{\kappa_2} + \frac{g_1(\zeta)}{\kappa_1})\right)
\]  

(20)

and the time evolution of the demons is, as above, deduced from the requirement that \(f\) is a solution of a generalized Liouville equation in the phase space. We obtain

\[
\frac{d}{dt} \zeta = \kappa_1 \left( \frac{p}{m} G \frac{e^{\beta \hbar \omega} - 1}{\beta \hbar \omega} - \frac{1}{\beta} \frac{\partial G}{\partial p} \right) ,
\]  

(21)

\[
\frac{d}{dt} \xi = \kappa_2 \left( m\omega^2 r F \frac{e^{\beta \hbar \omega} - 1}{\beta \hbar \omega} - \frac{1}{\beta} \frac{\partial F}{\partial r} \right) .
\]  

(22)

Again, it is interesting to notice that

\[
\frac{1}{\beta} \left\langle \left\langle \frac{\partial G}{\partial p} \right\rangle \right\rangle = \left\langle \left\langle \frac{p}{m} G \frac{e^{\beta \hbar \omega} - 1}{\beta \hbar \omega} \right\rangle \right\rangle ,
\]  

(23)

i.e. the ratio of the canonical averages of the quantities that determine the time derivative of the demons is \(\beta\), just as in the classical case. The quantity
\[
H^* = \left( \frac{p^2}{2m} + \frac{1}{2}m\omega^2 r^2 \right) e^{\beta \hbar \omega} - \frac{1}{\beta \hbar \omega} - \frac{g_2(\xi)}{\kappa_1} - \frac{g_1(\zeta)}{\kappa_2}
\]

\[
+ \frac{1}{\beta} \int_t^{t'} dt' \left( \frac{\partial G}{\partial p} g'_1 + \frac{\partial F}{\partial r} g'_2 \right)
\]

is conserved during the time evolution defined by (19), (21), (22).

In principle, since the choice of the functions \(F, G, g_1, g_2\) is arbitrary, this method offers a lot of freedom. The most prominent coupling scheme recommended by KBB is the so-called cubic coupling scheme with the following choice of functions [5]

\[
g_1 = \frac{1}{2} \xi^2, \quad g_2 = \frac{1}{4} \zeta^4, \quad F = r^3, \quad G = p,
\]

which leads in the quantum case to the special set of equations of motion

\[
\frac{d}{dt} r = \frac{p}{m} - \xi r^3, \quad \frac{d}{dt} p = -m\omega^2 r - \zeta p
\]

\[
\frac{d}{dt} \xi = \kappa_1 \left( \frac{p^2}{m} e^{\beta \hbar \omega} - \frac{1}{\beta} e^{\beta \hbar \omega} \right) - \frac{1}{\beta} \frac{\partial G}{\partial p} g'_1
\]

\[
\frac{d}{dt} \zeta = \kappa_2 \left( m\omega^2 r^4 e^{\beta \hbar \omega} - \frac{1}{\beta} \frac{\partial F}{\partial r} g'_2 \right)
\]

that we have investigated taking \(\kappa_1 = \kappa_2 = 1\). Finally, we note that (27), (28) may easily be linked to the equations of motion proposed by Kusnezov in [9]. \(w_{qm}\) plays the role of Kusnezov’s \(\rho(Q, P)\). However, while Kusnezov’s approach is limited to quantum systems of finite dimensionality, our method works for this system with a Hilbert space of infinite dimensionality because we take advantage of the properties of coherent states.

3 Results

In classical molecular dynamics simulations of the harmonic oscillator, the simple Nosé-Hoover method features ergodicity problems, while the Nosé-Hoover chain method and the demon approach of KBB work well [4,5]. The correct classical phase space density is perfectly reproduced both by the chain and by the demon dynamics.

Formally, the only difference between the quantum phase space density \(w_{qm}\), eq. (10), and its classical counterpart is given by the factor \(\left( e^{\beta \hbar \omega} - 1 \right) / (\beta \hbar \omega)\).
This also applies to the respective equations of motion of the different dynamics. Since this factor is only a number that depends on temperature, but not on the phase space variables \( r \) and \( p \), we anticipate that it does not influence the overall characteristics of the dynamics. Therefore, we expect that ergodicity problems in the quantum case will arise under the same circumstances as in the classical case.

We show results for the set of parameters chosen in [4] to enable a direct comparison. We took \( m = 1, \omega = 1 \) and initial conditions \( r(0) = 1, p(0) = 1 \) with \( 1/\beta = 1.0 \). The numerical integration of the equations of motion was carried out with a fourth-order Runge-Kutte algorithm with a step size that ensured conservation of pseudoenergy to more than seven significant figures. All runs were made over a total integration time of \( 2000 \tau \), where \( \tau = 2\pi/\omega \).

3.1 Nosé-Hoover and Nosé-Hoover chain method, KBB method with cubic coupling scheme

The left panel of figure 1 presents a \((r, p)\)-density map and the projected distribution functions for the simple Nosé-Hoover dynamics. We find a result that is very similar to the classical case [4]: The dynamics does not fill the phase space with the correct weight and, moreover, we find that the obtained distributions strongly depend on the initial conditions and values of the parameters chosen (not shown). Thus, the dynamics is not ergodic.

The situation changes radically with the introduction of a second thermostating variable acting upon the first pseudofriction coefficient \( p_\eta \), see figure 1, right panel. The distribution functions sampled by this time evolution reproduce \( w_{qm} \) extremely well, and changes of the initial conditions and parameters do not have a noticeable effect on the results. The dynamics generated in this way is obviously ergodic. The addition of further thermostating variables does not influence the results.

We point out that the statistics obtained by time averaging over the quantum time evolution is the quantum statistics of the harmonic oscillator. To make this evident, we present plots of the partially integrated distribution function \( w_{qm}(r, p)/Z \) along with plots of its classical limit

\[
w_{cl}(r, p) = \lim_{\frac{\beta \hbar \omega}{\beta \hbar \omega - 1} \to 1} \frac{1}{Z(\beta)} w_{qm}(r, p) = \beta \hbar \omega \exp \left( -\beta \left( \frac{p^2}{2m} + \frac{1}{2} m \omega^2 r^2 \right) \right)
\]

which is proportional to the classical canonical ensemble distribution function. Since \( (e^{\beta \hbar \omega} - 1)/(\beta \hbar \omega) > 1 \) for all \( \beta \), the quantum distribution function is always narrower compared to its classical limit.
Fig. 1. Left panel: Simple Nosé-Hoover dynamics of a quantum harmonic oscillator, right panel: Nosé-Hoover chain dynamics. From above: \((r,p)\)-density plot, momentum distribution, position distribution. The solid line depicts the exact quantum result given by the respective partially integrated function \(w_{qm}/Z\) (e.g., \(f(r) = \frac{1}{Z} \int \frac{dp}{\sqrt{2\pi\hbar}} w_{qm}(r,p)\)), the dashed line represents the corresponding classical distribution \(w_{cl}\) normalized to the same value (see (29)). The distributions sampled by time averaging are presented as histograms.

Figure 2 presents the results obtained from a KBB-demon-dynamics using the cubic coupling scheme. The results are similar to the case of the chain dynamics, in particular, the dynamics is also ergodic.

3.2 Mean values of selected observables

Finally, the results of time averaging are compared to the analytical ensemble averages for two typical observables, the internal energy and its variance. The
analytical formulas are briefly given:

\[ U(\beta) = \left\langle H \right\rangle = \frac{\hbar \omega}{2} + \frac{\hbar \omega}{e^{\beta \hbar \omega} - 1} \quad (30) \]

\[ \text{var}(H) = \left\langle H^2 \right\rangle - \left\langle H \right\rangle^2 = \left( \frac{\hbar \omega}{2 \sinh(\frac{1}{2} \beta \hbar \omega)} \right)^2 \]

Figure 3 shows the excellent agreement between the exact results and the results obtained by time averaging with a KBB dynamics. The small deviations are clearly of statistical origin. They increase at higher temperatures because we kept the total sampling time constant, although the volume of the relevant phase space increases with temperature. Therefore, to cover it with the same accuracy at a higher temperature, one would need a longer sampling time.
Fig. 3. Values of the internal energy and its variance of the harmonic oscillator obtained from time averaging with a KBB dynamics (crosses) compared to the exact quantum canonical ensemble result (solid line).

4 Discussion and outlook

This article presents a straightforward, yet non-trivial extension of the powerful methods of heat bath coupling in classical molecular dynamics simulations to a genuine quantum system of infinite dimensionality. The application of the method to a quantum system of many distinguishable particles or a three-dimensional harmonic potential is a simple generalization.

Since the knowledge of \(w_{qm}\) is indispensable for the setting up of the equations of motion for the pseudofriction coefficients, the method is limited to systems where \(w_{qm}\) is known. Therefore, also non-interacting identical fermions, both moving freely or contained in a harmonic oscillator potential, can be thermalized using the respective distribution functions [11]. Moreover, by coupling one of the solvable systems to a more complex system of interacting particles, one can possibly determine its equilibrium properties. This idea that permits to evaluate ensemble averages by time averaging is potentially very powerful, since efficient approximate quantum dynamics methods (Time-Dependent Hartree-Fock, Fermionic Molecular Dynamics, etc. [13]) are available that are applicable also for indistinguishable fermions. Thus, a new method of calculating thermodynamic properties of interacting Fermion systems seems conceivable that might also work where other methods fail.

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