Herman–Kluk allows analysis of quantum discrete breathers in higher dimensional systems

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1. Introduction

The understanding of the localization of energy on a quantum level in non-linear many-body systems [1–3] is important for advances in a variety of research fields that range from medicine [4] and nano-science [5–8] to quantum computers [9] and materials used for harvesting of solar energy [6]. From classical mechanic perspective, this phenomenon is known as discrete breathers (DBs) or internal localized modes [1,2]. Examples of DBs in mathematical models of systems that have a practical importance include micromechanical cantilever arrays [7], interacting Josephson junctions [9], Bose–Einstein condensates [10], crystals [8], layered high-$T_c$ superconductors [11], antiferromagnetic structures [12], coupled non-linear optical wave-guides [5], quantum dots [6], molecules [13–18] and biological polymers [4]. These systems all have many degrees of freedom and are of a scale where quantum effects are important in certain instances. The quantum mechanical equivalent of DBs are quantum DBs (QDBs), also known as internal localized modes or tunnelling modes.

Current approaches to modelling QDBs are limited to exact quantum dynamics (EQD). The computational cost of EQD scales exponentially with respect to the number of the degrees of freedom in the system. Quantum calculations on the systems of non-linear coupled oscillators that have QDBs are limited to 2–3 degrees of freedom [19–22]. Some models account for only a few levels on each site and can scale up in the number of sites [23,24]. These truncated EQD models are capable of extracting some conceptual information about the phenomenon. Studying QDBs in a practical application requires a better scalability of the computational cost with respect to the system’s complexity and size than what truncated EQD models can offer. One solution is to use a classical trajectory and propagate quantum information, such as the width of the wavepacket, along this trajectory [25,26]. Using this strategy, we applied a low-order Quantized Hamilton Dynamics (QHD) approach [27] in order to account for important quantum effects in DBs, such as the finite width of the transient region, in which delocalized normal-mode behaviour switches to localized motion, and zero-point energy [28]. However, accurate modelling of the evolution of a wavefunction, which is important for spectral properties of the system, is beyond the reach of low level QHDs.

Semi-classical techniques provide a more accurate alternative to EQD. In particular, the Herman–Kluk (HK) approximation constitutes a simple and effective method to study quantum many-body systems [29–33].
HK has proved itself as a robust tool in modelling dynamics of atoms and molecules [33,34], calculating optical response functions [35], and investigating chaotic systems [36]. In concept, the HK method is based on path-integrals. It propagates coherent states with the stationary phase approximation. Closely related path-integral based methods that are based on propagation of coherent states include Coupled Coherent States [37], higher order HK [32], complex semiclassical dynamics [38,39] and Matching-Pursuit/Split-Operator Fourier Transform [40,41]. An alternative approach with similar performance characteristics is Multilayer Multiconfiguration Time-Dependent Hartree Theory [42]. Prior to this publication, there were no documented results showing spectral signatures of QDBs in quantum systems with more than three dimensions and a large number of on-site states.

Recognizing both the promise and the inherent limitations of the truncated EQD and QHD methods, we applied the semi-classical HK approach [29,32] to compensate for the deficiencies of QHD and truncated EQD. By performing a simulation of the quantum wave-function dynamics using HK in a seven-dimensional system with more than 30 occupied levels on each site, we show that studying QDBs in many-body systems becomes feasible. Given the fundamental nature of the HK approximation in semi-classical dynamics, similar or improved accuracy and performance is to be expected with other semi-classical path-integral based approaches.

The ability to accurately model QDBs in large systems significantly advances current work in the field of QDBs. Previously, realistic DBs have been studied using the classical mechanics equation of motion. The classical mechanical approach misrepresents many properties of QDBs, including the transient region, the ground level energy, and tunnelling modes [28]. Our work shows that quantum effects can be added to the models. Furthermore, the Hamiltonian used in the current study has practical importance. It is similar to the Hamiltonian in micro-mechanical cantilever arrays [7,43]. In this paper we document that semi-classical methods are capable of advancing research on QDBs to large and complex systems.

This paper is organized as follow: Section 2 (Methods) describes the computational setup, including the Hamiltonian of the system, the initial conditions, the HK propagator, and its numerical implementation. In Section 3 (Results) we discuss and analyse the results of our calculations. We compare the HK and EQD calculations of the spectrum of a small model system, and illustrate the advantages of HK by calculating the signatures of QDB modes in a larger, seven-site system. In Section 4 (Conclusion) we summarize our findings and consider the broader ranging implications and applicability of the semi-classical approaches.

2. Methods

We analyse the dynamics of QDBs by considering the evolution of an initial state in time. The simplest way to observe this evolution is to consider the projection of the time-dependent wave-packet $|\psi(t)\rangle$ onto the initial state $|\psi_0\rangle$. In the time-domain, one obtains the correlation function $\langle \psi(t)|\psi_0\rangle$. The Fourier transform of the correlation function gives a spectrum in the frequency domain.

$$I(\omega) = \int \exp(i\omega t) \cdot \langle \psi(t)|\psi_0\rangle dt. \quad (1)$$

The system is described by a linearly coupled quartic dimer with a Hamiltonian:

$$H = c_h (p_1^4 + p_2^4 + q_1^4 + q_2^4) + c_a (q_1^2 + q_2^2) + c_c q_1 q_2. \quad (2)$$

We treat the two-dimensional system with both EQD and HK in order to test the accuracy of the HK technique. The multi-dimensional generalization of Hamiltonian Equation (2) is

$$H = c_h \sum_{i=1}^{j} (\hat{p}_i^4 + \hat{X}_i^4) + c_a \sum_{i=1}^{j} \hat{X}_i^4 + c_c \sum_{i=1}^{j} \hat{X}_i \hat{X}_{(i \mod j)+1}. \quad (3)$$

We study the dynamics of the seven-dimensional model of QDBs using the HK approach.

Semi-classically, the correlation function is computed using the HK propagator [29,32]. HK and other semi-classical methods rely on approximating quantum dynamics around classical trajectories. According to Lagrangian mechanics, classical dynamics follows a path of stationary action. In semi-classical mechanics, the quantum time-evolution operator is expanded around these paths of stationary action. HK propagates a wave function $|\psi_0\rangle$ to $|\psi_t\rangle$. To do that, it expands the initial state in terms of coherent states $|p_0, q_0\rangle$ and propagates these states along classical trajectories, while capturing quantum effects. Integrating over this set of initial states adds up the quantum contribution from all of the paths in the set. In theory, the set is infinite, but in practice only the states that have a good overlap with the initial wave-function $|\psi_0\rangle$ contribute and other states can be neglected. Since we are studying the dynamics on the initial state $|\psi_0\rangle$, we
first expand it in terms of \(|x\rangle\), followed by expansion in terms of \(|p, q\rangle\):

\[
|\psi\rangle = \int dx|x\rangle\langle x|\psi\rangle = \int dp \int dq \int dx|pq\rangle\langle pq|x\rangle\langle x|\psi\rangle.
\]

(4)

In HK, auto-correlation of a position state \(|x_0\rangle\) is expressed as:

\[
\langle x|\langle x_0| \exp(-iHt)|x_0\rangle 
\approx \left(\frac{1}{2\pi}\right)^4 \int dp_0 \int dq_0 \langle x_0|p,q_0\rangle R(p_0,q_0,t) 
\times \exp[iS(p_0,q_0,t)][p_0q_0|x_0].
\]

(5)

\(f\) represents the number of the degrees of freedom. \(p_0\) and \(q_0\) respectively represent the momentum and position coordinates of the initial propagated coherent states. \(p_0 = p(p_0,q_0,t)\) and \(q_0 = q(q_0,p_0,t)\) represent the final momentum and position coordinates of the propagated coherent state. The coordinates follow Hamilton’s equations of motion:

\[
\dot{p} = -\frac{\partial H}{\partial q},
\]

(6)

\[
\dot{q} = \frac{\partial H}{\partial p}.
\]

(7)

The integral in Equation (5) is evaluated numerically. There are two parts to the calculation: propagating classical paths \((p_0, q_0)\), and integrating over \(dp_0dq_0\).

For the first task we use the Dormand–Prince eighth-order Runge–Kutta [44] ODE integrator. In the second task, the integral is computed using importance sampling Monte Carlo integration [44]. The main contribution to the integrals comes from \(|pq\rangle\) that are around \(|x_0\rangle\). Therefore, we use the \(|x_0pq\rangle\) probability distribution for the importance sampling.

The attractiveness of HK over EQD comes from the slow increase in computational costs as the system grows in size. A propagation of each trajectory requires computing path-integrals over \(N\) coordinate and \(N\) momentum variables, the action, and the \(2N \times 2N\) elements of the monodromy matrix, i.e. partial derivatives of the final phase space point with respect to the initial one, found in Equation (10). In addition, we neglect trajectories that have a highly divergent value of \(R\) (Equation (10)); they are physically unrealistic and come from errors in numerical integration. The overall procedure leads to \(O(N^3)\) scaling of numerical effort for the integration of each individual trajectory.

The main limitation in the application of the HK propagator to a complex quantum system is a slow convergence of the phase space integral in Equation (5). The longer the propagation time \(t\), the more sensitive is the action \(S(p_0,q_0,t)\) to the initial conditions \((p_0,q_0)\), and the more oscillatory is the exponential term \(\exp[iS(p_0,q_0,t)]\) in the integrand. Numerically, this leads to a strong interference and cancellation in an ensemble of classical trajectories used to perform the integration of Equation (5). The pre-exponential factor Equation (10) grows with time and ensures an approximate conservation of normalization by the propagator. Its average absolute value \(|R(p_0,q_0,t)|\) can be thought of as a measure of numerical efficiency (the greater the expansion coefficients the less efficient is the decomposition into a basis set of coherent states). It has been shown that in many systems the divergence of the HK correlation function results from a few individual trajectories that have unusually large values of \(R\). Such trajectories occur naturally in classical mechanics. If a trajectory passes close to an unstable point in phase space, its sensitivity matrix diverges. One method of improving the convergence of HK consists of simply discarding trajectories that have \(R\) greater than a chosen value and adjusting the normalization constant.

The EQD calculation for the two-dimensional system was performed by finding the eigenstates of the Hamiltonian Equation (2) in the harmonic
oscillator basis, and then projecting the initial state $|\psi_0\rangle$ onto the eigenstates.

3. Results

We start with a side-by-side comparison of HK and EQD applied to the two-dimensional non-linear Hamiltonian Equation (2). Advantages of the HK approach become apparent in the second part of the result section, where we discuss tunnelling modes in a system with a larger number of sites, Equation (3). These large systems cannot be modelled using conventional quantum dynamics methods. The section ends with the discussion of limitations of HK for application to highly non-linear systems.

3.1. Comparison between Herman–Kluk and exact quantum dynamics

Figure 1 compares in detail calculation of the spectra with HK and EQD. Figure 2 zooms in on critical features of the plots. One can see that the methods are congruent in identifying the eigenenergies. The calculated spectra are Fourier transforms of the correlation functions computed with HK (bottom) and EQD (top), Equation (1). The system was propagated in time according to the Hamiltonian Equation (2) with the coherent state on one of the oscillators initially displaced to 3.5 au and the other one being in the ground state. The Hamiltonian parameters are $c_h = 0.5$, $c_c = 0.2$ and $c_a = 0.02$. The error in the height of the intensities can be attributed to the truncated basis set in EQD and a limited number of trajectories in HK.

The spectrum shown in Figure 1 contains several qualitatively distinct regions that are attributed to different dynamical behaviour of the non-linear system. See our earlier work [28] for additional discussion of these regimes. Figure 2 presents a closer comparison between the EQD and HK data for each region. The low energy part of the spectrum (below 15) resembles a spectrum of linear coupled oscillators. It consists of groups of same-quanta states that are split due to coupling. We refer to the states in these groups as non-tunnelling states. In the higher energy part of the spectrum (above 10) one can find local states – the tunnelling states. These isolated peaks are, essentially, pairs of symmetric and anti-symmetric states. The states are localized on the sites, rather than delocalized between the sites, which is the case of the non-tunnelling states. The tunnelling and non-tunnelling states correspond to localized and delocalized vibrational modes in classical mechanics. The regions of localized and delocalized modes are separated by a distinct value of energy. Above that value, the classical energy cannot transfer from one site to the other. In quantum mechanics, energy can always transfer between the sites, which is the case of the non-tunnelling states. The tunnelling and non-tunnelling states correspond to localized and delocalized vibrational modes in classical mechanics. The regions of localized and delocalized modes are separated by a distinct value of energy. Above that value, the classical energy cannot transfer from one site to the other. In quantum mechanics, energy can always transfer between the sites. The transfer time is short at low energies exhibiting non-tunnelling states. The transfer time is exponentially large at high energies involving tunnelling modes. While the transition between the localized and delocalized regimes is abrupt in classical mechanics, in quantum mechanics both tunnelling and non-tunnelling modes can co-exist, and the transition
is gradual. This (transient) region can be seen in Figure 1 at energies from 10 to 15 au. In high energies, the primary tunnelling modes are accompanied by satellite peaks. They can be referred to as higher order tunnelling modes (HOTM) due to the fact that they involve considerable contributions from several states [28].

It should be noted that we do not study the quantum tunnelling process itself, but rather, we identify correctly the energy and intensity of the tunnelling modes. The HK approach is based on purely classical trajectories, and therefore, cannot describe tunnelling correctly. The latter requires times that are much longer than the simulation time used here.

3.2. Application of Herman–Kluk to higher-dimensional systems

The two-dimensional example considered above showed that accurate signatures of QDBs can be obtained with the HK semi-classical approach for a minor computational effort. Here we extend the application of HK to a system that presents significant challenges to EQD. Figure 3 compares the spectra for two, four, and seven coupled anharmonic oscillators, Equation (3), calculated with the HK method.

The parameters of the multi-dimensional Hamiltonian Equation (3) are the same as for the two-dimensional case Equation (2): $c_h = 0.5$, $c_c = 0.2$ and $c_a = 0.02$; and the initial conditions for the four- and seven-dimensional cases are obtained similarly to the two-dimensional case by displacing the ground coherent state of one of the oscillators 3.5 au.

The comparison of the data shown in the three panels of Figure 3 allows us to draw conclusions about energy localization in systems of increasing dimensionality. As the number of sites grows, the intensity of the spectral lines corresponding to the tunnelling modes increases, while the intensities of individual non-tunnelling modes decrease. At the same time, the density of non-tunnelling modes grows, while the density of tunnelling modes remains virtually unchanged.

In the non-tunnelling regime at the low energy region of the spectrum the coupling dominates the anharmonicity, Equation (3), and the properties of the spectrum can be understood by considering a set of bi-linearly coupled harmonic oscillators. Tunnelling modes appear when the anharmonicity is large. In the non-tunnelling regime the energy is exchanged between the sites, while in the tunnelling regime, the energy remains localized within the site with the displaced initial condition. Therefore, the high energy part of the
spectrum originates from a single (anharmonic) oscillator, since the other oscillators remain in their ground states.

In the low energy parts of the spectra energy levels split due to the bi-linear coupling between the sites. The number of peaks grows rapidly with increasing number of sites, and the spectral features become less resolved. At high energies only the displaced site contributes to the spectrum, and the number of peaks is independent of the number of sites. The intensity of the high energy peaks is higher than the intensity of the lower energy peaks. At low energies the intensity is shared between many peaks, while at high energies it is concentrated within essentially a single peak. The absolute spectral intensity grows with increasing number of sites, because displacement of a single site results in a higher energy excitation due to the contributions of the coupling terms, Equation (3).

Application of the HK approach to the higher-dimensional systems deserves further comments. The seven-site system did present additional challenges compared to the smaller-scale calculation. About 30% of the trajectories diverged numerically due to unphysically large $R (>10)$ from Equation (5). These trajectories were filtered out. Neglecting such a significant portion of all trajectories can lead to incorrect results, due to damaging the initial distribution. In order to obtain a converged spectrum, we had to truncate the correlation function with a Gaussian that had a standard deviation of 50 time units, while the trajectories ran for 100 time units. Nevertheless, HK is clearly advantageous compared to EQD. The Monte Carlo nature of HK is beneficial in this case. The HK calculation has been done using only $10^5$ trajectories. This number should be compared to a typical size of the basis set needed in an EQD calculation. For instance, using 30 harmonic oscillator basis functions per site, which is the least amount needed to obtain reasonable results over the considered energy range, leads to $30^7 \approx 10^{10}$, which is five orders of magnitude higher than the number of trajectories used in the semiclassical calculations.

HK is drastically cheaper computationally than EQD. It accurately represents the spectrum of non-tunnelling and tunnelling modes, higher-order tunnelling modes, and a transient region. At the same time, the spectrum obtained by HK lacks sufficient resolution to show splitting between tunnelling states. The correlation function has to be much longer for this purpose. Improving the resolution becomes exponentially harder for HK, because it requires inclusion of a larger number of trajectories to calculate the correlation function for a longer time. Furthermore, HK is a semi-classical approximation to EQD. It cannot properly represent quantum mechanical tunnelling. Therefore, it will not be able to accurately describe the separation between the tunnelling states.

4. Conclusions
Our calculations showed that HK is a viable method for modelling non-tunnelling modes in QDB systems with the precision of an EQD approach.
HK accurately predicted the position of both non-tunnelling and tunnelling modes, the transient region, and higher-order tunnelling modes. The advantage of using HK is clear when studying systems that are too large for an EQD approach. We showed that with an increased number of sites, the energy of non-tunnelling modes is shared by a larger number of states, while the energy of tunnelling modes stays localized.

This work is meant as a proof of concept that even simple semi-classical methods, such as HK, offer conceptual insights into the quantum mechanical aspects of DBs. There exist many ways to improve on the performance of HK: action-angle variables, generalized Filinov transformation, forward–backward initial value representation, log-derivative formulation of the prefactor, symplectic integrators or expansion to higher orders. In applications, most of the potentials will involve fewer eigenstates on each site, allowing for an even larger number of sites. Furthermore, there are a number of more modern path-integral based methods that will further improve on performance. Although basic HK already proved to be a capable method, these additional improvements will allow further theoretical insight on the contribution of quantum effects to localization and tunnelling in non-linear quantum-scale systems.

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