Wave-packet dynamics by optimized polynomials methods

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An optimized and numerically stable method, based on the formalism of orthogonal polynomials, is proposed to solve the time-dependent Schrödinger equation and to construct energy-filtered wave-packets. It is compared to other methods (Runge-Kutta, Second-Order Differencing scheme). A method is also devised to study quantum diffusion in finite systems with periodic boundary conditions, which allows to suppress boundary effects. As an illustration of our methods, electronic localization is studied in a random magnetic flux lattice.

Introduction

Computing the time-evolution of wave-functions gives many valuable information about electronic properties, such as localization lengths, wave-packet dynamics or transport coefficients.[1–3] In order to perform these studies it is generally necessary to produce first a state with a sufficiently well-defined energy and to determine its evolution by solving the time-dependent Schrödinger equation. In this paper, we prove that these operations can be performed by general algorithms derived from the theory of orthogonal polynomials.[4,5] These algorithms lead to optimum accuracy and are numerically stable in any situation (smooth or more irregular and fractal spectra). A comparison with other propagation schemes is given. Besides, a simple way to study diffusion properties in a system with periodic boundary conditions is presented, allowing to suppress the effect of boundaries. An application to a system of great current interest is given.

Polynomial methods

Hereafter, we consider quantum systems described by time-independent tight-binding hamiltonians $\hat{H}$ with finite bandwidth W. There exists several methods to compute the evolution of a state in such systems. A first possibility is exact diagonalization, but it is not applicable to large systems, except very particular cases. The Trotter methods [1] treat a broader class of systems, whose hamiltonians can be split into two parts $\hat{H}_1$ and $\hat{H}_2$ such that it is numerically workable to change from the diagonalization basis of $\hat{H}_1$ to the diagonalization basis of $\hat{H}_2$. Some other methods are valid for any type of hamiltonian. The Runge-Kutta and the Second-Order Differencing (SOD) schemes, as well as their improvements,[6] discretize time into steps Δt much smaller than $\hbar/W$. The evolution operator $\hat{U}(\Delta t) = \exp(-i\hat{H}\Delta t/\hbar)$ is expanded in the first powers of $\hat{H}$, and iterated many times. Some other methods allow direct propagation on a larger time t by using a high-order expansion of $\hat{U}(t)$: the Lanczos scheme is commonly used up to the tenth order,[7,8] while the Chebyshev scheme is used at still larger order (several tens).

Apart from diagonalization and Trotter methods, an important common feature of these methods is that they can be viewed as approximations of $\hat{U}(t)$ with a complex polynomial of degree N :

$$\hat{U}(t) \simeq R_N(\hat{H}, t)$$

obtained after several Runge-Kutta or SOD steps, or after a single step with high-order methods. More generally, many operators of the form $f(\hat{H})$ may be approximated with an appropriate polynomial of degree N :

$$f(\hat{H}) \simeq R_N(\hat{H})$$

In this paper, we will focus essentially on two cases. For the evolution operator, $f(\hat{H}) = \exp(-i\hat{H}t/\hbar)$. For a gaussian energy-filtering operator, $f(\hat{H}) = \exp[-(\hat{H} - E_0)^2/\Delta^2]$ and one can check, by spectral decomposition, that $f(\hat{H})$ enlarges the relative weight of energy components close to E_0.

Optimized polynomials

The main question considered here is which polynomial R_N gives the best accuracy at a given numerical cost. Let us consider first the numerical cost. In order to compute $R_N(\hat{H})|\psi\rangle$, there are 3 types of operations: additions of vectors, multiplications by scalars and applications of $\hat{H}$. Detailed analysis shows that the most time-consuming part is the application of $\hat{H}$, particularly when each orbital is coupled to a great number of other orbitals. Thus the total numerical cost essentially depends on the number of iterations of $\hat{H}$, equal to the degree N of R_N, and is nearly independent of the coefficients of R_N. Now the question is to give a definition for accuracy in order to compare two polynomials of same degree. We define the error as the norm of the vector $|\delta\psi_N\rangle = R_N(\hat{H})|\psi\rangle - f(\hat{H})|\psi\rangle$. Errors due to finite computer precision are not taken into account at this stage. By spectral decomposition, $\langle\delta\psi_N|\delta\psi_N\rangle = \int dE n_\psi(E)\big|R_N(E) - f(E)\big|^2$, where $n_\psi(E) = \langle\psi|\delta(E - \hat{H})|\psi\rangle$ is the local density of states (LDoS), independent of time. The best polynomial R_N should be obtained by minimizing $\langle\delta\psi_N|\delta\psi_N\rangle$ but, as shown below, it is useful to consider more generally the minimization of

$$\Delta_n(R_N, f) = \int dE n(E)\big|R_N(E) - f(E)\big|^2 \qquad (1)$$

where $n(E)$ is non-zero on the whole spectrum of $\hat{H}$, and may be chosen close to $n_\psi(E)$, in the sense that their respective moments are close to each other.

The minimization of $\Delta_n(R_N, f)$ is done within the formalism of orthogonal polynomials.[9] We consider a positive weight-function $n(E)$ which is non-zero only on a finite interval and such that $\int dE n(E) = 1$. If one defines a hermitian product in the space of complex functions of E as $\langle f|g\rangle = \int dE n(E) f^*(E) g(E)$, the integral $\Delta_n(R_N, f)$ is the squared distance between f and R_N. There exists a system of real orthonormal polynomials $\{P_n\}_{n\geq 0}$ such that degree$[P_n] = n$. These polynomials satisfy a three-term recurrence relation $EP_n(E) = a_n P_n(E) + b_{n-1} P_{n-1}(E) + b_n P_{n+1}(E)$, where the a_n, b_n are real coefficients, and $P_{-1} = 0$. The system of polynomials is unique, provided that a sign convention is chosen for the b_n (usually $b_n > 0$).

The polynomial of degree N (or less) which minimizes the integral $\Delta_n(R_N, f)$ is easily found for any function f such that $\langle f|f\rangle$ is finite. It is the orthogonal projection of f on the subspace of polynomials with degree $\leq N$:

$$R_N^{(0)}(E) = \sum_{n=0}^{N} \langle P_n|f\rangle P_n(E) \tag{2}$$

and, from general theory of developments in orthogonal polynomials, $\Delta_n(R_N^{(0)}, f) \to 0$ when $N \to +\infty$, therefore $R_N^{(0)}$ converges to f with respect to the distance $\sqrt{\Delta_n}$.

This leads, at least formally, to a polynomial development of $f(\hat{H})|\psi\rangle$:

$$\begin{aligned} R_N^{(0)}(\hat{H}) &= \sum_{n=0}^{N} \langle P_n|f\rangle P_n(\hat{H}) \\ f(\hat{H})|\psi\rangle &= \lim_{N\to+\infty} \sum_{n=0}^{N} \langle P_n|f\rangle P_n(\hat{H})|\psi\rangle \end{aligned} \tag{3}$$

First, let us study the convergence of this series towards $f(\hat{H})|\psi\rangle$, in the sense that $\langle \delta\psi_N|\delta\psi_N\rangle$ must tend to zero, where $|\delta\psi_N\rangle = R_N^{(0)}(\hat{H})|\psi\rangle - f(\hat{H})|\psi\rangle$. It will be proven elsewhere [10] that, for the evolution and gaussian energy-filtering operators, convergence is obtained with any regular $n(E)$ which is non-zero where $n_\psi(E)$ is non-zero. The proof is simple if there exists a constant λ such that $n_\psi(E) < \lambda n(E)$ for all E, which is the case for many systems. Then $\Delta_{n_\psi}(R_N^{(0)}, f) < \lambda \Delta_n(R_N^{(0)}, f) \to 0$, and since $\Delta_{n_\psi}(R_N^{(0)}, f) = \langle \delta\psi_N|\delta\psi_N\rangle$, Eq. (3) is demonstrated. Second, let us consider the coefficients $\langle P_n|f\rangle = \int dE n(E) P_n(E) f(E)$ which can be calculated either from their integral form, or by a more convenient method described elsewhere. [10] In the case of the evolution operator ($f(E) = \exp(-iEt/\hbar)$), one can prove the majoration $|\langle P_n|f\rangle| \leq \alpha^n/(1-\alpha/n)n!$, where $n > \alpha = Bt/\hbar$ and B is the half-width of the energy interval where $n(E)$ is non-zero. Finally, from the recurrence relations between orthogonal polynomials, the norm of the vectors $P_n(\hat{H})|\psi\rangle$ is bounded by A^n, where A is a constant. Thus, in the case of the evolution operator, the n^{th} term of the series (3) has a norm bounded by $(A\alpha)^n/(1-\alpha/n)n!$, and the series converges absolutely and quickly : this allows a good summation where neglected terms are very small. Fast convergence is also obtained in the case of the gaussian energy-filtering function $f(E) = \exp[-(E-E_0)^2/\Delta^2]$.

Focusing on the evolution operator, several choices for $n(E)$, corresponding to different polynomial expansions (3), are now discussed. The most popular polynomial expansion is based on Chebyshev polynomials of the first kind, associated to a weight-function $n(E) = 1/(2\pi b\sqrt{1-(E-a)^2/4b^2})$ defined on an interval $[a-2b, a+2b]$ which contains the whole spectrum of $\hat{H}$. It is numerically stable and efficient for computing evolution of states with broad LDoS in systems with single-band regular DoS. But it is well-known that this method is not efficient for states with peaked LDoS such as energy-filtered states.[1]

From the integral expression of the error, $\langle \delta\psi_N|\delta\psi_N\rangle = \Delta_{n_\psi}(R_N, f)$, best accuracy is obtained with a development on the orthogonal polynomials associated to the LDoS $n_\psi(E)$. This choice is equivalent to the Lanczos method: the states $P_n(\hat{H})|\psi\rangle$ form an orthonormal basis which can be obtained by Lanczos tridiagonalization of $\hat{H}$, while the recurrence coefficients a_n, b_n are just the corresponding matrix elements. But despite its optimal character for any LDoS, the Lanczos method is subject to numerical instabilities, due to finite computer precision. We have tested it at large order N on various tight-binding hamiltonians. The long-time evolution was computed by iterating the algorithm with the same polynomial: $|\psi(kt)\rangle \simeq [R_N^{(0)}(\hat{H}, t)]^k |\psi(0)\rangle$. An instability was shown to occur after several time-steps t, because of band-edge components of $|\psi(0)\rangle$ growing exponentially and thus leading to non-conservation of unitarity. Reasons were identified as roundoff errors combined with a bad interpolation of $\exp(-iEt/\hbar)$ by $R_N^{(0)}(E, t)$ close to the band-edges.

The method proposed here combines the advantages of Chebyshev and Lanczos schemes. It is close to the Lanczos scheme, hence nearly optimized, but it is numerically stable in any situation. A new weight-function $n(E)$ is derived from $n_\psi(E)$ by convolution with a narrow semi-elliptic function centered around $E = 0$. As $n(E)$ is close to $n_\psi(E)$, the error $\langle \delta\psi_N|\delta\psi_N\rangle$ is nearly minimal, but each energy band is slightly enlarged, yielding to a good polynomial interpolation at the band-edges. The corresponding recurrence coefficients a_n, b_n are directly calculated from the initial a_n^ψ, b_n^ψ as described elsewhere.[10,11] It has been tested at large order $N \simeq 50$ on many tight-binding systems with regular, singular or fractal spectra : 2D and 3D disordered or quasiperiodic lattices and random magnetic flux lattices. The method is stable, and numerical efficiency is as good as the ideal Lanczos method. To test accuracy, conservation of the LDoS has been checked. Moreover, no significant difference was observed when using double machine precision or when increasing the number of terms in Eq. (3).

For sake of clarity, let us recapitulate the numerical algorithms used for propagation and energy-filtering. Both are based on the development (3) of an operator $f(\hat{H})$. First a weight-function $n(E)$ and its associated orthogonal polynomials P_n are derived by convolution of the LDoS. For propagation during a time t, $f(E) = \exp(-iEt/\hbar)$, and the coefficients $\langle P_n|f\rangle$ are computed. Using the recurrence relation between polynomials, each vector $P_n(\hat{H})|\psi\rangle$ is evaluated recursively from the two previous vectors, and summed in the series (3). For large systems, most of the computer memory used by the algorithm is occupied by these three quantum states, and the N iterations of $\hat{H}$ done during the recurrence

are the most time-consuming steps. Since $|\psi(t)\rangle$ has the same LDoS than $|\psi(0)\rangle$, propagation at next time $2t$ is obtained by doing exactly the same recursive summation based on the same weight-function $n(E)$, but starting from the initial state $|\psi(t)\rangle$, and so on. Gaussian energy-filtering is based on the same principles. We start from an extended random-phase state, whose LDoS is close to the total DoS. An energy E_0 and a width Δ are chosen, and the coefficients $\langle P_n|f\rangle$ associated to $f(E) = \exp[-(E-E_0)^2/\Delta^2]$ are computed. Then the partial series (3) is summed recursively with a sufficient number N of terms, as for the propagation algorithm, and a filtered-state is obtained.

Comparison with other propagation methods

The optimized propagation algorithm is used with a polynomial of degree $N = 50$, and compared to the second-order Runge-Kutta and SOD methods. The system considered here is a semi-infinite chain with nearest-neighbour hopping and we study the evolution of a state initially localized on the first site. In fact, by tridiagonalization of the hamiltonian, any propagation problem in 2D or 3D can be reduced to such a propagation on a semi-infinite chain. Thus the test performed here in 1D gives also a good estimate of the accuracy of each method in 2D or 3D, but without taking into account errors due to finite machine precision.

The semi-infinite chain considered here corresponds to the propagation of an initially localized state on an infinite periodic 1D chain. Thus onsite energies are zero, the first hopping energy is $s\sqrt{2}$, and all other hopping energies are s. The propagation is computed until time $T = 100\hbar/s$, and the error is defined as the norm of $|\delta\psi\rangle = |\psi\rangle_{computed} - |\psi\rangle_{exact}$. For each method, accuracy is improved by using smaller time-steps and thus increasing the numerical cost, defined as the total number N_{it} of iterations of $\hat{H}$. The results are presented in Fig. 1. First we notice that for the second order Runge-Kutta and SOD methods, the error scales as $1/N_{it}^2$, in agreement with the following rough estimate. The error at each time-step Δt is of order Δt^3. The total error is the sum of errors due to N_{it} time-steps $\Delta t \sim 1/N_{it}$, thus it is proportional to $1/N_{it}^2$. With the optimized polynomial method, the error decreases much more quickly and it is generally much more efficient than second-order methods. 350 iterations of $\hat{H}$ are sufficient to obtain an accuracy of 10^{-6}, while more than 800000 iterations are needed with the SOD method.

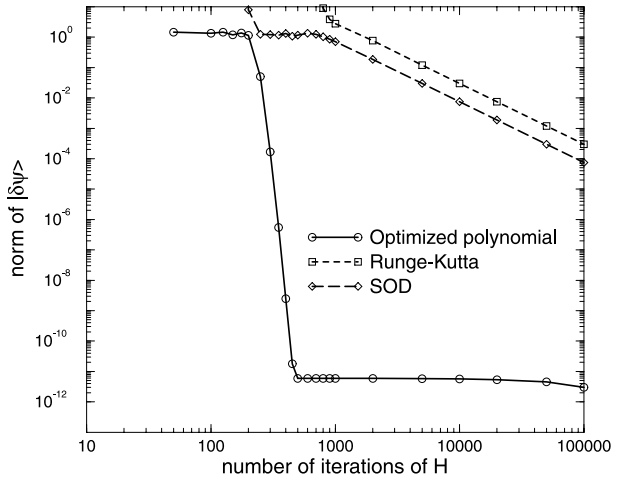

Fig. 1. Comparison of three different propagation methods in a 1D periodic system : accuracy versus numerical cost.

In this case, the optimized polynomial method is not more efficient than the Chebyshev one, because the state is not energy-filtered. Comparison with the Chebyshev method will be presented elsewhere [10] for energy-filtered wave-packets.

Periodic boundary conditions

To complete our presentation of the numerical method, problems related to finite sample size are discussed. When studying wave-packet dynamics, boundaries of the system are quickly reached, especially when the state is energy-filtered, in which case it is often initially extended to the whole system. Finite-size effects can not be removed, but a general way to suppress boundary effects is to use periodic boundary conditions (PBC). In order to study the diffusivity $D_\psi(t) = \langle\psi|(\hat{X}(t) - \hat{X}(0))^2|\psi\rangle/t$, we introduce the operator :

$$\hat{A}(t) = \exp\left(\frac{2i\pi\hat{X}(t)}{L}\right)\exp\left(\frac{-2i\pi\hat{X}(0)}{L}\right) - \mathbb{1} \qquad (4)$$

where L is the system length along the x direction, and $\exp(2i\pi\hat{X}(t)/L) = \exp(i\hat{H}t/\hbar)\exp(2i\pi\hat{X}/L)\exp(-i\hat{H}t/\hbar)$ (note that a similar operator has been introduced by R. Resta in order to study polarization effects). [12] In the limit of short times t or large L, when the wave-packet has spread on a small distance compared to L, $\hat{A}(t)$ is close to $2i\pi(\hat{X}(t) - \hat{X}(0))/L$, as proved in Ref. 10. Introducing the function

$$I_\psi(t) = \langle\psi|\hat{A}^+(t)\hat{A}(t)|\psi\rangle \qquad (5)$$

the diffusivity can be approximated as :

$$D_\psi(t) \simeq \frac{1}{t}\frac{L^2}{4\pi^2}I_\psi(t) \qquad (6)$$

and the equality holds in the large L limit. Since boundary effects have been suppressed, the L-behaviour of the approximate diffusivity (6) is expected to be more regular, and reliable extrapolations for $L \to +\infty$ can be made.

Application : random-flux lattice

We consider the so-called 2D random magnetic systems. The nature of eigenstates in such systems is still a serious matter of concern since the different numerical approaches which have been used [13,14] lead to contradictions. But all these studies gave similar results concerning the localization of states close to the band-edges. Such states are considered here in order to test our method, with the same model parameters as in reference. [14] The lattice is $200a \times 200a$ large with PBC and the hopping terms have modulus s. For different configurations of random fluxes, we have obtained filtered states with a very small energy-width ranging from 0.3% to 1% of the total bandwidth. The long-time evolution of $I_\psi(t) = \langle\psi|\hat{A}^+(t)\hat{A}(t)|\psi\rangle$, related to the diffusivity (Eq. (6)), is shown in Fig. 2 for states filtered at $E = -3.35\,\mathrm{s}$

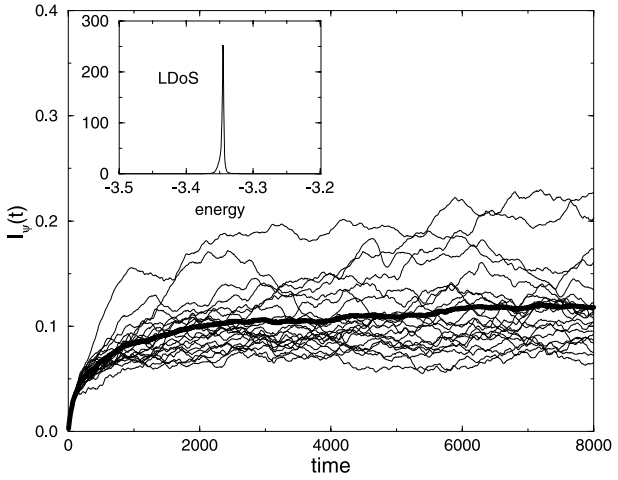

Fig. 2. Time-evolution of $I_\psi(t) \simeq 4\pi^2\langle(\hat{X}(t)-\hat{X}(0))^2\rangle/L^2$ for energy-filtered states on a random-flux lattice. The bold curve gives the average over 21 configurations and a typical LDoS is shown in inset. Time is in $\hbar/s$ units.

and 21 configurations of random-flux. The calculated localization length, $\xi = 13a$, is much smaller than the length $L = 200a$ of the system, which justifies the use of approximation (6). This value is in good agreement with,[13,14] and the scaling of ξ with energy has also been verified. In conclusion, a sharp energy-filtering has been obtained, much better than in Ref. 14. Since the filtered states are linear combinations of many localized eigenstates, they are extended to the whole sample, but the use of the function $I_\psi(t)$ has suppressed boundary effects.

Conclusion

We have developed new tools to study quantum wave-packet dynamics for tight-binding hamiltonians. These tools allow to produce energy-filtered states with high energy resolution, and to compute the evolution of wave-packets in an optimized and numerically stable manner. We have also shown how to suppress boundary effects for calculating diffusivity, by using periodic boundary conditions. These methods can be generalized to the study of propagation of other type of waves, acoustic or electromagnetic for example, provided that the system is spatially discretized.

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