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Berry phase and time-dependent wave operators

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Abstract

The present paper analyses the relation between the theory of the time-dependent wave operator and the Berry phase concept. It is proved that the wave operator approach is consistent with the non-adiabatic (Aharonov–Anandan) Berry phase, given that the wave operator and the parallel transport commute. It is then demonstrated that the non-Abelian Aharonov–Anandan phase can be calculated by working inside a reduced active space in the framework of wave operator theory. Finally an adiabatic transport formula is derived in the wave operator context and the influence of this effective Hamiltonian theory on the Berry phase is analysed. The theoretical results concerning the non-adiabatic Berry phase are confirmed numerically by considering a photodissociation process in the framework of the generalized Floquet theory.

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1. Introduction

The adiabatic transport formula is a standard approximation in quantum mechanics and the well-known simple adiabatic theorem [1] appears as a basic concept in theoretical physics. The generic form of an adiabatic theorem is the following. Let $U(t,0)$ be the evolution operator of a dynamical quantum system, and let $P_m(t)$ be an instantaneous spectral projector. If appropriate conditions reflecting the adiabatic limit are satisfied, then

$$U(t,0)P_m(0) = P_m(t)U(t,0). \tag{1}$$

In 1984 Berry [2] found that if the adiabatic theorem conditions are satisfied then the wavefunction of a quantum dynamical system issuing from $\psi(0) = |i,0\rangle$ at $t = 0$ is given by the adiabatic transport formula

$$\psi(t) = \mathrm{e}^{-\imath\hbar^{-1}\int_0^t \lambda_i(t')\mathrm{d}t' - \int_0^t \langle i,t'|\partial_{t'}|i,t'\rangle \mathrm{d}t'}|i,t\rangle, \tag{2}$$

where $|i,t\rangle$ is the eigenvector associated with a non-degenerate instantaneous eigenvalue $\lambda_i(t)$ which is permanently isolated from the rest of the Hamiltonian spectrum. The extra phase term $\mathrm{e}^{-\int_0^t \langle i,t'|\partial_{t'}|i,t'\rangle \mathrm{d}t'}$ is called the Berry phase.

Next, Wilczek and Zee [3] proved that if the eigenvalue is M-fold degenerate then the wavefunction issuing from $\psi(0) = |i,a,0\rangle$ is

$$\psi(t) = \sum_{a=1}^{M} \mathrm{e}^{-\imath\hbar^{-1}\int_0^t \lambda_i(t')\mathrm{d}t'} \left[\mathbb{T}\,\mathrm{e}^{-\int_0^t A(t')\mathrm{d}t'}\right]_{ba} |i,b,t\rangle \tag{3}$$

where $(|i,a,t\rangle)_{a=1,\ldots,M}$ is an orthonormal basis of the eigensubspace of $\lambda_i(t)$, $\mathbb{T}$ is the time-ordering operator, and A is a matrix with elements $A_{ab}(t) = \langle i,a,t|\partial_t|i,b,t\rangle$. The unitary matrix $\mathbb{T}\,\mathrm{e}^{-\int_0^t A(t')\mathrm{d}t'}$ is called a non-Abelian Berry phase.

More generally, let $H(t)$ be a time-dependent Hamiltonian with eigenvalues $\{\lambda_i(t)\}_i$ and eigenvectors $\{|i,t\rangle\}_i$ (the case of a n-fold degenerate eigenvalue is not excluded, but in this case $\exists i_1 \neq \cdots \neq i_n$ such that $\forall t \lambda_{i_1} = \cdots = \lambda_{i_n}$ and $\{|i_1,t\rangle,\ldots,|i_n,t\rangle\}$ is then an arbitrary orthonormal basis set of the eigensubspace). Suppose that there exists a set of indices I such that $P_m(t) = \sum_{i\in I} |i,t\rangle\langle i,t|$ satisfies the adiabatic theorem (equation (1)). If $\psi(0) = |i,0\rangle$, then

$$\psi(t) = \sum_{j\in I} \left[\mathbb{T}\,\mathrm{e}^{-\imath\hbar^{-1}\int_0^t E(t')\mathrm{d}t' - \int_0^t A(t')\mathrm{d}t'}\right]_{ji} |j,t\rangle \tag{4}$$

where $\forall i,\ j \in I$, $E_{ij}(t) = \lambda_i(t)\delta_{ij}$ and $A_{ij}(t) = \langle i,t|\partial_t|j,t\rangle \cdot \mathrm{Ran}P_m(t)$ is called an adiabatic active subspace.

The selection of the adiabatic active subspace within which the dynamics is confined is a central problem. In most of the applications this subspace is spanned by the eigenvectors whose eigenvalues are related to the initial eigenvalue $\lambda_i(0)$ by crossings (or avoided crossings) appearing at one or more times during the dynamical process. This result merits further comment. First, the selection of the active subspace involves knowing $\{\lambda_i(t)\}_i$ for all t; this represents a large amount of numerical work. Second, such an active subspace is, at each time, globally isolated from the rest of the spectrum in agreement with equation (1). Nevertheless this subspace is generally quite different from a pure degenerate eigenspace such as that involved in equation (3). It thus appears necessary to adapt the pure adiabatic formulation to take into account the real composition of the adiabatic active subspace. The Bloch wave operator theory possesses some interesting features which facilitate the solution of such a problem. This theory is an effective Hamiltonian theory which was developed many years ago within nuclear physics and quantum chemistry [4–6] to overcome some limitations of *ab initio* methods. It was extended recently to time-evolution processes and the time-dependent wave operator concept [7] was introduced to solve the Schrödinger equation in large Hilbert spaces $\mathcal{H}$. The main concepts of the wave operator theory are the existence of a suitably chosen small subspace S_0 of $\mathcal{H}$ (called the wave operator active subspace) within which the Schrödinger equation is integrated by using an effective Hamiltonian, and the existence of the wave operator which reconstructs the exact solution in the full space when it is applied to the projected solution.

The efficiency of the wave operator theory for the description of adiabatic processes has been revealed in a previous paper [8], which introduced an adiabatic theorem for the time-dependent wave operator. This theorem identifies the adiabatic active subspace at $t = 0$ with the wave operator active subspace. The active subspace $\mathrm{Ran}P_m$ then appears as central and should be selected at the beginning of a calculation. Usually the wave operator active subspace incorporates the strong and the fast part of the dynamics only, but at the adiabatic limit it is defined as the space within which the full dynamics is confined. This constraining condition

increases the size of the required active space. This feature can be a strong handicap. For instance, in chaotic processes exhibiting strong non-adiabatic couplings, the wavefunction 'explores' the major part of the Hilbert space, which makes the use of the adiabatic wave operator theory very difficult. We continue here the analysis of the time-dependent wave operator theory, while assuming that such a fully chaotic situation is not present.

In this context, the mathematical framework of our study can be summarized as follows. Let $t \mapsto P(t)$ be a curve in the space of projectors satisfying the Schrödinger–von Neumann equation :

$$\imath\hbar \frac{\mathrm{d}P(t)}{\mathrm{d}t} = [H(t), P(t)]. \tag{5}$$

The Berry phase phenomenon appears as a horizontal lift of this curve in the Hilbert space fibred over the projector space. Suppose that the Hilbert space is n-dimensional, $\mathcal{H} = \mathbb{C}^n$. For the Abelian case, the space of projectors is the complex projective space $\mathbb{C}P^{n-1}$, and the Berry phase phenomenon is related to the principal bundle with base space $\mathbb{C}P^{n-1}$ and with the group of phases $U(1)$ as typical fibre. In the non-Abelian case, the space of M-fold projectors is known to be the complex Grassmanian manifold $G_M(\mathbb{C}^n)$. The principal bundle related to the non-Abelian Berry phase phenomenon then has as total space $\mathbb{C}^n$, with $U(M)$ as typical fibre. We find the wavefunction by considering the parallel transport of an initial state, associated with the horizontal lift, in the associated vector bundle. In the adiabatic limit, the projectors involved in the projector space are only spectral projectors associated with the eigenvectors selected by the adiabatic theorem. In the generic situation of an infinite-dimensional Hilbert space a projector space obtained by inductive limit can be defined; we denote it by $\mathbb{C}P^{\infty}$ or $G_M(\mathbb{C}^{\infty})$. This mathematical structure was first described by Simon [9]; a complete exposition of the fibre bundle theory applied to the Berry phase concept can be found in [10]. The important fact is that the Berry phenomenon involves two ingredients, the time-dependent projector $P(t)$ sited in the base space, and the time-dependent phase (or unitary matrix) associated with the horizontal lift sited in the total space of the bundle. In [8] we proved that the wave operator is a succession of instantaneous Bloch wave operators at the adiabatic limit, demonstrating in this way that the wave operator is rigorously related to spectral projectors at the adiabatic limit. This result is related to the behaviour of the wave operator treatment in the projector space, but it does not elucidate the relationship between the wave operator and the Berry phase. In particular, it is important to know if the wave operator treatment induces an additional phase factor and if it is possible to compute the Berry phase by using the effective Hamiltonian of the wave operator theory in place of the true Hamiltonian. Moreover, the Berry phase phenomenon is not limited to the adiabatic case; for a generic cyclic evolution it appears a non-adiabatic Berry phase, the so-called Aharonov–Anandan phase. The relation between the wave operator and the Berry phase thus must be discussed in a more general context than the adiabatic limit. That is the subject of this paper.

This paper is organized as follows. Section 2 is devoted to a short summary of the wave operator theory. Section 3 analyses the non-adiabatic Berry phase phenomenon which appears in the wave operator method and illustrates our Berry phase equations by considering a photodissociation process in the framework of the generalized Floquet theory. Finally, section 4 comments further on the adiabatic transport concept as treated by the wave operator theory.

We note that there is some ambiguity in the use of the term 'adiabatic'. Sometimes a system is called adiabatic if it can be described by using a small number of states, in other words if the dynamics can be projected into a small active space isolated from the rest of the Hilbert space. However, the term is also used in the literature in a more restrictive sense, to imply that the adiabatic theorem is satisfied. In this case the time-variations of the external

perturbations are slow with respect to the proper quantum time and the active subspace is then a spectral subspace. The two different senses of 'adiabatic' are used (and distinguished) in the present paper. In section 3 we consider a system (the H_2^+ molecule submitted to a laser pulse) which does not satisfy any adiabatic theorem but for which there exists an isolated active space. The Berry phases are called 'non-adiabatic' because they are not the consequence of an adiabatic theorem. By contrast, a system which satisfies an adiabatic theorem is considered in section 4 and the subsequent Berry phase is called adiabatic. To remove any ambiguity we will distinguish the systems which satisfy an adiabatic theorem by using the expression 'adiabatic in the strong sense'. For systems solely characterized by an isolated active space, the expression 'adiabatic in the weak sense' will be adopted. In this context the expression 'adiabatic limit' introduced when the interaction duration tends to infinity refers to adiabaticity in the strong sense.

2. A review of the wave operator theory

This section gives a brief review of the wave operator approach. More details can be found in [4, 5, 11–14] and in the review papers [6, 7]. Stationary Bloch wave operator theory and time-dependent wave operator theory are closely related. Both are effective Hamiltonian theories which solve the basic equations (eigenequations and time-dependent evolution equations) in small active subspaces before reconstituting the full exact solutions in the full Hilbert space.

Consider, for example, the eigenvalue problem $H\psi = \lambda\psi$, where H is an operator in a separable Hilbert space $\mathcal{H}$. Let S_0 and S be two subspaces of $\mathcal{H}$ such that $S_0 \cap S^\perp = S_0^\perp \cap S = \{0\}$. We call them active and target subspaces and we denote the projectors of these spaces by P_0 and P, respectively. We are interested in eigenvectors included in S such that $P\psi = \psi$. We reduce the problem to one within the active subspace by solving the new equation

$$H^{\text{eff}}\psi_0 = \lambda\psi_0, \tag{6}$$

where the effective operator is defined by $H^{\text{eff}} = P_0 H\Omega$. The wave operator is formally defined by the expression $\Omega = P(P_0PP_0)^{-1}$, where $(P_0PP_0)^{-1}$ is the inverse of P within S_0. It connects the projected and the exact solutions ($\psi = \Omega\psi_0$) and is itself the solution of a nonlinear generalized eigenvalue equation

$$H\Omega = \Omega H\Omega = \Omega H_{\text{eff}}. \tag{7}$$

In the time-dependent case the dynamical system is described by a time-dependent Hamiltonian $H(t)$ in a separable Hilbert space $\mathcal{H}$ and by the evolution operator $U(t)$ ($\forall t \in [0, T]$) which is a solution of the Schrödinger equation. The effective Hamiltonian (which describes the approximate dynamics in S_0) is defined by $H^{\text{eff}}(t) = P_0H(t)\Omega(t)$ and the target space is $S(t) = P_0U(t)S_0 \subset S_0$, where P_0 is the projector on S_0. The time-dependent wave operator $\Omega(t) = P_0 + X(t)$ with ($X = Q_0XP_0$, Q_0 being the projector on the orthogonal supplement of S_0) is given by the formal expression

$$\Omega(t) = U(t)(P_0U(t)P_0)^{-1}, \tag{8}$$

where $(P_0U(t)P_0)^{-1} = P_0(P_0U(t)P_0)^{-1}P_0$ represents an inversion within S_0 ($\text{Dom}(P_0U(t)P_0)^{-1} = S_0$).
In the active space, the projected equation

$$\imath\hbar\partial_t\psi_0(t) = H^{\text{eff}}(t)\psi_0(t) \tag{9}$$

takes the place of the Schrödinger equation $\imath\hbar\partial_t\psi(t) = H(t)\psi(t)$, and $\Omega(t)$ connects the two solutions $\psi(t) = \Omega(t)\psi^0(t) = \psi^0(t) + X(t)\psi^0(t)$ in such a way that $X(t)\psi^0(t)$ is the component of $\psi(t)$ orthogonal to S_0.

The evolution of the reduced wave operator $X(t)$ is governed by the nonlinear equation [7]

$$\imath\hbar\frac{\partial X}{\partial t} = Q_0(1-X)H(1+X)P_0, \tag{10}$$

which can be rewritten as [7]

$$(H - \imath\hbar\partial_t)\Omega = \Omega(H^{\text{eff}} - \imath\hbar\partial_t). \tag{11}$$

Equation (11) is identical to the fundamental equation of the Bloch wave operator theory (equation (6)) if the Floquet Hamiltonian $H_F = H - \imath\hbar\partial_t$ (acting in a generalized Hilbert space) is taken in place of the usual Hamiltonian H. This shows the close relationship between time-dependent and stationary wave operator formulations and also the consistency of the time-dependent wave operator theory with Floquet theory.

Various iterative methods have been proposed for the integration of the wave operator equation, e.g., the recursive distorted wave approximation (RDWA) and the single cycle method (SCM). These procedures are robust and are able to work with Hamiltonians which are rotated in the complex plane or which include added complex absorbing potentials. Details concerning these algorithms can be found in [6, 7, 12, 15].

The RDWA algorithm is used here in conjunction with the *wave operator sorting algorithm* (WOSA) proposed by Wyatt [16] as a technique to select the active subspace. At the adiabatic limit, the model space includes all the selected states reordered by WOSA and with corresponding Floquet eigenstates which possess a non-negligible overlap with the initial state.

In a previous work [7], we conjectured that the adiabatic limit of the time-dependent wave operator is given by a succession of instantaneous Bloch wave operators. In other words,

$$\lim_{T\to+\infty}\Omega_T(s) = P(s)(P(0)P(s)P(0))^{-1}, \tag{12}$$

where $\sigma(t)$ is a spectral group of discrete eigenvalues with the projector $P(t)$ and labelled by the set of indices I. We choose $\text{Ran}P(0)$ as the active space S_0 and $\text{Ran}P(s)$ as the target space $S(s)$ (s is the reduced time on the interval $[0, T]$, i.e. $s = t/T$). $U_T(s)$ is the evolution operator and $\Omega_T(s) = U_T(s)(P(0)U_T(s)P(0))^{-1}$ is the time-dependent wave operator. The conjectured result (12) was actually later demonstrated (in [8]).

This expression reveals that the adiabatic limit of the wave operator is a pure stationary operator without any rapid phase terms, by contrast with the equivalent adiabatic limit of the wavefunction, which includes both dynamical and Berry phases. Rapid phases are nevertheless present in the wave operator formulation, but they are separated from the stationary part and are incorporated into the effective Hamiltonian. This splitting makes the calculation of the wave operator more easy. Moreover the wave operator theory provides a consistent approach to the adiabatic transport problem (in the strong sense of adiabaticity). We analyse in the next sections how the Berry phase can be derived by working inside the reduced active space and how a generalized non-adiabatic non-Abelian Berry phase appears in the wave operator theory when the adiabatic conditions are satisfied in multi-dimensional spaces.

3. Non-adiabatic Berry phase and wave operators

In 1987, Aharonov and Anandan [17] found that the Berry phase is not an exclusive feature of the adiabatic approximation but also characterizes a cyclic evolution. Let $t \mapsto \psi(t)$ be the wavefunction, a solution of the Schrödinger equation of a quantum dynamical system with

Hamiltonian $H(t)$. We suppose that the evolution is cyclic, i.e. $\psi(T) = \mathrm{e}^{\imath\varphi}\psi(0)$. In this case there exists a gauge transformation $\tilde{\psi}(t) = \mathrm{e}^{-\imath\gamma(t)}\psi(t)$ such that $\tilde{\psi}(T) = \tilde{\psi}(0)$, namely

$$\psi(t) = \mathrm{e}^{-\imath\hbar^{-1}\int_0^t \langle\tilde{\psi}(t')|H(t')|\tilde{\psi}(t')\rangle \mathrm{d}t' - \int_0^t \langle\tilde{\psi}(t')|\partial_{t'}|\tilde{\psi}(t')\rangle \mathrm{d}t'}\tilde{\psi}(t). \tag{13}$$

The phase factor $\mathrm{e}^{-\int_0^t \langle\tilde{\psi}(t')|\partial_{t'}|\tilde{\psi}(t')\rangle \mathrm{d}t'}$ is called the non-adiabatic Berry phase (or Aharonov–Anandan phase).

Equation (13) is also a parallel transport formula. Indeed, consider $t \mapsto P(t) = |\tilde{\psi}(t)\rangle\langle\tilde{\psi}(t)|$ as a map in the complex projective space $\mathbb{C}P^\infty$. $t \mapsto \tilde{\psi}(t)$ is an arbitrary section of the line bundle with base space $\mathbb{C}P^\infty$, total space $\mathcal{H}$ and typical fibre $\mathbb{C}$. This bundle is endowed with a connection defined by the gauge potential which is expressed, in the convention of this section, as

$$A(t) = \imath\hbar^{-1}\langle\tilde{\psi}(t)|H(t)|\tilde{\psi}(t)\rangle\,\mathrm{d}t + \langle\tilde{\psi}(t)|\partial_t|\tilde{\psi}(t)\rangle\,\mathrm{d}t. \tag{14}$$

The parallel transport of the state $\psi(0)$ which is in the fibre $\mathbb{C}$ over $P(0)$, i.e. the horizontal lift of the curve $t \mapsto P(t)$ passing by $\psi(0)$, takes the form (13). A discussion about the connection inducing both the Berry phase and the dynamical phase can be found in [18].

Our study analyses the compatibility of the wave operator theory with the Berry phase concept. More precisely, we want show that it is possible to compute the total (Abelian or non-Abelian) phase defining the parallel transport by using only operators of the wave operator effective theory in place of the true operators.

3.1. The one-dimensional case

Let $\psi^0(t)$ be the solution of the effective Schrödinger equation (9) with $P_0 = |i\rangle\langle i|$. We suppose that $\psi^0(T) = \mathrm{e}^{\imath\varphi}\psi^0(0)$. We can then introduce a parallel transport equation into the active space. Let $\mathrm{e}^{-\imath f(t)}$, so that $\tilde{\psi}^0(t) = \mathrm{e}^{-\imath f(t)}\psi^0(t)$ satisfies $\tilde{\psi}^0(T) = \tilde{\psi}^0(0)$. The gauge transformation is determined by requiring $\mathrm{e}^{\imath f(t)}\tilde{\psi}^0$ to be a solution of the effective Schrödinger equation

$$\imath\hbar\partial_t\psi^0(t) = H^{\mathrm{eff}}(t)\psi^0(t) \tag{15}$$

so that

$$-\hbar f'(t)\tilde{\psi}^0(t) + \imath\hbar\partial_t\tilde{\psi}^0(t) = H^{\mathrm{eff}}\tilde{\psi}^0(t). \tag{16}$$

By projecting this equation onto $\tilde{\psi}^0(t)$ we have

$$\imath f'(t) = -\frac{\langle\tilde{\psi}^0(t)|\partial_t|\tilde{\psi}^0(t)\rangle}{\langle\tilde{\psi}^0(t)|\tilde{\psi}^0(t)\rangle} - \imath\hbar^{-1}\frac{\langle\tilde{\psi}^0(t)|H^{\mathrm{eff}}|\tilde{\psi}^0(t)\rangle}{\langle\tilde{\psi}^0(t)|\tilde{\psi}^0(t)\rangle}. \tag{17}$$

$\psi^0(t) = \mathrm{e}^{-\imath\hbar^{-1}\int_0^t \frac{\langle\tilde{\psi}^0(t')|H^{\mathrm{eff}}|\psi^0(t')\rangle}{\langle\tilde{\psi}^0(t')|\tilde{\psi}^0(t')\rangle}\mathrm{d}t' - \int_0^t \frac{\langle\tilde{\psi}^0(t)|\partial_{t'}|\tilde{\psi}^0(t')\rangle}{\langle\tilde{\psi}^0(t')|\tilde{\psi}^0(t')\rangle}\mathrm{d}t'}\tilde{\psi}^0(t)$ is a parallel transport associated with the unnormalized section $\tilde{\psi}^0(t)$. The question is, since ψ^0 is a parallel transport associated with the section $\tilde{\psi}^0$, is $\Omega(t)\psi^0(t)$ a parallel transport and (if so) for which section?

Let $\Omega(t)$ be the wave operator; and define $|\psi(t)\rangle$ as being the normalized wavefunction computed from $\psi^0(t)$ by $\Omega(t) : |\psi(t)\rangle = \Omega(t)|\psi^0(t)\rangle$. It is clear that

$$\psi(t) = \mathrm{e}^{-\imath\hbar^{-1}\int_0^t \frac{\langle\tilde{\psi}^0(t')|H^{\mathrm{eff}}|\tilde{\psi}^0(t')\rangle}{\langle\tilde{\psi}^0(t')|\tilde{\psi}^0(t')\rangle}\mathrm{d}t' - \int_0^t \frac{\langle\tilde{\psi}^0(t)|\partial_{t'}|\tilde{\psi}^0(t')\rangle}{\langle\tilde{\psi}^0(t')|\tilde{\psi}^0(t')\rangle}\mathrm{d}t'}\Omega(t)\tilde{\psi}^0(t). \tag{18}$$

This expression does not seem to be a parallel transport. Its phase is described by using $\tilde{\psi}^0$, although the reference section seems to be $\Omega\tilde{\psi}^0$. But we will prove that, in fact, the phase computed with $\tilde{\psi}^0$ is equal to the phase computed with $\Omega\tilde{\psi}^0$. By the definition of the wave operator, we have $\Omega\psi^0 = \psi$, thus

$$\Omega(t) = \frac{|\psi(t)\rangle\langle i|}{\langle i|\psi(t)\rangle} \tag{19}$$

and

$$\Omega^{\dagger}\Omega = \frac{|i\rangle\langle i|}{\langle\psi^0|\psi^0\rangle}. \tag{20}$$

Since $\tilde{\psi}^0$ differs from ψ^0 only by a phase factor, we have

$$\Omega^{\dagger}\Omega = \frac{|i\rangle\langle i|}{\langle\tilde{\psi}^0|\tilde{\psi}^0\rangle}. \tag{21}$$

By using the equation $(H - \imath\hbar\partial_t)\Omega = \Omega(H^{\text{eff}} - \imath\hbar\partial_t)$ we find that

$$-\imath f'(t) = \imath\hbar^{-1}\frac{\langle\tilde{\psi}^0(t)|H^{\text{eff}}(t)|\tilde{\psi}^0(t)\rangle}{\langle\tilde{\psi}^0(t)|\tilde{\psi}^0(t)\rangle} + \frac{\langle\tilde{\psi}^0(t)|\partial_t|\tilde{\psi}^0(t)\rangle}{\langle\tilde{\psi}^0(t)|\tilde{\psi}^0(t)\rangle} \tag{22}$$

$$= \imath\hbar^{-1}\langle\tilde{\psi}^0(t)|\Omega^{\dagger}\Omega H^{\text{eff}}(t)|\tilde{\psi}^0(t)\rangle + \langle\tilde{\psi}^0(t)|\Omega^{\dagger}\Omega\partial_t|\tilde{\psi}^0(t)\rangle \tag{23}$$

$$= \imath\hbar^{-1}\langle\tilde{\psi}^0(t)|\Omega^{\dagger}\Omega(H^{\text{eff}}(t) - \imath\hbar\partial_t)|\tilde{\psi}^0(t)\rangle \tag{24}$$

$$= \imath\hbar^{-1}\langle\tilde{\psi}^0(t)|\Omega^{\dagger}(H(t) - \imath\hbar\partial_t)\Omega|\tilde{\psi}^0(t)\rangle \tag{25}$$

$$= \imath\hbar^{-1}\langle\Omega(t)\tilde{\psi}^0(t)|H(t)|\Omega(t)\tilde{\psi}^0(t)\rangle + \langle\Omega(t)\tilde{\psi}^0(t)|\partial_t|\Omega(t)\tilde{\psi}^{(}t)\rangle \tag{26}$$

$$\tag{27}$$

and finally

$$|\psi(t)\rangle = \mathrm{e}^{-\imath\hbar^{-1}\int_0^t\langle\Omega(t')\tilde{\psi}^0(t')|H(t')|\Omega(t')\tilde{\psi}^0(t')\rangle\mathrm{d}t' - \int_0^t\langle\Omega(t')\tilde{\psi}^0(t')|\partial_{t'}|\Omega(t')\tilde{\psi}^0(t')\rangle\mathrm{d}t'}|\Omega(t)\tilde{\psi}^0(t)\rangle. \tag{28}$$

We find a structure similar to that described by equation (13). We conclude that if $\psi^0(t)$ is a parallel transport associated with the section $\tilde{\psi}^0(t)$, then $\psi(t) = \Omega(t)\psi^0(t)$ is a parallel transport associated with the section $\Omega(t)\tilde{\psi}^0(t) \equiv \tilde{\psi}(t)$. Moreover, we can compute the true phase by using the effective Hamiltonian.

The parallel transport in the effective Hamiltonian case involves another bundle structure than that for the usual parallel transport considering in (13), because it uses a non-selfadjoint Hamiltonian H^{eff} and an unnormalized function ψ^0. Moreover, ψ^0 is defined within S_0, which is one dimensional. The new total space of the bundle is $\mathbb{C} = S^0$ (the vector space generated by $|i\rangle$). $|\psi^0(t)\rangle\langle\psi^0(t)|$ is not an orthonormal projector, and the new base space $\mathbb{C}/U(1) = \mathbb{R}$ takes the place of $\mathbb{C}P^{\infty}$ ($|\psi^0(t)\rangle\langle\psi^0(t)|$ is in fact an unnormalized vector of $\mathbb{C}$ without phase, it represents the magnitude of ψ^0). If we consider the equation

$$\Omega(t)|\psi^0(t)\rangle\langle\psi^0(t)|\Omega(t)^{\dagger} = |\psi(t)\rangle\langle\psi(t)|, \tag{29}$$

we see that Ω transforms a path in $\mathbb{R}$ into a path in $\mathbb{C}P^{\infty}$. Let R^{eff} be the parallel transport operation with the effective Hamiltonian and R be the parallel transport operation in the whole Hilbert space defined by equation (13). Finally if we denote by $X^{[0,T]}$ the set of maps from $[0, T]$ to a space X, the equality (26) states that the following diagram commutes:

$$\begin{array}{ccc} \mathbb{C}^{[0,T]} & \xrightarrow{\Omega} & \mathcal{H}^{[0,T]} \\ {\scriptstyle R^{\text{eff}}}\uparrow & & \uparrow{\scriptstyle R} \\ \mathbb{R}^{[0,T]} & \xrightarrow{\Omega\cdot\Omega^{\dagger}} & (\mathbb{C}P^{\infty})^{[0,T]} \end{array}.$$

This diagram displays the fact that the parallel transport and the wave operator operations are commutative.

A simple illustration of the non-adiabatic Berry phase is given by the generalized Floquet Berry phase. Consider, for example, the interaction between a molecule and a short laser pulse such that the interaction vanishes at $t = 0$ and at $t = T$. One can transform this cyclic

system into a periodic system by repeating the interaction from $[0, T]$ into $[T, 2T]$, $[2T, 3T]$, etc. This mathematical artefact, together with the introduction of the Floquet Hamiltonian $H_F(t) = H(t) - \imath\hbar\partial_t$ in the new Hilbert space $\mathcal{H} \otimes L^2([0, T], \mathrm{d}t)$, constitutes the framework of the generalized Floquet theory [19]. The time-independent quasi-energies χ_i and the quasi-eigenvector $|\mu_i(t)\rangle$ constructed in the first Brillouin zone are defined by

$$H_F(t)|\mu_i(t)\rangle = \chi_i|\mu_i(t)\rangle. \tag{30}$$

If we suppose that $\psi(0) = |\mu_i(0)\rangle$ then we have

$$\psi(t) = \mathrm{e}^{-\imath\hbar^{-1}\chi_i t}|\mu_i(t)\rangle. \tag{31}$$

Following Moore and Steedman [20] this Floquet phase can be written as the sum of a dynamical phase and a geometrical Berry phase:

$$\imath\hbar^{-1}\chi_i T = \imath\hbar^{-1}\int_0^T \langle\mu_i(t')|H(t')|\mu_i(t')\rangle\,\mathrm{d}t' + \int_0^T \langle\mu_i(t)|\partial_t|\mu_i(t)\rangle\,\mathrm{d}t. \tag{32}$$

We thus conclude that the Floquet Berry phase is a particular case of the non-adiabatic Berry phase. The condition of periodicity which is added to the Aharonov–Anandan cyclic scheme imposes a total phase $(\chi_i t)$ whose time derivative is constant. To illustrate this non-adiabatic Berry phase in a one-dimensional space we consider the photodissociation of H_2^+, involving an excitation and a dissociation step:

$$H_2^+\left(^2\Sigma_g^+, v = 0, J = 0\right) + n\hbar\omega_o \rightarrow H_2^+\left(^2\Sigma_u^+\right) \rightarrow H^+ + H(1s). \tag{33}$$

The Hilbert space describing the H_2^+ molecule is $L^2(\mathbb{R}^+, \mathrm{d}r) \otimes \mathbb{C}^2 \otimes L^2([0, T], \mathrm{d}t)$, where $L^2(\mathbb{R}^+, \mathrm{d}r)$ is the Hilbert space describing the vibration of the molecule, r being the distance between the two nuclei. $\mathbb{C}^2$ is the Hilbert space of the electron. In this model we consider only the fundamental electronic state $^2\Sigma_g^+$ and the first excited state $^2\Sigma_u^+$. $L^2([0, T], \mathrm{d}t)$ is the Hilbert space which describes the time. The dressed Hamiltonian of H_2^+ interacting with the field is then

$$\begin{aligned} H_F = -\frac{\hbar^2}{2m}\frac{d^2}{\mathrm{d}r^2} \otimes I_2 \otimes I_T + \left(V_{^2\Sigma_g^+}(r) \otimes \left|^2\Sigma_g^+\right\rangle\!\left\langle^2\Sigma_g^+\right| + V_{^2\Sigma_u^+}(r) \otimes \left|^2\Sigma_u^+\right\rangle\!\left\langle^2\Sigma_u^+\right|\right) \otimes I_T \\ + \mu(r) \otimes \left(\left|^2\Sigma_u^+\right\rangle\!\left\langle^2\Sigma_g^+\right| + \left|^2\Sigma_g^+\right\rangle\!\left\langle^2\Sigma_u^+\right|\right) \otimes E(t)\cos(\omega_o t) - \imath\hbar I_R \otimes I_2 \otimes \partial_t \end{aligned} \tag{34}$$

where I_R, I_2 and I_T are the identity operators of $L^2(\mathbb{R}^+, \mathrm{d}r)$, $\mathbb{C}^2$ and $L^2([0, T], \mathrm{d}t)$, μ is the electric dipole moment, m is the reduced mass of the diatom, and $V_{^2\Sigma_{u/g}^+}(r)$ are potentials for the two electronic energy surfaces.

The laser is characterized by its carrier wave frequency $\omega_o = 0.295\,868$ au and by its pulse shape $E(t)$, defined as the following gaussian envelope function:

$$E(t) = \begin{cases} \epsilon_o \exp\left[-\left(\frac{t-t_1}{\tau}\right)^2\right] & \text{for} \quad t \leqslant t_1 \\ \epsilon_o & \text{for} \quad t_1 \leqslant t \leqslant t_2 \\ \epsilon_o \exp\left[-\left(\frac{t-t_2}{\tau}\right)^2\right] & \text{for} \quad t \geqslant t_2. \end{cases} \tag{35}$$

Previous studies have observed that the photoreactive relaxation issuing from the ground vibrational state $|v = 0\rangle$ of Σ_g^+ has a strongly adiabatic character (in the weak sense) over a wide field-intensity range, in the sense that only one quasi-eigenvector participates, i.e. $\psi(0) = |\mu_1(0)\rangle = |v = 0\rangle$. Note that this weak adiabatic condition implies that asymptotically the quasi-eigenvector has a single component: $\langle v|\mu_1(0)\rangle = \langle v|\mu_1(T)\rangle = \delta_{v,0}$.

In the framework of the generalized Floquet Berry phase, the function $\tilde{\psi}$ should be identified with the Floquet eigenvector, namely $\tilde{\psi}(t) = |\mu_1(t)\rangle$ and $\tilde{\psi}^0(t) = (\langle v = 0|\mu_1(t)\rangle)|v = 0\rangle$. By including these identities into equation (26), the formula $\gamma'(t) = f'(t)$

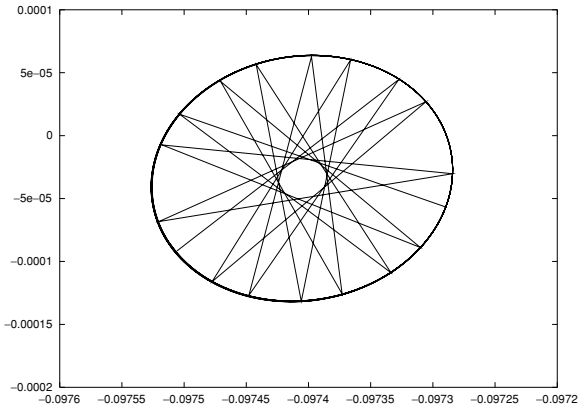

Figure 1. The trajectories in the complex plane of the two members of equation (36). The left-hand side is deduced from the quasi-eigenstate $|\mu_1(t)\rangle$ calculated at the discrete values of a DVR time basis set. The right-hand side is deduced from equation (38) by integration of the Schrödinger equation.

gives

$$\chi_1 + \mathrm{i}\hbar \frac{\langle \tilde{\psi}^0(t)|\partial_t \tilde{\psi}^0(t)\rangle}{\langle \tilde{\psi}^0(t)|\tilde{\psi}^0(t)\rangle} = \frac{\langle \tilde{\psi}^0|H^{\mathrm{eff}}|\tilde{\psi}^0(t)\rangle}{\langle \tilde{\psi}^0(t)|\tilde{\psi}^0(t)\rangle} \tag{36}$$

$$= \frac{\langle v=0|H(t)\mu_1(t)\rangle}{\langle v=0|\mu_1(t)\rangle} \tag{37}$$

$$= \frac{\langle v=0|H(t)\psi(t)\rangle}{\langle v=0|\psi(t)\rangle}. \tag{38}$$

Figure 1 represents the two members of this equation over the range ($t = 700$ au, t = 800 au) situated on the constant part of the envelope ($E(t) = \epsilon_0$) and for an intensity $I = 10^{12}$ W cm^{-2}. The left-hand side of (36) is calculated at the discrete values t_i of a DVR basis and corresponds to the tips of the straight segments. The right-hand side is calculated using equation (38) and gives the continuous elliptic curve. This figure reveals that equation (36) is perfectly obeyed. This proves both the almost adiabatic character (in the weak sense) of the process and the preservation of the gauge transformation after projection into the active space. A second proof of the adiabaticity (in the weak sense) is the periodicity of the trajectory ($\langle v=0|H(t)|\psi(t)\rangle/\langle v=0|\psi(t)\rangle$, constituted from a superposition of a few almost identical closed loops (see figure 1). This reflects the periodicity of the unique quasi-eigenvector $|\mu_1(t)\rangle$ which participates in the phase (equation (37)) when the amplitude and the frequency of the electric field are constant. The adiabaticity in the weak sense is confirmed by figure 2, which reveals an adiabatic photodissociation [21] (caused by the presence of complex energies) and not a non-adiabatic photodissociation (the bound states ($v > 0$) are never significantly populated).

The adiabatic character is destroyed for stronger amplitudes, $I = 10^{13}$ W cm^{-2} and $I = 10^{14}$ W cm^{-2}. Figures 3 and 4 make evident the non-periodicity of the function $\langle v=0|H\psi\rangle/\langle v=0|\psi\rangle$. This non-periodicity is the signature of a dynamics involving numerous quasi-eigenvectors. Note also that the size of the curves increases dramatically with the intensity of the field.

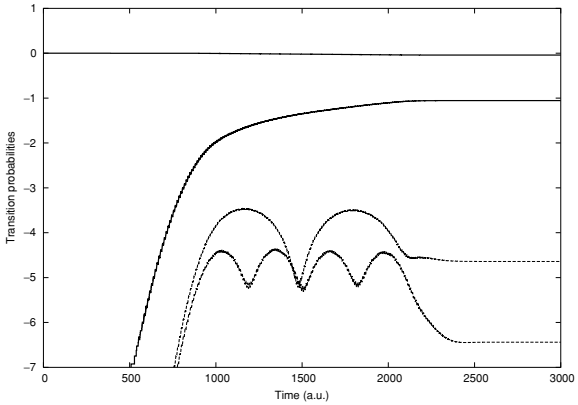

Figure 2. Dissociation probability and vibrational transition probabilities on a logarithmic scale as a function of time. Solid lines: survival probability $P_{0\to 0}$ and dissociation probability; dashed lines: $P_{0\to 1}$ and $P_{0\to 2}$. The small decreasing of the survival probability mainly supplies the dissociation probability.

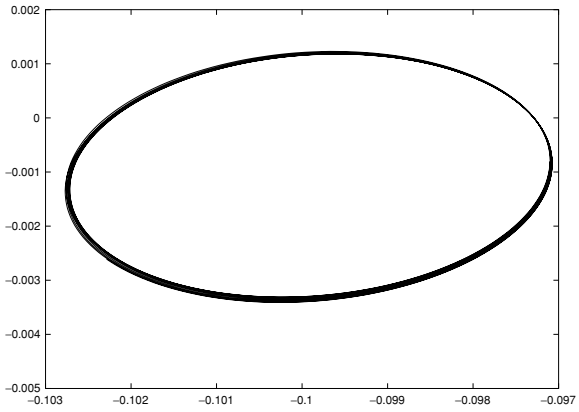

Figure 3. The trajectories in the complex plane corresponding to $\langle v=0|H(t)\psi(t)\rangle/\langle v=0|\psi(t)\rangle$ for an intensity $I = 10^{13}$ W cm^{-2}.

3.2. The multi-dimensional case

The molecular processes generally involve multi-dimensional active spaces. Thus it is important to generalize in these cases the commutativity rules demonstrated in the previous section.

We prove that the non-Abelian effective phase is a usual non-Abelian phase expressed with a section obtained by the wave operator transformation.

Let $P_0 = \sum_i |i\rangle\langle i|$ be the projector associated with the active space $S_0.\forall i$, let $\psi_i^0(t)$ be the solution of the effective Schrödinger equation with initial condition $\psi_i^0(t) = |i\rangle$. Let $\psi_i^0(t) = \sum_j U_{ji}(t)\tilde{\psi}_j^0(t)$ be a unitary gauge transformation such that $\tilde{\psi}_i^0(0) = |i\rangle$ and $\tilde{\psi}_i^0(T) = \tilde{\psi}_i^0(0)$. With a calculation analogous to the previous one we obtain

$$\imath\hbar \sum_k \dot{U}_{ki}\langle\tilde{\psi}_j^0|\tilde{\psi}_k^0\rangle + \imath\hbar \sum_k U_{ki}\langle\tilde{\psi}_j^0|\partial_t|\tilde{\psi}_k^0\rangle = \sum_k U_{ki}\langle\tilde{\psi}_j^0|H^{\text{eff}}|\tilde{\psi}_k^0\rangle. \tag{39}$$

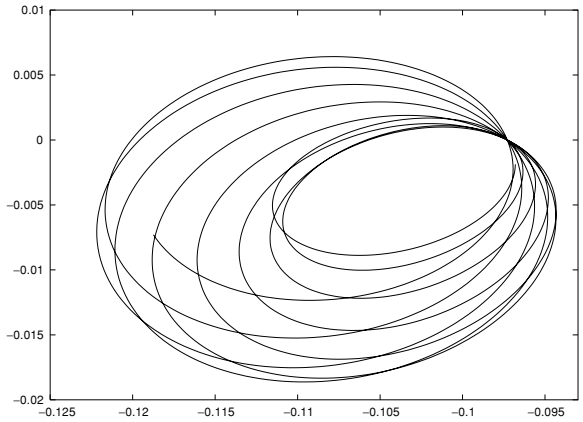

Figure 4. Same as figure 3 but for an intensity $I = 10^{14}$ W cm^{-2}.

The introduction of the overlap matrix $T_{jk} = \langle\tilde{\psi}^0_j|\tilde{\psi}^0_k\rangle$ gives

$$\imath\hbar(T\dot{U})_{ji} + \imath\hbar\sum_k U_{ki}\langle\tilde{\psi}^0_j|\partial_t|\tilde{\psi}^0_k\rangle = \sum_k U_{ki}\langle\tilde{\psi}^0_j|H^{\rm eff}|\tilde{\psi}^0_k\rangle. \tag{40}$$

The use of this expression leads to the result

$$(\dot{U}U^{-1})_{ml} = (T^{-1}T\dot{U}U^{-1})_{ml} \tag{41}$$

$$= \sum_{j,i} T^{-1}_{mj}(T\dot{U})_{ji}U^{-1}_{il} \tag{42}$$

$$= -\imath\hbar^{-1}\sum_{i,j,k} T^{-1}_{mj}U_{ki}U^{-1}_{il}\langle\tilde{\psi}^0_j|H^{\rm eff}|\tilde{\psi}^0_k\rangle - \sum_{i,j,k} T^{-1}_{mj}U_{ki}U^{-1}_{il}\langle\tilde{\psi}^0_j|\partial_t|\tilde{\psi}^0_k\rangle \tag{43}$$

$$= -\imath\hbar^{-1}\sum_{j} T^{-1}_{mj}\langle\tilde{\psi}^0_j|H^{\rm eff}|\tilde{\psi}^0_l\rangle - \sum_{j} T^{-1}_{mj}\langle\tilde{\psi}^0_j|\partial_t|\tilde{\psi}^0_l\rangle. \tag{44}$$

The gauge transformation of the wavefunction inside the model space is then

$$\psi^0_i(t) = \sum_j \left[\mathbb{T}\,{\rm e}^{-\imath\hbar^{-1}\int_0^t E^{\rm eff}(t')\mathrm{d}t' - \int_0^t A^{\rm eff}(t')\mathrm{d}t'}\right]_{ji}\tilde{\psi}^0_j(t) \tag{45}$$

with $E^{\rm eff}_{ij}(t) = \sum_k T^{-1}_{ik}\langle\tilde{\psi}^0_k(t)|H^{\rm eff}(t)|\tilde{\psi}^0_j(t)\rangle$ and $A^{\rm eff}_{ij}(t) = \sum_k T^{-1}_{ik}\langle\tilde{\psi}^0_k(t)|\partial_t|\tilde{\psi}^0_j(t)\rangle$, this second term being responsible for a non-adiabatic non-Abelian Berry phase.

We want show that $\psi_i(t) = \Omega(t)\psi^0_i(t)$ is also a parallel transport. We define $\tilde{\psi}_i(t)$ as being $\Omega(t)\tilde{\psi}^0_i(t)$. By definition $\Omega\psi^0_i = \psi_i$. We write then $\Omega = \sum_{jk} c_{jk}|\psi_j\rangle\langle\psi^0_k|$, then $\langle\psi_j|\Omega|\psi^0_i\rangle = \delta_{ij} \iff \sum_k c_{jk}\langle\psi^0_k|\psi^0_i\rangle = \delta_{ij}$. But

$$\langle\psi^0_j|\psi^0_k\rangle = \sum_{il} U_{kl}U^{-1}_{ji}\langle\tilde{\psi}^0_i|\tilde{\psi}^0_k\rangle = (U^{-1}TU)_{jk} \tag{46}$$

so that

$$\Omega(t) = \sum_{jk}(U^{-1}TU)^{-1}_{jk}|\psi_j\rangle\langle\psi^0_k|. \tag{47}$$

Then we have

$$\Omega^{\dagger}\Omega = \sum_{jkn} (U^{-1}TU)^{-1}_{nj} (U^{-1}TU)^{-1}_{jl} \left|\psi^0_n\right\rangle\left\langle\psi^0_k\right| \tag{48}$$

$$= \sum_{jkn} (TU)^{-1}_{nj} (U^{-1}T)^{-1}_{jl} \left|\psi^0_n\right\rangle\left\langle\psi^0_k\right| \tag{49}$$

so that

$$\left\langle\tilde{\psi}^0_i\right|\Omega^{\dagger}\Omega = \sum_{jknl} (TU)^{-1}_{nj} (U^{-1}T)^{-1}_{jk} U_{il} \left\langle\psi^0_l\middle|\psi^0_n\right\rangle\left\langle\psi^0_k\right| \tag{50}$$

$$= \sum_{jknl} (TU)^{-1}_{nj} (U^{-1}T)^{-1}_{jk} U_{il} (U^{-1}TU)_{ln} \left\langle\psi^0_k\right| \tag{51}$$

$$= \sum_{jkn} (TU)_{in} (TU)^{-1}_{nj} (U^{-1}T)^{-1}_{jk} \left\langle\psi^0_k\right| \tag{52}$$

$$= \sum_{k} (U^{-1}T)^{-1}_{ik} \left\langle\psi^0_k\right| \tag{53}$$

$$= \sum_{kl} T^{-1}_{il} U_{lk} \left\langle\psi^0_k\right| \tag{54}$$

$$= \sum_{l} T^{-1}_{il} \left\langle\tilde{\psi}^0_l\right| \tag{55}$$

and, finally,

$$(\imath\hbar^{-1}E^{\mathrm{eff}} + A^{\mathrm{eff}})_{ij} = \sum_k T^{-1}_{ik} \left\langle\tilde{\psi}^0_k(t)\middle|H^{\mathrm{eff}}(t)\middle|\tilde{\psi}^0_j(t)\right\rangle + \sum_k T^{-1}_{ik} \left\langle\tilde{\psi}^0_k(t)\middle|\partial_t\middle|\tilde{\psi}^0_j(t)\right\rangle \tag{56}$$

$$= \imath\hbar^{-1} \left\langle\tilde{\psi}^0_i\middle|\Omega^{\dagger}\Omega(H^{\mathrm{eff}} - \imath\hbar\partial_t)\middle|\tilde{\psi}^0_j\right\rangle \tag{57}$$

$$= \imath\hbar^{-1} \langle\tilde{\psi}_i|(H - \imath\hbar\partial_t)|\tilde{\psi}_j\rangle \tag{58}$$

$$= \imath\hbar^{-1} \langle\tilde{\psi}_i|H|\tilde{\psi}_j\rangle + \langle\tilde{\psi}_i|\partial_t|\tilde{\psi}_j\rangle \tag{59}$$

$$\equiv (\imath\hbar^{-1}E + A)_{ij}. \tag{60}$$

In other words, if $\left\{\psi^0_i(t)\right\}_i$ is a non-Abelian parallel transport associated with the section $\left\{\tilde{\psi}^0_i\right\}_i$, then $\left\{\Omega\psi^0_i\right\}_i$ is a non-Abelian parallel transport associated with the section $\left\{\Omega\tilde{\psi}^0_i\right\}$. The wave operator method is then compatible with the calculation of the non-Abelian Aharonov–Anandan phases.

Let $P_0(t)$ and $P(t)$ be defined by

$$P_0(t) = \sum_i \left|\tilde{\psi}^0_i(t)\right\rangle\left\langle\tilde{\psi}^0_i(t)\right| \qquad P(t) = \sum_i |\tilde{\psi}_i(t)\rangle\langle\tilde{\psi}_i(t)|. \tag{61}$$

It is clear that $\Omega(t)P_0(t)\Omega(t)^{\dagger} = P(t)$. P_0 is not an orthogonal projector. Let $G_M(\mathbb{C}^{\infty})$ be the space of M-fold orthogonal projectors of $\mathcal{H}$. $P_0(t)$ is an element of $S_0/U(M)$. Let R^{eff} be the non-Abelian horizontal lift operation associated with the effective Hamiltonian and R be the non-Abelian horizontal lift operation associated with the true Hamiltonian. We have

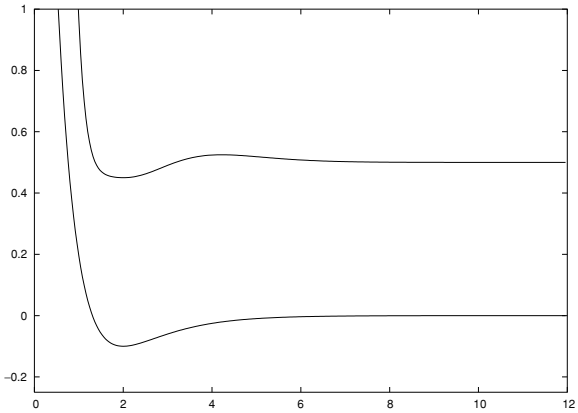

Figure 5. Representation of the two electronic potential surfaces of our model system.

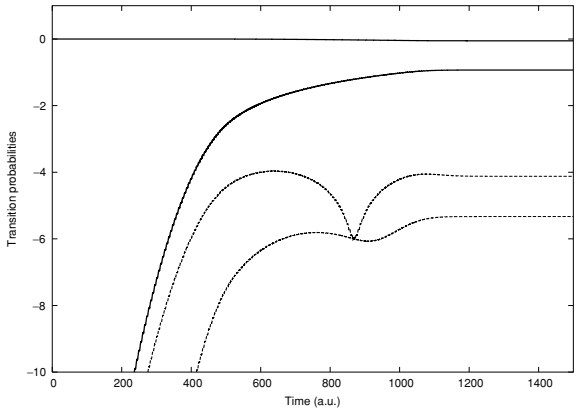

Figure 6. Vibrational transition probabilities on a logarithmic scale as a function of time. Solid lines: survival probability $P_{v=0\to v=0}$ and $P_{v=0\to v'=0}$; dashed lines: $P_{v=0\to v'=1}$ and $P_{v=0\to v=1}$.

the following commutative diagram:

$$\begin{array}{ccc} S_0^{[0,T]} & \xrightarrow{\Omega} & \mathcal{H}^{[0,T]} \\ {\scriptstyle R^{\text{eff}}}\uparrow & & \uparrow{\scriptstyle R} \\ (S_0/U(M))^{[0,T]} & \xrightarrow{\Omega\cdot\Omega^{\dagger}} & (G_M(\mathbb{C}^{\infty}))^{[0,T]} \end{array}.$$

To illustrate this result, we consider a generalized Floquet Berry phase, as in the one-dimensional case. The H_2^+ system is selected again, the ground surface is a Morse potential and the upper surface is taken to be the potential of Hulburt and Hirschfelder [22] in place of the structureless surface $\left(^2\Sigma_u^+\right)$ (figure 5). Four bound states exist in the well of this upper surface. The laser is tuned to the transition from the ground state of the ground surface $|v=0\rangle$ to the ground state of the upper surface $|v'=0\rangle$, i.e. $\omega_o = 0.546$ au, and the laser intensity is reduced to $I = 4.10^{10}$ W cm^{-2}.

Figure 5 presents the two electronic surfaces involved in the process. The survival probability and some inelastic transitions are presented in figure 6. Contrary to the H_2^+ example,

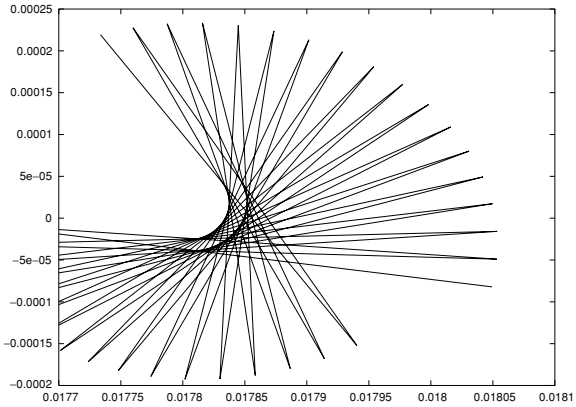

Figure 7. Matrix element $\Delta_{1,1}$ (cf equation 48) calculated at discrete values of the time over the range ($t = 700$ au, t = 800 au). The values correspond to the tips of the straight segments. For this figure the model space is specially reduced to a one-dimensional space: $P_0 = |v = 0\rangle\langle v = 0|$.

an inelastic transition probability, namely $P_{v=0\to v'=0}$, takes important values. The dynamics is not purely adiabatic (in the weak sense) and its correct description requires the selection of a degenerate active space, for example, by including in the model space the vibrational states $|v = 0\rangle$, $|v' = 0\rangle$ and $|v' = 1\rangle$. The size of the model space is arbitrary but not its method of construction. The states which belong to this space are selected by using the wave operator sorting algorithm proposed by Wyatt and Iung [16]. One can note that this procedure selects the three states which are the most populated after the action of the field–matter interaction (figure 6). We thus take $P_0 = |v = 0\rangle\langle v = 0| + |v' = 0\rangle\langle v' = 0| + |v' = 1\rangle\langle v' = 1|$.

To satisfy the condition $\tilde{\psi}_j(T) = \tilde{\psi}_j(0)$, one can involve (as in the H_2^+ case) the generalized Floquet eigenstates; nevertheless the condition $\tilde{\psi}_j(t = 0) = \psi_j(t = 0)$ cannot now be satisfied simply by identifying $\tilde{\psi}$ with a unique Floquet eigenstate. By working with an active space of dimension three, we implicitly assume that the initial values of the wavefunctions $\psi_1(0) = |v = 0\rangle$, $\psi_2(0) = |v' = 0\rangle$ and $\psi_3(0) = |v' = 1\rangle$ can be expressed as a linear combination of the initial values of three Floquet eigenstates, $|\mu_1(0)\rangle$, $|\mu_2(0)\rangle$ and $|\mu_3(0)\rangle$. More exactly, by introducing the (3×3) matrix $\mathbf{B}$, such that

$$(\mathbf{B}^{-1})_{i,j} = \langle i|P_0\mu_j(t = 0)\rangle, \tag{62}$$

one obtains $\tilde{\psi}_i(t) = \sum_j \mu_j(t) \times (\mathbf{B})_{j,i}$, where the indices i and j denote the three vibrational states $|v = 0\rangle$, $|v' = 0\rangle$ and $|v' = 1\rangle$ which participate in the definition of P_0. The three Floquet eigenstates are calculated by acting on the model space using a degenerate Bloch wave operator algorithm. By introducing the $(N \times 3)$ matrix $\mu = (|\mu_1\rangle, |\mu_2\rangle, |\mu_3\rangle)$, equations (57) and (58) can be rewritten as follows at each instant t:

$$\Delta_{i,j} = (\mathbf{B}^\dagger\mu^\dagger H_F\mu\mathbf{B})_{i,j} - \sum_k (\mathbf{B}^\dagger\mu^\dagger P_0\mu\mathbf{B})^{-1}_{i,k} \times (\mathbf{B}^\dagger\mu^\dagger P_0 H^{\text{eff}} P_0\mu\mathbf{B})_{k,j}$$
$$+\imath\hbar \sum_k (\mathbf{B}^\dagger\mu^\dagger P_0\mu\mathbf{B})^{-1}_{i,k} \times (\mathbf{B}^\dagger\mu^\dagger P_0\partial_t P_0\mu\mathbf{B})_{k,j} - \imath\hbar\mathbf{B}^\dagger\mu^\dagger\partial_t\mu\mathbf{B} = 0. \tag{63}$$

The defects $\Delta_{i,j}$ are represented in figures 7–9 for three active spaces of increasing size. In the non-degenerate case (figure 7) the defect $\Delta_{1,1}$ is important, with a modulus of about 10^{-2}. By including into the active space the two Floquet eigenstates associated with the vibrational states $|v = 0\rangle$ and $|v' = 0\rangle$ (figure 8), the defects decrease by three order of magnitude. The introduction of a third state $|v' = 1\rangle$ into the model space (figure 9) decreases again the moduli

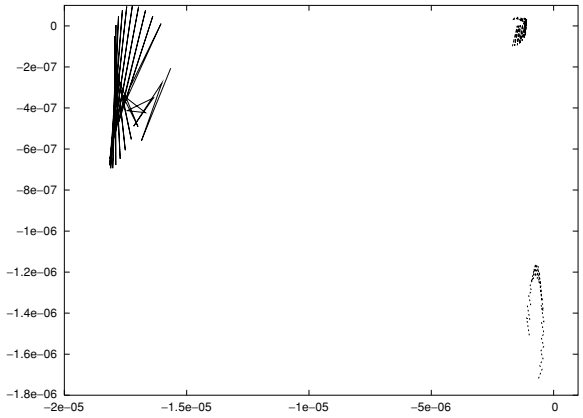

Figure 8. Matrix elements $\Delta_{1,1}$ (solid lines), $\Delta_{1,2}$ (dotted lines) and $\Delta_{2,2}$ (dashed lines) (equation 48) calculated at discrete values of the time over the range ($t = 700$ au, t = 800 au). The model space is reduced to a two-dimensional space: $P_0 = |v = 0\rangle\langle v = 0| + |v' = o\rangle\langle v' = 0|$.

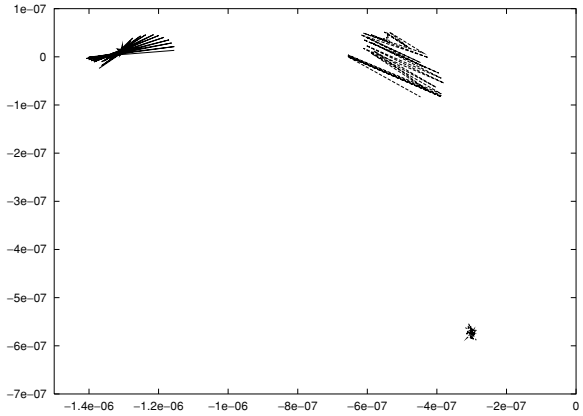

Figure 9. Same as figure 8 but the model space includes three vibrational states $|v = 0\rangle$, $|v' = 0\rangle$ and $|v' = 1\rangle$.

of the matrix elements $\Delta_{1,1}$, $\Delta_{1,2}$ and $\Delta_{2,2}$ by one order of magnitude. Thus one observes, as expected, a rapid convergence of the matrix Δ to zero. The small defects which subsist in figure 9 have moduli of about 10^{-6}. This modifies the inelastic transition probabilities by a factor of about 10^{-12}, a factor which is negligible compared to the probabilities values (figure 6). One can then assert that this Floquet Berry phase example obeys equations (56)–(60) and thus confirms that the non-Abelian Aharonov–Anandan phase can be calculated by using the effective Hamiltonian in the active space.

The direct relation between the existence of small defects (figure 9) and the non-completeness of the reduced basis is confirmed by figure 10. This figure represents the orthonormality factors for the two states $\tilde{\psi}_1(t)$ and $\tilde{\psi}_2(t)$ calculated in a three-dimensional active space. The generalized Floqet eigenstates $|\mu\rangle$ which compose these states perfectly satisfy the orthonormality conditions on the full time interval $[0, T]$, namely with $s = 2\pi t/T$

$$\langle\langle \mu_i | \mu_j \rangle\rangle = \frac{1}{2\pi} \int_0^{2\pi} \langle \mu_i(s) | \mu_j(s) \rangle \, \mathrm{d}s = \delta_{i,j}, \tag{64}$$

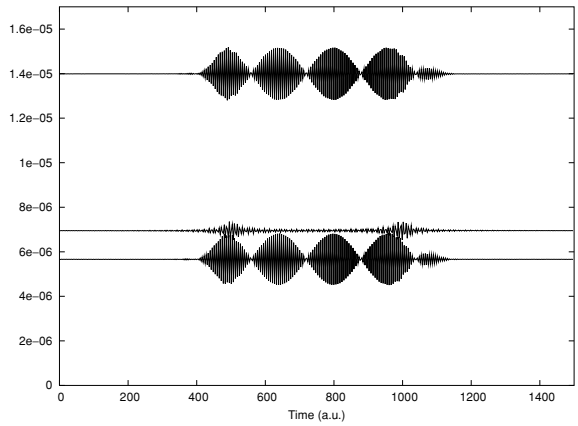

Figure 10. The instantaneous orthonormality factors $|\langle\tilde{\psi}_i(t)|\tilde{\psi}_j(t)\rangle - \delta_{i,j}|$ calculated as a function of time in the three-dimensional active space. The three states ($|i\rangle$, $i = 1$ to 3) which compose the model space are, respectively, $|v = 0\rangle$, $|v' = 0\rangle$ and $|v' = 1\rangle$. The three curves correspond (from top to bottom) to ($i = 1$, $j = 1$), ($i = 1$, $j = 2$) and ($i = 2$, $j = 2$).

but the same relationship is not satisfied at each instant by the functions $|\tilde{\psi}\rangle$. The errors revealed by figure 10 are about constant on the full time interval, with small modulations during the field–matter interaction between $t = 400$ and $t = 1100$. Asymptotically the states $|\tilde{\psi}_1(t = 0)\rangle$ and $|\tilde{\psi}_2(t = 0)\rangle$ should be equal to $|v = 0\rangle$ and $|v' = 0\rangle$, respectively, and the instantaneous orthogonality should be rigorously correct. The defects can thus be attributed to small components $\langle k|(1 - P_0)\mu_j(t = 0)\rangle$ outside the model space. One then recapitulates previously observed results [23], namely that the existence of the N-finite-dimensional active space is closely related to the asymptotic behaviour of the wavefunction. The wavefunction issuing from the model space should be included at the final time $t = T$ in the same model space. Under these conditions one can construct a group of N generalized Floquet eigenstates whose non-vanishing components are asymptotically (i.e. at $t = 0$ and $t = T$) incorporated in the model space.

4. Adiabatic transport with wave operators

The Berry phase was first introduced within a purely adiabatic approach (in the strong sense) [2] but using the adiabatic limit complicates the verification of the commutation of the wave operator with the parallel transport. To reach the adiabatic limit it is necessary to integrate over a very large time-interval, and moreover we do not obtain perfect adiabatic conditions when the non-perturbed energy spectrum is dense. It is then almost impossible to distinguish between errors due to the defects of the theory and those due to the residual non-adiabatic effects. For this reason the adiabatic case (in the strong sense) will be analysed solely theoretically in section 4.

The adiabatic limit of the time-dependent wave operator is a succession of Bloch wave operators; the adiabatic wave operator thus transforms the spectral active space at time 0 to the spectral active space at a time s. The true wavefunction is $\psi(s) = \Omega_\infty(s)U^{\text{eff}}(s)|i\rangle$. Since an adiabatic theorem can be applied, ψ is a parallel transport associated with the section $\{|i, s\rangle\}$, which is the set of instantaneous eigenvectors. But since Ω_∞ does not contain any phase (since it is expressed only in terms of projectors), the Berry and the dynamical phases could be contained in $U^{\text{eff}}(s)$. This is the subject of this section.

4.1. The adiabatic limit and the wave operator in the one-dimensional case

We consider the case of a one-dimensional subspace, $I = \{i\}$, where S_0 is generated by the eigenvector $|i,0\rangle \equiv |i\rangle$ and $S(s)$ is generated by $|i,s\rangle$. If T is in the neighbourhood of $+\infty$ then the wavefunction expressed in the adiabatic transport formula is

$$\psi_T(s) \sim \psi_\infty(s) = \mathrm{e}^{-\imath\hbar^{-1}T\int_0^s \lambda_i(s')\mathrm{d}s' - \int_0^s \langle i,s'|\partial_{s'}|i,s'\rangle \mathrm{d}s'}|i,s\rangle \tag{65}$$

where ψ_T is the solution of the Schrödinger equation before taking the adiabatic limit, i.e. a solution of the equation

$$\frac{\imath\hbar}{T}\partial_s\psi_T(s) = H(s)\psi_T(s). \tag{66}$$

Consider now the wave operator formulation. One can easily derive the adiabatic limit of the wave operator. We know that $\Omega_\infty(s) = \lim_{T\to+\infty}\Omega_T(s) = P_i(s)(P_i(0)P_i(s)P_i(0))^{-1}$ [8] with $P_i(0) = |i\rangle\langle i|$ and $P_i(s) = |i,s\rangle\langle i,s|$, so that

$$\begin{aligned} P_i(0)P_i(s)P_i(0) &= \langle i|i,s\rangle\langle i,s|i\rangle|i\rangle\langle i| \\ \Rightarrow \quad (P_i(0)P_i(s)P_i(0))^{-1} &= \frac{|i\rangle\langle i|}{\langle i|i,s\rangle\langle i,s|i\rangle} \end{aligned} \tag{67}$$

and then

$$\Omega_\infty(s) = \frac{|i,s\rangle\langle i,s|i\rangle\langle i|}{\langle i|i,s\rangle\langle i,s|i\rangle} = \frac{|i,s\rangle\langle i|}{\langle i|i,s\rangle}. \tag{68}$$

The introduction of the projected state $|i^0,s\rangle = P_i(0)|i,s\rangle = \langle i|i,s\rangle|i\rangle$ leads to the result

$$\Omega_\infty(s)|i^0,s\rangle = |i,s\rangle. \tag{69}$$

The effective Hamiltonian which corresponds to this wave operator is expressed as

$$H_T^{\mathrm{eff}}(s) = P_i(0)H(s)\Omega_T(t) = \frac{\langle i|H(s)|\psi_T(s)\rangle}{\langle i|\psi_T(s)\rangle}|i\rangle\langle i| \tag{70}$$

and its adiabatic limit is

$$\begin{aligned} H_\infty^{\mathrm{eff}} &= \lim_{T\to+\infty} H_T^{\mathrm{eff}} = P_i(0)H(s)\Omega_\infty(s) \\ &= \frac{\langle i|H(s)|i,s\rangle}{\langle i|i,s\rangle}|i\rangle\langle i| = \lambda_i(s)|i\rangle\langle i|. \end{aligned} \tag{71}$$

We cannot directly apply the adiabatic transport formula in S_0, because H_T^{eff} is not independent of T. We will proceed in a different way. Let $U_T(s)$ be the full evolution operator and $U_T^{\mathrm{eff}}(s)$ be the evolution operator in S_0 related to $H_T^{\mathrm{eff}}(s)$. By definition these two operators are related by the wave operator:

$$U(s)P(0) = \Omega_T(s)U_T^{\mathrm{eff}}(s)P(0). \tag{72}$$

The adiabatic limit of U_T^{eff} can be derived by first expressing the solution of the Schrödinger equation in the active subspace. As the model space is non-degenerate, $U_T^{\mathrm{eff}}(s)$ is a pure exponential; by taking into account equation (70) and the identity $\langle i|H(s)|\psi_T(s)\rangle = \frac{\imath\hbar}{T}\langle i|\partial_s\psi_T(s)\rangle$, we obtain

$$U_T^{\mathrm{eff}}(s) = \mathrm{e}^{-\imath\hbar^{-1}T\int_0^s H_T^{\mathrm{eff}}(s')\mathrm{d}s'} \tag{73}$$

$$= \mathrm{e}^{\int_0^s \frac{\langle i|\partial_{s'}|\psi_T(s')\rangle}{\langle i|\psi_T(s')\rangle}\mathrm{d}s'}|i\rangle\langle i| \tag{74}$$

$$= \mathrm{e}^{\int_0^s \partial_{s'}\ln\langle i|\psi_T(s')\rangle\mathrm{d}s'}|i\rangle\langle i| \tag{75}$$

$$= \mathrm{e}^{\ln\langle i|\psi_T(s)\rangle}|i\rangle\langle i| \tag{76}$$

$$= \langle i|\psi_T(s)\rangle|i\rangle\langle i| \tag{77}$$

and if T is in a neighbourhood of $+\infty$, then

$$U_T^{\mathrm{eff}}(s) \sim \langle i|\psi_\infty(s)\rangle|i\rangle\langle i| \tag{78}$$

so that

$$\psi_\infty^0(s) = U_\infty^{\mathrm{eff}}(s)|i\rangle = \langle i|\psi_\infty(s)\rangle|i\rangle \tag{79}$$

$$= \mathrm{e}^{-\imath\hbar^{-1}T\int_0^s \lambda_i(s')\mathrm{d}s' - \int_0^s \langle i,s'|\partial_{s'}|i,s'\rangle \mathrm{d}s'}\langle i|i,s\rangle|i\rangle \tag{80}$$

$$= \mathrm{e}^{-\imath\hbar^{-1}T\int_0^s \lambda_i(s')\mathrm{d}s' - \int_0^s \langle i,s'|\partial_{s'}|i,s'\rangle \mathrm{d}s'}|i^0,s\rangle. \tag{81}$$

The geometric and the dynamical phases are thus correctly represented in the effective evolution operator.

Finally, using equations (65), (69) and (81) gives the result

$$\Omega_\infty(s)\psi_\infty^0(s) = \psi_\infty(s). \tag{82}$$

Equations (81) and (82) prove that the phase of the wavefunction is preserved by working with H^{eff} in the model space, i.e., the phase of ψ_∞^0 is equal to the sum of the Berry phase and the dynamical phase. It thus emerges that the wave operator theory provides a consistent approach to the adiabatic transport problem.

We should stress that the difficulty of the demonstration associated with equation (78) is due to the non-uniform character of the adiabatic limit, which requires that

$$\lim_{T\to\infty}\int_0^s H_T^{\mathrm{eff}}(s')\,\mathrm{d}s' \neq \int_0^s \lim_{T\to\infty} H_T^{\mathrm{eff}}(s')\,\mathrm{d}s'. \tag{83}$$

The left-hand side of equation (83) contains the dynamical phase and the Berry phase, while the right-hand side involves the dynamical phase exclusively.

4.2. Adiabatic transport with wave operators in the multi-dimensional case

As in the one-dimensional case, one can prove that the non-Abelian phase terms are contained in the effective evolution operator. In conformity with the adiabatic limit we assume that the dynamics is confined in the space spanned by $\{|i,s\rangle\}_{i\in I}$, where I represents a finite set of indices. The adiabatic subspace of this problem is then $S_0 = \langle|i,0\rangle\rangle_{i\in I}$ and we denote $|i,0\rangle$ by $|i\rangle$. In the same way as for the one-dimensional case, we set

$$\psi_\infty^0(s) = U_\infty^{\mathrm{eff}}(s)|i\rangle \sim \mathbb{T}\,\mathrm{e}^{-\imath\hbar^{-1}\int_0^s T H_T^{\mathrm{eff}}(s')\mathrm{d}s'}|i\rangle. \tag{84}$$

As in the one-dimensional case, we have

$$\Omega_T(s)U_T^{\mathrm{eff}}(s)P(0) = U_T(s)P(0) \tag{85}$$

and consequently, at the adiabatic limit,

$$\Omega_\infty(s)U_\infty^{\mathrm{eff}}(s)P(0) = U_\infty(s)P(0) = P(s)U_\infty(s). \tag{86}$$

Equation (86) leads to the result

$$\Omega_\infty(s)\psi_\infty^0(s) = P(s)U_\infty(s)|i\rangle$$

$$= \psi_\infty(s) \tag{87}$$

$$= \sum_{j\in I}[\mathbb{T}\,\mathrm{e}^{-\imath\hbar^{-1}T\int_0^s E(s')\mathrm{d}s' - \int_0^s A(s')\mathrm{d}s'}]_{ji}|j,s\rangle \tag{88}$$

so that

$$\Omega_\infty(s)\psi^0_\infty(s) = \sum_{j\in I}[\mathbb{T}\,\mathrm{e}^{-\imath\hbar^{-1}T\int_0^s E(s')\mathrm{d}s' - \int_0^s A(s')\mathrm{d}s'}]_{ji}|j,s\rangle \tag{89}$$

or, by projecting into the model space,

$$\sum_{j,k\in I}[\Omega_\infty(s)]_{kj}\left[U^{\mathrm{eff}}_\infty(s)\right]_{ji}|k\rangle = \sum_{j,k\in I}[\mathbb{T}\,\mathrm{e}^{-\imath\hbar^{-1}T\int_0^s E(s')\mathrm{d}s' - \int_0^s A(s')\mathrm{d}s'}]_{ji}\langle k|j,s\rangle|k\rangle. \tag{90}$$

The adiabatic limit of the wave operator can be expanded as follows:

$$\Omega_\infty(s) = P(0) + Q(0)P(s)(P(0)P(s)P(0))^{-1}. \tag{91}$$

This implies that

$$\langle k|\Omega_\infty(s)|j\rangle = \delta_{jk}. \tag{92}$$

By introducing equation (92) into equation (90) one gets

$$\left[U^{\mathrm{eff}}_\infty(s)\right]_{ki} = \sum_{j\in I}\langle k|j,s\rangle[\mathbb{T}\,\mathrm{e}^{-\imath\hbar^{-1}T\int_0^s E(s')\mathrm{d}s' - \int_0^s A(s')\mathrm{d}s'}]_{ji}. \tag{93}$$

Finally, the expression of the adiabatic transport inside S_0 can be written in the form

$$\psi^0_\infty(s) = \sum_{k\in I}\left[U^{\mathrm{eff}}_\infty(s)\right]_{ki}|k\rangle \tag{94}$$

$$= \sum_{k,j\in I}[\mathbb{T}\,\mathrm{e}^{-\imath\hbar^{-1}T\int_0^s E(s')\mathrm{d}s' - \int_0^s A(s')\mathrm{d}s'}]_{ji}\langle k|j,s\rangle|k\rangle \tag{95}$$

$$= \sum_{j\in I}[\mathbb{T}\,\mathrm{e}^{-\imath\hbar^{-1}T\int_0^s E(s')\mathrm{d}s' - \int_0^s A(s')\mathrm{d}s'}]_{ji}\sum_{k\in I}|k\rangle\langle k|j,s\rangle \tag{96}$$

$$= \sum_{j\in I}[\mathbb{T}\,\mathrm{e}^{-\imath\hbar^{-1}T\int_0^s E(s')\mathrm{d}s' - \int_0^s A(s')\mathrm{d}s'}]_{ji}|j^0,s\rangle. \tag{97}$$

This expression confirms, for the multi-dimensional case, the consistency of the wave operator theory with the adiabatic transport formulation.

4.3. Standard phases versus effective phases

We want to return to the difficulty of defining effective phases at the adiabatic limit. This effect is due to the use of a more subtle definition of the effective dynamical phase at the adiabatic limit. We will analyse this point in more detail by considering again the non-adiabatic one-dimensional case and by taking its adiabatic limit.

A 'naive' expression of the effective dynamical phase, plausible at first sight, would be

$$\lambda^{\mathrm{eff}}_{\mathrm{naive}}(s) = \frac{\left\langle\psi^0_\infty(s)\middle|H^{\mathrm{eff}}_\infty(s)\middle|\psi^0_\infty(s)\right\rangle}{\left\langle\psi^0_\infty(s)\middle|\psi^0_\infty(s)\right\rangle}. \tag{98}$$

However, we have seen that $H^{\mathrm{eff}}_\infty(s) = \lambda_i(s)|i\rangle\langle i|$; thus $\lambda^{\mathrm{eff}}_{\mathrm{naive}}(s) = \lambda_i(s)$. Consequently the naive expression is not correct, because we have $A^{\mathrm{eff}}(s) = \frac{\langle i^0,s|\partial_s|i^0,s\rangle}{\langle i^0,s|i^0,s\rangle} \neq \langle i,s|\partial_s|i,s\rangle$, so that the sum of the dynamical and Berry phases is not preserved.

Consider the wavefunction of the non-adiabatic Berry phase

$$\psi^0_T(s) = \mathrm{e}^{-\imath\hbar^{-1}T\int_0^s \frac{\langle\tilde\psi^0_T(s')|H^{\mathrm{eff}}_T(s)|\tilde\psi^0_T(s')\rangle}{\langle\tilde\psi^0_T(s')|\tilde\psi^0_T(s')\rangle}\mathrm{d}s' - \int_0^s \frac{\langle\tilde\psi^0_T(s')|\partial_{s'}|\tilde\psi^0_T(s')\rangle}{\langle\tilde\psi^0_T(s')|\tilde\psi^0_T(s')\rangle}\mathrm{d}s'}\tilde\psi^0_T(s) \tag{99}$$

with $\lim_{T\to+\infty}\tilde\psi^0_T(s) = |i^0,s\rangle$.

The dynamical phase which is generated by the term H_T^{eff} is not equal to the 'naive' phase because of the non-uniform character of the adiabatic limit, namely

$$\lim_{T\to+\infty}\int_0^s \frac{\langle\psi_T^0(s')|H_T^{\mathrm{eff}}(s)|\psi_T^0(s')\rangle}{\langle\psi_T^0(s')|\psi_T^0(s')\rangle}\,\mathrm{d}s' \neq \int_0^s \lim_{T\to+\infty}\frac{\langle\psi_T^0(s')|H_T^{\mathrm{eff}}(s)|\psi_T^0(s')\rangle}{\langle\psi_T^0(s')|\psi_T^0(s')\rangle}\,\mathrm{d}s' \tag{100}$$

$$\neq \int_0^s \frac{\langle\psi_\infty^0(s')|H_\infty^{\mathrm{eff}}(s')|\psi_\infty^0(s')\rangle}{\langle\psi_\infty^0(s')|\psi_\infty^0(s')\rangle}\,\mathrm{d}s' \tag{101}$$

$$\neq \int_0^s \lambda_{\mathrm{naive}}^{\mathrm{eff}}(s')\,\mathrm{d}s'. \tag{102}$$

The true effective dynamical phase is then

$$\lambda_\infty^{\mathrm{eff}}(s) = \frac{\partial}{\partial s}\lim_{T\to+\infty}\int_0^s \frac{\langle\psi_T^0(s')|H_T^{\mathrm{eff}}(s)|\psi_T^0(s')\rangle}{\langle\psi_T^0(s')|\psi_T^0(s')\rangle}\,\mathrm{d}s', \tag{103}$$

and as $\lim_{T\to+\infty}\tilde{\psi}_T^0(s) = |i^0, s\rangle$, we finally obtain

$$\psi_\infty^0(s) \sim \mathrm{e}^{-\imath\hbar^{-1}T\int_0^s \lambda_\infty^{\mathrm{eff}}(s')\mathrm{d}s' - \int_0^s \frac{\langle i^0,s'|\partial_{s'}|i^0,s'\rangle}{\langle i^0,s'|i^0,s'\rangle}\mathrm{d}s'}|i^0, s\rangle. \tag{104}$$

By considering this expression, the appendix demonstrates that the sum of the effective dynamical and effective geometric phases is equal to the sum of the two corresponding standard phases, i.e.

$$\imath\hbar^{-1}T\lambda_\infty^{\mathrm{eff}}(s) + \frac{\langle i^0, s|\partial_s|i^0, s\rangle}{\langle i^0, s|i^0, s\rangle} \sim \imath\hbar^{-1}T\lambda_i(s) + \langle i, s|\partial_s|i, s\rangle. \tag{105}$$

An interesting result follows concerning this effective phase. Let $\hat{H}_\infty^{\mathrm{eff}}(s) = \lambda_\infty^{\mathrm{eff}}(s)|i\rangle\langle i| \neq \lambda_i(s)|i\rangle\langle i| = H_\infty^{\mathrm{eff}}(s)$. We see that

$$\hat{H}_\infty^{\mathrm{eff}}(s)|\psi_\infty^0(s)\rangle = \lambda_\infty^{\mathrm{eff}}(s)\langle i|\psi_\infty^0(s)\rangle|i\rangle \tag{106}$$

$$\sim \frac{\imath\hbar}{T}\frac{\langle i|\partial_s|\psi_\infty(s)\rangle}{\langle i|\psi_\infty(s)\rangle}\langle i|\psi_\infty^0(s)\rangle|i\rangle \tag{107}$$

$$\sim \frac{\imath\hbar}{T}\langle i|\partial_s|\psi_\infty(s)\rangle|i\rangle \tag{108}$$

$$\sim \frac{\imath\hbar}{T}\partial_s\langle i|\psi_\infty(s)\rangle|i\rangle \tag{109}$$

$$\sim \frac{\imath\hbar}{T}\partial_s|\psi_\infty^0(s)\rangle \tag{110}$$

and simultaneously

$$H_\infty^{\mathrm{eff}}(s)|\psi_\infty^0(s)\rangle = \lambda_i(s)|\psi_\infty^0(s)\rangle. \tag{111}$$

$\psi_\infty^0(s)$ is then an instantaneous eigenvector for H_∞^{eff}, although it is a solution of the Schrödinger equation for $\hat{H}_\infty^{\mathrm{eff}} = \partial_s\lim_{T\to\infty}\int_0^s H_T^{\mathrm{eff}}$. This shows the difficulty of treating the adiabatic limit in the effective Hamiltonian approximation. With a Hamiltonian independent of T, the adiabatic wavefunction is simultaneously an eigenvector and a solution of the Schrödinger equation for this Hamiltonian, whereas in the effective theory, where H_T^{eff} is dependent on T, this property is conserved only if we introduce two effective Hamiltonians, one for which it is an eigenvector and one for which it is solution of the Schrödinger equation.

The demonstration can be generalized without difficulty to the multi-dimensional case. In this case we find

$$E^{\mathrm{eff}}_{\infty}(s)_{ij} = \frac{\partial}{\partial s} \lim_{T\to\infty} \int_0^s \sum_k T^{-1}_{ik,T}(s') \left\langle \tilde{\psi}^0_{k,T}(s') \middle| H^{\mathrm{eff}}_T(s') \middle| \tilde{\psi}^0_{j,T}(s') \right\rangle \mathrm{d}s' \qquad (112)$$

where $\lim_{T\to\infty} \tilde{\psi}^0_{i,T}(s) = |i^0, s\rangle$, and

$$\imath\hbar^{-1} T E^{\mathrm{eff}}_{\infty}(s)_{ij} + \sum_k T^{-1}_{ik}(s) \langle k^0, s|\partial_s|j^0, s\rangle = \imath\hbar^{-1} T \lambda_i(s)\delta_{ij} + \langle i, s|\partial_s|j, s\rangle \qquad (113)$$

with $T_{ik}(s) = \langle i^0, s|k^0, s\rangle$.

5. Conclusion

The time-dependent wave operator theory is an effective Hamiltonian theory which treats the dynamics of a quantum system by projecting the evolution equations into active subspaces before reconstituting the exact solution in the full Hilbert space. A previous study revealed that this theory is relevant to the analysis of the adiabatic limit (in the strong sense of adiabatic). This study examined more closely the wave operator itself and proved that its adiabatic limit is a temporal succession of instantaneous Bloch wave operators. For systems monitored by adiabatic evolutions of periodic Hamiltonians, the same study proposed new specific evolution equations constructed in the framework of the two-time variables Floquet theory, essentially a version of the (t, t') theory adapted to the wave operator concept.

In the present study we analyse more carefully the phase term $U(t, 0; H^{\mathrm{eff}})$ which participates with $\Omega(t)$ in the time evolution of the system. This analysis is made successively in non-adiabatic cyclic situations and at the adiabatic limit by looking at the transformations which this effective Hamiltonian theory produces in the geometric and the dynamical phases. The principal results of this study are the following. In the case of non-adiabatic cyclic evolutions, the wave operator and the parallel transport commute and the gauge transformation is preserved. As a consequence, the dynamical phase and the Berry phase can be obtained by working with the effective Hamiltonian in the active space. The treatment of a photodissociation process produced by a short laser pulse confirms these theoretical results and reveals that the Floquet Berry phase is a particular case of the non-adiabatic Berry phase characterized by a total phase which is a linear function of time. In this case the parallel transport gives rise to a periodic function $\tilde{\psi}(t)$, which is a linear combination of generalized Floquet eigenstates. A careful analysis of this expansion proves that the existence of an N-dimensional active space requires that N generalized Floquet eigenstates should be projected at the initial time into an N-dimensional model space which includes the initial free state.

At the adiabatic limit the wave operator theory remains consistent with the adiabatic transport formulation. The sum of the dynamical and geometric phase is preserved. Nevertheless, a difficulty appears in the calculation of the dynamical phase, owing to the non-uniform character of the adiabatic limit. A 'naive' expression of this phase is proved to be false.

In summary, we have seen that it is possible to make simultaneous use of the parallel transport formulation and the wave operator theory, because the wave operator and the parallel transport are commutative operations. Nevertheless it should be noted that the wave operator method conserves only the sum of the Berry phase and the dynamical phase and not each phase separately. This is an important point because physicists usually eliminate the dynamical phase by using a gauge transformation in the one-dimensional case or in the cases where the dynamical phase and the Berry phase commute. If we apply the wave operator method,

this elimination is forbidden. By using the language of the fibre bundle theory, which is a usual geometric framework to interpret the Berry phase phenomenon, one can say that the wave operator method is not gauge invariant for the Berry connection A but is gauge invariant for the extended connection $A + \imath\hbar^{-1}E\,\mathrm{d}t$.

Appendix

The calculation of the effective dynamical phase (section 4.3) requires some expressions or equations which were given in the previous sections, i.e. $H_T^{\mathrm{eff}}(s) = P(0)H(s)\Omega(s)$, $\Omega_T(s) = \frac{|\psi_T(s)\rangle\langle i|}{\langle i|\psi_T(s)\rangle}$, $\left|\psi_T^0(s)\right\rangle = \langle i|\psi_T(s)\rangle|i\rangle$, $|i^0, s\rangle = \langle i|i, s\rangle|i\rangle$ and $\imath\hbar T^{-1}\partial_s\psi_T(s) = H(s)\psi_T(s)$.

The calculation of $\lambda_\infty^{\mathrm{eff}}(s)$ gives

$$\imath\hbar^{-1}T\int_0^s \lambda_\infty^{\mathrm{eff}}(s')\,\mathrm{d}s' \sim \imath\hbar^{-1}T\int_0^s \frac{\left\langle\psi_T^0(s')\middle|H_T^{\mathrm{eff}}(s)\middle|\psi_T^0(s')\right\rangle}{\left\langle\psi_T^0(s')\middle|\psi_T^0(s')\right\rangle}\,\mathrm{d}s' \tag{A.1}$$

$$\sim \imath\hbar^{-1}T\int_0^s \frac{\left\langle\psi_T^0(s')\middle|H(s')\Omega_T(s')\middle|\psi_T^0(s')\right\rangle}{\left\langle\psi_T^0(s')\middle|\psi_T^0(s')\right\rangle}\,\mathrm{d}s' \tag{A.2}$$

$$\sim \imath\hbar^{-1}T\int_0^s \frac{\langle\psi_t(s')|i\rangle\langle i|H(s')|\psi_T(s')\rangle\langle i|\psi_T(s')\rangle}{\langle i|\psi_T(s')\rangle\langle\psi_T(s')|i\rangle\langle i|\psi_T(s')\rangle}\,\mathrm{d}s' \tag{A.3}$$

$$\sim \imath\hbar^{-1}T\int_0^s \frac{\langle i|H(s')|\psi_T(s')\rangle}{\langle i|\psi_T(s')\rangle}\,\mathrm{d}s' \tag{A.4}$$

$$\sim \imath\hbar^{-1}T\int_0^s \frac{\imath\hbar}{T}\frac{\langle i|\partial_{s'}|\psi_T(s')\rangle}{\langle i|\psi_T(s')\rangle}\,\mathrm{d}s' \tag{A.5}$$

$$\sim \imath\hbar^{-1}T\int_0^s \frac{\imath\hbar}{T}\frac{\partial}{\partial s'}\ln\langle i|\psi_T(s')\rangle\,\mathrm{d}s' \tag{A.6}$$

$$\sim -\ln\langle i|\psi_T(s)\rangle \tag{A.7}$$

$$\sim -\ln\langle i|\psi_\infty(s)\rangle \tag{A.8}$$

$$\imath\hbar^{-1}T\lambda_\infty^{\mathrm{eff}}(s) \sim -\frac{\partial}{\partial s}\ln\langle i|\psi_\infty(s)\rangle \tag{A.9}$$

$$\sim -\frac{\langle i|\partial_s|\psi_\infty(s)\rangle}{\langle i|\psi_\infty(s)\rangle} \tag{A.10}$$

$$\sim \imath\hbar^{-1}T\lambda_i(s) + \langle i, s|\partial_s|i, s\rangle - \frac{\langle i|\partial_s|i, s\rangle}{\langle i|i, s\rangle} \tag{A.11}$$

$$\sim \imath\hbar^{-1}T\lambda_i(s) + \langle i, s|\partial_s|i, s\rangle - \frac{\langle i, s|i\rangle\langle i|i, s\rangle\langle i|i\rangle}{\langle i|i, s\rangle\langle i, s|i\rangle} \tag{A.12}$$

$$\sim \imath\hbar^{-1}T\lambda_i(s) + \langle i, s|\partial_s|i, s\rangle - \frac{\langle i^0, s|\partial_s|i^0, s\rangle}{\langle i^0, s|i^0, s\rangle} \tag{A.13}$$

so that finally

$$\imath\hbar^{-1}T\lambda_\infty^{\mathrm{eff}}(s) + \frac{\langle i^0, s|\partial_s|i^0, s\rangle}{\langle i^0, s|i^0, s\rangle} \sim \imath\hbar^{-1}T\lambda_i(s) + \langle i, s|\partial_s|i, s\rangle. \tag{A.14}$$

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