Berry’s phases for arbitrary spins nonlinearly coupled to external fields: application to the entanglement of $N > 2$ non-correlated 1/2-spins

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Received 15 June 2010, in final form 14 September 2010
Published 28 October 2010
Online at stacks.iop.org/JPhysA/43/465302

Abstract

We derive the general formula giving Berry’s phase for an arbitrary spin, having both magnetic-dipole and electric-quadrupole couplings with external time-dependent fields. We assume that the ‘effective’ electric and magnetic fields remain orthogonal during the quantum cycles. This mild restriction has many advantages. It provides simple symmetries leading to useful selection rules. Furthermore, the Hamiltonian parameter space coincides with the density matrix space for a spin $S = 1$. This implies a mathematical identity between Berry’s phase and the Aharonov–Anandan phase, which is lost for $S > 1$. We have found that new physical features of Berry’s phases emerge for integer spins $\geq 2$. We provide explicit numerical results of Berry’s phases for $S = 2, 3, 4$. For any spin, one easily finds well-defined regions of the parameter space where instantaneous eigenstates remain widely separated. We have decided to stay within them and not deal with non-Abelian Berry’s phases. We present a thorough and precise analysis of the non-adiabatic corrections with separate treatment for periodic and non-periodic Hamiltonian parameters. In both cases, the accuracy for satisfying the adiabatic approximation within a definite time interval is considerably improved if one chooses for the time derivatives of the parameters a time-dependence having a Blackman pulse shape. This has the effect of taming the non-adiabatic oscillation corrections which could be generated by a linear ramping. For realistic experimental conditions, the non-adiabatic corrections can be kept below the 0.1% level. For a class of quantum cycles, involving as the sole periodic parameter the precession angle of the electric field around the magnetic field, the corrections odd upon the reversal of the associated rotation velocity can be canceled exactly if the quadrupole to dipole coupling is chosen appropriately (‘magic values’). The even ones are eliminated by taking the difference of the Berry phases generated...
by two ‘mirror’ cycles. We end by indicating a possible application of the theoretical tools developed in this paper. We propose a way to perform a holonomic entanglement of \(N\) non-correlated \(1/2\)-spins by performing adiabatic cycles governed by a Hamiltonian given as a nonlinear function of the total spin operator \(S_z\), defined as the sum of the \(N\) individual spin operators. The basic idea behind this proposal is the mathematical fact that any non-correlated states can be expanded into eigenstates of \(S^2\) and \(S_z\). The same eigenvalues appear several times in the decomposition when \(N > 2\) but all these states differ by their symmetry properties under the \(N\)-spin permutations. The case \(N = 4\) and \(S_z = 1\) is treated explicitly and a maximum entanglement is achieved.

PACS numbers: 03.65.Vf, 03.67.Bg, 03.75.Dg, 37.25.+k

(Some figures in this article are in colour only in the electronic version)

1. Introduction

About 25 years ago, geometry made a new entrance in quantum mechanics with the discovery of geometric phases [1–3]. These developments hinge upon the simple fact that all the physical information relative to a pure quantum state \(|\Psi\rangle\) is contained in the density matrix \(\rho = |\Psi\rangle\langle\Psi|\). This mathematical object has two important properties: (i) it is invariant upon the Abelian gauge transformation \(|\Psi\rangle \rightarrow \exp(i\chi)|\Psi\rangle\) where \(\chi\) is an arbitrary real number, and (ii) it satisfies the nonlinear relation \(\rho^2 = \rho\). The density matrix space \(\mathcal{E}(\rho)\) has clearly a non-trivial topology. A geometric phase is acquired when the quantum system performs a time evolution along a closed circuit on \(\mathcal{E}(\rho)\) and satisfies at every instant \(t\) the so-called parallel transport condition \(\langle\frac{\partial}{\partial t}|\rho\rangle = 0\). The above terminology reflects the fact that the set of quantum vector states associated with a given density matrix \(\rho\) can be viewed as the ‘fiber’ of a linear ‘fiber bundle’ constructed above the ‘base space’ \(\mathcal{E}(\rho)\).

Our work deals mainly with adiabatic quantum cycles performed within a definite time interval \(0 \leq t \leq T\). They are generated by a Hamiltonian depending on a set of parameters \(X(t)\), assumed to be a system of coordinates for a differential manifold \(M\). In this way, an adiabatic quantum cycle generates a mapping of a closed circuit drawn upon the parameter space \(M\) onto a closed loop upon the density matrix space \(\mathcal{E}(\rho)\). The Berry phases can then be viewed as the geometric phases associated with this particular class of quantum cycles.

At this point, a question arises naturally: how can one extract the geometric phase since \(\rho\) is phase independent? The answer lies within the superposition principle of quantum mechanics: any linear combination of two quantum states \(|\Psi_1\rangle\) and \(|\Psi_2\rangle\) relative to a given quantum system \(|\Psi_{12}\rangle = c_1|\Psi_1\rangle + c_2|\Psi_2\rangle\) is an accessible state for the system. In this context, such a construction will be achieved via the interaction of the system with specific classical radio-frequency fields, using the so-called Ramsey pulses [4]. The density matrix associated with \(|\Psi_{12}\rangle\) is \(\rho_{12} = |\Psi_{12}\rangle\langle\Psi_{12}| = |c_1|^2\rho_1 + |c_2|^2\rho_2 + \Delta\rho_{12}\). The crossed contribution \(\Delta\rho_{12} = c_1 c_2^*|\Psi_1\rangle\langle\Psi_2| + \text{h.c.}\) contains all the information needed to obtain the difference of the geometric phases acquired by the states \(|\Psi_1\rangle\) and \(|\Psi_2\rangle\) during an adiabatic quantum cycle. In some experimental schemes considered in a forthcoming paper [5] the geometric phase acquired by \(|\Psi_2\rangle\) will be \textit{a priori} zero, and the measurement will yield directly the phase acquired by \(|\Psi_1\rangle\).

A huge amount of work has been triggered by the publication of M Berry’s paper [1] on the phase acquired by a quantum state at the end of its adiabatic evolution along a closed cycle. Among the flurry of following papers, two selections have been presented
in comprehensive reviews [6, 7] providing guides to the extensive literature on the subject. There are also pedagogical presentations and a thorough review of manifestations giving many examples drawn from a great variety of physical systems [8–10]. More recently, Berry’s phase has become a topic of renewed interest, both theoretical and experimental, with regard to improved information processing and more specifically for its potential use in quantum computing [11, 12]. At the same time, its unwanted manifestations in fundamental precision measurements have to be carefully investigated [13–15]. Conversely, in other settings, Berry’s phase might be just the right tool to detect still unobserved effects, such as parity violation in atomic hydrogen [16, 17].

This paper presents a detailed study of Berry’s phases resulting from adiabatic quantum cycles performed by an arbitrary spin, interacting nonlinearly with external electromagnetic fields. We assume that the Hamiltonian, governing the time evolution, involves both linear and quadratic couplings of the spin operator $S$. The addition of a quadratic spin coupling generates new features which, as we shall see, can show up for $S > 1$. The first purpose of this paper is to develop the formalism to calculate them, whatever the value of $S$, integer or half-integer. In addition, we shall give explicit numerical results for several spin values. Throughout this paper, we shall deal with the following set of quadratic spin Hamiltonians:

$$H(B(t), E(t)) = \gamma_S S \cdot B(t) + \gamma_Q (S \cdot E(t))^2.$$  

(1)

We have found in the literature only a few papers dealing with Berry’s phase for a spin submitted to a time-varying quadrupole interaction [18, 19]. These deal with nuclear quadrupole resonance spectra in a magnetic resonance experiment involving sample rotation. However, the absence of any magnetic interaction is assumed, which leads to level-degeneracy. These papers generalize Berry’s phase to the adiabatic transport of degenerate states. In this case, the geometric phase is replaced by a unitary matrix given by the Wilson loop integral of a $SU(n)$ non-Abelian gauge potential where $n$ is the dimension of the eigenspace associated with a given degenerate quantum level [20–22]. In contrast, in our work, we are interested in situations where the energy levels of the instantaneous Hamiltonian stay widely separated.

We impose on the model the extra constraint $E \cdot B = 0$. As shown previously by one of the authors (CB) and G Gibbons [24, 25], the parameter space of $H(B, E)$ with $E \cdot B = 0$ is isomorphic to the space $E(\rho)$ relative to spin-1 states, namely the complex projective plane $CP^2$. In [25], Berry’s phase generated by the Hamiltonian (1) for $S = 1$ is found to be mathematically identical to the Aharonov–Anandan (AA) phase, if one uses an appropriate parametrization of $CP^2$. However, the physical contents are in general different, since, in contrast to Berry’s phase, the AA phase is not restricted to adiabatic quantum cycles. Hereafter, we derive an expression for Berry’s phase relative to (1) for spins $S > 1$. In this case one loses the identity with the AA phase, which involves now closed circuits drawn upon the larger projective complex spaces $CP^{2S}$.

It is convenient to introduce the rotation $R(t)$ which brings the $x$ and $z$ axes along the $E$ and $B$ fields and the associated unitary transformation $U(R(t))$. This allows us to rewrite the spin Hamiltonian (1) as $H(E, B) = \gamma_3 B U(R(t)) \mathcal{H}(\lambda) U^\dagger(R(t))$, with the dimensionless Hamiltonian $\mathcal{H}(\lambda)$ given by $\mathcal{H}(\lambda) = S_3 \hbar^{-1} + \lambda S_2^2 \hbar^{-2}$. The parameter $\lambda = \hbar \gamma_Q E^2 / (\gamma_S B)$, combined with the Euler angles $\theta, \varphi, \alpha$ relative to $R(t)$ leads to a convenient set of coordinates for $CP^2$. A quantum cycle within the time interval $0, T$ satisfies the boundary conditions $\theta(T) = \theta(0)$, $\varphi(T) = \varphi(0) + 2n_\varphi \pi$, $\lambda(T) = \lambda(0)$, $\alpha(T) = \alpha(0) + n_\alpha \pi$, where $n_\varphi$ and $n_\alpha$ are arbitrary integers. The eigenstates of $\mathcal{H}(\lambda)$, $\Psi(\lambda, m)$ are labeled with a magnetic number $m$ by requiring that the analytical continuation of $\Psi(\lambda, m)$ toward $\lambda = 0$ coincides with the angular momentum eigenstates $|S, m\rangle$. Thanks to the constraint $E \cdot B = 0$, $\mathcal{H}(\lambda)$ has several discrete symmetries we shall use throughout this paper. The ‘$m$’ parity $(-1)^{S+m}$, associated with a
π -rotation around ẑ, is a good quantum number for \( \mathcal{H}(\lambda) \). Therefore, \( \mathcal{H}(\lambda) \) can mix states \( \tilde{\psi}(\lambda, m_1) \) and \( \tilde{\psi}(\lambda, m_2) \) if and only if \( |m_1 - m_2| \) is an even integer. This selection rule implies that \( \tilde{\psi}(\lambda, m) \) can be written as a direct sum of even and odd blocks. This greatly simplifies the construction of the eigenstates of \( \mathcal{H}(\mathbf{E}, \mathbf{B}) \) and the Berry-phase determination.

After some manipulations involving group theory techniques, Berry’s phase is written as a loop integral in the parameter space \( \mathbb{C}P^2 \):

\[
\beta(m, \lambda) = \oint p(m, \lambda)(\cos \theta d\phi + d\alpha) - m(d\phi + d\alpha),
\]

where \( \hbar p(m, \lambda) \) is the average value of \( \mathbf{S} \) along the \( \mathbf{B} \) field direction. This quantity is obtained by taking the gradient of the eigenenergies of \( \mathcal{H}(\mathbf{E}, \mathbf{B}) \) with respect to \( \gamma_S \mathbf{B} \). We have performed an explicit calculation of the eigenvalues \( \mathcal{E}(m, \lambda) \) of \( \mathcal{H}(\lambda) \) for the spins \( S = 2, 3 \) and 4 for \( \lambda \) values of physical interest. The results for \( \mathcal{E}(m, \lambda) \) and \( p(m, \lambda) \) are displayed in figure 1.

A superficial look at the above expression of \( \beta(m, \lambda) \) may give the impression that our result is, after all, not so different from the case of a pure dipole coupling. However, there are specific features associated with the quadrupole coupling which appear more easily for special cycles where \( \lambda \) and \( \alpha \) are the only time-varying parameters. The particular case \( S = 2, m = 0 \) is especially instructive in this respect. We recall that Berry’s phase associated with a pure dipole coupling is vanishing for an arbitrary cycle with the \( m = 0 \) state. Performing the above simple cycle with the state \( S = 2, m = 0 \) and assuming a constant quadrupole coupling, \( \lambda(t) = 1 \), one obtains from a look at curves of figure 1 the quite remarkable result \( \beta(m = 0, \lambda) = \oint p(0, \lambda)d\sigma = \pi \mod 2\pi \).

Our motivation for this paper is to make contact with experiment, having in mind the spectacular progress of atomic interferometry. Then one has to face the problem of the practical realization of a quadrupole coupling, having a magnitude comparable to the magnetic dipole one. For alkali atoms this is unrealistic if the Stark shift arises from a static E-field. The ac Stark shift induced by a light beam [23] is much more flexible. However, in most cases, reaching values of \( \lambda \) close to 1 requires the tuning of the beam frequency to be so close to an atomic line that the ac Stark effect induces an instability of the ‘dressed’ atomic ground state. In typical cases this implies stringent constraints upon the duration of Berry’s cycle. As a result, the question of the validity of the adiabatic approximation becomes crucial. This has led us to devote a full section of this paper to the precise evaluation of the non-adiabatic corrections. Our theoretical analysis is illustrated by numerical results for a few relevant cases. It provides guidelines for our forthcoming paper [5], devoted to experimental proposals.

Our analysis of the non-adiabatic corrections proceeds in two steps. We first deal with the corrections associated with the time derivatives of the Euler angles \( \theta, \phi, \alpha \). A convenient approach is the study of Berry’s cycle in the rotating frame attached to the \( \mathbf{B} \) and \( \mathbf{E} \) fields, by performing upon the laboratory quantum state \( \Phi(t) \) the unitary transformation \( \Phi(t) = U^{-1}(R(t))\Phi(t) \). The corresponding Hamiltonian is obtained by adding to \( \gamma_S \mathbf{B} \mathcal{H}(\lambda) \) the extra term \( \gamma_S \mathbf{S} \cdot \Delta \mathbf{B}(t) \), where \( \Delta \mathbf{B}(t) \) is the magnetic field generated by the Coriolis effect. The longitudinal component \( \Delta \mathbf{B}_{\|} = -\gamma_S^{-1}(\cos \theta \phi + \alpha) \hat{z} \) gives rise to a pure dynamical phase shift at the end of Berry’s cycle. As expected it incorporates \( \beta(m) - m \oint (d\phi + d\alpha) \) at its lowest order contribution with respect to \( \eta = -\Delta \mathbf{B}_{\|}/(\gamma_S \mathbf{B}) \). When \( \alpha \) is the sole varying Euler parameter, the higher order terms in \( \eta \) give the complete set of non-adiabatic corrections.

In addition, we show that the odd-order ones \( \propto \eta^{2n+1} \) vanish exactly if one chooses for \( \lambda \) the ‘magic’ value \( \lambda^*(\eta) \). On the other hand the transverse component \( \Delta \mathbf{B}_{\perp} \), proportional to \( \sin \theta \phi \), presents risks: it involves a linear combination of the spin operators \( S_z \) and \( S_y \) and induces a mixing with opposite \( m \)-parity states (\( \Delta m = \pm 1 \)), possibly nearly degenerate unless stringent constraints are imposed upon \( \lambda \). As regards the non-adiabatic corrections associated with \( \lambda \), we concentrate our attention on the ramping process of \( \lambda(t) \) from \( \lambda(0) = 0 \)
Figure 1. Reduced energies $\mathcal{E}(m, \lambda)$ and polarizations $p(m, \lambda)$ versus $\lambda$ for $S = 2, 3$ and 4. Since $\mathcal{E}(m, \lambda = 0) = p(m, \lambda) = m$, intersection of the curves with the vertical axis $\lambda = 0$ indicates their $m$ labeling. The $S = 2$ curves exhibit the symmetry relations $\mathcal{E}(m, \lambda) = -\mathcal{E}(-m, -\lambda)$ and $p(m, \lambda) = -p(-m, -\lambda)$, derived in the text. For $m > 0$ there is a clear evidence for the degeneracies expected when $\lambda \gg 1$.

to $\lambda(T) \sim 1$, the Euler angles keeping fixed values. Our method can be viewed as an extension of the rotating frame approach. We introduce the unitary transformation which makes $\mathcal{H}(\lambda)$ diagonal within the $|S\, m\rangle$ basis. The time evolution equation acquires an extra non-diagonal matrix $\Delta \mathcal{H}(t) \propto \dot{\lambda}(t)$ which has the same effect as an rf pulse with sharp edges if $\dot{\lambda}(t)$ increases linearly with $t$. This leads to rather large oscillating non-adiabatic corrections exhibited in our work (see figure 5). The standard procedure to tame them out is to give to $\dot{\lambda}(t)$ a Blackman pulse shape [27].
In the last section, we propose a ‘holonomic’ procedure for the entanglement of \( N \) non-correlated \( 1/2 \)-spins (or \( N \) Qbits.) The basic tools are Berry’s cycles generated by a Hamiltonian, formally identical to \( H(B(t), E(t)) \), except that, now, \( S \) is meant to be the total spin operator of the \( N \) spins, \( \sum_{i=1}^{N} s_i \). The method is based on a known mathematical property: any non-correlated \( N 1/2 \)-spin state can be expanded into a sum of \( S_z \) and \( S_y \) eigenstates. A given eigenvalue of \( S_z \) will appear several times if \( N > 2 \), but all the angular momentum states \( \psi_{S,M} \) have different symmetry properties under permutations of the \( N \) spins, which leave \( H(B(t), E(t)) \) invariant by construction. Thus, the states \( \psi_{S,M} \), organized into an orthogonal basis, behave as if they were associated with isolated spins. An initial non-correlated state, written as the sum \( \sum_i s_i \psi_{S,M} \), is transformed at the end of Berry’s cycle into \( \sum_i s_i \psi_{S,M} \exp i\beta(S_M) \). With an appropriate choice of cycle, we have been able to achieve maximum entanglement for \( N = 4, S_z = 1 \).

2. The instantaneous eigenfunctions of \( H(B, E) \) for a given adiabatic cycle

In this section we construct the instantaneous eigenfunctions of \( H(B, E) \) for an arbitrary adiabatic cycle. The result is put under a form well adapted to the calculation of Berry’s phase by group theoretical methods. Our method applies to both integer and half-integer spins.

2.1. Instantaneous spin Hamiltonian: symmetry properties

As a preliminary step, it is convenient to study the particular field configuration where \( E \) and \( B \) are along the \( x \) - and \( z \) -axes, respectively, \( \hat{n} = \hat{x}, \hat{b} = \hat{z} \), and write equation (1) as

\[
\hat{H}(B, E) = \gamma_s B S_x + \gamma_Q E^2 S_z.
\]

(3)

For the explicit calculations to be performed in the cases \( S > 1 \), it is convenient to introduce the dimensionless Hamiltonian \( \hat{H}(\lambda) \):

\[
\hat{H}(B, E) = \gamma_s B \hbar \mathcal{H}(\lambda);
\]

\[
\mathcal{H}(\lambda) = S_z/\hbar + \lambda(S_x/\hbar)^2;
\]

(4)

\[\lambda = \hbar \gamma_Q E^2/(\gamma_s B).\]

It is important to note that \( \mathcal{H}(\lambda) \) is invariant under three transformations. The first, \( T_1 \), corresponds to the reflection with respect to the \( x \), \( y \) plane, which changes \( S_x, S_y \) into \( -S_x, -S_y \), respectively, while leaving \( S_z \) unaltered.

\[
T_1^{-1} S_x T_1 = -S_x, \quad T_1^{-1} S_y T_1 = -S_y, \quad T_1^{-1} S_z T_1 = S_z.
\]

(5)

The second transformation, \( T_2 \), is the product of the reflection with respect to the \( y, z \) plane by the time reversal operation. The transformation of the spin operator under \( T_2 \) obeys the relations

\[
T_2^{-1} S_x T_2 = -S_x, \quad T_2^{-1} S_y T_2 = S_y, \quad T_2^{-1} S_z T_2 = S_z.
\]

(6)

The third transformation is a rotation of \( \pi \) around the \( z \) axis, which changes \( S_x \) and \( S_y \) into \( -S_x \) and \( -S_y \), respectively, while leaving \( S_z \) unaltered.

\[
T_3^{-1} S_x T_3 = -S_x, \quad T_3^{-1} S_y T_3 = -S_y, \quad T_3^{-1} S_z T_3 = S_z.
\]

(7)

(Note that \( T_1 \) and \( T_2 \) have the same effect on pseudo-vectors but opposite effects on vectors.)

Let us now discuss some consequences of these invariance properties. To this end, we introduce the eigenvectors \( \psi(m, \lambda) \) of the Hamiltonian \( \mathcal{H}(\lambda) \), together with their eigenenergies \( E(m, \lambda) \):

\[
\mathcal{H}(\lambda) \psi(m, \lambda) = E(m, \lambda) \psi(m, \lambda).
\]

(8)
Throughout this work the eigenenergies are supposed non-degenerate. In the limit \( \lambda \to 0 \), the states \( \hat{\psi}(m, \lambda) \) have to coincide with the angular momentum states \( |S, m\rangle: S^2 |S, m\rangle = \hbar^2 S(S + 1) |S, m\rangle \) and \( S_z |S, m\rangle = \hbar m |S, m\rangle \). The relations (5) imply that the quantum average of \( \Sigma = S/\hbar \) relative to \( |\psi(m, \lambda \neq 0)\rangle \), i.e. the polarization of the quantum state \( \rho(m, \lambda) \), lies along the \( z \)-axis:

\[
\langle \hat{\psi}(m, \lambda) | \Sigma | \hat{\psi}(m, \lambda) \rangle = \{0, 0, p(m, \lambda)\}.
\]

The invariance under \( T_1 \) and \( T_2 \) requires that the off-diagonal elements of the alignment tensor vanish:

\[
A_{ij} = \frac{i}{2} \langle \hat{\psi}(m, \lambda) | [\Sigma_i, \Sigma_j] | \hat{\psi}(m, \lambda) \rangle = \delta_{i,j} A_{ii}.
\]

A third important consequence of the above invariance properties is that the eigenvectors \( \hat{\psi}(m, \lambda) \) may be taken as real. This can be verified by noting that the matrix associated with \( \mathcal{H}(\lambda) \) in the angular momentum basis \( |S, m\rangle \), using the standard phase convention, is real and symmetric, so that its eigenvectors are real.

Equation (7) implies that the operator associated with the rotation of \( \pi \) around \( \hat{z} \), \( T_3 = \exp(-i \pi S_z) \), commutes with the Hamiltonian \( \mathcal{H}(\lambda) \). For convenience, let us introduce the \( m \)-Parity, \( P_3 = \exp(iS_z) T_3 = \exp(-i\pi(S - S_z)) \). The \( m \)-Parity of the angular momentum eigenstate \( |S, m\rangle \) is \((-1)^{(S-m)}\). Within the angular momentum basis, the Hamiltonian \( \mathcal{H}(\lambda) \) can then be written as the direct sum of two matrices acting respectively on the states even and odd with respect to the operator \( P_3 \):

\[
\mathcal{H}(\lambda) = \mathcal{H}_{\text{even}}(\lambda) \oplus \mathcal{H}_{\text{odd}}(\lambda).
\]

As a conclusion, we would like to stress that the field orthogonality condition plays an essential role in making the mathematical problem tractable for spins \( S > 1 \). Otherwise, the problem would become rapidly complicated and any insight into Berry’s phase physics gets blurred by the algebra.

2.2. The instantaneous eigenfunctions of \( H(B(t), E(t)) \)

Since we are going to use group theory arguments, it is appropriate to recall some basic facts about the rotation group in quantum mechanics. One introduces the unitary operator \( U(\mathcal{R}(\hat{u}, \chi)) \) associated with the rotation \( \mathcal{R}(\hat{u}, \chi) \) acting on the spin state vectors

\[
U(\mathcal{R}(\hat{u}, \chi)) = \exp \left( -i \frac{\hat{u} \cdot S}{\hbar} \chi \right),
\]

where \( \mathcal{R}(\hat{u}, \chi) \) stands for the rotation \( R \) around the unit vector \( \hat{u} \) by an angle \( \chi \). This operator provides a unitary representation of the rotation group in the sense that it obeys the multiplication rule: \( U(R_1) U(R_2) = U(R_1 \cdot R_2) \), together with the unitarity relation \( U^{-1}(R) = U^\dagger(R) \). Applying the above rule to the case where \( R_2 \) is an infinitesimal rotation, one derives the important relation

\[
U^{-1}(\mathcal{R}(\hat{u}, \chi)) \cdot S U(\mathcal{R}(\hat{u}, \chi)) = \mathcal{R}(\hat{u}, \chi) \cdot S,
\]

which expresses the fact that the unitary transformation \( U(\mathcal{R}(\hat{u}, \chi)) \) rotates the spin observables.

Let us associate with the orthogonal vectors \( E \) and \( B \) the trihedron \( (\hat{e}, \hat{b} \land \hat{e}, \hat{b}) \) which can be constructed by applying the rotation \( R_E(\theta, \varphi, \alpha) \) to the fixed coordinate trihedron \( (\hat{x}, \hat{y}, \hat{z}) \), with \( \theta, \varphi, \alpha \) denoting the usual Euler angles

\[
R_E(\theta, \varphi, \alpha) = \mathcal{R}(\tilde{z}, \varphi) \mathcal{R}(\tilde{y}, \theta) \mathcal{R}(\tilde{x}, \alpha).
\]
To ensure the validity of the adiabatic approximation for the quantum cycles generated by $H(B(t), E(t))$, we shall assume that the Euler angles are slowly varying functions of time. More precisely, we shall require that their time derivatives $\dot{\alpha}, \dot{\theta}, \dot{\phi}$—together with the time derivative of the Stark–Zeeman coupling ratio $\dot{\lambda}$—are much smaller than the rate $\Delta E_{\text{min}}/\hbar$, where $\Delta E_{\text{min}}$ stands for the minimum distance between the energy levels of $\hat{H}(B(t), E(t))$. The adiabatic cycle within the time interval $0 \leq t \leq T$ is specified by the boundary conditions involving the two finite integers $n_{\phi}$ and $n_{\alpha}$:

$$\phi(T) = \phi(0) + 2n_{\phi}\pi; \quad \alpha(T) = \alpha(0) + n_{\alpha}\pi. \quad (14)$$

Note that in contrast to the periodic variables $\phi(t)$ and $\alpha(t)$, $\lambda(t)$ and $\theta(t)$ recover their initial values at the end of the cycle.

To proceed it is convenient to introduce the unitary operator $U(R(t))$ associated with the rotation $R(t)$, given by

$$R(t) = R(\hat{z}, \phi(t))R(\hat{y}, \theta(t))R(\hat{z}, \alpha(t)). \quad (15)$$

Using the relation (12) with $R(\hat{u}, \chi) = R(t)^{-1}$ and the identity $R(t)^{-1}a \cdot b = a \cdot R(t)b$, it is straightforward to derive the important relation

$$U(R(t))\hat{H}(B(t), E(t))U(R) = H(B(t), E(t)). \quad (17)$$

As a consequence, the wavefunctions $\Psi(m,t)$ defined as

$$\Psi(m,t) = U(R(t))\hat{\psi}(m,\lambda(t)) \quad (18)$$

are instantaneous eigenfunctions of $H(B(t), E(t))$ with eigenvalues $E(m, B(t), E(t))$:

$$H(B(t), E(t))\Psi(m,t) = U(R(t))\hat{H}(B(t), E(t))\hat{\psi}(m,\lambda(t))$$

$$= E(m, B(t), E(t))\Psi(m,t) \quad (19)$$

$$E(m, B(t), E(t)) = \gamma_3\hbar B(t)\bar{E}(m, \lambda(t)). \quad (20)$$

We would like to stress that the instantaneous wavefunctions $\Psi(m,t)$ have, by construction, a well-defined phase since, as shown previously, the state vectors $\hat{\psi}(m,\lambda(t))$ can all be taken as real.

3. Explicit calculation of the eigenfunction parameters for Berry’s phase evaluation

We present for $S = 2, 3$ and $4$, explicit calculations of the eigenenergies $\bar{E}(m, \lambda)$ and the polarization of the eigenstates, which, for symmetry, is along the $B$ direction $p(m, \lambda) = \langle \hat{\psi}(m,\lambda)|S_z|\hat{\psi}(m,\lambda)\rangle$. (In this section, for the sake of simplicity we use a unit system where $\hbar = 1$.) We construct the matrix associated with $\hat{\mathcal{H}}(S, \lambda)$ within the angular momentum basis: $|S m\rangle$. It is convenient to rewrite the Hamiltonian in terms of the operators $S_\pm, S^2 = S_x + iS_y$, and $S^- = S_x - iS_y$:

$$\hat{\mathcal{H}}(S, \lambda) = S_z + \lambda S_x^2,$$

$$\equiv S_z - \frac{\lambda}{2} S_x^2 + \frac{\lambda}{4} ((S^+)^2 + (S^-)^2) + \frac{\lambda}{2} S(S + 1).$$

Using textbook formulas, for the matrix elements of $(S^\pm)^2$ in the $|S m\rangle$ basis, it is easy to write $\hat{\mathcal{H}}(S, \lambda)$ in the matrix form. Using the invariance under the symmetry $T_3$ introduced
in subsection 2.1, \( \mathcal{H}(S, \lambda) \) can be written as a direct sum of two matrices acting respectively upon the states \(|Sm\rangle \) with even and odd values of \( S-m \), \( \mathcal{H}(S, \lambda) = \mathcal{H}_{\text{even}}(S, \lambda) \oplus \mathcal{H}_{\text{odd}}(S, \lambda) \), one of order \( S \) and the other of order \( (S+1) \) depending on the parity of \( S \). For \( S = 1 \) and 2 one easily finds

\[
\mathcal{H}_{\text{odd}}(1, \lambda) = \begin{pmatrix}
\lambda/2 + 1 & \lambda/2 \\
\lambda/2 & \lambda/2 - 1
\end{pmatrix},
\]

\[
\mathcal{H}_{\text{even}}(2, \lambda) = \begin{pmatrix}
\lambda + 2 & \sqrt{3/2} \lambda & 0 \\
\sqrt{3/2} \lambda & 3\lambda & \sqrt{3/2} \lambda \\
0 & \sqrt{3/2} \lambda & \lambda - 2
\end{pmatrix},
\]

\[
\mathcal{H}_{\text{odd}}(2, \lambda) = \begin{pmatrix}
5\lambda/2 + 1 & 3\lambda/2 \\
3\lambda/2 & 5\lambda/2 - 1
\end{pmatrix}.
\]

The Hamiltonians \( \mathcal{H}_{\text{even}}(3, \lambda) \) and \( \mathcal{H}_{\text{odd}}(3, \lambda) \) as well as \( \mathcal{H}_{\text{even}}(4, \lambda) \) and \( \mathcal{H}_{\text{odd}}(4, \lambda) \) are given in the appendix. The eigenvalues are obtained in a standard way by solving the two polynomial equations:

\[
det(x I - \mathcal{H}_{\text{even}}(2, \lambda)) \equiv x^3 - 5x^2\lambda + 4x\lambda^2 - 4x + 12\lambda = 0,
\]

\[
det(x I - \mathcal{H}_{\text{odd}}(2, \lambda)) \equiv x^2 - 5x\lambda + 4\lambda^2 - 1 = 0.
\]  

(22)

The eigenvalue equations for \( S = 3 \) and \( S = 4 \) can be found in the appendix. Mathematica codes yield explicit expressions for the eigenenergies \( E(m, \lambda) \). Although the formulas are rather complex, they lead to very accurate numerical results for all the values of \( \lambda \) of interest. The accuracy is better than \( 10^{-12} \). This result has been checked using numerical Mathematica codes, obtained directly from the matrix expression. To calculate the polarization \( p(m, \lambda) \) we have used the fact that it is given by the derivative of the eigenenergy with respect to \( \gamma \lambda \) (Hellmann–Feynman theorem). Writing the eigenvalue of \( \hat{H}(B, E) \) as \( E(m, B, E) = \gamma S \mathcal{E}(m, \lambda) \), with \( \lambda = \frac{h\gamma B E}{\gamma S} \), one immediately gets

\[
p(m, \lambda) = \frac{\partial E(m, B, E)}{\partial B} = \mathcal{E}(m, \lambda) - \lambda \frac{\partial \mathcal{E}(m, \lambda)}{\partial \lambda}.
\]

(23)

The results are given in figure 1 for both the energies and the polarizations. The eigenenergies \( \mathcal{E}(m, \lambda) \) can be labeled unambiguously since for small values of \( \lambda \) the eigenvalues are equal to \( m \), up to corrections of the order of \( \lambda^2 \). In figure 1 there is evidence for near-degeneracies of opposite-parity level pairs: \( \mathcal{E}(m, \lambda) \) is approaching \( \mathcal{E}(m - 1, \lambda) \) when \( \lambda \gg 1 \) for \( m = S - q \), where \( q \) is an even integer such that \( 0 \leq q < S \). This was expected since in this limit \( \mathcal{H}(\lambda) \) is dominated by the term \( \propto S_0^2 \). The convergence is much slower for \( m < 0 \) since the positive term \( \lambda S_0^2 \) has then to fight against a negative Zeeman effect. There are \( S \) degenerate doublets with \( \mathcal{E}(m, \lambda) / \lambda \cong m^2 \), as expected in the limit \( \lambda \gg 1 \). In addition, by looking at the even–odd (or odd–even) pairs, one sees clearly that the pair \( \mathcal{E}(S, \lambda), \mathcal{E}(S - 1, \lambda) \) converges, without crossing, to the degenerate doublet \( \mathcal{E}(S, \lambda) \cong \mathcal{E}(S - 1, \lambda) \cong \lambda S^2 \). The next lower pair ends as the degenerate doublet having the energy \( \cong \lambda (S - 1)^2 \) and so on until one reaches the isolated level \( m = -S \), which has no possibility other than converging toward the non-degenerate level with energy \( \lambda \times 0 \). While this behavior is clearly exhibited in the simple case \( S = 2 \), only its two first steps are clearly apparent for \( S = 3 \) and 4, but we have verified that the above picture is valid for all values of \( m \) by computing the ratios \( \mathcal{E}(m, \lambda) / \lambda \) when \( \lambda \gg 1 \).

A striking feature of the case \( S = 2 \) in figure 1 is the symmetry of the plotted curves under the transformation \( m \rightarrow -m, \lambda \rightarrow -\lambda \), involving both the eigenenergies and the polarizations: \( \mathcal{E}(m, \lambda) \rightarrow -\mathcal{E}(-m, -\lambda), p(m, \lambda) \rightarrow -p(-m, -\lambda) \). To prove
these symmetry properties in the general case, it is convenient to perform upon the spin
system a rotation of angle $\pi$ around the $x$ axis, $R(\hat{x}, \pi)$. By introducing the spin unitary
operator $U(R(\hat{x}, \pi)) = \exp(-i S_x \pi)$, associated with the rotation $R(\hat{x}, \pi)$, one can write
the transformation law for the Hamiltonian $\mathcal{H}(\lambda)$:

$$U(R(\hat{x}, \pi))\mathcal{H}(\lambda)U^{-1}(R(\hat{x}, \pi)) = -S_z + \lambda S_x^2 = -\mathcal{H}(-\lambda).$$

Applying $U(R(\hat{x}, \pi))$ to both sides of the eigenvalue equation $\mathcal{H}(\lambda)\hat{\psi}(m, \lambda) = E(m, \lambda)\hat{\psi}(m, \lambda)$, one can write
$$U(R(\hat{x}, \pi))\mathcal{H}(\lambda)\hat{\psi}(m, \lambda) = -\mathcal{H}(-\lambda)U(R(\hat{x}, \pi))\hat{\psi}(m, \lambda) = E(m, \lambda)U(R(\hat{x}, \pi))\hat{\psi}(m, \lambda).$$

The state $U(R(\hat{x}, \pi))\hat{\psi}(m, \lambda)$ is an eigenstate of $\mathcal{H}(-\lambda)$ with the eigenenergy $-E(m, \lambda)$. It coincides, up to a phase factor, with the eigenstate $\hat{\psi}(-m, -\lambda)$, since the effect of the rotation $R(\hat{x}, \pi)$ is to flip the spin component along the $z$ axis. This completes the proof that $E(m, \lambda) = -E(-m, -\lambda)$ with the similar relation for $p(m, \lambda)$ as a consequence of equation (23).

In addition, the quantum-averaged polarizations $p(m, \lambda)$ satisfy the sum rule
$$\sum_m p(m, \lambda) = 0 \quad (25)$$
valid for any value of $S$ and $\lambda$. It is readily derived using the fact that $p(m, \lambda)$ can be evaluated as the partial derivative of the energy with respect to $\gamma S_B$ (equation (23)) and noting the general structure of the polynomial equation whose solutions provide the eigenenergies.

To conclude this section we would like to say a few words concerning the state $m = 0$ which is of particular interest for integer spins $S \geq 2$. The fact that a second-order Stark effect can induce a polarization in an initially unpolarized state was somewhat unexpected. To gain greater insight we have performed a perturbation computation in powers of $\lambda$. Although one has to go to third order to find a non-zero effect the final result is given by the rather elegant formula
$$p(0, \lambda) = \frac{1}{8} \lambda^3 S(S + 2)(S^2 - 1)(1 + O(\lambda^2)).$$

(26)

It gives a quite accurate result for $S = 2$ if $|\lambda| \leq 0.4$, but the domain of validity gets smaller for $S = 3$: $|\lambda| \leq 0.12$. Finally we would like to point out the smooth behavior of $p(0, \lambda)$ for $S = 2$ in the vicinity of $\lambda = 1$ and the remarkably simple exact values taken by the reduced energy and the polarization at $\lambda = 1$, namely $\mathcal{E}(0, 1) = 2$ and $p(0, 1) = 1$. The time of writing, it is unclear whether this is a mere numerical accident or an indication of something more profound.

4. Berry’s phase for adiabatic cycles generated by $H(B(t), E(t))$ acting
on arbitrary spins

We start this section with a mini-review introducing the basic physical and mathematical
features of Berry’s phase concept. In particular we derive the general formula giving
Berry’s phase in terms of the instantaneous eigenfunctions of the time-dependent Hamiltonian
generating the adiabatic quantum cycles. Introducing this formula, the results of the previous
section and relying upon group theory arguments, we perform an explicit construction of
Berry’s phase relative to $H(B(t), E(t))$, valid for arbitrary spins. The final result is expressed
as a loop integral in the $\mathbb{C}P^2$ space, using as coordinates the Euler angles and the parameter $\lambda$. We show that for the case $S = 2$ a circular loop drawn upon a spherical subspace of $\mathbb{C}P^2$ leads to a loop integral very different from that of the case $S = 1$ which involves a magnetic monopole Aharonov–Bohm phase.
4.1. Berry’s phase as a physical observable and topological concept: a mini-review

In this introductory subsection, we are going to follow, in several places, a presentation by the late Leonard Schiff, in tribute to his memory. He devoted few pages of his venerable textbook [26] a correct and precise treatment of the adiabatic approximation, involving a non-integrable phase. To study the adiabatic quantum cycles generated by the Schrödinger equation,

$$i\hbar \frac{\partial}{\partial t} \Phi(t) = H(t) \Phi(t)$$

where \( H(t) = H(B(t), E(t)) \),

it is convenient to expand the solution \( \Phi(t) \) in terms of the eigenstates of \( H(t) \):

$$\Phi(t) = \sum_n a_n(t) \exp \left( \frac{i}{\hbar} \chi(n, t) \right) \Psi_1(n, t),$$

where

$$\chi(n, t) = \gamma(n, t) - \int_0^t \mathrm{d}t_1 E(n, B(t_1), E(t_1)).$$

The first term \( \gamma(n, t) \) in (28) is a phase that vanishes for \( t = 0 \), but is otherwise arbitrary. Its value will be determined by the reasoning leading to the adiabatic approximation. The second term,

$$\Phi_D(n) = -\frac{1}{\hbar} \int_0^t \mathrm{d}t_1 E(n, B(t_1), E(t_1)),$$

known as the ‘dynamical phase’, produces a contribution which cancels \( H(t) \Phi(t) \) in the wave equation. We shall take as the initial condition \( \Phi(t_0) = \Psi_1(m, 0) \) or, in other words, \( a_n(0) = \delta_{n,m} \). In this case we can replace the exact Schrödinger equation by the system of differential equations:

$$i\hbar \dot{a}_n(t) = a_n(t) \left( \dot{\gamma}(n, t) - \langle \Psi(n, t) | i\hbar \frac{\partial}{\partial t} \Psi(n, t) \rangle \right)$$

$$- \sum_{k \neq n} a_k(t) \exp \left( \chi(k, t) - \chi(n, t) \right) \langle \Psi(n, t) | i\hbar \frac{\partial}{\partial t} \Psi(k, t) \rangle.$$ 

(30)

Since the adiabatic condition requires that \( a_n(t) \approx 1 \) whatever \( t \), a necessary condition to ensure its validity is to cancel the coefficient of \( a_n(t) \) on the rhs of equation (30). This is achieved if we make the following choice for the ‘gauge’ \( \gamma(n, t) \):

$$\gamma(n, t) = \int_0^t \mathrm{d}t_1 \langle \Psi(n, t_1) | i\hbar \frac{\partial}{\partial t} \Psi(n, t_1) \rangle.$$ 

(31)

Later on, we shall see that this non-integrable gauge is a basic ingredient in the mathematical expression of Berry’s phase in terms of the instantaneous wavefunctions \( \Psi(m, t) \). The next step to validate the adiabatic approximation is to find appropriate conditions allowing the sum \( \sum_{n \neq m} |a_n(t)|^2 \) to remain below a predefined level for \( t > 0 \). This task will be performed in detail in section 5 but for the moment, let us assume that it is achieved. Within the adiabatic approximation, the solution of the Schrödinger equation \( i\hbar \frac{\partial}{\partial t} \Phi(t) = H(t) \Phi(t) \), with the initial condition \( \Phi(0) = \Psi(m, 0) \), is given by

$$\Phi_{\text{ADB}}(m, t) = \exp \left( \frac{i}{\hbar} \left( \gamma(m, t) - \int_0^t \mathrm{d}t_1 E(m, B(t_1), E(t_1)) \right) \right) \Psi(m, t).$$

(32)
We can now calculate the phase shift of the wavefunction $\Phi_{ADB}(m, t)$ at the end of the adiabatic cycle:

$$\arg\left(\frac{\Phi_{ADB}(m, T)}{\Phi_{ADB}(m, 0)}\right) = \arg\left(\frac{\Psi(m, T)}{\Psi(m, 0)}\right) + \frac{1}{\hbar}\int_0^T \text{d}t E(m, B(t), E(t)),$$

$$= \frac{1}{\hbar}\int_0^T \text{d}t E(m, B(t), E(t)) + \beta(m), \quad (33)$$

where we have made Berry’s phase $\beta(m)$ stand out on the rhs of the above equation. Using equation (31), one immediately gets the basic formula giving $\beta(m)$ in terms of the instantaneous wavefunctions $\Psi_1(m, t)$:

$$\beta(m) = \int_0^T \text{d}t \langle \Psi(m, t) | i\frac{\partial}{\partial t} \Psi(m, t) \rangle + \phi(m), \quad (34)$$

$$\phi(m) = \arg\left(\frac{\Psi_1(m, T)}{\Psi_1(m, 0)}\right).$$

It is crucial to note that the ‘dynamical phase’ $\phi_D(m)$ and $\beta(m)$ obey different scaling laws under the transformation $B \to \xi B$, involving an arbitrary real parameter $\xi$. If one remembers that $E(m, B(t), E(t)) = \gammaSB \mathcal{E}(m, \lambda(t))$, one sees immediately that $\phi_D(m)$ is multiplied by $\xi$, while $\beta(m)$, being geometric, is unchanged. In principle, this scaling difference could be used to separate the two phases. However, a more practical way to isolate Berry’s phase consists in measuring the phase for a given adiabatic cyclic evolution and that associated with the ‘image’ circuit obtained by performing on the Hamiltonian parameters the transformations: $\alpha(t) \to -\alpha(t)$, $\psi(t) \to -\psi(t)$, while keeping the other two unchanged. The two competing phases are transformed as $\phi_D(m) \to \phi_D(m)$, $\beta(m) \to -\beta(m)$, so that the dynamical phase can be eliminated by subtraction.

To end this mini-review, we would like to give, within the present context, a simplified description of the topological interpretation of Berry’s phase, from Simon [2]. We have just shown that $\Phi_{ADB}(m, t)$ is a physical state obeying the Schrödinger equation within the adiabatic approximation. For our purpose, it is convenient to introduce the mathematical vector state $\Phi_\parallel(m, t) = \exp\left(-i\frac{\phi_D(m, t)}{\hbar}\right)\Phi_{ADB}(m, t)$ and to calculate the differential form

$$\langle \Phi_\parallel | d\Phi_\parallel \rangle = \langle \Phi_{ADB} | -i\frac{\partial}{\partial t} \Phi_{ADB} \rangle + \langle \Phi_{ADB} | \frac{1}{\hbar} H(t) \Phi_{ADB} \rangle dt = 0.$$  

The evolution of the state vector $\Phi_\parallel$ along the closed loop is then said to satisfy the ‘parallel transport’ condition $\langle \Phi_\parallel | d\Phi_\parallel \rangle = 0$. If the state $\Phi_\parallel(t)$ is injected into the general formula for Berry’s phase, one immediately finds that

$$\beta(m) = \arg\left(\frac{\Phi_\parallel(m, T)}{\Phi_\parallel(m, 0)}\right), \quad (35)$$

in the case of parallel transport.

Let us now give a rather elementary introduction to the mathematical concepts behind the above notion of ‘parallel transport’ applied to the evolution of quantum states. This arises rather naturally from a linear fiber bundle interpretation of quantum mechanics. The linear fiber bundle associated with the quantum state space is constructed from the ‘base space’ $E(\rho)$ of the ‘pure’ state density matrix $\rho = |\Psi\rangle\langle\Psi|$. Assuming for simplicity that the states $\Psi$ have unit norms, one finds that $\rho$ satisfies the simple nonlinear relation $\rho^2 = \rho$. This implies clearly that $E(\rho)$ has a non-trivial topology. The ‘fiber’ is the one-dimensional space associated with a definite $\rho$. The vector states of the fiber are given by $\Phi = \exp(i\phi)|\Psi\rangle$, where $\phi$ is an arbitrary
phase and $\Psi$ a representative state of the fiber. The infinitesimal variation $d\Phi$ during the quantum cycle is said to be ‘vertical’ if it takes place along the fiber: $d\Phi = d\Phi$, where $d\Phi$ is an infinitesimal $c$-number. Conversely, it is called ‘horizontal’ or ‘parallel’ if $\langle\Phi|d\Phi\rangle = 0$.

The fact that Berry’s phase can be viewed as a displacement respective to the fiber, associated with $\rho(0) = \rho(T)$—in our case a phase shift resulting from a parallel transport along a closed loop drawn upon the base space $E(\rho)$—emphasizes its topological character.

4.2. Berry’s phase as a loop integral in the Hamiltonian $H(B(t), E(t))$ parameter space

Our starting point is the formula (34) giving Berry’s phase for quantum adiabatic cycles associated with the instantaneous wavefunction $\Psi(m, t)$.

A group theoretical derivation. To evaluate the expression (34), it is convenient to write $\beta(m)$ in terms of $\hat{\Psi}(m, \lambda)$:

$$\beta(m) - \phi(m) = \int_0^T dt \langle\hat{\Psi}(m, \lambda(t))|\frac{\partial}{\partial t}\hat{\Psi}(m, \lambda(t))\rangle + \int_0^T dt \langle\hat{\Psi}(m, \lambda(t))|U^{-1}(R(t))i\frac{\partial}{\partial t}(U(R(t)))|\hat{\Psi}(m, \lambda(t))\rangle. \quad (36)$$

Since $|\hat{\Psi}(m, \lambda(t))\rangle$ is a real vector in the $|S, m\rangle$ basis, the first term of the integral can be written as

$$\frac{i}{2}\frac{\partial}{\partial t}\left\{\sum_{\mu=-S}^{S}\langle[S, \mu]|\hat{\Psi}(m, \lambda(t))\rangle^2\right\}.$$ If the cyclic condition $\lambda(T) = \lambda(0)$ is satisfied, the line integral of this term is zero. If one now uses the explicit form of $U(R(t))$ in terms of the spin operator $S$ (equation (16)), it is easily seen that the second term of equation (36) is a linear combination of time derivatives of the Euler angles:

$$i\hbar U^{-1}(R(t))\frac{\partial}{\partial t}(U(R(t))) = \dot{\alpha} D_{\alpha} + \dot{\theta} D_{\theta} + \dot{\phi} D_{\phi}. \quad (37)$$

Let us consider $D_{\alpha}$:

$$D_{\alpha} = \exp\left(\frac{i}{\hbar}S_{\alpha}(t)\right) \cdot \exp\left(\frac{i}{\hbar}S_{\theta}(t)\right) \cdot \exp\left(\frac{i}{\hbar}S_{\phi}(t)\right) \cdot \exp\left(-\frac{i}{\hbar}S_{\alpha}(t)\right) \cdot S_{\alpha} \cdot \exp\left(-\frac{i}{\hbar}S_{\theta}(t)\right) \cdot \exp\left(-\frac{i}{\hbar}S_{\phi}(t)\right).$$

Since $S_{\alpha}$ commutes with the exponential $\exp(-\frac{i}{\hbar}S_{\alpha}(t))$, $D_{\alpha}$ reduces to

$$D_{\alpha} = U^{-1}(R(t))U(R(t))S_{\alpha} = S_{\alpha}. \quad (38)$$

For the two remaining terms such as $D_{\phi}$ the calculation is not so simple. Using the fact that $S_{\phi}$ commutes with $\exp(-\frac{i}{\hbar}S_{\phi}(t))$, one can write

$$D_{\phi} = \exp\left(\frac{i}{\hbar}S_{\phi}(t)\right) \cdot \exp\left(-\frac{i}{\hbar}S_{\phi}(t)\right) S_{\phi} \cdot \exp\left(\frac{i}{\hbar}S_{\theta}(t)\right) \cdot \exp\left(-\frac{i}{\hbar}S_{\theta}(t)\right) \cdot \exp\left(-\frac{i}{\hbar}S_{\alpha}(t)\right).$$
Using equation (12), one can derive the commutation relation

\[ S_z \cdot \exp \left( -\frac{i}{\hbar} S_z \theta(t) \right) = \exp \left( -\frac{i}{\hbar} S_z \theta(t) \right) \cdot (S_z \cos \theta - S_y \sin \theta). \]

After repeating a similar operation to push \( \exp \left( -\frac{i}{\hbar} S_z \alpha(t) \right) \) to the left of \( (S_z \cos \theta - S_y \sin \theta) \), one arrives finally at the following expression:

\[ D_\psi = S_z \cos \theta + \sin \theta(-S_x \cos \alpha + S_y \sin \alpha). \]

In a similar way one obtains

\[ D_\theta = S_y \cos \alpha + S_z \sin \alpha, \]

but this term will not lead to any contribution to \( \beta \) because of the boundary condition \( \theta(T) = \theta(0) \). Since the quantum average of \( \mathbf{S} \) relative to the state \( \hat{\psi}(m, \lambda) \) has a single component along \( \hat{z} \), we end up with the compact expression

\[ \beta(m) - \phi(m) = \int_0^T p(m, \lambda(t))(\cos \theta \dot{\psi} + \dot{\theta}) dt. \]

We now have to calculate the phase shift \( \phi(m) = \arg \{\Psi(m, T)/\Psi(m, 0)\} \) appearing in equation (34). A preliminary step is to rewrite the unitary transformation \( U(R(t)) \) under a modified form:

\[
\begin{align*}
U(R(t)) &= V(t) \exp \left( -\frac{i}{\hbar} S_z (\varphi(t) + \alpha(t)) \right), \\
V(t) &= \exp \left( -\frac{i}{\hbar} S_y \varphi(t) \right) \exp \left( -\frac{i}{\hbar} S_z \theta(t) \right) \exp \left( \frac{i}{\hbar} S_x \varphi(t) \right).
\end{align*}
\]

The transformation law given by equation (12) can be extended to any tensor operator \( S_i, S_j, S_k \ldots \), and consequently to any analytical function of \( S_z \). Using the extended law, one can write \( V(t) \) in the compact form:

\[ V(t) = \exp \left( -\frac{i}{\hbar} \theta(t)(\cos \varphi(t)S_z + \sin \varphi(t)S_y) \right). \]

Using the boundary conditions \( \theta(T) = \theta(0), \lambda(T) = \lambda(0), \alpha(T) = \alpha(0) + n_\varphi \pi \) and \( \varphi(T) = \varphi(0) + 2n_\varphi \pi \), one obtains the relation \( V(T) = V(0) \). Introducing for convenience the notation \( U(\hat{z}, u) = \exp \left( -\frac{i}{\hbar} S_y u \right) \) it is then possible to rearrange \( \Psi(m, T) \) as follows:

\[
\begin{align*}
\Psi(m, T) &= V(0)U(\hat{z}, \varphi(T) + \alpha(T))\hat{\psi}(m, \lambda(0)), \\
&= V(0)U(\hat{z}, \varphi(0) + \alpha(0))U(\hat{z}, \Delta \varphi + \Delta \alpha)\hat{\psi}(m, \lambda(0)), \\
\text{with} \quad \Delta \varphi + \Delta \alpha &= \varphi(T) + \alpha(T) - \varphi(0) - \alpha(0), \\
&= (2n_\varphi + n_\alpha)\pi.
\end{align*}
\]

The next step is to prove that \( \hat{\psi}(m, \lambda(0)) \) is an eigenstate of \( U(\hat{z}, \Delta \varphi + \Delta \alpha) \) using its expansion in terms of the angular momentum state vectors \( |S, q \rangle \), eigenstates of \( S_z \) of eigenvalue \( \hbar q \):

\[ \hat{\psi}(m, \lambda(0)) = \sum_{|m - 2n| \leq S} C_{m,n}(\lambda(0))|S, m - 2n \rangle. \]

Using equations (44), one sees immediately that, as a consequence of the quantum cycle boundary conditions, each state \( |S, m - 2n \rangle \) appearing in the above sum is an eigenstate of \( U(\hat{z}, \Delta \varphi + \Delta \alpha) \) with the eigenvalue \( \exp(-i\pi (2n_\varphi + n_\alpha)) \). This leads to the basic relation connecting \( \Psi(m, T) \) and \( \Psi(m, 0) \):

\[
\begin{align*}
\Psi(m, T) &= \exp(-im\pi (2n_\varphi + n_\alpha))V(0)U(\hat{z}, \varphi(0) + \alpha(0))\hat{\psi}(m, \lambda(0)) \\
&= \exp(-im(\Delta \varphi + \Delta \alpha))\Psi(m, 0).
\end{align*}
\]
One immediately obtains the phase shift $\phi(m)$ appearing on the rhs of equation (34):

$$
\phi(m) = \arg \frac{\Psi(T)}{\Psi(0)} = -m(\varphi(T) - \varphi(0) + \alpha(T) - \alpha(0)) = -m \int_0^T (\varphi(t) + \alpha(t)) \, dt.
$$

(47)

By combining equations (41) and (47), we arrive at the expression of Berry’s phase for an arbitrary quantum adiabatic cycle generated by the Hamiltonian $H(B(t), E(t))$ of equation (1) with $E(t) \cdot B(t) = 0$:

$$
\beta(m) = -\int_0^T [(m - p(m, \lambda(t))) \cos \theta(t)] \varphi(t) \, dt - \int_0^T [m - p(m, \lambda(t))] \alpha(t) \, dt.
$$

(48)

**Berry’s phase as a loop integral of an Abelian gauge field.**  Berry’s phase $\beta(m)$ given by equation (48) can be written as a loop integral around a closed curve $C$ drawn in the parameter space $\mathbb{C}P^2$, using $\lambda, \theta, \varphi, \alpha$ as coordinates:

$$
\beta(m) = -\oint_C ((m - p(m, \lambda)) \cos \theta) d\varphi + (m - p(m, \lambda)) d\alpha).
$$

(49)

The above formula suggests that $\beta(m)$ could be written as a loop line integral of an Abelian gauge field $A_i(x, t)$, defined in the parameter space. The non-vanishing components of the gauge field candidate, $A_\varphi$ and $A_\alpha$, are then given by

$$
A_\varphi = -m + p(m, \lambda) \cos \theta, \quad A_\alpha = -m + p(m, \lambda).
$$

(50)

Thus, Berry’s phase looks like the Aharonov–Bohm phase [29], for the Abelian gauge field $(A_\varphi, A_\alpha)$:

$$
\beta(m) = \oint_C (A_\varphi d\varphi + A_\alpha d\alpha).
$$

(51)

What remains to be proved is that the above expression is indeed invariant under the gauge transformation of the Abelian field:

$$
A_\varphi \to A_\varphi + \frac{\partial}{\partial \varphi} g(\varphi, \theta, \alpha, \lambda), \quad A_\alpha \to A_\alpha + \frac{\partial}{\partial \alpha} g(\varphi, \theta, \alpha, \lambda).
$$

(52)

The crucial point is that $g(\varphi, \theta, \alpha, \lambda)$ must be a single-valued function of $B$ and $E \otimes E$, like the gauge field itself. This implies that $g(\varphi, \theta, \alpha, \lambda)$ must be a periodic function of $\varphi$ and $\alpha$, with periods respectively of $2\pi$ and $\pi$. Remembering the boundary conditions (14) for the quantum cycle $C$, one finds immediately that the gauge contributions to $\beta(m)$, $g(\varphi(0) + 2n, \theta(0), \alpha(0) + n, \lambda(0)) - g(\varphi(0), \theta(0), \alpha(0), \lambda(0))$ do indeed vanish. This completes the identification of $\beta(m)$ as a Aharonov–Bohm phase.

**Berry’s phase geometry generated by nonlinear spin Hamiltonians for $S > 1$.**  For the sake of simplicity, in the next sections we shall concentrate on Berry’s cycles where $\lambda$ and $\alpha$ are the only time-varying parameters. The question then arises as to whether we shall not lose in this way most of the new features introduced with the nonlinearity of $H(t)$. In order to make clear the cause for our concern, let us perform the change of variables upon the two leftover parameters which map the associated 2D manifold onto a 2D sphere: $\lambda = -2\cot(\bar{\varphi})$ with $0 < \bar{\theta} < \pi$ and $\bar{\varphi} = 2\alpha$. In [24] we have given an explicit form of the $\mathbb{C}P^2$ metric obtained with these kind of coordinates. For the sub-manifold associated with $\bar{\varphi} = \bar{\theta} = 0$ it turns out
that this metric $ds^2 = \frac{1}{4} (d\tilde{\phi}^2 \sin^2 \theta + d\tilde{\theta}^2)$ is identical to the one associated with a 2D sphere having a radius $1/2$ (this factor results from the choice: $\tilde{\phi} = 2\alpha$).

To answer the question, we can now consider the component of the gauge field $A_\alpha$ for two different cases. For $S = 1, m = 1$, the gauge field is given by the same expression as the gauge field associated with a linear Hamiltonian $A_\alpha(\tilde{\theta}) = \cos \tilde{\theta} - 1$. However, the situation is totally different for the case $S = 2, m = 0$, where $A_\alpha(\tilde{\theta})$ exhibits the peculiar shape shown in figure 2. This implies that the geometry involved in Berry’s phase for the sub-manifold differs from its usual interpretation in terms of the solid angle defined by a closed loop drawn upon a 2D sphere. The origin of this phenomenon lies in the fact that the density matrix space for $S = 2$ is not $\mathbb{C}P^2$ but rather $\mathbb{C}P^4$. Unlike the case $S = 1$, there is no longer a one to one correspondence between the parameter space and the density matrix space. It is thus not surprising that the geometry involved in Berry’s phase for a nonlinear Hamiltonian should differ from the linear case, even for the simple cycles involving only the variations of the two parameters $\lambda$ and $\alpha$.

5. Non-adiabatic corrections in Berry’s cycles using the rotating frame Hamiltonian

For quantum cycles of finite duration $T$, the time derivatives of the Hamiltonian parameters cannot take arbitrary small values. For instance, in the experiments discussed in a separate paper [5], the quadratic spin coupling generated by an ac Stark effect induces an atomic instability when $\lambda \approx 1$. As a result, the measurement of Berry’s phases studied by interferometry experiments on alkali atoms will require good control of the non-adiabatic corrections arising from the time-dependent external $B, E$ fields.

We shall consider separately the effect coming from the periodic parameters $\alpha$ and $\phi$ in subsections 5.1 and 5.2, and the non-periodic $\lambda$ in subsection 5.3. Our method involves the time-dependent Schrödinger equation in the rotating frame. We use an adiabatic approximation within this frame, the slowly varying parameters being, then, the time derivatives of the periodic parameters $\dot{\alpha}$ and $\dot{\phi}$.
We set $\Phi_1(t) = U(R(t))\tilde{\Phi}(t)$, and write the wave equation relative to $\tilde{\Phi}_1(t)$ in the rotating frame:

$$
\left( i\hbar \frac{d}{dt} - \hat{H}(B(t), E(t)) \right) \tilde{\Phi}_1(t) = -U^{-1}(R(t))i\hbar \frac{d}{dt} U(R(t))\tilde{\Phi}(t) = -(D\dot{\alpha} + D\dot{\psi} + D\dot{\theta})\tilde{\Phi}(t) = \gamma_S S \cdot \Delta B(t) \tilde{\Phi}(t).
$$

The rotating frame Hamiltonian can be written as

$$
\tilde{H}(t) = \hat{H}(B(t), E(t)) + \gamma_S S \cdot \Delta B(t),
$$

where $\Delta B(t)$ is an additional effective magnetic field generated by the Coriolis effect. It can be decomposed into a longitudinal component $\Delta B//_L(t)$ and a transverse one $\Delta B\perp L(t)$. Using the explicit expressions of $D\alpha$ and $D\phi$ in subsection 3.2 and assuming here—for the sake of simplicity—that $\dot{\theta} = 0$, one arrives at the following expressions for of the effective field:

$$
\begin{align*}
\Delta B//_L &= -\gamma_S^{-1}(\cos \theta \dot{\phi} + \dot{\alpha}) \hat{z} \\
\Delta B\perp L &= -\gamma_S^{-1} \sin \theta \dot{\phi}(-\cos \alpha \hat{x} + \sin \alpha \hat{y}) \\
&= \gamma_S^{-1} \sin \theta \dot{\phi} R(\hat{z}, -\alpha) \hat{x}.
\end{align*}
$$

The role of those two components will be examined separately.

5.1. Berry’s phase and its non-adiabatic corrections involving the longitudinal effective magnetic field $(B(t) + \Delta B//_L(t)) \hat{z}$

We consider, first, the part of the Hamiltonian governing the evolution of $\tilde{\Phi}_1(t)$ associated with $\Delta B//_L$, which is the sole present if $\dot{\phi} = 0$. This part can be written very simply in terms of $\hat{H}(B, E)$:

$$
\tilde{H}//_L(t) = \hat{H}(B(t) + \Delta B//_L(t), E(t)).
$$

To proceed, it is convenient to introduce the dimensionless parameter $\eta$:

$$
\eta = -\frac{\Delta B//_L}{B} = \frac{\cos \theta \dot{\phi} + \dot{\alpha}}{\gamma_S B}.
$$

Using $\tilde{B} = (1 - \eta)B$, the eigenvalues of $\tilde{H}//_L(t)$ are simply

$$
E(m, B + \Delta B//_L, E) = \gamma_S \tilde{B} \mathcal{E}(m, \lambda) = \gamma_S B(1 - \eta)\mathcal{E} \left( m, \frac{\lambda}{1 - \eta} \right).
$$

Since the quantum cycles generated by $\tilde{H}//_L(t)$ are devoid of any topology, we can expect Berry’s phase relative to the laboratory frame to be buried inside the dynamical phase of $\hat{H}(t)$:

$$
\tilde{\phi}_{D}//_L(\lambda, \eta, m) = -\int_0^T dt \gamma_S B(1 - \eta)\mathcal{E} \left( m, \frac{\lambda}{1 - \eta} \right).
$$

Indeed, it is easily seen that Berry’s phase is given by the first-order contribution in the $\eta$-series expansion of the above integral:

$$
\begin{align*}
\tilde{\phi}_{D}//_L^{(1)}(\lambda, \eta, m) &= -\int_0^T dt \gamma_S B \left( \mathcal{E}(m, \lambda) - \lambda \frac{\partial \mathcal{E}(m, \lambda)}{\partial \lambda} \right) \\
&= \int_0^T dt \ p(m, \lambda)(\cos \theta \dot{\phi} + \dot{\alpha}), \\
&= \beta(m) - \phi(m).
\end{align*}
$$
where we have used equation (23) of section 3. The phase \( \phi(m) \) is recovered by returning to the laboratory frame. The even-order contributions \( \propto \eta^2 \) are eliminated by subtracting term by term those coming from image circuits associated with opposite signs of the \( \eta \)-parameter (i.e. \( \varphi \rightarrow -\varphi \) and \( \alpha \rightarrow -\alpha \)).

We now show that it is possible to find a magic value of \( \lambda = \lambda^*(\eta) \) that leads to cancellation of all the contributions of the dynamical phase \( \propto \eta^{2p+1} \). More precisely let us define

\[
\Delta p(m, \lambda, \eta) = \frac{1}{2} \eta \left( \frac{\eta}{1+\eta} \right) - \frac{1}{2} \eta \right) - p(m, \lambda),
\]

where

\[
\Delta \beta^{(2)}(m) = \int_0^T dt \left( \cos \theta \dot{\phi} + \dot{\alpha} \right) q(m, \lambda) \eta^2.
\]

The above formula seems to suggest that the integrand in the expression giving \( \Delta \beta^{(2)}(m) \) is singular when \( \eta = \pm 1 \). In fact it is easily seen that this is not the case. Indeed, the ratios \( \mathcal{E}(m, \lambda)/\lambda \) converge toward the eigenvalues of \( (S_x/\hbar)^2 : 0, 1, 4, \ldots, S^2 \), as \( \lambda \rightarrow \infty \). As a preliminary step, let us consider the lowest order expansion of the rhs of \( \Delta p(m, \lambda, \eta) \) with respect to \( \eta \)

\[
\beta \left( \frac{2}{m} \right) = \int_0^T dt \left( \cos \theta \dot{\phi} + \dot{\alpha} \right) \frac{1}{6} \left( \lambda^3 \mathcal{E}''(m, \lambda) + 3 \lambda^2 \mathcal{E}'(m, \lambda) \right).
\]

Figure 3 shows the variations of \( q(m, \lambda) \) versus \( \lambda \) in the cases \( S = 2 \) and \( S = 4 \) for \( m = 0 \). A quite remarkable effect, clearly visible in figure 3, is the vanishing of \( q(0, \lambda) \) for a particular value of \( \lambda \), in the vicinity of the maximum of \( \frac{\partial}{\partial \lambda} p(0, \lambda) \). This should allow for rather accurate determinations of \( \beta(0) \), as explained in the caption of figure 3.

In the case where \( \alpha \) is the sole time-dependent parameter we have found that a similar property holds in fact to all orders in \( \eta \). More precisely, we have shown that there exists for a given value of \( \eta \) a ‘magic’ value \( \lambda^*(S, \eta) \) such that the non-adiabatic correction to \( p(m, \lambda) \) cancels exactly:

\[
\Delta p(0, \lambda^*(S, \eta), \eta) = 0.
\]

The magic values can be accurately represented by the polynomial fits given below:

\[
\lambda^*(2, \eta) = 0.838213 - 0.0837823 \eta - 0.0431478 \eta^2 - 0.0231887 \eta^3 + 0.0207986 \eta^4
\]

\[
\lambda^*(4, \eta) = 0.509982 - 0.0900927 \eta - 0.0349985 \eta^2 - 0.0436495 \eta^3 + 0.0373634 \eta^4.
\]

As it is apparent from the above formulas, the ‘magic’ values \( \lambda^*(S, \eta) \) are slowly varying functions of \( \eta \) within the interval \( 0 \leq \eta \leq 0.5 \). We have verified that they both satisfy equation (64) with a precision better than \( 3 \times 10^{-7} \). In conclusion, the above results open the road to measurements of Berry’s phase \( \beta(0) \) for \( S = 2 \) and \( S = 4 \), for quantum cycles where \( \alpha \) is the sole varying Euler angle, under conditions where the non-adiabatic corrections can be kept below the one ppm level.

5.2. Non-adiabatic corrections induced by the effective transverse magnetic field \( \Delta B_\perp(t) \)

As a first step, we calculate the corrections to Berry’s phase relative to the laboratory frame, odd under \( \eta \)-reversal, coming from the dynamical phase induced by \( \Delta B_\perp(t) \) within the rotating
Figure 3. Comparison of Berry’s phase to its lowest order non-adiabatic corrections, when $\alpha$ is the only time-dependent Euler angle, for a quantum cycle involving the spin configurations $S = 2$ or $4$ and $m = 0$. Berry’s phase is given by $\beta(0) = \int_0^T dt p(0, \lambda(t)) \dot{\alpha}(t)$. Using the results of subsection 5.1, one finds that the non-adiabatic correction is given by $\Delta_1(\beta(0)) = \int_0^T dt q(0, \lambda(t)) \dot{\alpha}(t) \left( \frac{\dot{\alpha}}{\gamma_S B(t)} \right)^2$. The explicit expression of $q(m, \lambda)$ in terms of the second- and third-order derivatives of $E(m, \lambda)$ with respect to $\lambda$ can be found in equation (63). One sees clearly that, for $S = 2$, if the parameter $\lambda$ is required to stay within the small interval $0.8 \pm 0.2$, then $\Delta_1(\beta(0))/\beta(0)$ will be $< 10^{-3}$ provided $\dot{\alpha}(t)$ is kept smaller than one-tenth of the Larmor angular frequency $\gamma_S B(t)$. Similarly, for $S = 4$, there is a value of $\lambda$, for which $q(0, \lambda)$ cancels. The partial derivative $\partial p(0, \lambda)/\partial \lambda$ is governing the response of $\beta(0, \lambda)$, say, to a small variation of the $B$ field, and are nearly at their maximum when $q(0, \lambda)$ is close to 0. Note also the fourfold increase of the sensitivity for $S = 4$ compared to $S = 2$. 

5.2.1. Correction to the dynamical phase. It is convenient to introduce the dimensionless parameters $\mu$ and $\tilde{\mu}$ and the dimensionless operator $\Sigma = \hbar^{-1} S$:

$$\mu = -\frac{\Delta B_\perp}{B} = \frac{\sin \theta \dot{\phi}}{\gamma_S B}, \quad \tilde{\mu} = \frac{\mu}{1 - \eta}. \quad (66)$$

We rewrite $\tilde{H}(t)$ as follows:

$$\tilde{H}(t) = \gamma_S \tilde{B} h (\tilde{\mathcal{H}}(\tilde{\lambda}) - \tilde{\mu} (-\cos \alpha \Sigma_x + \sin \alpha \Sigma_y)). \quad (67)$$

To proceed, it is convenient to introduce the perturbation expansion of the eigenenergies of $\tilde{H}(t)$:

$$\tilde{E}(\tilde{\lambda}, \tilde{\mu}) = \gamma_S \tilde{B} h (E(m, \tilde{\lambda}) + \tilde{\mu}^2 E_{\perp}^{(2)}(m, \tilde{\lambda}) + O(\tilde{\mu}^4)). \quad (68)$$
The second-order energy shift caused by the $\mu$-contribution to $\tilde{H}(t)$ is given, to all orders in $\eta$, by

$$\mathcal{E}^{(2)}_{\perp}(m, \tilde{\lambda}) = \sum_{|n-m|\text{odd}} \frac{|\langle \psi(n, \tilde{\lambda}) | \cos \alpha \Sigma_x - \sin \alpha \Sigma_y | \psi(m, \tilde{\lambda}) \rangle|^2}{\mathcal{E}(m, \lambda) - \mathcal{E}(n, \tilde{\lambda})}. \quad (69)$$

The symmetry properties of $H(\lambda)$ (section 2) have two consequences: (i) only $|n-m|$-odd terms contribute to the sum and (ii) the cross terms involving $\Sigma_x \Sigma_y$ cancel out. This means that there is no contribution proportional to $\sin \alpha \cos \alpha$. We are left with the terms proportional to $\cos^2 \alpha$ and $\sin^2 \alpha$. We shall assume that the velocity $\dot{\lambda}$ is a slowly varying function of $t$ during the cycle, so that $\cos^2 \alpha$ and $\sin^2 \alpha$ can be replaced by their average values of $1/2$.

At this point we have found convenient to solve numerically two auxiliary problems, namely the search of the eigenstates of the two Hamiltonians:

$$\mathcal{H}_x(\lambda, \mu) = \mathcal{H}(\lambda) - \mu \Sigma_i \quad \text{for} \quad i = x \quad \text{and} \quad i = y.$$  

Let $\mathcal{E}_{x,y}(\lambda, \mu)$ be the individual eigenenergies of $\mathcal{H}_{x,y}$ which are even functions of $\mu$. To lowest order in $\mu$, they take the form

$$\mathcal{E}_{x,y}^{(2)}(\lambda, \mu) = \mathcal{E}(\lambda) + \mu^2 \mathcal{E}_{x,y}^{(2)}(\lambda) + \mathcal{O}(\mu^4), \quad (70)$$

where $\mathcal{E}_{x,y}^{(2)}(\lambda)$ are obtained by an interpolation toward $\mu = 0$. The overall second-order energy shift of equation (69) can be expressed as

$$\mathcal{E}^{(2)}_{\perp}(m, \tilde{\lambda}) = \frac{1}{2} (\mathcal{E}_{x}^{(2)}(\tilde{\lambda}) + \mathcal{E}_{y}^{(2)}(\tilde{\lambda})). \quad (71)$$

We have now all we need to write the contribution to the dynamical phase associated with $\tilde{H}(t)$ to second order in $\mu$ and all orders in $\eta$:  

$$\phi^{(2)}_{D,\perp} = -\int_0^T dt \gamma_S \tilde{B} \mu^2 \mathcal{E}^{(2)}_{\perp}(m, \tilde{\lambda}),$$

$$= \int_0^T dt \gamma_S B \left[ \frac{\mu^2}{1 - \eta} \mathcal{E}^{(2)}_{\perp}(m, \frac{\lambda}{1 - \eta}) \right]. \quad (72)$$

In practice, we concentrate on the contribution to the dynamical phase relative to $\tilde{H}(t)$ of the order of $\mu^2 \eta$, which is easily derived from the above equation. In order to highlight the close connection with Berry’s phase we replace $\eta$ in the integrand by its explicit expression of equation (57):

$$\phi^{(2,1)}_{D,\perp}(m) = \int_0^T dt \mu^2 (\cos \theta \psi' + \dot{\psi} \mu^2 \mathcal{E}^{(2)}_{\perp}(m, \lambda),$$

with $p^{(2)}(m, \lambda) = \left( 1 + \frac{\partial}{\partial \lambda} \right) \mathcal{E}^{(2)}_{\perp}(m, \lambda)). \quad (73)$

Despite the sign difference in the $\lambda$ derivative, our notation underlines the analogy of the above correction with Berry’s phase, but the presence of the factor $\mu^2 \sim \psi^2$ spoils its geometric character. The quantity $p^{(2)}(m, \lambda)$ has been plotted in figure 4 for the cases $S = 2$, $m = 0$ and $m = -1$ over the interval $0.7 < \lambda < 1.2$. As expected, the non-adiabatic corrections are $\lesssim \mu^2$ for $m = 0, -1$. We have performed a similar computation for $m = 1$ and found, in contrast, that $p^{(2)}(m = 1, \lambda)$ blows up to values $\gtrsim 100$.

5.2.2. Berry’s phase associated with $\Delta B_\perp$. The full Hamiltonian $\tilde{H}(t) = \tilde{H}_{//}(t) + \gamma_S \mathbf{S} \cdot \Delta \mathbf{B}_\perp$ generates quantum cycles endowed with a non-trivial geometry since the effective transverse magnetic field $\Delta \mathbf{B}_\perp$ rotates about the $\hat{z}$ axis with the angular velocity $\dot{\alpha}$. As above we shall
limit ourselves to the second-order contribution with respect to \( \mu \). We proceed by writing the eigenfunctions of \( \tilde{H}(t) \) in the first-order expansion with respect to the parameter \( \tilde{\mu} \):
\[
\hat{\psi}(m, \tilde{\lambda}) = \tilde{\mu} \hat{\psi}^{(1)}(m, \tilde{\lambda}) + O(\tilde{\mu}^2).
\]
In a similar way to before, it is convenient to introduce the first-order expansion of the eigenfunctions \( \hat{\psi}_{x,y}(m, \lambda) \) of \( H_{x,y}(\lambda, \mu) \):
\[
\hat{\psi}^{(1)}_{x,y}(m, \lambda) = \sum_{|n-m|\text{odd}} \frac{\langle \hat{\psi}(n, \lambda) | \Sigma_{x,y} | \hat{\psi}(m, \lambda) \rangle}{E(m, \lambda) - E(n, \lambda)} \hat{\psi}(n, \lambda).
\]
Using equation (71), one finds that the first-order contribution to the \( \tilde{H}(t) \) eigenfunction (divided by \( \tilde{\mu} \)) can be written as
\[
\hat{\psi}^{(1)}(m, \tilde{\lambda}) = -\cos \alpha \hat{\psi}^{(1)}_y(m, \tilde{\lambda}) + \sin \alpha \hat{\psi}^{(1)}_x(m, \tilde{\lambda}).
\]
It is then easily seen that Berry’s phase appears only to second order in \( \mu \) and involves the familiar time derivative product: \( \int_0^T dt \langle \hat{\psi}^{(1)}(m, \tilde{\lambda}) | i \hbar \frac{\partial}{\partial t} \hat{\psi}^{(1)}(m, \tilde{\lambda}) \rangle \), which is proportional to \( \dot{\alpha} \). If the other parameters in the integrand vary slowly during the closed cycle, the only terms surviving are those proportional to \( \cos^2 \alpha \) and \( \sin^2 \alpha \) which can be replaced by their average values of \( 1/2 \). This leads to the following correction to Berry’s phase:
\[
\Delta^{(2)} \beta_{\perp}(\tilde{\lambda}, \tilde{\mu}) = \int_0^T dt \tilde{\mu}^2 \alpha \Im \{ \langle \hat{\psi}^{(1)}_y(m, \tilde{\lambda}) | \hat{\psi}^{(1)}_x(m, \tilde{\lambda}) \rangle \},
\]
a result valid to all orders in \( \eta \). It is important to note that, unlike the case of the dynamical phase (72), this correction involves the product of \( \tilde{\mu}^2 \) by \( \dot{\alpha} \). Therefore, it is the contribution odd in \( \eta \) that is eliminated by the parameter reversal, so that the dominant contribution corresponds to
Figure 5. Effect of taming the non-adiabatic oscillations generated by an abrupt application of the quadratic spin interaction, thanks to the use of a Blackman pulse-shaped time-derivative, oscillation taming (OT) procedure. The diagram shows non-adiabatic corrections to $\langle S_z \rangle$ at the end of the transition from $\lambda(0) = 0$ to $\lambda(T) = \lambda_0$, starting from an initial state which is an angular momentum eigenstate $|S = 2, m\rangle$ (left, $\lambda_0 = 1, m = -1$; right, $\lambda_0 = \lambda^*(0) = 0.838, m = 0$). The big black dot denotes the adiabatic prediction for $\langle S_z \rangle$. The red curves are for a linear increase of $\lambda(t)$, while the black ones correspond to the OT choice, i.e. $\dot{\lambda}(t) = f(\frac{t}{T})/f(0)$, where $f(s)$ is a Blackman pulse. The rather strong oscillating non-adiabatic corrections to $\langle S_z \rangle$ are clearly swept away by this latter choice, provided $\gamma_S B T \geq 25$.

The limit $\eta \to 0$ i.e. $\tilde{\lambda} \to \lambda$, $\tilde{\mu} \to \mu$. The quantity $C_{x,y}(m, \lambda) = \Im\{\langle \hat{\psi}_{x,y}^{(1)}(m, \lambda) | \hat{\psi}_{x,y}^{(1)}(m, \lambda) \rangle\}$ is plotted together with $p^{(2)}(m, \lambda)$ in figure 5, leading to the same conclusions as those reached at the end of the previous paragraph.

5.3. Non-adiabatic corrections associated with ramping-up the quadratic interaction

In previous subsections, Berry’s phases were induced by cyclic variations of the periodic Euler angles performed in an initial state $\hat{\psi}(Sm; \lambda_0)$. In fact, to prepare this eigenstate, one has to start from an eigenstate of $S$ and $S_z$ at a certain time $t = 0$. During the time interval $0 \leq t \leq T$, the evolution of the spin system is governed by $\mathcal{H}(\lambda(t)) = S_z/\hbar + i\lambda(t)(S_x/\hbar)^2$. Writing $\lambda(t) = \lambda_0 g(t/T)$, we assume that $g(s)$ increases ‘slowly’ from 0 to 1. We now ask whether, for given values of the rising time $T$, there might be a better choice for $g(s)$ than just $g(s) = s$ when $\lambda_0 \sim 1$, if one wishes to keep the non-adiabatic corrections below a predetermined level. Up to the end of this subsection, we have chosen $1/(\gamma_S B)$ as the time unit: all the time-dependent physical quantities and their time derivatives are functions of $\tau = t \gamma_S B$.

5.3.1. The two-level spin system $S = 2, m = \pm 1$. To get some insight into this problem, let us first consider the simple case of the adiabatic evolution of the state $S = 2, m = \pm 1$
adiabatic corrections are much less sensitive to the time dependence of the phase shift, which is purely dynamical in this case. As expected, the non-adiabatic corrections are an order of magnitude smaller.

We have solved numerically the Schrödinger equation for the evolution of the Hamiltonian \( \hat{H}_2(t) \) which governs the adiabatic processes involving the mixing of more than two levels. As noted in section 2.1, the non-adiabatic correction to \( \langle \Phi_1(\tau) | \Phi_1(0) \rangle \) is formally identical, but with one significant difference, namely \( \tan \zeta = 1/2 \).

In section 6, we study the time evolution of a 4-spin system governed by the direct sum of the two Hamiltonians: \( \hat{H}_1(t) \oplus \hat{H}_2(t) \) relative to the same value of \( \lambda \). We will have to use the two different expressions of \( \lambda \), for \( S = 2, \zeta_2 = 6\lambda/(9\lambda^2+4) \), and for \( S = 1, \zeta_1 = 2\lambda/(\lambda^2+4) \).

If \( \lambda[S] \) is rising linearly, the \( \lambda[S] \) pulses are replaced by trapezoidal Ramsey pulses with sharp edges leading also to non-adiabatic oscillating corrections.

There is a standard way to wash them out which is used in nuclear magnetic resonance and atomic interferometry experiments. This consists of using the so-called Blackman pulse shape [27]. Adapted to the present context, it leads to the following choice of \( \dot{\lambda}(t) \):

\[
\dot{\lambda}_{\text{BM}}(t) = \lambda_0 f \left( \frac{t}{T} \right) \int f(0),
\]

where \( f(s) = 0.42 - 0.5 \cos (2\pi s) + 0.08 \cos (4\pi s) \).

We have solved numerically the Schrödinger equation for \( \lambda_0 = 1 \) for the two different choices, Blackman and trapezoidal pulses for \( \dot{\lambda}(t) \). The important quantity for our purpose is \( \langle S_1(T) \rangle \), since it governs the magnitude of Berry’s phase induced by the cyclic variation of the Euler-angles. It is important to note that \( \langle S_1 \rangle \) is affected to first order in \( \dot{\lambda}(t) \) in contrast with the dynamical phase which is modified only to second order.

We have plotted \( \langle S_1(T) \rangle \) in figure 5 (left-hand curves), as a function of \( \gamma_S B T \). The big (black) dot on the ordinate axis shows the adiabatic prediction. The use of the Blackman pulse for \( \dot{\lambda}(t) \) leads to spectacular convergence toward the adiabatic limit: for \( \gamma_S B T \geq 25 \) the non-adiabatic correction to \( \langle S_1(T) \rangle \) plunges down below the 0.02% level. We have also computed the phase shift, which is purely dynamical in this case. As expected, the non-adiabatic corrections are much less sensitive to the time dependence of \( \dot{\lambda}(t) \) and we find that in both cases \( |\phi_D(\text{exact}) - \phi_D(\text{adiab})| < 0.008 \) when \( \gamma_S B T \geq 25 \). Similar calculations for \( S = |m| = 1 \) and the initial value \( m = -1 \) for \( \lambda_0 = 1 \) indicate that corrections (not shown) are an order of magnitude smaller.

5.3.2. More than two levels. We now extend the foregoing ‘rotating’ frame method to non-adiabatic processes involving the mixing of more than two levels. As noted in section 2.1, the Hamiltonian \( \hat{H}(\dot{\lambda}(t)) = S_i/\hbar + \dot{\lambda}(t)(S_i/\hbar)^2 \) is described in the angular momentum basis \( |S m \rangle \) by a real symmetric matrix. This implies that its eigenstates \( \psi(S m; \dot{\lambda}) \) can be written as real...
vectors. An evident consequence is the following identity, obtained by taking the derivative of the normalization condition of the real eigenvectors:

$$\left\langle \hat{\psi}(Sm; \lambda) \frac{\partial}{\partial \lambda} \hat{\psi}(Sm; \lambda) \right\rangle = 0.$$  \hspace{1cm} (80)

It implies that the adiabatic phase is purely dynamical. Let us now introduce the transformation matrix:

$$V_S(\lambda) = \sum_m \left\langle \psi(Sm; \lambda) \right| Sm \rangle.$$  \hspace{1cm} (81)

One can directly verify that $V_S(\lambda)$ transforms $\mathcal{H}(Sm; \lambda)$ into a diagonal matrix within the basis $\left\langle Sm \right|$, by writing $V_S(\lambda) \left\langle Sm \right| = \sum_m \hat{\psi}(Sm; \lambda) \left\langle Sm \right| Sm \rangle = \hat{\psi}(Sm; \lambda)$. The time evolution in the ‘rotating’ frame is then governed by the following Hamiltonian:

$$\tilde{H}_S(t) = V_S^\dagger \mathcal{H}(Sm; \lambda) V_S - i \dot{\lambda} V_S^\dagger \frac{\partial}{\partial \lambda} V_S,$$

$$= \tilde{H}_S(\lambda) + \Delta \tilde{H}_S(t).$$  \hspace{1cm} (82)

The expression $\tilde{H}_S(\lambda) = \sum_m \mathcal{E}(Sm; \lambda) \left\langle Sm \right|$ follows from equation (81), while the identity (80) implies that $\Delta \tilde{H}_S(t)$ is non-diagonal:

$$\Delta \tilde{H}_S(t) = \sum_{m_1 \neq m_2} \left\langle Sm_2 \right| \left\langle Sm_1 \right| - i \dot{\lambda} \left\langle \psi(Sm_1; \lambda) \right| \left\langle \psi(Sm_2; \lambda) \right|.$$  \hspace{1cm} (83)

Using an identity obtained from the time derivative of the eigenvalue equation (8), we can rewrite the rhs of the above equation as

$$\Delta \tilde{H}_S(t) = -i \dot{\lambda}(t) \sum_{m_1 \neq m_2} \frac{\left\langle Sm_2 \right| \left\langle Sm_1 \right|}{\mathcal{E}(Sm_1; \lambda) - \mathcal{E}(Sm_2; \lambda)} \left( \hat{\psi}(Sm_1; \lambda) \right| \left( S_e / \hbar \right)^2 | \hat{\psi}(Sm_2; \lambda) \rangle.$$  

The above formula can be considered an adaptation of a more general one to be found in [28] and references therein. One must stress that energy denominators $\mathcal{E}(Sm_1; \lambda) - \mathcal{E}(Sm_2; \lambda)$ involve levels for which $m_1 - m_2$ is an even integer. For instance, in the cases $S = 2$, $m = 0, \pm 2$, the two energy differences involved are varying slowly within the interval $0 \leq \lambda \leq \lambda_0 = 0.838$. It follows that the time dependence of $\Delta \tilde{H}_S(t)$ is dominated by $-i \dot{\lambda}(t)$. Let us assume that $\lambda(t)$ is increasing linearly within the time interval $0 \leq t \leq T$ : $\lambda(t) = \lambda_0 T$. The effect of $\Delta \tilde{H}_S(t)$, as in the previous cases, is equivalent to an rf trapezoidal pulse with sharp edges. In the right-hand curve of figure 5, we have plotted the average polarization as a function of the rising time $T$ for $\dot{\lambda}(t)$, toward the value $\lambda_0 = \lambda^\ast(0) = 0.838$. There is a very rapid damping of the oscillating non-adiabatic corrections, passing below the level of 0.2% for $\gamma_S BT \geq 25$ when $\dot{\lambda}(t)$ has a Blackman pulse shape. This is in strong contrast with the case of a linearly rising $\dot{\lambda}(t)$ where, for $\gamma_S BT \geq 25$, the non-adiabatic oscillating corrections have an amplitude of about 10%, decreasing only very slowly for longer rising times. The conspicuous chaotic behavior in the 3-level case reflects the fact that the two frequencies involved have in general an irrational ratio.

5.4. Concluding remarks

Let us sum up the results of this section. We have shown that if $\dot{\lambda}(t)$ is a linear function within the finite time interval $0 < t < T$, vanishing elsewhere, oscillating non-adiabatic corrections are generated by the sharp jumps of $\dot{\lambda}(t)$ at $t = 0$ and $t = T$. We have found that the remedy is to choose for $\dot{\lambda}(t)$ a Blackman pulse shape. It is clear that one should use the same
prescription for $\dot{\alpha}(t), \dot{\psi}(t)$, for taming analogous unwanted oscillations. By solving exactly the Schrödinger equation governed by the Hamiltonian $\hat{H}/(t)$ of equation (56), as illustrated in section 6 and in a separate paper [5] on two definite examples, we have indeed found that this remedy works beautifully.

It should be stressed that the near-degeneracies of opposite parity level pairs $(\xi(m, \lambda), \xi(m - 1, \lambda))$, which occur when $m > 0$ and $\lambda > 0$, do not affect the validity of the adiabatic approximation for Berry’s cycles if $\alpha(t)$ and $\lambda(t)$ are the only time-dependent parameters. This simplification is a consequence of the symmetry properties of the Hamiltonian $\hat{H}(\mathbf{B}(t), \mathbf{E}(t))$ resulting from the $\mathbf{E}$ and $\mathbf{B}$ orthogonality.

Furthermore, choosing for $\lambda(t)$ the ‘magic’ expression $\lambda^*(\frac{m + 1}{2\gamma_S})$ given by equation (65) makes the difference of Berry phases relative to the mirror cycles $(\alpha(t), \lambda(t))$ and $(-\alpha(t), \lambda(t))$ free of non-adiabatic corrections: all the correcting terms $\propto (\dot{\alpha}(t)/\gamma_S)^{2q+4}$, vanish whatever $q > 0$.

6. Holonomic entanglement of $N$ non-correlated 1/2-spins

6.1. Introduction

The purpose of this section is to show that Berry’s cycles studied in this paper are able to entangle $N$ non-correlated 1/2-spin states, while such an effect would not be present with linear spin couplings. Since the analysis presented here is exploratory, we shall concentrate upon the $N = 4$ case which already involves the basic features of our entanglement procedure.

It is convenient to classify the set of the four states with respect to the eigenvalues of $S_z = \sum_{i} s_{iz}$. The case $M = 2$ is trivial. The case $M = 1$ is the most interesting one for our purpose and will be the main subject of this paper. The case $M = 0$ will be discussed briefly at the end of this section. The results for negative values of $M$ are readily obtained from the positive ones by using the reflection laws introduced in section 3. The vector space generated by the linear combinations of non-correlated spin states with $M = 1$ has dimension 4 and the natural choice for a basis is the set of the four orthogonal states:

$$\Phi^{(1)} = \mid -\frac{1}{2} \rangle \otimes \mid \frac{1}{2} \rangle \otimes \mid \frac{1}{2} \rangle \otimes \mid \frac{1}{2} \rangle,$$

$$\Phi^{(2)} = \mid \frac{1}{2} \rangle \otimes \mid -\frac{1}{2} \rangle \otimes \mid \frac{1}{2} \rangle \otimes \mid \frac{1}{2} \rangle,$$

$$\Phi^{(3)} = \mid \frac{1}{2} \rangle \otimes \mid \frac{1}{2} \rangle \otimes \mid \frac{1}{2} \rangle \otimes \mid -\frac{1}{2} \rangle,$$

$$\Phi^{(4)} = \mid \frac{1}{2} \rangle \otimes \mid \frac{1}{2} \rangle \otimes \mid -\frac{1}{2} \rangle \otimes \mid -\frac{1}{2} \rangle.$$  

(84)

We shall assume that the time evolution of the 4-spin state $\Psi_4(t)$ is governed by the time-dependent Hamiltonian $H_N(t) = H(\mathbf{B}(t), \mathbf{E}(t))$, which looks formally like the quadratic spin Hamiltonian of equation (1) discussed extensively in the present paper:

$$H(\mathbf{B}(t), \mathbf{E}(t)) = U(R(t))\hat{H}(\mathbf{B}(t), \mathbf{E}(t))U^{-1}(R(t)),$$

$$\hat{H}(\mathbf{B}(t), \mathbf{E}(t)) = \gamma_S B(t)(S_z/h + \lambda(t)(S_z/h^2)).$$

The crucial difference lies in the fact that $\mathbf{S}$ is meant to be the total spin operator $\mathbf{S} = \sum_{i=1}^{N} s_i$.

We shall take as the initial state for $\Psi_4(t)$, one of the non-correlated states appearing in equation (84), say $\Psi_4(0) = \Phi^{(1)}$. By expanding $H_N(t)$ in terms of single-spin operators one gets a sum of spin–spin interactions: $\propto s_i \cdot \vec{n} \cdot s_j \cdot \vec{n}$, where $\vec{n}$ is given by $\vec{n} = U(R(t))\hat{\mathbf{e}}$. This indicates that $H_N(t)$ can create spin correlations in the state $\Psi_4(t)$. As a final remark, we stress that, by construction, $H_N(t)$ is invariant under all permutations of the $N = 4$ spins. This invariance property will play an essential role in our entanglement procedure.
6.2. Expansion of a non-correlated 4-spin state into a sum of the $S^2$ eigenstates

As a first step, we shall construct four angular momentum eigenstates which are linear combinations of the four states $\Phi^{(i)}$. Using the rules of addition of quantum angular momenta, one finds that the possible eigenvalues of $S^2$, $\hbar^2 S(S + 1)$, correspond to $S = 2$ and $S = 1$. There is a unique way to construct the state $S = 2$. It is obtained by applying the operator $S_+ = S_1 - iS_2$ upon the state $S = 2, M = 2$, i.e. $\Psi_{2,2} = \frac{1}{\sqrt{2}} (|\frac{1}{2} \rangle \otimes |\frac{1}{2} \rangle \otimes |\frac{1}{2} \rangle \otimes |\frac{1}{2} \rangle)$. One immediately gets

$$\Psi_{21} = \frac{i}{4} (\Phi^{(1)} + \Phi^{(2)} + \Phi^{(3)} + \Phi^{(4)}). \quad (85)$$

The above state is clearly invariant under all permutations of the four-spin states.

One must now construct three orthogonal states with $S = 1$ which will have different symmetries under the permutations of the four spins. If one ignores the orthogonal condition, it is easy to get three $S = 1$ states which are linearly independent. The method consists of taking a linear combination of two states $\Phi^{(i)}$ in such a way as to factor out the two-spin singlet state: $(|\frac{1}{2} \rangle \otimes |\frac{1}{2} \rangle - |\frac{1}{2} \rangle \otimes |\frac{1}{2} \rangle)/\sqrt{2}$, which is invariant under the rotation group. Let us give a typical example of such a construction:

$$\Phi^{(1,2)} = (\Phi^{(1)} - \Phi^{(2)})/\sqrt{2},$$

$$= \frac{i}{\sqrt{2}} (|\frac{1}{2} \rangle \otimes |\frac{1}{2} \rangle - |\frac{1}{2} \rangle \otimes |\frac{1}{2} \rangle) \otimes |\frac{1}{2} \rangle \otimes |\frac{1}{2} \rangle) \quad (86)$$

is a good candidate for an $S = M = 1$ state. Two similar $S = M = 1$ states $\Phi^{(1,3)}$, $\Phi^{(1,4)}$ are obtained from $\Phi^{(1,2)}$ by performing the cyclic permutation (234). These three states are linearly independent but a basis with orthogonal states would be more convenient. Since symmetry will be the basic tool in this subsection, let us start with the more symmetric $S = M = 1$ state: $\frac{1}{2} (\Phi^{(1)} - \Phi^{(2)} + \Phi^{(3)} - \Phi^{(4)})$. The two other states are obtained by applying the cyclic permutation (234). One then arrives at the following basis for the $S = M = 1$ subspace, easily seen to be orthogonal:

$$\Psi_{1,1} = \frac{1}{2} (\Phi^{(1)} - \Phi^{(2)} + \Phi^{(3)} - \Phi^{(4)}),$$

$$\Psi_{1,1}^2 = \frac{1}{2} (\Phi^{(1)} - \Phi^{(3)} + \Phi^{(4)} - \Phi^{(2)}), \quad (87)$$

$$\Psi_{1,1}^3 = \frac{1}{2} (\Phi^{(1)} - \Phi^{(4)} + \Phi^{(2)} - \Phi^{(3)}).$$

The above three states have different symmetry properties under the three permutations (23), (24), (34), which are listed below:

$$(23), (24), (34)|\Psi_{1,1}^1 = (1, -1, 1)\Psi_{1,1}^1,$$

$$(23), (24), (34)|\Psi_{1,1}^2 = (1, -1, 1)\Psi_{1,1}^2,$$

$$(23), (24), (34)|\Psi_{1,1}^3 = (1, -1, 1)\Psi_{1,1}^3.$$

The four eigenstates $\Psi_{1,1}^1$ and $\Psi_{2,1}$ constitute a complete orthonormal basis for the four spin-1/2 states with $M = 1$. The expansion of the four non-correlated states $\Phi^{(i)}$ are readily obtained from equations (84) and (85):

$$\Phi^{(i)} = \frac{1}{2} \left( \sum_{j=1}^{n=3} a_{ij} \Psi_{1,1}^i + \Psi_{2,1} \right), \quad (88)$$

where

$$a_{ij} = \{1, 1, 1\}, \quad a_{2j} = \{-1, -1, 1\},$$

$$a_{3j} = \{1, -1, -1\}, \quad a_{4j} = \{-1, 1, -1\}. \quad (89)$$
6.3. The adiabatic evolution of four non-correlated, 1/2-spin states governed by $H(B(t), E(t))$

The Hamiltonian $H_4(t) = H(B(t), E(t))$, as noted before, is invariant under either one of the three ‘parities’ $\epsilon_{(23)}, \epsilon_{(24)}, \epsilon_{(34)}$, associated with the permutations $(23), (24)$ and $(34)$. As a consequence, the matrix elements $\langle \Psi_{1M}' | H_4(t) | \Psi_{1M}' \rangle$ for $M = M'$ vanish if $i \neq j$. Moreover, the same result holds if $M = \pm 1, M' = \pm 1$ and $M \neq M'$. If $U_4(t)$ denotes the unitary operator associated with the quantum evolution governed by $H_4(t)$, the four states $U_4(t)\Psi_{2M}$ and $U_4(t)\Psi_{1M}'$ will have the same permutation symmetries as their parent states. In conclusion, the four states $\Psi_{2M}, \Psi_{1M}'$ behave vis-à-vis the Hamiltonian $H_4(t)$, as if they were associated with isolated spins $S$ and we can apply to them the results derived in the previous sections.

Our Berry’s cycle is organized in three steps. At $t = 0$, $H_4(0) = g_S B(t) S_z$. The first step $0 \leq t \leq T$ involves the adiabatic ramping of $\lambda(t)$ from 0 to $\lambda_0 \simeq -1$. In the second step $T \leq t \leq 3T$, one proceeds to the rotation of $E$ around $B$ by an angle $3\pi$, while keeping $\lambda(t) = \lambda_0$. For the third step $3T \leq t \leq 4T$, $\lambda(t)$ makes an adiabatic return to its initial value $\lambda = 0$. During the whole cycle the time dependences of $\lambda(t)$ and $\dot{\lambda}(t)$ are described by Blackman functions. The effect of this choice is to tame the non-adiabatic oscillating corrections which would be generated, if these parameters had discontinuous time derivatives.

We have performed an exact theoretical analysis of the cycle by solving the corresponding Schrödinger equation in the rotating frame with $g_S B T = 25$. We have found that the adiabatic approximation is working to better than 0.1%. It turns out that somewhat accidentally the difference of the dynamical phases for $S = 2, M = 1$ and $S = 1, M = 1$ is close to 0 modulo $2\pi$. Slightly tuning the $\lambda$-rising (-lowering) times can improve the cancellation to the 0.1% level. The cyclic evolution of the four spins, initially non-correlated, appears then as completely governed by Berry’s phases $\beta(2, 1; \lambda_0)$ and $\beta(1, 1; \lambda_0)$.

6.4. The final construction of the 4-spin holonomic entangled state

At the end of Berry’s cycle, $\Phi^{(i)}_f$ is transformed into the following state:

$$\Phi^{(i)}_{BP}(\lambda_0) = \frac{1}{2} \left\{ \exp(i\beta(21; \lambda_0)) \Psi_{21} + \exp(i\beta(11; \lambda_0)) \sum_{j=1}^{3} a_{ij} \Psi_{1j} \right\}. \quad (90)$$

Using equation (88) to express the sum $\sum_{j=1}^{3} a_{ij} \Psi_{1j}$ in terms of $\Phi^{(i)}_f$ and $\Psi_{21}$, one obtains the final expression

$$\Phi^{(i)}_{BP}(\lambda_0) = \exp(i\beta(11; \lambda_0)) \left( \Phi^{(i)}_f - \frac{1}{2} \Psi_{21} \right) + \frac{1}{2} \exp(i\beta(21; \lambda_0)) \Psi_{21}. \quad (91)$$

(We can verify in this expression that if $\Delta \beta(\lambda_0) = \beta(21; \lambda_0) - \beta(11; \lambda_0) = 0$, then $\Phi^{(i)}_{BP}$ coincides with $\Phi^{(i)}_f$ up to a phase, as one expects.)

From the energy curves given in figure 1 one sees that the region $\lambda > 0$ should be, in principle, avoided since the nearly crossing levels $S = 2, M = 2$ and $S = 2, M = 1$ could spoil the validity of the adiabatic approximation, while the region $\lambda < 0$ is much more favorable. However, in the present context where $\lambda$ and $\alpha$ are the only time-dependent parameters, this is only a protection against stray magnetic fields orthogonal to the main field,
since $H(t)$ has no $\Delta M = \pm 1$ matrix elements. The two Berry’s phases are given explicitly by rather simple expressions:

\[
\beta(21; \lambda_0) = 3\pi \left(\frac{2}{\sqrt{\lambda_0^2 + 4}} - 1\right),
\]

\[
\beta(11; \lambda_0) = 3\pi \left(\frac{2}{\sqrt{\lambda_0^2 + 4}} - 1\right),
\]

(92)

In order to obtain the maximum entanglement of $\Phi_{BP}^{(i)}$ after a rotation of $3\pi$, it is interesting to choose $\lambda_0 = \lambda_{\max} = -0.97$ which leads to $\Delta \beta(\lambda_{\max}) = -\pi$. Replacing $\Psi_{21}$ by its expression in terms of $\Phi^{(i)}$ (equation (85)) we arrive at the final expression of the four quantum entangled states, generated by Berry’s cycles with $\Delta \beta(\lambda_0) = -\pi$, from any of the four non-correlated states with $M = 1$ listed in equation (84):

\[
\Phi_{BP}^{(i)}(\lambda_{\max}) = \exp(i\chi) \left(\Phi^{(i)} - \frac{1}{2} \sum_{j=1}^{4} \Phi^{(j)}\right),
\]

(93)

where $\chi = \beta(11; \lambda_{\max})$. In order to make more apparent the holonomic entanglement resulting from Berry’s cycle, let us rewrite $\Phi_{BP}^{(i)}(\lambda_{\max})$ using the explicit forms of the non-correlated states $\Phi^{(i)}$ given in equation (84):

\[
\Phi_{BP}^{(i)}(\lambda_{\max}) = \frac{\exp(i\chi)}{2} \left(\sum_{j=1}^{4} \Phi(j) - \frac{1}{2} \sum_{j=1}^{4} \Phi(j)\right)
\]

\[
= \sum_{j=1}^{4} \Phi(j) - \frac{1}{2} \sum_{j=1}^{4} \Phi(j)
\]

\[
= \left|\Phi_2\right| \otimes \left|\Phi_1\right| - \frac{1}{2} \left[\left|\Phi_2\right| \otimes \left|\Phi_1\right| + \left|\Phi_1\right| \otimes \left|\Phi_2\right|\right].
\]

(94)

A similar approach can be used to treat the case of four non-correlated spins with $M = \sum_{i=1}^{4} m_i = 0$. Among the three angular momentum states $S = 2, 1, 0; M = 0$, only the state $S = 2, M = 0$ has a non-vanishing Berry’s phase. As a consequence, one gets an entanglement which is weaker than in the above $M = 1$ case where it is maximum. The case of eight 1/2-spins (or eight Qbits) with $M = 1$ is of particular interest since it involves four non-vanishing Berry’s phases for $S = 1, 2, 3, 4$ but it will require more powerful group theory tools, like the Young operators [30]. However, the basic ideas of the entanglement procedure would remain the same.

The reader might have the impression that the above method of entanglement is constrained by the fact that Berry’s phases depend upon few parameters. However one should keep in mind that $\beta(SM; \lambda)$ is actually given by a Aharonov–Bohm integral involving an Abelian gauge field along a closed loop drawn upon the four-dimensional manifold $CP^2$, which implies a considerable freedom (see equation (49)).

From a practical point of view entanglement methods based on geometric phases are known to present advantages of robustness: they are resilient to certain types of errors in the control of the parameters driving the quantum evolution [11].

We end this section by giving an example where the collective spin–spin interaction between a few spins, similar to that described by $H_N(t)$, has been successively implemented. This concerns the case of two or four alkali-like ions, without nuclear spin, in an ion-trap. The idea is to realize illumination of the $N$ ions (N Qbits) with two lasers fields having two different frequencies so that the two-photon process, exciting any pair of ions in the trap, is resonant, but neither of the frequencies are resonant with the single excitation of an ion [31, 32]. Realization performed for two and four ions in a trap [33] can be generalized to more ions. In the present context it would be necessary to match the magnitude of the dc
magnetic field and the radiation fields. According to the analysis given above, rotating the linear polarization of the radiation fields around the $B$-field should make it possible to generate holonomic entanglement of the Qbits.

7. Summary and perspectives

7.1. Synopsis of the paper

The purpose of this paper is the theoretical study of Berry’s phases, generated in cyclic evolutions of isolated spins of arbitrary values. The spins are assumed to be nonlinearly coupled to time-dependent external electromagnetic fields (possibly effective) via the superposition of a dipole and a quadrupole couplings. Configurations leading to degenerate instantaneous eigenvalues are avoided. In other words non-Abelian Berry’s phases are not considered. We also assume that the two effective fields are orthogonal, a mild restriction but with many advantages. It implies several discrete symmetries of the spin Hamiltonian which considerably simplify the algebra. For instance, two angular momentum states having different quantum numbers $m_1$ and $m_2$ are coupled if, and only if, $m_1 - m_2$ is an even integer. Furthermore, for $S = 1$, the geometric space of the Hamiltonian parameters is isomorphic to the density matrix space $\mathbb{C}P^2$ which makes Berry’s and Aharonov–Anandan geometric phases mathematically identical.

Using rotation group theory, and the aforementioned discrete symmetries, we obtain compact expressions for the instantaneous eigenfunctions of the Hamiltonian, labeled by the magnetic quantum number $m$, valid in the limit of small quadrupole coupling. We derive an explicit compact expression for Berry’s phase $\beta(m)$ in terms of the usual Euler angles $\phi, \theta, \alpha$ associated with the trihedron defined by the $B, E$ directions, and an extra dimensionless parameter $\lambda$ giving the quadrupole to dipole coupling ratio. The result is given as a loop integral of an Abelian field along a closed circuit drawn upon the parameter space $\mathbb{C}P^2$:

$$ \beta(m) = \oint_{\mathbb{C}} p(m, \lambda)(\cos \theta d\phi + d\alpha) - m(d\phi + d\alpha), $$

where $\bar{p}(m, \lambda)$, the average value of the spin angular momentum taken along the $B$ field, is just the gradient of the eigenenergy with respect to $B$. The apparent simplicity of the above formula conceals the geometry contained in $p(m, \lambda)$. This important feature is clearly exhibited in Berry’s cycles where $\lambda$ and $\alpha$ are the sole varying parameters. The case $S = 2, m = 0$, where Berry’s phase is vanishing for a linear spin Hamiltonian, is particularly spectacular: $p(0, \lambda)$ is an odd function of $\lambda$ and takes the value 1 when $\lambda = 1$. This peculiar geometry is best illustrated in figure 2 where $A_\phi = p(0, \lambda)$ is plotted against the spherical coordinate $\tilde{\theta}$ defined by $\lambda = -2 \cot \tilde{\theta}$ with $0 < \tilde{\theta} < \pi$.

The non-adiabatic corrections associated with the Euler angles derivatives have been analyzed within the rotating frame attached to the time-varying fields. The Coriolis effect generates an extra magnetic field $\Delta B$ which involves a linear combination of the Euler-angle time derivatives. The longitudinal component along the $B$ field is the only one which survives when $\alpha$ is the sole time-dependent Euler angle. The corresponding Hamiltonian $\tilde{H}_{ij}$ is devoid of any geometry. As a consequence, the phase shift acquired at the end of the cycle $\phi(B + \Delta B)$ is purely dynamical. Berry’s phase is incorporated into the dynamical phase under the form of its first-order contribution with respect to $\eta = -\Delta B_j / B = (\cos \theta \dot{\phi} + \dot{\alpha})/(\gamma S B)$. The higher order terms give all the non-adiabatic corrections associated with $\dot{\alpha}$ when it is the only varying periodic parameter. We have also shown that the subset of these corrections, odd under a reversal of $\eta$, cancels exactly for ‘magic’ values $\lambda = \lambda^*(\eta)$. This cancellation is implemented
in the experimental project described in [5]. The case of the non-adiabatic corrections induced by the transverse magnetic field $\Delta B_\perp / B$ is somewhat more involved since it introduces a non-trivial geometry and, as a consequence, Berry’s phase contribution to be added to the one coming from the transverse dynamical phase. An explicit evaluation of the complete lowest order total correction $\Delta \beta_\perp / \beta_\perp$ is found to be $\propto (\varphi / \gamma_\perp B)^2$ up to a numerical coefficient computed explicitly for $S = 2$, $m = 0$ and $-1$. The results are displayed in figure 4.

This discussion hinges upon the implicit assumption that the finite value of $\lambda$ is reached by an adiabatic ramping governed by $\mathcal{H}(\lambda(t)) = S_z / h + \lambda(t) (S_z / h)^2$, leading to a pure eigenstate $\hat{\psi}(\lambda)$, free of any non-adiabatic pollution. To analyze this problem, it was convenient to perform the time-dependent unitary transformation $V(\lambda(t))$ which transform $\mathcal{H}(\lambda(t))$ into a diagonal matrix within the angular momentum basis. It follows from the symmetry properties of $\mathcal{H}(\lambda(t))$ that $V(\lambda(t))$ can be taken as real and symmetric. The Hamiltonian, which governs the spin evolution in the transformed basis, contains the non-diagonal term $i \hat{\lambda}(t) V^T (\lambda(t)) \frac{d}{dt} V(\lambda(t))$. Since its time dependence is dominated by the time derivative $\dot{\lambda}(t)$, a linear increase of $\lambda(t)$ would be equivalent to an rf pulse with sharp edges leading to the oscillating non-adiabatic corrections exhibited in figure 5. The standard procedure to smooth them out is to use for $\lambda(t)$ a Blackman pulse shape [27]. There is a second assumption implicit in the rotating frame analysis, namely that the adiabatic approximation be valid for the rotating frame Hamiltonian $\tilde{H}(t)$. By solving numerically the Schrödinger equation describing the whole cycle, we have verified (see [5] for explicit numerical results) that, indeed, the adiabatic approximation works beautifully provided one also uses a Blackman-pulse shape for the angular speed, $\alpha(t)$.

In the last section, we propose a procedure to entangle a system of $N$ non-correlated 1/2-spins (or $N$ Qbits). It involves Berry’s cycles generated by a Hamiltonian formally identical to one given in equation (1), but with an important change: $S$ stands now for the total spin operator $S = \sum_{i=1}^{\mathcal{N}} s_i$. We have used a method which is well adapted to the simple configuration of four 1/2-spins. In view of possible extensions to $N$ spins with $N > 4$, we would like to rephrase our approach within a more general framework. A configuration of $N$ non-correlated 1/2-spins can be described by a factorizable spin tensor of order $N$. It constitutes a reducible representation of the $S(U_2)$ group. There is a general procedure to decompose this tensor into irreducible tensors, associated with eigenstates of $S^2$ and $S_z$. With the exception of the trivial case $N = 2$, the angular momentum state $\Psi_S M$ can appear several times in the decomposition. According to a known theorem of group theory [30], all the states $\Psi^j_{S M}$ have different symmetry properties under the permutation of the $N$ spins and they can be organized into an orthogonal basis, $\{ \Psi_{S M}^j \} = \delta_{S,S} \delta_{M,M} \delta_{i,j}$. Let us now consider the set of Hamiltonians $\mathcal{H}(S)$ which are given by a nonlinear series expansion with respect to the spin components $s_x, s_y, s_z$. By construction, $\mathcal{H}(S)$ is invariant upon any permutation of the $N$ spins. As a consequence, $\mathcal{H}(S)$ is diagonal within the basis $\Psi_{S M}^j$: $\{ \Psi_{S M}^j \} | \mathcal{H}(S) | \Psi_{S M}^j \} = \delta_{S,S} \delta_{M,M} \delta_{i,j} | \mathcal{H}(S) | \Psi_{S M}^j \}$. The above equation expresses the simple fact that $\mathcal{H}(S)$ is acting upon the states $\Psi_{S M}^j$ as if they were isolated spins $S$. Any initial state of $N$ non-correlated spins can then be written as $| \Psi_N(t = 0) \rangle = \sum_m \sum_i \alpha_{S M}^i | \Psi_{S M}^i \rangle$. One now performs an adiabatic Berry’s cycle along a closed loop in the parameter space such that $\mathcal{H}(S, 0) = \mathcal{H}(S, T) = \gamma S \cdot B$. At the end of the cycle the $N$ spin system is given by the following state vector: $| \Psi_N(T) \rangle = \sum_m \sum_i \alpha_{S M}^i \exp i \beta(S M) | \Psi_{S M}^i \rangle$.

With a suitable choice of Berry’s cycle, we have shown in the particular case $N = 4$, $M = 1$ that the final state is endowed with a maximal entanglement. Thus, extension to higher values of $N$ is worth pursuing, remembering that according to quantum computing experts: ‘entanglement, as with most good things, it is best consumed in moderation’ [34].
7.2. Experimental perspectives

In a separate work, we have explored the possibility of cold alkali atoms in their ground state to measure Berry’s phase with atomic interferometers. The spin operator is then identified with the total angular momentum $F = s + I$. The ac-Stark shift induced by a linearly polarized light beam tuned off-resonance of one resonance line can induce the quadratic interaction if one accepts a few experimental compromises. The candidate for our spin system is the ground state hyperfine (hf) level $5S_{1/2}$, $F = 2$, of $^{87}$Rb. We have found it judicious to tune the laser frequency midway between the two lines $5S_{1/2}$, $F = 2 \rightarrow 5P_{1/2}$, $F = 1$ and $F = 2$. The effective quadrupole coupling takes then the simple form $H_Q = \frac{\hbar}{\Omega^2}(F \cdot \hat{e})^2$, where $\Lambda W_p$ is the $5P_{1/2}$ hf splitting, $\Omega$ the Rabi frequency relative to the transition $5S_{1/2} \rightarrow 5P_{1/2}$ and $\hat{e}$ the light polarization. The price to be paid arises from the instability of the ‘dressed’ ground state described by its decay rate $\Gamma_{\text{dec}} = \frac{4\Omega^2}{\Lambda^2 W_p} \Gamma_{5P_{1/2}}$ (where $\Gamma_{5P_{1/2}}$ is the spontaneous decay rate of the excited state). Although, with our choice of detunings, this effect does not affect Berry’s phase value, it can seriously perturb the measurement process. One is clearly facing a difficult optimization problem, if one wants also to keep the non-adiabatic correction below the 0.1% level. We give here some features of the solution described in [5]. With an external magnetic field of 1 mG, one can adjust $\Omega^2$ (i.e. the light beam intensity) in order to get a quadrupole to dipole coupling ratio close to 1. The off-diagonal density matrix element holding Berry’s phase decays to half its initial value, but adjusting the interferometer parameters can compensate for this effect and keep the interference contrast close to its maximum.

On the above example the role of both the hyperfine interaction and instability of the atomic excited states is clearly exhibited. This gives a clear illustration of the atomic internal structure contribution to the spin dynamics. Although these effects upon Berry’s cycles can be accounted for by choosing appropriate values of the effective $B, E$ fields, they lead to severe experimental constraints. It looks possible to satisfy these constraints with $^{87}$Rb atoms, but with light alkalis this is far from obvious [5]. On the other hand, the external degrees of freedom, i.e. the atomic nuclei coordinates, have been supposed to be fixed physical quantities, an assumption which would obviously fail for ultracold atoms belonging to a fermionic or bosonic quantum gas. These coordinates become truly quantum variables which appear explicitly in the density matrix of the system. In such a case, the problem of Berry’s phase generated by an adiabatic cycle of the coupled external fields becomes well beyond the scope of this paper.

Acknowledgments

The authors would like to thank Dr M D Plimmer for helpful discussions.

Appendix. Some useful formulas and algebraic results for $S = 3$ and $S = 4$

$$H_{\text{odd}}(3, \lambda) = \begin{pmatrix} \frac{3\lambda}{2} + 3 & \frac{\sqrt{15}\lambda}{2} & 0 & 0 \\ \frac{\sqrt{15}\lambda}{2} & \frac{11\lambda}{2} + 1 & 3\lambda & 0 \\ 0 & 3\lambda & \frac{11\lambda}{2} - 1 & \frac{\sqrt{15}\lambda}{2} \\ 0 & 0 & \frac{\sqrt{15}\lambda}{2} & \frac{3\lambda}{2} - 3 \end{pmatrix}$$ (A.1)
\[
\mathcal{H}_{\text{even}}(3, \lambda) = \begin{pmatrix}
4\lambda + 2 & \sqrt{\frac{15}{2}}\lambda & 0 \\
\sqrt{\frac{15}{2}}\lambda & 6\lambda & \sqrt{\frac{15}{2}}\lambda \\
0 & \sqrt{\frac{15}{2}}\lambda & 4\lambda - 2
\end{pmatrix}
\] (A.2)

\[
\det(\mathcal{H}_{\text{odd}}(3, \lambda) - x I) = x^4 - 14x^3\lambda + 49x^2\lambda^2 - 10x^2 - 36x\lambda^3 + 102x\lambda - 216\lambda^2 + 9,
\] (A.3)

\[
\det(\mathcal{H}_{\text{even}}(3, \lambda) - x I) = x^3 - 14x^2\lambda + 49x\lambda^2 - 4x - 36\lambda^3 + 24\lambda.
\] (A.4)

\[
\mathcal{H}_{\text{even}}(4, \lambda) = \begin{pmatrix}
2\lambda + 4 & \sqrt{7}\lambda & 0 & 0 & 0 \\
\sqrt{7}\lambda & 8\lambda + 2 & 3\sqrt{\frac{5}{2}}\lambda & 0 & 0 \\
0 & 3\sqrt{\frac{5}{2}}\lambda & 10\lambda & 3\sqrt{\frac{5}{2}}\lambda & 0 \\
0 & 0 & 3\sqrt{\frac{5}{2}}\lambda & 8\lambda - 2 & \sqrt{7}\lambda \\
0 & 0 & 0 & \sqrt{7}\lambda & 2\lambda - 4
\end{pmatrix}
\] (A.5)

\[
\mathcal{H}_{\text{odd}}(4, \lambda) = \begin{pmatrix}
\frac{11\lambda}{2} + 3 & \frac{15\sqrt{7}\lambda}{2} & 0 & 0 \\
\frac{15\sqrt{7}\lambda}{2} & \frac{19\lambda}{2} + 1 & 5\lambda & 0 \\
0 & 5\lambda & \frac{19\lambda}{2} - 1 & \frac{15\sqrt{7}\lambda}{2} \\
0 & 0 & \frac{15\sqrt{7}\lambda}{2} & \frac{11\lambda}{2} - 3
\end{pmatrix}
\] (A.6)

\[
\det(\mathcal{H}_{\text{even}}(4, \lambda) - x I) = x^5 - 30x^4\lambda + 273x^3\lambda^2 - 20x^3 - 820x^2\lambda^3 + 472x^2\lambda^2 + 182x\lambda + 576\lambda^4 - 712\lambda^2 + 9.
\] (A.7)

\[
\det(\mathcal{H}_{\text{odd}}(4, \lambda) - x I) = x^4 - 30x^3\lambda + 273x^2\lambda^2 - 10x^2 - 820x\lambda^3 + 182x\lambda^4 - 712\lambda^2 + 9.
\] (A.8)

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