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Classification of cyclic initial states and geometric phase for the spin-j system

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Abstract. Quantum states which evolve cyclically in their projective Hilbert space give rise to a geometric (or Aharonov–Anandan) phase. An aspect of primary interest is stable cyclic behaviour as realized under a periodic Hamiltonian. The problem has been handled by use of time-dependent transformations treated along the lines of Floquet's theory as well as in terms of exponential operators with a goal to examine the variety of initial states exhibiting cyclic behaviour. A particular case of special cyclic initial states is described which is shown to be important for nuclear magnetic resonance experiments aimed at the study of the effects of the geometric phase. An example of arbitrary spin j in a precessing magnetic field and spin $j = 1$ subject to both axially symmetric quadrupolar interaction and a precessing magnetic field are presented. The invariant (Kobe's) geometric phase is calculated for special cyclic states.

1. Introduction

The discovery of the adiabatic topological phase by Berry [1] initiated intense interest, which has grown in recent years. Aharonov and Anandan [2] generalized Berry's result to a geometric phase associated with closed evolution paths in projective Hilbert space. Essentially, a connection was drawn between a quantum mechanical observable and the topological structure embedded in a Hilbert space. Recently, Anandan [3] presented a geometrical formulation of quantum mechanics principles. This can be regarded as an alternative picture of quantum mechanics providing a different outlook for a variety of quantum effects. Due to the general nature of the geometric approach, it allows for analogy to theories such as quantum field theory [4]. At the same time it provides for easily visualizable models, such as the generalized Bloch sphere [5].

The most popular approach in the study of the geometric phase addresses the evolution under a Lie algebraic Hamiltonian [6–8]. In this case the compact version of the projective Hilbert space can be generated, spanned by raising–lowering operators of the given algebra. This approach provides us with direct geometric insight. However, from the viewpoint of specific quantum mechanical calculations the applicability of this method is restricted to low-rank algebras since the Schrödinger equation is converted to a system of nonlinear differential equations. These become intractable for algebras of higher rank.

Another approach has been formulated by Moore [9] for evolution under a periodic Hamiltonian. The coordinate system implanted into the projective Hilbert space is represented by a unitary matrix. Cyclic trajectories are given by periodic matrices $S^I(t)$, the existence of which follow from Floquet theory. The computational aspects are somewhat

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less critical for this approach since the problem can be reduced to an eigenproblem for a Floquet Hamiltonian in a procedure that can be regarded as a version of Fourier transformation for the Schrödinger equation [10]. Nevertheless, the method faces difficulties in practical applications, and analytic results can be obtained only for a limited number of Hamiltonians.

In the present analysis we relax the periodicity condition imposed on the Floquet matrix $S'(t)$. As shown below, this is of convenience in the treatment of certain systems and therefore expands the boundaries of Floquet formalism in geometric phase studies. In the particular case when the Hamiltonian is periodic and belongs to the $SU(2)$ algebra, it allows a direct connection to be drawn between the Lie algebraic and Floquet approaches. At the same time, the price paid for the ease of calculation is the loss of the gauge potential formulation for the geometric phase.

The paper is organized as follows. In section 2 we present the classification of cyclic initial states (CIS) for the general case of finite-dimensional systems subject to periodic Hamiltonians. In section 3 the example of an arbitrary spin j in a precessing magnetic field is treated. In section 4 we examine the case of $j = 1$ subject to a precessing magnetic field and axially symmetric quadrupolar interaction. Generalization to arbitrary spin j is also presented.

2. Cyclic behaviour of quantum states

Consider the evolution of a quantum system as given by

$$|\psi(t')\rangle = U(t')|\psi(0)\rangle. \tag{1}$$

Taking the eigenvectors of $U(t')$ as the initial states $|\psi(0)\rangle$, we can verify that the evolution during the period of time t' is cyclic, giving rise to a multiplicative phase factor. Thus, $|\psi(0)\rangle$ are cyclic initial states. This cyclic behaviour, however, is not stable since it cannot be expected that the $|\psi(0)\rangle$'s remain eigenvectors of $U(t)$ at times $2t', 3t', \ldots, nt'$. Such unstable cyclic behaviour is not of interest in the study of the geometric phase; in contrast, stable cyclic behaviour is the tacit primary target for geometric phase studies to date.

Consider a quantum system described by the wavefunction $\Psi(t)$ which evolves under the periodic Hamiltonian $H(t + \tau) = H(t)$. A time-dependent transformation $S^{-1}(t)$,

$$\tilde{\Psi}(t) = S^{-1}(t)\Psi(t) \tag{2}$$

transforms the wavefunction to $\tilde{\Psi}(t)$, which obeys the Schrödinger equation with an accordingly transformed Hamiltonian $\tilde{H}$:

$$\mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}\tilde{\Psi}(t) = \tilde{H}\tilde{\Psi}(t) \tag{3}$$

where $\tilde{H}$ satisfies the equation

$$S(t)\tilde{H} = H(t)S(t) - \mathrm{i}\frac{\mathrm{d}}{\mathrm{d}t}S(t). \tag{4}$$

At this point $\tilde{H}$ is required to be time-independent. Under this condition, equation (4) is identified as a system of linear homogeneous first-order differential equations with periodic coefficients which can be solved with respect to the elements of $S(t)$. According to

Floquet theory, the fundamental matrix $\hat{S}(t)$ composed of linearly independent solutions of equation (4) can be represented as [11]

$$\hat{S}(t) = \hat{G}(t)e^{i\hat{Q}t} \tag{5}$$

where $\hat{G}(t)$ is periodic, $\hat{G}(t+\tau) = \hat{G}(t)$, and $\hat{Q}$ is constant and diagonal. Therefore, it follows that the set of solutions given by equation (5) is characterized by the periodicity property

$$S(t+\tau) = S(t)e^{iq\tau} \tag{6}$$

where q is the eigenvalue of $\hat{Q}$.

The significance of Floquet's results is that we can always find a solution $S(t)$ leading to a time-independent Hamiltonian, $\tilde{H}$, in a form which satisfies the criterion (6). Using equations (2) and (3) the form of evolution operator $U(t)$ can be specified:

$$\Psi(t) = U(t)\Psi(0) \tag{7}$$

$$U(t) = S(t)\tilde{U}(t)S^{-1}(0) = S(t)e^{-i\tilde{H}t}S^{-1}(0). \tag{8}$$

It should be noted that the transformation $S(t)$ can be chosen as non-unitary, giving rise to a non-Hermitian operator $\tilde{H}$ (cf equation (4)), with the proviso that the evolution operator (8) must be unitary. We confine ourselves, however, to the case of unitary $S(t)$, so that the resulting $\tilde{H}$ is Hermitian and can be interpreted as an effective Hamiltonian.

The above results can be compared with those due to Moore [9, 10]. In his work, Moore made use of Floquet theory in order to construct the following representation for the evolution operator $U(t)$:

$$U(t) = S'(t)e^{-i\tilde{H}'t} \tag{9}$$

where $\tilde{H}'$ is constant, $S'(t)$ is unitary and periodic, $S'(t+\tau) = S'(t)$, and $S'(0) = I$. Hence, $S'(t)$ is a matrix of time-dependent transformation which is subject to more restrictive requirements than those given by equation (6).

Now we embark on the construction of cyclic initial states (CIS) starting from equation (8). Following Moore [12], the first type of CIS is based on the set of eigenvectors of $\tilde{H}$:

$$\tilde{H}|\tilde{x}^{(n)}\rangle = \tilde{\lambda}_n|\tilde{x}^{(n)}\rangle \tag{10}$$

$$\Psi_n^c(0) = S(0)|\tilde{x}^{(n)}\rangle. \tag{11}$$

Using equations (8) and (6) we can verify that $\Psi_n^c(0)$ are indeed CIS obeying

$$\Psi_n^c(\tau) = \exp(i\varphi_n(\tau))\Psi_n^c(0) \tag{12}$$

where

$$\varphi_n(\tau) = -\tilde{\lambda}_n\tau - i\ln(\langle\tilde{x}^{(n)}|S(\tau)S^{-1}(0)|\tilde{x}^{(n)}\rangle). \tag{13}$$

Since $S(t)$ is taken to be unitary, which holds for any time t, it follows from equation (6) that the second contribution on the RHS of equation (13) is real. Similarly, the eigenvalues of the Hermitian $\tilde{H}$ are real, and, hence, the overall phase $\varphi_n(\tau)$ given by equation (13) is also real. The case when $\tilde{H}$ contains degeneracies was treated by Moore [12].

The result for the geometric phase stems from the expression given by Aharonov and Anandan [2]:

$$\beta_n^{\mathrm{G}}(\tau) = \varphi_n(\tau) + \mathrm{i}\int_0^\tau \langle \Psi_n^{\mathrm{c}}(t)|\frac{\mathrm{d}}{\mathrm{d}t}|\Psi_n^{\mathrm{c}}(t)\rangle\,\mathrm{d}t. \tag{14}$$

Using equations (8) and (13), one obtains

$$\beta_n^{\mathrm{G}}(\tau) = -\mathrm{i}\ln(\langle\tilde{x}^{(n)}|S(\tau)S^{-1}(0)|\tilde{x}^{(n)}\rangle) + \mathrm{i}\int_0^\tau \langle\tilde{x}^{(n)}|S^{-1}(t)\frac{\mathrm{d}}{\mathrm{d}t}S(t)|\tilde{x}^{(n)}\rangle\,\mathrm{d}t. \tag{15}$$

This class of CIS (equation (12)) always exists providing that the system in question is finite-dimensional. Thus, we can refer to the CIS $\Psi_n^{\mathrm{c}}(0)$ as natural cyclic states (nCIS).

Now we investigate an alternative choice for CIS, which, although subject to more restrictive requirements, nevertheless are important for practical analyses of spin-j systems. First, consider the diagonalizing unitary transformation R:

$$\tilde{U}(t) = \mathrm{e}^{-\mathrm{i}\tilde{H}t} = R^{-1}\mathrm{e}^{-\mathrm{i}\tilde{\lambda}t}R \tag{16}$$

and demand for the certain time, T, that

$$\tilde{\lambda}_m T = \tilde{\lambda}_k T + 2\pi N_{mk} \tag{17}$$

for all allowed values of m and k. Here N_{mk} specifies a matrix of arbitrary integers. Under these conditions it can be seen that $\tilde{U}(t)$ at $t = T$ is reduced to a phase-shift operator:

$$\tilde{U}(T) = \exp(-\mathrm{i}\tilde{\lambda}_m T)I \tag{18}$$

where I is the identity. Notice, that the index m of $\tilde{\lambda}_m$ is taken as a convention and can be replaced for any allowed value $m = k$.

The criterion (17) is equivalent to $\aleph - 1$ independent conditions, where $\aleph$ is the dimensionality of Hilbert space. We can regard these conditions as imposed on a set of parameters $\{T\} \otimes \{\tilde{h}_i\}$, where $\{\tilde{h}_i\}$ is a set of parameters which characterizes the Hamiltonian $\tilde{H}$.

If conditions (17) are met, the evolution operator taken at $t = T$ can be reduced to

$$U(T) = \exp(-\mathrm{i}\tilde{\lambda}_m T)S(T)S^{-1}(0). \tag{19}$$

Secondly, it is required that

$$S(t'')S^{-1}(t') = f(t'' - t') \tag{20}$$

implying the property of invariance with respect to time translation. With equation (20) it is straightforward to show that $U(T)$ commutes with $U(2T), \ldots, U(nT)$ and, therefore, gives rise to stable cyclic behaviour.

The eigenvectors of $S(T)S^{-1}(0)$ can then be chosen for the role of CIS, viz

$$S(T)S^{-1}(0)|z^{(m)}\rangle = \mu_m|z^{(m)}\rangle \tag{21}$$

$$\Phi_m^{\mathrm{c}}(0) = |z^{(m)}\rangle \tag{22}$$

$$\Phi_m^{\mathrm{c}}(T) = \exp(\mathrm{i}\varphi_m(T))\Phi_m^{\mathrm{c}}(0) \tag{23}$$

$$\varphi_m(T) = -\tilde{\lambda}_m T - \mathrm{i}\ln(\langle z^{(m)}|S(T)S^{-1}(0)|z^{(m)}\rangle). \tag{24}$$

Similarly, with $S(t)$ being a unitary operator, we find the overall phase $\varphi_m(T)$ to be real. The geometric phase is derived by use of equation (14) as

$$\beta_m^G(T) = -\tilde{\lambda}_m T - i\ln(\langle z^{(m)}|S(T)S^{-1}(0)|z^{(m)}\rangle) + \langle z^{(m)}|\tilde{H}|z^{(m)}\rangle T + i\int_0^T \langle z^{(m)}|e^{i\tilde{H}t}\left(S^{-1}(t)\frac{d}{dt}S(t)\right)e^{-i\tilde{H}t}|z^{(m)}\rangle\,dt. \tag{25}$$

Since the criteria for $\Phi_m^c(0)$ being CIS are stringent (equations (17) and (20)), we shall designate them as special cyclic initial states (sCIS). It was pointed out [12] that time-independent Hamiltonians can give rise to CIS of this type. Here we formulated the conditions for the existence of sCIS under a general periodic Hamiltonian. It is shown below that the sCIS are of primary importance for the problem of a spin j in a precessing magnetic field.

3. Spin j in a precessing magnetic field

The transformation $S(t)$ can be found by solving equation (4), which is essentially equivalent to solving the original Schrödinger equation for the system under consideration. However, for the problem of a spin in a precessing magnetic field, a rotating-frame transformation can be recognized as a realization of $S(t)$. This belongs to a class of 'cranking transformations' as described by Wang [13, 14].

The Hamiltonian for a spin exposed to a precessing magnetic field is

$$H(t) = \omega_0 I_z + \omega_1[I_x\cos(\omega t + \phi) + I_y\sin(\omega t + \phi)] \tag{26}$$

which by use of the rotating-frame transformation in the form

$$S(t) = \exp[-i(\omega t + \phi)I_z] \tag{27}$$

is transformed to

$$\tilde{H} = \Delta I_z + \omega_1 I_x \tag{28}$$

where $\Delta = \omega_0 - \omega$, and the period of $H(t)$ is $\tau = 2\pi/\omega$.

Using the properties of exponential operators [15] we can represent $\tilde{H}$ as

$$\tilde{H} = \exp(-i\theta I_y)\Omega I_z \exp(i\theta I_y) \tag{29}$$

where

$$\theta = \tan^{-1}\left(\frac{\omega_1}{\Delta}\right) \tag{30}$$

$$\Omega = \sqrt{\Delta^2 + \omega_1^2}. \tag{31}$$

This is known as a doubly rotated frame transformation. Consequently, the evolution operator is represented by

$$U(t) = \exp[-i(\omega t + \phi)I_z]\exp(-i\theta I_y)\exp(-i\Omega t I_z)\exp(i\theta I_y)\exp(i\phi I_z). \tag{32}$$

On the basis of equations (11) and (29) nCIS are identified as

$$\Psi_m^c(0) = \exp(-i\phi I_z)\exp(-i\theta I_y)|m\rangle \tag{33}$$

the overall phase is

$$\varphi_m(\tau) = -2\pi m\left(1 + \frac{\Omega}{\omega}\right) \tag{34}$$

and the geometric phase is found to be

$$\beta_m^G(\tau) = -2\pi m(1 - \cos\theta) = -2\pi m\left(1 - \frac{\Delta}{\Omega}\right). \tag{35}$$

The nCIS of spin j in a varying magnetic field have been described in numerous works [8, 13, 14, 16], and a similar situation has recently been detailed in [17]. Other authors [18] address the problem without specifying the form of the CIS. Our definition of nCIS differs from that used in these works by the factor $\exp(-i\phi I_z)$. Expression (35) recovers the results of Wang [13, 14] and Cui [8, 16].

Proceeding with the analysis of the geometrical meaning of $\beta_m^G(\tau)$, using equations (32) and (33) and rearranging the result, the expression for evolving nCIS is found:

$$\Psi_m^c(t) = \exp[-im(\Omega t + \omega t + \phi)]\{\exp[-i(\omega t + \phi)I_z]\exp(-i\theta I_y)\exp[i(\omega t + \phi)I_z]\}|m\rangle. \tag{36}$$

This is recognized to be a spin coherent state representation [6] with the operator in braces allowing for the standard mapping onto the two-sphere $S^2 = SU(2)/U(1) = SO(3)/SO(2)$. The representing vector is found to precess in a cone of angle θ around the z-axis at angular frequency ω (see figure 1). Hence, the evolution path is specified by the pair of angles $(\theta, \omega t)$ and S^2 is a realization of the projective Hilbert space. The geometric phase (35) is m times the solid angle enclosed by the trajectory, which is a well known result.

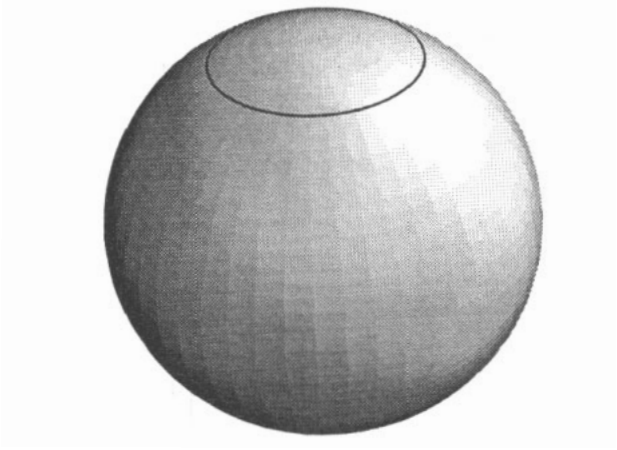


Figure 1. The evolution of nCIS as represented in the projective space S^2.

Consider now the SCIS. It can be seen immediately that the rotating-frame transformation (27) meets the requirement of translational invariance (20). Eigenvalues of $\tilde{H}$, $\tilde{\lambda}_m$, are found from equation (29) to be

$$\tilde{\lambda}_m = m\Omega. \tag{37}$$

Thus, the conditions (17) are

$$m\Omega T = (m+1)\Omega T + 2\pi N_{m,m+1} \qquad m = -j, -j+1, \ldots, j-1 \tag{38}$$

which can be satisfied by a proper choice of T. The minimum T is $T = 2\pi/\Omega$ and the operator

$$S(T)S^{-1}(0) = \exp\left[-\mathrm{i}2\pi\left(\frac{\omega}{\Omega}\right)I_z\right] \tag{39}$$

follows from equation (27).

Thus the SCIS are represented in the general case by the $|m\rangle$ basis states,

$$\Phi_m^c(0) = |m\rangle. \tag{40}$$

Notice, that if additional restrictions are placed on the parameters of the Hamiltonian, specifically, requiring that ω and Ω are commensurable, i.e. $\omega = n\Omega$, then the set of SCIS is given by a variety of arbitrary initial states.

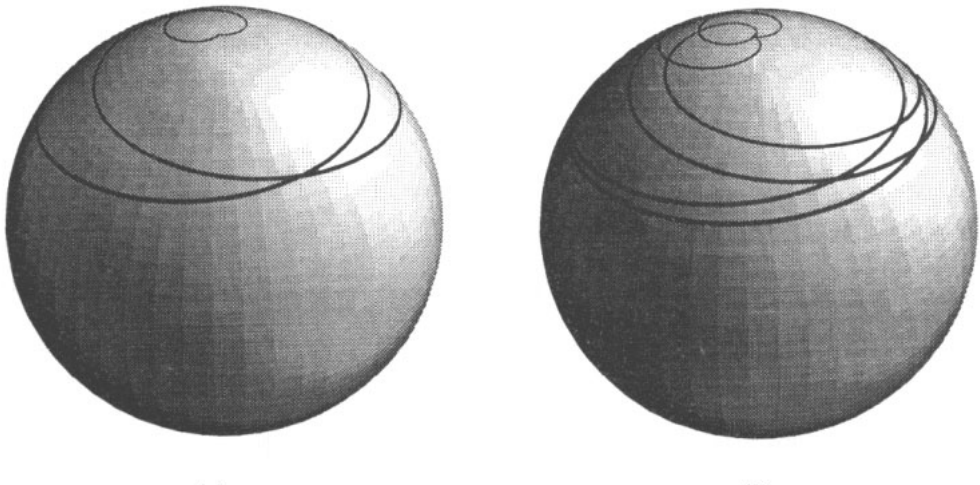
The overall and geometric phases for SCIS (equation (40)) are obtained as

$$\varphi_m(T) = -2\pi m\left(1 + \frac{\omega}{\Omega}\right) \tag{41}$$

$$\beta_m^G(T) = -2\pi m\left(1 - \frac{\Delta}{\Omega} + \frac{\omega}{\Omega}\frac{\omega_1^2}{\Omega^2}\right). \tag{42}$$

A geometrical interpretation is also available on the basis of equation (32). Taking S^2 as a projective space, we can see that the evolution of the vectors representing SCIS is given by a superposition of two rotations. One rotation is around the z-axis and the second is around the axis oriented as specified by the angles (θ, φ). The trajectories are shown in figure 2. The geometric phase (42) preserves its meaning as m times the solid angle enclosed by these spiral trajectories [19].

It is appropriate to analyse here the dimensionality of projective Hilbert spaces. Page [20] and Bouchiat and Gibbons [21] propose versions of the projective space described by $2\aleph - 2$ real parameters, where $\aleph$ is the dimensionality of Hilbert space. In the situation when the dynamics of the system is described within a framework of Lie algebra, there is a projective Hilbert space of dimensionality $\mathfrak{N} - \mathfrak{I}$ available, where $\mathfrak{N}$ and $\mathfrak{I}$ are the dimensionalities of Lie algebra and its Cartan subgroup, respectively [6–8]. For the problem in question this is S^2 of dimensionality two. It follows from the present analysis, however, that under favourable circumstances the dimensionality of projective Hilbert space can be reduced further. For the case under consideration the projective Hilbert space is one-dimensional, S^1 being parametrized by a single angle (Ωt). This should be regarded as a consequence of the coordinate singularity inherent in $SU(2)$. Construction of compact realizations of projective Hilbert space is, we believe, a key point in the calculation of geometric phases for specific quantum mechanical systems.



(*a*) (*b*)

Figure 2. The evolution of SCIS as represented in the projective space S^2. (*a*) The evolution curve for one period T, $\omega/\Omega = 2.2$. (*b*) The evolution curve for two periods T, $\omega/\Omega = 2.2$. Self cross section points shown by these curves correspond to the cycles of evolution, which can be characterized as unstable. If the ratio ω/Ω is given by an irrational number, each of these cycles occurs only once for an infinitely long evolution.

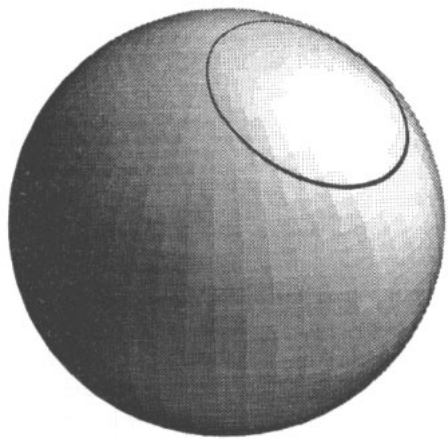


Figure 3. The evolution of SCIS in the 'rotated frame' of [5] as represented in the projective space S^2.

To the best of our knowledge, SCIS for the problem in question were investigated only by Layton *et al* in their paper [5]. Analysis of [5], however, involves a transformation which is referred to as a 'rotating-frame transformation'. The problem of geometric phase invariance under time-dependent unitary transformations was originally posed and explored by Kobe [22]. Recently, it was discussed by Kendrick [23] and Liang and Müller-Kirsten [24]. It was demonstrated that the geometric phase is invariant under time-dependent unitary transformations which are properly defined by equations (2)–(4). At the same time, the transformation of the Hamiltonian (4) alone, without the corresponding transformation of the wavefunction (2), breaks the Schrödinger equation and, therefore, changes the dynamics

of the system, giving different results for the overall and geometric phases [24]. This type of partial transformation was invoked in the analysis of [5], which is equivalent to setting $S(t) = 1$ in equation (2) and, consequently in equation (39). With this choice, the class of SCIS is expanded to a whole variety of initial states, and the trajectory is depicted as a closed loop (see figure 3). The results for the geometric phase for the state $|m\rangle$ then becomes

$$\gamma_m^{\mathrm{G}}(T) = -2\pi m\left(1 - \frac{\Delta}{\Omega}\right). \tag{43}$$

This result was obtained for the state $|-j\rangle$ in [5] and can be compared with our expression (42) for the state $|m\rangle$.

The difference between equations (42) and (43) requires a comment from the viewpoint of practical nuclear magnetic resonance (NMR) where the spin response is recorded in a rotating frame. The concept exploited by experimental NMR is different from that used here. In the former case it implies that the phase accumulated by a carrier wave of an NMR receiver is subtracted from the observable phase accumulated by a density matrix element. This is not the origin of the difference between equations (42) and (43).

4. Spin $j = 1$ subject to an axially symmetric quadrupole interaction in a precessing magnetic field

Spin $j = 1$ as an example of a system exhibiting geometric phase effects was investigated by Bouchiat and Gibbons [21]. These authors elaborated the parametrization for CIS with the parameters allowing for a geometrical interpretation. It was also demonstrated how a time-dependent unitary transformation can be used to construct a variety of Hamiltonians and associated CIS starting from a particular Hamiltonian with known CIS (cf equations (2) and (4)). Following this, Bouchiat [25] examined the adiabatic Berry's phase exhibited by a $j = 1$ system subject to a quadrupolar interaction. The non-Abelian adiabatic phase for spin $j = \frac{3}{2}$ was recently treated by Kwon *et al* [26].

The present treatment is believed to be the first calculation of the non-adiabatic geometric phase for a spin system evolving under a Hamiltonian containing terms that are quadratic in spin operators. We consider the case of an axially symmetric quadrupolar interaction with its symmetry axis directed along the static component of the precessing magnetic field. A good example of such a case is found in typical NMR experiments performed on a pair of dipolar coupled spins of $\frac{1}{2}$ in a liquid crystalline environment [27]. The Hamiltonian' is given by

$$H(t) = \omega_0 I_z + \omega_1[I_x\cos(\omega t + \phi) + I_y\sin(\omega t + \phi)] + \omega_{\mathrm{Q}}(I_z^2 - \tfrac{2}{3}E) \tag{44}$$

where ω_{Q} is a quadrupolar coupling constant and E is the identity. Using the transformation (27), $\tilde{H}$ is obtained as

$$\tilde{H} = \Delta I_z + \omega_1 I_x + \omega_{\mathrm{Q}}(I_z^2 - \tfrac{2}{3}E). \tag{45}$$

The eigenvalues of the Hamiltonian $\tilde{H}$ are given by a cubic equation [28]:

$$\lambda^3 - a\lambda - b = 0 \tag{46}$$

with

$$a = \Delta^2 + \omega_1^2 + \tfrac{1}{3}\omega_Q^2$$

$$b = \tfrac{1}{3}\omega_Q(2\Delta^2 - \omega_1^2 - \tfrac{2}{9}\omega_Q^2)$$

which leads to the eigenvalues

$$\lambda_n = 2(a/3)^{1/2}\cos[\tfrac{1}{3}\cos^{-1}(c) + 2(n+1)\pi/3] \qquad (n = -1, 0, 1) \tag{47}$$

$$c = \tfrac{3}{2}(3/a)^{1/2}(b/a)$$

and eigenvectors

$$\begin{aligned} x_{-1}^{(n)} &= -\frac{1}{\sqrt{2}}\omega_1(\tfrac{1}{3}\omega_Q - \lambda_n + \Delta)/P \\ x_0^{(n)} &= [(\tfrac{1}{3}\omega_Q - \lambda_n)^2 - \Delta^2]/P \\ x_1^{(n)} &= -\frac{1}{\sqrt{2}}\omega_1(\tfrac{1}{3}\omega_Q - \lambda_n - \Delta)/P \\ P^2 &= \omega_1^2[(\tfrac{1}{3}\omega_Q - \lambda_n)^2 + \Delta^2] + [(\tfrac{1}{3}\omega_Q - \lambda_n)^2 - \Delta^2]^2. \end{aligned} \tag{48}$$

Applying the procedure presented in section 2, the nCIS are given by

$$\Psi_n^c(0) = \exp(i\phi I_z)|x^{(n)}\rangle. \tag{49}$$

Then the overall phase accumulated during the period $\tau = 2\pi/\omega$ is found to be

$$\varphi_n(\tau) = -2\pi\left(1 + \frac{\lambda_n}{\omega}\right). \tag{50}$$

Likewise, the geometric phase is calculated from equation (15), giving

$$\beta_n^G(\tau) = -2\pi\left(1 + \sum_m m|x_m^{(n)}|^2\right). \tag{51}$$

In the limit of $\omega_Q = 0$ this result reduces to equation (35).

Calculation of phases for sCIS starts with

$$\Phi_m^c(0) = |m\rangle. \tag{52}$$

The conditions (17) read as

$$\begin{aligned} (\lambda_{-1} - \lambda_0)T &= 2\pi N_{-10} \\ (\lambda_0 - \lambda_1)T &= 2\pi N_{01}. \end{aligned} \tag{53}$$

In terms of NMR it can be identified as the generalized Hartmann–Hahn match condition. The relationship

$$\lambda_{-1} + \lambda_0 + \lambda_1 = 0 \tag{54}$$

is also invoked, thereby yielding

$$\begin{aligned}\lambda_{-1} &= \frac{1}{3}\frac{2\pi l}{T} \\ \lambda_0 &= \frac{1}{3}\frac{2\pi l}{T} + \frac{2\pi k}{T} \\ \lambda_1 &= \frac{1}{3}\frac{2\pi l}{T} - \frac{2\pi(l+k)}{T}\end{aligned} \tag{55}$$

with

$$k = N_{01}$$

$$l = -(2N_{-10} + N_{01}).$$

The period T and the Hamiltonian parameters necessary to meet the conditions (53) can now be determined. The cubic equation (46) gives

$$\begin{aligned}\lambda_{-1}\lambda_0 + \lambda_0\lambda_1 + \lambda_{-1}\lambda_1 &= -a \\ \lambda_{-1}\lambda_0\lambda_1 &= b.\end{aligned} \tag{56}$$

Combining equations (55) and (56) yields

$$\begin{aligned}\Delta^2 + \omega_1^2 + \frac{1}{3}\omega_Q^2 &= \frac{1}{3}\left(\frac{2\pi}{T}\right)^2 (l^2 - 3kl + 3k^2) \\ \omega_Q\left(\Delta^2 - \frac{1}{2}\omega_1^2 - \frac{1}{9}\omega_Q^2\right) &= -\frac{1}{18}\left(\frac{2\pi}{T}\right)^3 l(l-3k)(3k-2l).\end{aligned} \tag{57}$$

These equations generate surfaces in the four-dimensional parameter space $\{\Delta, \omega_1, \omega_Q, T\}$. Intersections of the two surfaces give a variety of $\{\Delta^c, \omega_1^c, \omega_Q^c, T^c\}$, which give rise to the SCIS. Rearranging equations (57) gives

$$\frac{(\Delta^2 + \omega_1^2 + \frac{1}{3}\omega_Q^2)^3}{\omega_Q^2(\Delta^2 - \frac{1}{2}\omega_1^2 - \frac{1}{9}\omega_Q^2)^2} = 12\frac{[1 - 3k/l + 3(k/l)^2]^3}{(1-3k/l)^2(3k/l-2)^2} \tag{58}$$

and

$$T = \frac{\pi l}{3}\frac{(1-3k/l)(2-3k/l)}{[1-3k/l+3(k/l)^2]}\frac{\Delta^2 + \omega_1^2 + \frac{1}{3}\omega_Q^2}{\omega_Q(\Delta^2 - \frac{1}{2}\omega_1^2 - \frac{1}{9}\omega_Q^2)}. \tag{59}$$

Equation (58) imposes the restriction on the set $\{\Delta^c, \omega_1^c, \omega_Q^c\}$, whereas equation (59) determines T in terms of $\{\Delta^c, \omega_1^c, \omega_Q^c\}$. Since k and l are arbitrary integers, it would appear that by taking $k \to \infty$ and $l \to \infty$ we shall be in a position to satisfy equation (58) for arbitrary values of $\Delta, \omega_1, \omega_Q$. However, it would also lead to $T \to \infty$. In practice, condition (58) can easily be met by proper choices of k and l and adjustment of the experimental parameters Δ and ω_1.

Consider now the calculation of phases. The overall phase is found from equations (55) to be

$$\varphi_m(T) = -\frac{2\pi l}{3} - m\omega T \tag{60}$$

where l and T are fixed in accordance with equations (58) and (59).

In order to calculate the geometric phase (equation (25)), we apply to the Hamiltonian $\tilde{H}$, the diagonalizing transformation as determined by its eigenvectors (48). Performing the integration and using equations (53) results in the following expression:

$$\beta_m^G(T) = -\frac{2\pi l}{3} - m\omega T + m\Delta T + (m^2 - \tfrac{2}{3})\omega_Q T + \left(\sum_{m',m'',m'''} \delta_{0,N_{m',m''}} m'' x_m^{(m')} x_{m'}^{(m'')} x_{m''}^{(m''')} x_{m'''}^{(m)} \right) \omega T. \tag{61}$$

This can be simplified by requiring

$$k/l \notin \{0, -\tfrac{1}{2}, -1\} \tag{62}$$

(see equations (55)). In this way, the last term of equation (61) is reduced to $(\sum_{m',m''} m''|x_{m'}^{(m)}|^2|x_{m'}^{(m'')}|^2)\omega T$. These results are consistent with those in section 3 for the limiting case of $\omega_Q = 0$. Conditions (58) and (59) introduce singularities in this case which are, however, easily handled, so that the result (42) is recovered.

Equation (61), along with equations (48), give the geometric phase for $j = 1$. This calculation can be readily generalized for spins with $j > 1$. The overall phase for nCIS is obtained as

$$\varphi_n(\tau) = -2\pi \left(j + \frac{\lambda_n}{\omega} \right) \tag{63}$$

in agreement with equation (50) for $j = 1$, while the geometric phase is given by

$$\beta_n^G(\tau) = -2\pi \left(j + \sum_m m|x_m^{(n)}|^2 \right). \tag{64}$$

Thus the problem is reduced to finding the eigenvectors and eigenvalues of $\tilde{H}$ for increasing values of j.

In case of sCIS the complete system of linear equations for $\lambda_{-j}, \lambda_{-j+1}, \ldots, \lambda_j$ can be constructed in analogy to equations (53) and (54), leading to

$$\begin{aligned} \lambda_{-j} &= \frac{2\pi}{T}\frac{l}{2j+1} \\ \lambda_{-j+1} &= \frac{2\pi}{T}\frac{l}{2j+1} + \frac{2\pi}{T}k_1 \\ \lambda_{-j+2} &= \frac{2\pi}{T}\frac{l}{2j+1} + \frac{2\pi}{T}k_2 \\ &\ldots \\ \lambda_j &= \frac{2\pi}{T}\frac{l}{2j+1} - \frac{2\pi}{T}\left(l + \sum_i k_i \right). \end{aligned} \tag{65}$$

Using these values of λ_j, we obtain, in analogy to equations (60) and (61), the phases as

$$\varphi_m(T) = -\frac{2\pi l}{2j+1} - m\omega T \tag{66}$$

$$\beta_m^G(T) = -\frac{2\pi l}{2j+1} - m\omega T + m\Delta T + (m^2 - \tfrac{2}{3})\omega_Q T + \left(\sum_{m',m'',m'''} \delta_{0,N_{m',m''}} m'' x_m^{(m')} x_{m'}^{(m'')} x_{m''}^{(m''')} x_{m'''}^{(m)} \right) \omega T. \tag{67}$$

Since the eigenvalues are fixed by equations (65), the eigenvectors, $x^{(p)}$ in equation (67), can be found in analytical form by solving the corresponding system of linear equations.

As expected, equations (65) lead to $2j$ conditions imposed on $\{\Delta, \omega_1, \omega_Q\} \otimes \{T\}$. However, $2j$ integers $\{l, k_i\}$ can be interpreted as additional $(2j-1)$ continuous fitting parameters.

In conclusion, the present analysis provides a basis for a theoretical description of NMR experiments aimed at elucidation of the geometric phase, such as the experiment due to Suter *et al* [29]. The cyclic evolution as realized in their experiment starts from a steady state of a fictitious spin-$\frac{1}{2}$ system. Thus, the initial state can be described in terms $|\pm\frac{1}{2}\rangle$ vectors and, therefore, can be identified as SCIS, thereby giving a realizable example of their importance. The analysis presented in section 3 provides a theoretical basis for interpretation of Suter's experiment, assuming that the spin $j = 1$ system can be treated as two fictitious spins $\frac{1}{2}$. This approach allows for a direct connection with the 'solid angle' geometric phase description. The results of section 4 can be used for more thorough analysis. We will defer a more detailed consideration of this experiment to a later publication.

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References

[1] Berry M V 1984 *Proc. R. Soc.* A **392** 45
[2] Aharonov Y and Anandan J 1987 *Phys. Rev. Lett.* **58** 1593
[3] Anandan J 1991 *Found. Phys.* **21** 1265
[4] Moody J, Shapere A and Wilczek F 1986 *Phys. Rev. Lett.* **56** 893
[5] Layton E, Huang Y and Chu S-I 1990 *Phys. Rev.* A **41** 42
[6] Perelomov A M 1986 *Generalized Coherent States and Their Applications* (Berlin: Spinger)
[7] Brihaye Y, Giler S, Kosiński P and Maślanka P 1990 *J. Phys. A: Math. Gen.* **23** 1985
[8] Cui S-M 1993 *Phys. Rev.* A **47** 690
[9] Moore D J and Stedman G E 1990 *J. Phys. A: Math. Gen.* **23** 2049
[10] Moore D J 1990 *J. Phys. A: Math. Gen.* **23** L665
[11] Shirley J H 1965 *Phys. Rev.* A **138** 979
[12] Moore D J 1991 *Phys. Rep.* **210** 1
[13] Wang S-J 1990 *Phys. Rev.* A **42** 5103
[14] Wang S-J 1990 *Phys. Rev.* A **42** 5107
[15] Slichter C P 1990 *Principles of Magnetic Resonance* (Berlin: Springer)
[16] Cui S-M 1992 *Phys. Rev.* A **45** 5255
[17] Fernández C D J, del Olmo M A and Santander M 1992 *J. Phys. A: Math. Gen.* **25** 6409
[18] Gao X-C, Xu J-B and Qian T-Z 1991 *Phys. Lett.* **152A** 449
[19] Skrynnikov N R 1994 unpublished
[20] Page D N 1987 *Phys. Rev.* A **36** 3479
[21] Bouchiat C and Gibbons G W 1988 *J. Physique* **49** 187
[22] Kobe D H 1990 *J. Phys. A: Math. Gen.* **23** 4249
[23] Kendrick B 1992 *J. Phys. A: Math. Gen.* **25** 885
[24] Liang J-Q and Müller-Kirsten H J W 1992 *Ann. Phys.* **219** 42
[25] Bouchiat C 1989 *J. Physique* **50** 1041
[26] Kwon O, Kim S-K and Kim Y 1992 *Phys. Lett.* **164A** 374
[27] Keller A 1988 *Adv. Magn. Reson.* **12** 183
[28] Bowden G J, Hutchison W D and Separovic F 1988 *J. Magn. Reson.* **79** 413
[29] Suter D, Mueller K T and Pines A 1988 *Phys. Rev. Lett.* **60** 1218