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The Berry phase for a threefold degenerate state

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Abstract. Different expressions for calculating the Berry phase of adiabatic processes are reviewed and their limitations are discussed. These expressions are then applied to the case of a circuit surrounding a triple degeneracy. It is shown that the most general formula for the Berry phase requires the use of the full SU(3) invariance group of a T state coupled to $\varepsilon+\tau_1+\tau_2$ modes. The $T\times(\varepsilon+\tau_2)$ Jahn–Teller problem appears as a special case with SO(3) symmetry.

1. Introduction

The aim of this paper is to elucidate some aspects of the calculation of Berry's geometric phase (Berry 1984), with special attention to the $T\times(\varepsilon+\tau_2)$ Jahn–Teller problem. For a review on geometric phases in physics the reader is referred to a recent reprint volume, edited by Shapere and Wilczek (1989), and to papers by Aitchison (1988) and by Zwanziger *et al* (1990). Generalizations of the quantum phase to non-adiabatic cycles have been considered by Aharonov and Anandan (1987), and most recently by Moore and Stedman (1990a).

In section 2 we present a general outline of the Berry phase calculation. The case of the SO(2) and SU(2) Hamiltonians is revisited in section 3. The next section is devoted to the $T\times(\varepsilon+\tau_2)$ Jahn–Teller problem with SO(3) symmetry. In section 5 we address the general case of a threefold degenerate state with SU(3) symmetry.

2. Derivation of the Berry phase

Suppose we have a quantum system characterized by a time-independent Hamiltonian $\hat{H}(X)$ where $X\in\Omega$ is a parameter from a space Ω. Let $|n(X)\rangle$ be a certain non-degenerate and normalized eigenstate of the stationary Schrödinger equation

$$\hat{H}(X)|n(X)\rangle = E_n(X)|n(X)\rangle \qquad X\in\Omega \tag{1}$$

depending smoothly on X. In our notation we omit current variables (spatial, momentum, spin) on which $\hat{H}(X)$, $E_n(X)$ and $|n(X)\rangle$ depend, except for the parameter X.

Let us now assume that the Hamiltonian evolves from the time t_0 until t_1 along a smooth trajectory $X(t)$ in the parameter's space Ω. If the evolution is slow enough, instead of solving the time-dependent Schrödinger equation

$$\mathrm{i}\hbar\partial_t|\psi(t)\rangle = \hat{H}(t)|\psi(t)\rangle \tag{2}$$

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one can use the adiabatic approximation which states for non-degenerate $|n(X)\rangle$:

$$|\psi(t)\rangle \approx |n(X(t))\rangle \exp\left\{-\frac{\mathrm{i}}{\hbar}\int_0^t E_n(X(t'))\,\mathrm{d}t'\right\}. \tag{3}$$

In the context of this approximation the omitted (current) variables are called fast variables, whereas X is the slow variable as we assume that it changes slowly compared to them. The approximation (3) states that the wavefunction $|\psi\rangle$ changes in time like $|n(X)\rangle$ the stationary solution of (1), multiplied by a dynamical phase factor which registers the passage of time. In other words, $\hat{H}(t)$ does not change rapidly enough to allow transitions between different solutions $|m(X)\rangle$ of (1) and they evolve independently of each other.

The projection of (2) in the adiabatic approximation (3) onto $\langle n(X(t))|$ yields the rule

$$\langle n(X(t))|\partial_t|n(X(t))\rangle = 0 \qquad t_0 < t < t_1. \tag{4}$$

This equation describes the evolution in time of a given state $|n(X(t))\rangle$. Since $|n(X(t))\rangle$ depends on t only through $X(t)$ the rule can be converted into the form

$$\mathrm{Im}\langle n|\mathrm{d}n\rangle = 0. \tag{5}$$

Here the imaginary function was introduced for later calculational convenience. Its use is justified by the fact that the bracket must be purely imaginary as a result of normalization of $|n(X)\rangle$. Hence any state in real form necessarily obeys this rule of evolution.

The law in (5) is a crucial point in the derivation of the Berry phase. It defines a way of transporting the quantum state $|n\rangle$ along a curve $C = \{X(t): t_0 \leq t \leq t_1\}$ in accordance with the Schrödinger equation under an adiabatic constraint (Berry 1989).

In the language of differential geometry (5) defines a connection in the line bundle $(P, \mathrm{U}(1), C)$ with the bundle space P being a sum over $X \in C$ of $E_n(X)$ energy eigenspaces for $\hat{H}(X)$, with structural group U(1) and the base space $C \subset \Omega$. Let us call P' the principal bundle $(P', \mathrm{U}(1), \Omega)$ spanned over the whole space Ω. Expression (5) is thus a quantum parallel transport law for $|n(X)\rangle$.

Let us now consider the system that is transported in this way along a curve C which is closed ($X(t_0) = X(t_1) = X_0$) in Ω. After such a circuit, it may return as a state which is not identical with the initial one. The classical analogue of this is a parallel transport of a vector along a closed curve on a sphere. The final position of such a vector is rotated by some angle depending on the curve. Due to the assumption that the states $|n(X)\rangle$ are non-degenerate the final state can differ from the initial one in a given X_0 only by a phase $\gamma_n(C)$

$$|n(X_0^-)\rangle = \mathrm{e}^{\mathrm{i}\gamma_n(C)}|n(X_0^+)\rangle \tag{6}$$

where $|n(X_0^+)\rangle = \lim_{t\to t_0^+}|n(X(t))\rangle$ is the initial state and $|n(X_0^-)\rangle = \lim_{t\to t_1^-}|n(X(t))\rangle$ the final state. One way of calculating this phase is to take the solution $|n(X)\rangle$ of (1) in a form in which it obeys the parallel transport law (5) along $C\backslash\{X_0\}$. This can always be achieved by an appropriate choice of the gauge function $\mu(X)$ for $|n(X)\rangle$

$$|n^\mu(X)\rangle = |n(X)\rangle\,\mathrm{e}^{-\mathrm{i}\mu(X)} \qquad X \in C. \tag{7}$$

The Berry phase corresponding to the state $|n\rangle$ transported along the curve C is then

$$\gamma_n(C) = \mathrm{Im}\,\ln[\langle n^\mu(X_0^+)|n^\mu(X_0^-)\rangle] \tag{8}$$

where ln is a complex function. Expression (8) (or (6) in a gauge $\mu(X)$) is the most natural formula for the Berry phase, based entirely on an examination of the discontinuity of a wavefunction after parallel transport round a circuit. It exhibits the simple fact that the phase is a difference of phases of the same quantum stationary state $|n^{\mu}(X_0)\rangle$ after and before its evolution. The requirement connected with expression (8) is that the state $|n\rangle$ must be found in a gauge in which it obeys the parallel transport law (5) along C.

The next formula for the Berry phase can be derived if there exists another gauge function $\nu(X)=\mu(X)+\mu'(X)$, such that $\mu'(X)$ is differentiable in $C\backslash\{X_0\}$ and the vector field $|n^{\nu}(X)\rangle$ is continuous along C (including X_0). This implies that $|n^{\nu}(X)\rangle$ must be single-valued over the whole base space C. The field $|n^{\nu}(X)\rangle$ forms a frame of reference for the calculation of the phase. Although this gauge does not have to exist globally in P' one can always find it locally along a 1D curve C (in P). In terms of the function $\mu'(X)$ the Berry phase is

$$\gamma_n(C)=\mu'(X_0^-)-\mu'(X_0^+) \tag{9}$$

where $\mu'(X_0^-)$ and $\mu'(X_0^+)$ are final and initial point limits of $\mu'(X(t))$. Now we can write the second expression for the Berry phase which follows from (9) and (5) when the differential of (7) (for $\mu=\nu$) is projected onto $\langle n^{\nu}(X)|$

$$\gamma_n(C)=-\mathrm{Im}\oint_C\langle n^{\nu}|\mathrm{d}n^{\nu}\rangle. \tag{10}$$

In this form $\gamma_n(C)$ is a path integral of a 1-form $-\mathrm{Im}\langle n^{\nu}|\mathrm{d}n^{\nu}\rangle$. It is worth noting that using the expression (10) we do not have to know the actual form of the gauge $\mu(X)$ in which $|n^{\mu}(X)\rangle$ satisfies (5). Instead one should only find the gauge $\nu(X)$ in which the initial solution $|n(X)\rangle$ of (1) is continuous.

In the foregoing expressions for the Berry phase specific gauge transformations were seen to be required. This might make it appear that the phase itself is gauge dependent. Clearly this is not the case. The choice of specific gauges merely reflects the different conditions under which (8)-(10) are valid. The Berry phase itself is a geometric property of a closed curve in a projective Hilbert space and as such it is gauge invariant (Aharonov and Anandan 1987).

The difficulty of finding an appropriate gauge can be avoided when using Stokes' theorem and converting (10) into the surface integral of an exterior derivative of $\langle n^{\nu}|\mathrm{d}n^{\nu}\rangle$:

$$\gamma_n(C)=-\mathrm{Im}\iint_S\langle \mathrm{d}n^{\nu}|\wedge|\mathrm{d}n^{\nu}\rangle \tag{11}$$

where S is an arbitrary orientable surface such that $\partial S=C$ (its boundary is equal to C). In this form $|n^{\nu}(X)\rangle$ has to be differentiable as a function defined almost everywhere (i.e. except for the set of measure zero) on S. The most important feature of this expression is that it is invariant with respect to the gauge transformation (7). It follows then that one can use in formula (11) the wavefunction in its original form

$$\gamma_n(C)=-\mathrm{Im}\iint_S\langle \mathrm{d}n|\wedge|\mathrm{d}n\rangle \tag{12}$$

without referring to special gauges as in (8) and (10). The weak point of this formula is that the exterior derivative of $\langle n^{\nu}|\mathrm{d}n^{\nu}\rangle$ does not always exist, e.g. if Ω is 1D. Neither can one use this formula if the enclosed surface S is not orientable (Arnold 1978).

Note that derivations of (10) from (9) as well as (11) from (10) are consequences of Stokes' theorem expressing integrals of 0-forms by 1-forms, and 1-forms by 2-forms respectively.

The integrands in expressions (10) and (12) may be denoted as the connection form $\mathscr{A} := -\mathrm{Im}\langle n^{\nu}|\mathrm{d}n^{\nu}\rangle$ and the curvature from $\mathscr{B} := -\mathrm{Im}\langle \mathrm{d}n| \wedge |\mathrm{d}n\rangle$. There is an analogy between these forms and the vector potential $\boldsymbol{A}$ and the magnetic field $\boldsymbol{B}$ in the theory of electromagnetism. The formulae for the flux of a magnetic field through the surface S are analogous to expressions (10) and (12) for the Berry phase where the form $\mathscr{A}$ plays the role of the vector potential $\boldsymbol{A}$ and the form $\mathscr{B}$ corresponds to the field $\boldsymbol{B}$. Note that the connection form $\mathscr{A}$ yields a non-zero Berry phase only if the difference $\nu(X)-\mu(X)$ has a discontinuity at X_0 (cf. (9)). Thus the phase, as defined by (10) or (12), is due to some flux of the curvature form field $\mathscr{B}$ through C (cf. discussion at the end of the next section).

We note also that in his 1984 paper Berry proposed one additional expression for $\gamma_n(C)$ expanding the field $\mathscr{B}$ in terms of matrix elements of the operator $\nabla\hat{H}$, between different eigenfunctions of $\hat{H}$.

3. An example: the SO(2) and SU(2) Hamiltonians

In this section we briefly consider the case of a circuit near a point in parameter space at which two electronic states are degenerate. The purpose of this section is to provide an illustrative application of the general methods of section 2. Detailed treatments of this case are available in the literature, mainly in connection with the $E\times\varepsilon$ Jahn–Teller problem (Berry 1984, Ham 1987, Zwanziger *et al* 1990). Let $\hat{H}(R)$ be represented by a 2×2 diagonal matrix with eigenvalues $\frac{1}{2}R$ and $-\frac{1}{2}R$ and let $|\alpha\rangle$ and $|\beta\rangle$ be the corresponding eigenfunctions. These may be expressed by the respective column vectors

$$\begin{pmatrix}1\\0\end{pmatrix} \qquad \text{and} \qquad \begin{pmatrix}0\\1\end{pmatrix}.$$

T_1 in (13) is the generator of a real canonical transformation which leaves the eigenvalues invariant.

$$T_1 = \mathrm{e}^{-\mathrm{i}\theta\sigma_y} = \begin{pmatrix}\cos\theta/2 & -\sin\theta/2\\ \sin\theta/2 & \cos\theta/2\end{pmatrix} \qquad \theta\in[0,2\pi) \tag{13}$$

where

$$\sigma_y = \frac{1}{2}\begin{pmatrix}0 & -i\\ i & 0\end{pmatrix}$$

is the Pauli matrix, which generates a SO(2) subalgebra of SU(2). The corresponding Hamiltonian is given by

$$H_1 = T_1 H T_1^{-1} = \frac{R}{2}\begin{pmatrix}\cos\theta & \sin\theta\\ \sin\theta & -\cos\theta\end{pmatrix}. \tag{14}$$

Eigenfunctions corresponding to the $\frac{1}{2}R$ and $-\frac{1}{2}R$ roots are respectively:

$$|\alpha_1(\theta)\rangle = \cos\frac{\theta}{2}|\alpha\rangle + \sin\frac{\theta}{2}|\beta\rangle$$
$$|\beta_1(\theta)\rangle = -\sin\frac{\theta}{2}|\alpha\rangle + \cos\frac{\theta}{2}|\beta\rangle. \tag{15}$$

The parameter space of H_1 is $\Omega = \{(R, \theta): 0 < R, \theta \in [0, 2\pi)\}$, i.e. the plane without the degeneracy point $R = 0$. The states $|\alpha_1(\theta)\rangle$ and $|\beta_1(\theta)\rangle$ obey the parallel transport law (5) for $\theta \in (0, 2\pi)$ because they are real.

Although $\hat{H}_1$ depends smoothly on (R, θ) in Ω, this is not the case with the wavefunctions. Indeed in (15) the function $\cos\theta/2$ defined over the circle parametrized by $0 \leqslant \theta < 2\pi$ is not continuous at $\theta = 0$. By (8) we have for both states the same result

$$\gamma_{\beta_1}(C) = \begin{cases} \pi & \text{if } C \text{ encircles degeneracy} \\ 0 & \text{otherwise} \end{cases} \tag{16}$$

which is a particular case of a more general fact that the Berry phase is π if C lies in a plane through the degeneracy and encircles it.

Now we can construct a continuous basis $|n^\nu(X)\rangle$ that exists in this case globally over the space Ω and is given by the gauge $\nu(\theta) = \theta/2$:

$$|\alpha_1^\nu(\theta)\rangle = \mathrm{e}^{-\mathrm{i}(\theta/2)}|\alpha_1(\theta)\rangle$$
$$|\beta_1^\nu(\theta)\rangle = \mathrm{e}^{-\mathrm{i}(\theta/2)}|\beta_1(\theta)\rangle. \tag{17}$$

It allows us to use the expression (10) which reads

$$\gamma_n(C) = -\mathrm{Im}\sum_{i=1}^{N}\oint\left\langle n^\nu\left|\frac{\partial n^\nu}{\partial X_i}\right.\right\rangle \mathrm{d}X_i \tag{18}$$

where $X = (X_1, \ldots, X_N)$. It gives for paths encircling the degeneracy:

$$\gamma_{\beta_1}(C) = -\mathrm{Im}\int_0^{2\pi} -\frac{\mathrm{i}}{2}\,\mathrm{d}\theta = \pi \qquad \text{and} \qquad \gamma_{\alpha_1}(C) = \pi.$$

Strictly speaking one might argue that the function $\nu(\theta) = \theta/2$ is not a gauge in the classical sense, since it does not satisfy the condition $\oint \mathrm{d}\nu = 0$. As a result and contrary to the normal gauge, it produces a vector potential which gives rise to a 'fictitious' magnetic field. We note that this extended definition of a gauge is currently being used in modern field theory (Zwanziger *et al* 1990, Polonyi 1988).

An attempt to convert the integral (18) into a gauge-independent form (12) will not succeed in this case. This is because Stokes' theorem can only be used if the rank of the form is less than the space dimension (to make exterior differentiation possible). In the present case $-(\mathrm{i}/2)\,\mathrm{d}\theta$ is a 1-form in the 1D space C (it cannot be regarded as a 1-form in Ω because it is not linear in R). The present example shows that a phase angle of π can occur for a Hamiltonian and wavefunctions that are real. Apparently this result contrasts with a claim by Simon (1983) that the Berry phase is only present in magnetic fields or some other condition producing a non-real Hamiltonian. Simon's argument was based on the observation that the curvature form will vanish if one can choose the wavefunction to be real. Clearly this argument is not valid for the present example since a curvature form does not exist in this case.

Finally, to exemplify the surface integral formula for the Berry phase, we have to extend our model introducing a more-dimensional parameter space Ω by a further canonical transformation of H, generated by

$$T_2(\phi) = e^{-i\phi\sigma_z} = \begin{pmatrix} e^{-i(\phi/2)} & 0 \\ 0 & e^{i(\phi/2)} \end{pmatrix} \qquad \phi \in [0, 2\pi). \tag{19}$$

The transformed H is the SU(2) Hamiltonian and reads

$$H_2(R, \theta, \phi) = \frac{R}{2}\begin{pmatrix} \cos\theta & \sin\theta\, e^{-i\phi} \\ \sin\theta\, e^{i\phi} & -\cos\theta \end{pmatrix}. \tag{20}$$

Its eigenfunctions are

$$\begin{aligned} |\alpha_2(\theta, \phi)\rangle &= e^{-i(\phi/2)} \cos\frac{\theta}{2}|\alpha\rangle + e^{i(\phi/2)} \sin\frac{\theta}{2}|\beta\rangle \\ |\beta_2(\theta, \phi)\rangle &= -e^{-i(\phi/2)} \sin\frac{\theta}{2}|\alpha\rangle + e^{i(\phi/2)} \cos\frac{\theta}{2}|\beta\rangle. \end{aligned} \tag{21}$$

The full parameter space of the transformations $T_1(\theta)$ and $T_2(\phi)$ is the Cartesian product of circles $(\theta, \phi) \in S_1 \times S_1$, i.e. a torus.

However, in order to get a one-to-one correspondence between parameters and Hamiltonians different pairs (θ, ϕ) leading to the same matrices (20) should be regarded as one. This can be done by dividing the Cartesian product space by the equivalence relation $\sim$, defined as

$$(\theta, \phi) \sim (\theta', \phi')$$
$$\Leftrightarrow [(\theta, \phi) = (\theta', \phi') \vee (\theta' = 2\pi - \theta \wedge \phi' = \phi + \pi) \vee \theta = \theta' = 0 \vee \theta = \theta' = \pi].$$

This relation joins redundant parameters (cf figure 1). The resultant space $\Omega = (S_1 \times S_1)/\sim$ is then homeomorphic with the sphere S_2.

One can show that in the gauge (21) the parallel transport law (5) is obeyed by trajectories that form elongation lines and for the meridian of the sphere. Along those trajectories the wavefunctions have one discontinuity point where they change sign thus giving a Berry phase equal to π.

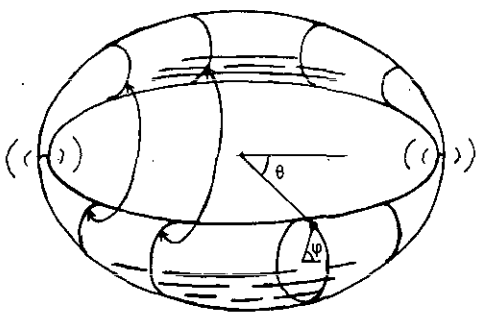

Figure 1. The torus $S_1 \times S_1$ is homeomorphic with the sphere S_2 if it is divided by the equivalence relation $\sim$. Under this relation points (θ, ϕ) and $(2\pi - \theta, \phi + \pi)$ become equivalent and circles corresponding to $\theta = 0$ $(\theta = \pi)$ are shrunk into a point.

To use formula (10) one should construct continuous vector fields $|n^{\nu}(X)\rangle$. In the present case it is not possible to find a gauge in which $|\beta_2^{\nu}(X)\rangle$ or $|\alpha_2^{\nu}(X)\rangle$ would be globally continuous on S_2. The most 'regular' gauge $\nu(\theta, \phi) = \phi/2$ yields

$$|\beta_2^{\nu}(\theta, \phi)\rangle = -e^{-i\phi} \sin\frac{\theta}{2}|\alpha\rangle + \cos\frac{\theta}{2}|\beta\rangle \tag{22}$$

in which (10) can be used for all trajectories that do not pass through the south pole $\theta = \pi$ of S_2 where $|\beta_2^{\nu}\rangle$ has a discontinuity. The connection form is then $\mathcal{A} = \frac{1}{2}(1 - \cos\theta)\,d\phi$. It corresponds to a flux line entering the sphere through its south pole.

For more general solutions we can now use the formula (12). It is practical to reformulate it into a form

$$\gamma_n(C) = -\mathrm{Im} \iint_S \sum_{i,j} \left\langle \frac{\partial n}{\partial X_i} \middle| \frac{\partial n}{\partial X_j} \right\rangle dX_i \wedge dX_j$$

$$= -2\,\mathrm{Im} \sum_{i<j} \iint_S \left\langle \frac{\partial n}{\partial X_i} \middle| \frac{\partial n}{\partial X_j} \right\rangle dX_i\, dX_j \tag{23}$$

where the summation runs over all pairs of components in X space. According to this the Berry phase for the state β_2 and along the circuit C is

$$\gamma_{\beta_2}(C) = \tfrac{1}{2} \iint_S \sin\theta\, d\theta\, d\phi = \tfrac{1}{2}\omega(C) \tag{24}$$

i.e. half the solid angle subtended by C at the centre of the sphere. This is the result which Berry obtained using the $\nabla\hat{H}$ formalism (Berry 1984).

Note that the field $|n(X)\rangle$, i.e. in this case $|\beta_2(\theta, \phi)\rangle$, is discontinuous on the poles of the parameter sphere (for $\theta = 0$ and π) and along the $\phi = 0$ elongation. Also in the most 'regular' gauge $|\beta_2^{\nu}(\theta, \phi)\rangle$ is discontinuous on the south pole $\theta = \pi$. But these irregular points can be excluded from the domain of the field because their set is of measure zero on the sphere S_2. On the remaining part of the domain the field $|n(X)\rangle$ is differentiable and the integral (24) gives the proper value of the Berry phase independently of the gauge chosen.

Going back to the analogy of the connection and curvature forms to the vector potential and the magnetic field, it is clear from (24) that if the Berry phase along C corresponds to a flux through S ($C = \partial S$) then the above example refers to the field of a monopole of strength $\frac{1}{2}$ situated at the centre of the parameter sphere. The field of a monopole is radial and spherically symmetric and thus cannot be described globally by a connection form $\mathcal{A}$, $\mathcal{B} = d\mathcal{A}$ (in our analogy, it corresponds to $\boldsymbol{B} = \mathrm{rot}\,\boldsymbol{A}$). This is the reason why we could not find the form $\mathcal{A}$ defined globally on the whole sphere. All existing vector potentials of a monopole introduce flux lines and break the spherical symmetry of the system.

4. The SO(3) Hamiltonian

In this section we apply the general formulae to the case of the $T \times (\varepsilon + \tau_2)$ Jahn–Teller Hamiltonian assuming linear and equal coupling to the τ_2 and ε phonon modes. This

Hamiltonian has SO(3) symmetry. The associated Berry phase has been studied by Chancey and O'Brien (1988), and by Judd (1989). Attention has also been devoted to the related $T\times\tau_2$ problem (O'Brien 1989, Ham 1990).

Let $\hat{H}(R)$ be represented by 3×3 matrix H with eigenvalues $+\frac{1}{2}q$, $+\frac{1}{2}q$, $-q(q>0)$, and let $|\xi\rangle$, $|\eta\rangle$, $|\zeta\rangle$ be the corresponding eigenfunctions. These may be written in a column vector notation as follows:

$$|\xi\rangle=\begin{pmatrix}1\\0\\0\end{pmatrix}\qquad |\eta\rangle=\begin{pmatrix}0\\1\\0\end{pmatrix}\qquad |\zeta\rangle=\begin{pmatrix}0\\0\\1\end{pmatrix}. \tag{25}$$

T_1 in (26) is a real canonical transformation which leaves the eigenvalues invariant:

$$T_1=\mathrm{e}^{-\mathrm{i}\phi L_3}\mathrm{e}^{-\mathrm{i}\theta L_2}=\begin{pmatrix}\cos\phi\cos\theta & -\sin\phi & \cos\phi\sin\theta\\ \sin\phi\cos\theta & \cos\phi & \sin\phi\sin\theta\\ -\sin\theta & 0 & \cos\theta\end{pmatrix}. \tag{26}$$

L_2 and L_3 are generator matrices of a SO(3) subalgebra of SU(3) (Wagner 1984):

$$L_2=\begin{pmatrix}0 & 0 & i\\ 0 & 0 & 0\\ -\mathrm{i} & 0 & 0\end{pmatrix}\qquad L_3=\begin{pmatrix}0 & -\mathrm{i} & 0\\ i & 0 & 0\\ 0 & 0 & 0\end{pmatrix}. \tag{27}$$

The transformed Hamiltonian is given by (O'Brien 1969):

$$H_1=T_1HT_1^{-1}=\begin{pmatrix}\frac{1}{2}q_\theta-\frac{\sqrt{3}}{2}q_\varepsilon & -\frac{\sqrt{3}}{2}q_\zeta & -\frac{\sqrt{3}}{2}q_\eta\\ -\frac{\sqrt{3}}{2}q_\zeta & \frac{1}{2}q_\theta+\frac{\sqrt{3}}{2}q_\varepsilon & -\frac{\sqrt{3}}{2}q_\xi\\ +\frac{\sqrt{3}}{2}q_\eta & -\frac{\sqrt{3}}{2}q_\xi & -q_\theta\end{pmatrix}. \tag{28}$$

with

$$q_\theta=\frac{q}{2}(3\cos^2\theta-1)\qquad q_\xi=\frac{\sqrt{3}}{2}q\sin 2\theta\sin\phi$$

$$q_\varepsilon=\frac{\sqrt{3}}{2}q\sin^2\theta\cos 2\phi\qquad q_\eta=\frac{\sqrt{3}}{2}q\sin 2\theta\cos\phi$$

$$q_\zeta=\frac{\sqrt{3}}{2}q\sin^2\theta\sin 2\phi.$$

The $(q_\theta, q_\varepsilon)$ parameters refer to the ε mode, while (q_ξ, q_η, q_ζ) constitute the τ_2 mode. The range of these parameters is restricted to the so-called (θ,ϕ) O'Brien sphere, which surrounds the triplet origin. The eigenfunction corresponding to the lowest root of H_1 is given by

$$|\zeta_1\rangle=T_1|\zeta\rangle=\sin\theta\cos\phi|\xi\rangle+\sin\theta\sin\phi|\eta\rangle+\cos\theta|\zeta\rangle. \tag{29}$$

The parameter space of this eigenfunction is a (θ,ϕ) torus with $(\theta,\phi)\in[0,2\pi)\times[0,2\pi)$. However in order to find the parameter space of physical importance, the coordinate space of the phonon modes should be preferred over the Hilbert space of the wavefunction. Physically distinguishable are parameters leading to different Hamiltonians. For

the H_1 Hamiltonian the relevant parameter is thus found to be the hemisphere $S_2/\sim$, where $\sim$ is the equivalence relation:

$$(\theta, \phi) \sim (\theta', \phi') \Leftrightarrow [(\theta, \phi) = (\theta', \phi') \vee (\theta' = \pi - \theta \wedge \phi' = \phi + \pi) \vee \theta' = -\theta = \pm\pi]$$

joining antipodal points of the sphere. The resultant space $S_2/\sim$ can be regarded as a family of directions at the centre of S_2. Exactly as in the case of the SO(2) Hamiltonian, the real state $|\zeta_1\rangle$ is seen to obey the quantum parallel-transport law (5). The associated Berry phase can thus be directly evaluated from the discontinuity of $|\zeta_1\rangle$ on a closed path trajectory using the formula (8). Clearly the parameter space allows two types of closed paths: C_1 paths start and finish at the same (θ, ϕ) point; C_2 paths start at (θ, ϕ) and finish at the (antipodal) inversion point $(\pi - \theta, \phi + \pi)$. Inspection of the wavefunction $|\zeta_1\rangle$ in (29) reveals that both paths induce different Berry phases:

$$\begin{aligned} \gamma(C_1) &= 0 \\ \gamma(C_2) &= -\pi. \end{aligned} \tag{30}$$

These conclusions are not dependent on whether or not the circuit encircles the north pole of the O'Brien sphere as suggested by Chancey and O'Brien (1988). This pole is distinguished only because of our choice of a reference frame. To use formula (10) one should again construct a continuous vector field $|\zeta_1^v\rangle$. Exactly as in the case of the SU(2) Hamiltonian (see (21)), it is not possible to find a gauge in which $|\zeta_1^v\rangle$ would be globally continuous on $S_2/\sim$. The gauge, chosen by Chancey and O'Brien (1988), is $\exp(i\phi)$. It must be realized that this choice only works for paths that avoid the $\theta = 0$ direction. For such paths one has

$$\begin{aligned} \gamma(C) &= -\oint_C \mathrm{Im}\langle\zeta_1^v|\mathrm{d}\zeta_1^v\rangle \\ &= -\oint_C \mathrm{d}\phi \end{aligned} \tag{31}$$

which yields the result in (30). It is noteworthy that the path integral in (31) cannot be converted into a gauge-invariant 2-form, because the 1-form $\langle n|\mathrm{d}n\rangle$ is a form in S_1, i.e. its domain is 1D. Hence if we want to apply Stokes' theorem, we must start from a more complete description of the threefold degeneracy in the centre. This can be done by using a SU(3) Hamiltonian.

5. The SU(3) Hamiltonian

In the previous section we reached the conclusion that a global gauge-invariant expression for the Berry phase of the SO(3) Hamiltonian could not be derived in a $\varepsilon + \tau_2$ subspace. Exactly as in the case of the twofold degeneracy, such an expression can only be found if one starts off from a more general description of the degenerate manifold, using a SU type Hamiltonian. The transformation which allows complete unitary freedom of one component vector of our manifold is the T_2 transformation, defined in (32):

$$\begin{aligned} T_2 &= \mathrm{e}^{\mathrm{i}/3(\alpha+\beta+\gamma)I}\, \mathrm{e}^{\mathrm{i}/3(\beta-\gamma)B_1}\, \mathrm{e}^{\mathrm{i}/3(\gamma-\alpha)B_2}\, \mathrm{e}^{\mathrm{i}/3(\alpha-\beta)B_3} \\ &= \begin{pmatrix} \mathrm{e}^{\mathrm{i}\alpha} & 0 & 0 \\ 0 & \mathrm{e}^{\mathrm{i}\beta} & 0 \\ 0 & 0 & \mathrm{e}^{\mathrm{i}\gamma} \end{pmatrix} \end{aligned} \tag{32}$$

with

$$I=\begin{pmatrix}1&0&0\\0&1&0\\0&0&1\end{pmatrix}\qquad B_1=\begin{pmatrix}0&0&0\\0&1&0\\0&0&-1\end{pmatrix}$$

$$B_2=\begin{pmatrix}-1&0&0\\0&0&0\\0&0&1\end{pmatrix}\qquad B_3=\begin{pmatrix}1&0&0\\0&-1&0\\0&0&0\end{pmatrix}.$$

The transformed ground state ket thus becomes

$$|\zeta_2\rangle = T_2|\zeta_1\rangle = \sin\theta\cos\phi\ \mathrm{e}^{\mathrm{i}\alpha}|\xi\rangle + \sin\theta\sin\phi\ \mathrm{e}^{\mathrm{i}\beta}|\eta\rangle + \cos\theta\ \mathrm{e}^{\mathrm{i}\gamma}|\zeta\rangle. \tag{33}$$

The parameter space of this $|\zeta_2\rangle$ function may be defined as: $S_2/8\times[0, 2\pi)^3$. Here $S_2/8$ is the eighth part of the S_2 sphere, with: $(\theta, \phi)\in[0, \pi/2]\times[0, \pi/2]$. In this real octant the θ and ϕ functions which appear in (33), are zero or positive. They may thus be identified as the absolute values of the eigenvector coefficients. The α, β, γ parameters are the three corresponding phase angles, each with a range of 2π. As a result the $|\zeta_2\rangle$ ket, defined by the five parameters $(\theta, \phi, \alpha, \beta, \gamma)$, is seen to address an arbitrary point in the U(3) Hilbert space of a threefold degeneracy.

The physical interactions, that can be represented in this space, may be found by transforming the H_1 Hamiltonian of (28):

$$H_2 = T_2 H_1 T_2^{-1} = \begin{pmatrix} \frac{1}{2}q_\theta - \frac{\sqrt{3}}{2}q_\varepsilon & -\frac{\sqrt{3}}{2}Q_\zeta & -\frac{\sqrt{3}}{2}Q_\eta^* \\ -\frac{\sqrt{3}}{2}Q_\zeta^* & \frac{1}{2}q_\theta + \frac{\sqrt{3}}{2}q_\varepsilon & -\frac{\sqrt{3}}{2}Q_\xi \\ -\frac{\sqrt{3}}{2}Q_\eta & -\frac{\sqrt{3}}{2}Q_\xi^* & -q_\theta \end{pmatrix} \tag{34}$$

with

$$Q_\xi = q_\xi\,\mathrm{e}^{\mathrm{i}(\beta-\gamma)}$$

$$Q_\eta = q_\eta\,\mathrm{e}^{\mathrm{i}(\gamma-\alpha)}$$

$$Q_\zeta = q_\zeta\,\mathrm{e}^{\mathrm{i}(\alpha-\beta)}.$$

This H_2 Hamiltonian is defined in a 8-dimensional space, comprising the five $\varepsilon+\tau_2$ phonon modes $(q_\theta, q_\varepsilon, \mathrm{Re}\,Q_\xi, \mathrm{Re}\,Q_\eta, \mathrm{Re}\,Q_\zeta)$ and three time-odd interactions $(\mathrm{Im}\,Q_\xi, \mathrm{Im}\,Q_\eta, \mathrm{Im}\,Q_\zeta)$ transforming in cubic symmetry as τ_1. Note that H_1 commutes with the totally symmetric part of the T_2 operator, $\exp[\mathrm{i}/3(\alpha+\beta+\gamma)I]$, which performs a global phase change of the Hilbert space. As a result H_2 contains only *differences* of the phase angles α, β, γ. It may thus be characterized as a $T\times(\varepsilon+\tau_1+\tau_2)$ Hamiltonian with SU(3) symmetry.

Several representations of the parameter space of H_2 are possible. As in (33) it is convenient to restrict the θ, ϕ variables to the positive $S_2/8$ octant. This choice puts no limitation on the range of q_θ and q_ε. On the other hand it confines the q_ξ, q_η, q_ζ parameters to zero or positive values. These parameters can therefore be identified with the absolute values of the complex Q parameters. Adopting a $Q=|Q|\exp(\mathrm{i}\,\mathrm{Arg}\,Q)$

format, one has

$$\begin{aligned} |Q_\xi| &= q_\xi \qquad \text{Arg } Q_\xi = \beta - \gamma \\ |Q_\eta| &= q_\eta \qquad \text{Arg } Q_\eta = \gamma - \alpha \\ |Q_\zeta| &= q_\zeta \qquad \text{Arg } Q_\zeta = \alpha - \beta. \end{aligned} \tag{35}$$

The Q arguments in (35) are seen to obey the following constraint:

$$\exp i(\text{Arg } Q_\xi + \text{Arg } Q_\eta + \text{Arg } Q_\zeta) = 1. \tag{36}$$

In the phase space of the Q parameters expression (36) defines a {111} periodic plane. Figure 2 shows a section of this plane in the $[0, 2\pi)^3$ Cartesian cube. This section contains two triangles and the (0, 0, 0) origin. The triangles can be represented by their projection on one of the phases of the cube, say Arg $Q_\zeta = 0$. The resulting phase space is the periodic square of the Q_ξ and Q_η arguments. It is topologically equivalent to a torus.

In summary, the parameter space Ω of the SU(3) Hamiltonian may be defined as $S_2/8 \times [0, 2\pi)^2$, where $S_2/8$ is the positive octant of a (θ, ϕ) sphere, and $[0, 2\pi)^2$ refers to the torus, formed by Arg Q_ξ and Arg Q_η. This space is represented schematically in figure 3. It should be noted that on the corners of the (θ, ϕ) octant the phase parameters are redundant, while along the sides only one phase parameter is required.

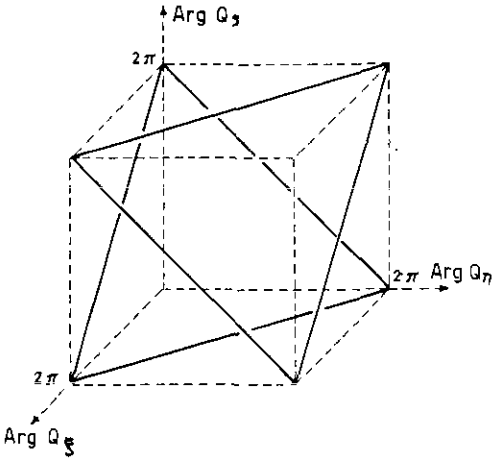

Figure 2. The phase space of the SU(3) Hamiltonian. The Q arguments are confined to the {111} periodic plane. In the Cartesian cube $[0, 2\pi)^3$ this plane consists of two triangles plus the point (0, 0, 0).

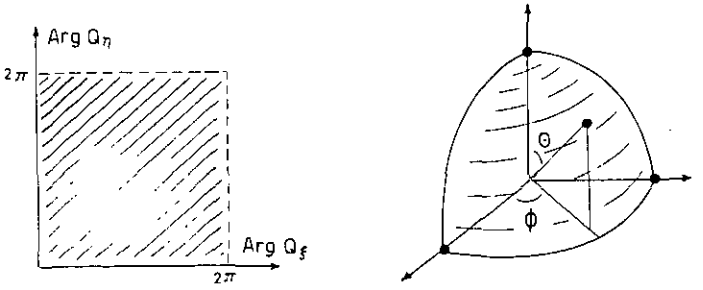

Figure 3. The parameter space Ω of the SU(3) Hamiltonian.

The space Ω allows different types of closed paths. The projection of a closed path in the (θ, ϕ) subspace must always be a closed curve, since this space does not contain redundancies. On the other hand in the phase space of the Q_ξ and Q_η arguments closed paths can be represented by a closed curve, or by a line connecting two opposite sides. A third kind of closure in this space involves a π-jump. This type of path will be discussed later in this section (*vide infra* (39) and figure 4).

The Berry phase associated with a closed path in Ω can now be caculated using both the path integral and surface integral methods. To apply the path integral expression we first extract a phase factor $e^{i\gamma}$ from $|\zeta_2\rangle$. The resulting function appears to be continuous in Ω. Following our earlier convention in (10) we will denote this function as $|\zeta_2^\nu\rangle$. One has

$$\begin{aligned}|\zeta_2^\nu\rangle &= e^{-i\gamma}|\zeta_2\rangle \\ &= \sin\theta\cos\phi\, e^{i(\alpha-\gamma)}|\xi\rangle + \sin\theta\sin\phi\, e^{i(\beta-\gamma)}|\eta\rangle + \cos\theta|\zeta\rangle \\ &= \sin\theta\cos\phi\, e^{-i\mathrm{Arg}Q_\eta}|\xi\rangle + \sin\theta\sin\phi\, e^{i\mathrm{Arg}Q_\xi}|\eta\rangle + \cos\theta|\zeta\rangle. \qquad (37)\end{aligned}$$

The Berry phase of this function follows at once from (10).

$$\begin{aligned}\gamma_{\zeta 2}(C) &= -\mathrm{Im}\oint_C \langle\zeta_2^\nu|\mathrm{d}\zeta_2^\nu\rangle \\ &= \oint_C \sin^2\theta[\cos^2\phi\ \mathrm{d}\,\mathrm{Arg}\,Q_\eta - \sin^2\phi\ \mathrm{d}\,\mathrm{Arg}\,Q_\xi]. \qquad (38)\end{aligned}$$

As an example we may consider closed paths in a subspace defined by $\phi = 0$. In this subspace the only non-vanishing interaction parameters are

$$q_\theta = \tfrac{1}{2}q(3\cos^2\theta - 1) \qquad q_\epsilon = \frac{\sqrt{3}}{2}q\sin^2\theta \qquad Q_\eta = \frac{\sqrt{3}}{2}q\sin 2\theta\exp(\mathrm{i}\,\mathrm{Arg}\,Q_\eta).$$

The q_θ and q_ϵ parameters may be recombined to

$$\tfrac{1}{2}q_\theta + \frac{\sqrt{3}}{2}q_\epsilon = \tfrac{1}{2}q$$

and

$$\frac{\sqrt{3}}{2}q_\theta - \tfrac{1}{2}q_\epsilon = \frac{\sqrt{3}}{2}q\cos 2\theta.$$

Hence in the 8D parameter space of the Hamiltonian, only three dimensions are relevant, viz.

$$\frac{\sqrt{3}}{2}q_\theta - \tfrac{1}{2}q_\epsilon \qquad \mathrm{Re}\,Q_\eta \qquad \mathrm{Im}\,Q_\eta.$$

In this subspace the SU(3) coupling conditions define a sphere with radius $(\sqrt{3}/2)q$. The corresponding polar angles are 2θ for latitude and $\mathrm{Arg}\,Q_\eta$ for longitude, as indicated in figure 4. The circular path C_1 in the figure is entirely in the real coordinate plane; it thus corresponds to closed path of the SO(3) Jahn–Teller Hamiltonian.

To calculate the associated Berry phase, one must realize that the phase of the Q_η parameter changes along this path. For positive real values of Q_η the phase angle is zero, but for negative real values the phase angle equals π. Hence $\mathrm{Arg}\,Q_\eta$ changes from 0 to π at the south pole of the path, and back from π to 0 at the north pole.

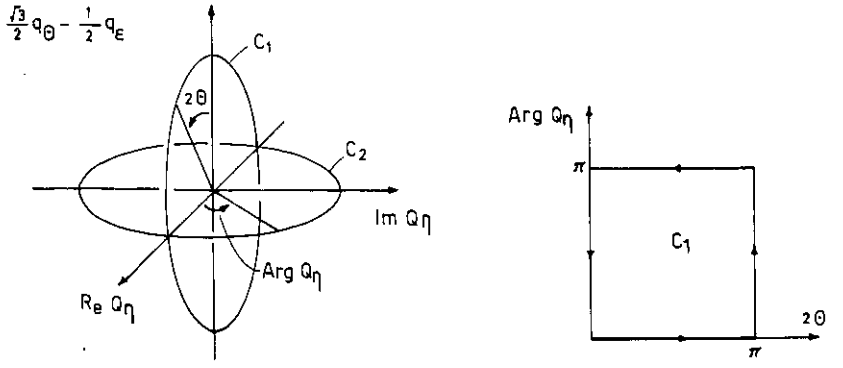

Figure 4. Examples of closed paths in Ω. The top of the figure shows the subspace defined by $\phi = 0$. The bottom shows a projection of the C_1 path in the space of angular coordinates.

This is illustrated more clearly in figure 4, where the path is plotted in the space of the angular coordinates. As can be seen from (38) the π-jump at the south pole ($2\theta = \pi$) will contribute to the Berry phase, yielding

$$\gamma_{\zeta_2}(C_1) = [\text{Arg } Q_\eta]_0^\pi = \pi. \tag{39}$$

This result of course concurs with the conclusions reached in the previous section. Another example is the circular path C_2 in the Argand plane of the Q_η coordinate (cf. figure 4). Along this path one has $2\theta = \pi/2$. Substituting this value in (38) again yields a Berry phase of π:

$$\gamma_{\zeta_2}(C_2) = \oint_{C_2} \sin^2\theta \, \text{d Arg } Q_\eta = \tfrac{1}{2}[\text{Arg } Q_\eta]_0^{2\pi} = \pi. \tag{40}$$

These results can be directly converted into a surface integral expression, by applying Stokes' theorem to the 1-form in (38). One obtains

$$\gamma_{\zeta_2}(C) = -\text{Im} \int\int_S \langle \text{d}\zeta_2^\nu | \wedge | \text{d}\zeta_2^\nu \rangle$$

$$= \int\int_S (\sin 2\theta \cos^2\phi \, \text{d}\theta - \sin 2\phi \sin^2\theta \, \text{d}\phi) \, \text{d Arg } Q_\eta$$

$$+ \int\int_S (-\sin 2\theta \sin^2\phi \, \text{d}\theta + \sin^2\theta \sin 2\phi \, \text{d}\phi) \, \text{d Arg } Q_\xi. \tag{41}$$

As an example we may again consider the $\phi = 0$ subspace of figure 4. In this case (41) reduces to one-half of the solid angle that C subtends on the spherical surface in the relevant subspace.

$$C \subset \{\phi = 0\}\colon \gamma(C) = \tfrac{1}{2} \int\int \sin 2\theta \, \text{d}2\theta \, \text{d Arg } Q_\eta = \tfrac{1}{2}\omega(C). \tag{42}$$

The result in (42) parallels the conclusions for closed paths encircling a twofold degeneracy. This should not be too surprising: by imposing the $\phi = 0$ condition on the Hamiltonian, one restricts its symmetry to a SU(2) subgroup and thus recovers Berry's treatment for a twofold degeneracy. In order to obtain a more transparent form of (41), which underlines the intrinsic symmetry of the result, it is indicated to replace the θ and ϕ coordinates by the more symmetrical Cartesian components $x = \sin\theta\cos\phi$, $y = \sin\theta\sin\phi$, $z = \cos\theta$, with the redundancy condition: $x^2 + y^2 + z^2 = 1$. In the positive (θ, ϕ) octant these Cartesian components are either zero or positive. The path integral of (38) may now be rewritten as

$$\gamma_{\zeta_2}(C) = \oint_C (x^2 \,\mathrm{d}\,\mathrm{Arg}\, Q_\eta - y^2 \,\mathrm{d}\,\mathrm{Arg}\, Q_\xi). \tag{43}$$

External differentiation over this 1-form yields the surface integral over a 2-form:

$$\gamma_{\zeta_2} = \iint_S (\mathrm{d}x^2 \,\mathrm{d}\,\mathrm{Arg}\, Q_\eta - \mathrm{d}y^2 \,\mathrm{d}\,\mathrm{Arg}\, Q_\xi). \tag{44}$$

This surface integral is seen to combine the four parameters that define the SU(3) Hamiltonian. By using the redundancy relations $\mathrm{d}(x^2+y^2+z^2) = 0$ and $\mathrm{d}(\mathrm{Arg}\, Q_\xi + \mathrm{Arg}\, Q_\eta + \mathrm{Arg}\, Q_\zeta) = 0$, it is possible to perform concomitant cyclic permutations of the xyz and $\xi\eta\zeta$ labels. Hence alternative forms of (44) read

$$\gamma_{\zeta_2}(C) = \iint_S (\mathrm{d}y^2 \,\mathrm{d}\,\mathrm{Arg}\, Q_\zeta - \mathrm{d}z^2 \,\mathrm{d}\,\mathrm{Arg}\, Q_\eta)$$

$$\gamma_{\zeta_2}(C) = \iint_S (\mathrm{d}z^2 \,\mathrm{d}\,\mathrm{Arg}\, Q_\xi - \mathrm{d}x^2 \,\mathrm{d}\,\mathrm{Arg}\, Q_\zeta). \tag{45}$$

These formulae provide the desired general expressions for the Berry phase of a threefold degenerate state.

6. Epilogue

As we have argued in this paper, the most natural formula for the evaluation of the geometric phase accompanying a cyclic evolution of a quantum system involves a direct examination of the wavefunction after parallel transport around a circuit. The only difficulty connected with this expression is that the state must be taken into a gauge in which it obeys the transport law. On the other hand, in order to use a gauge-independent formalism, based on Stokes' theorem, the circuit of interest must be defined in a sufficiently general parameter space. This observation connects the Berry phase concept to the group theory of the embedding of invariance groups.

Specifically for a threefold degeneracy, we have found that the $T \times (\varepsilon + \tau_2)$ Jahn–Teller problem with SO(3) symmetry must be embedded in the SU(3) parameter space, reflecting the full symmetry of interactions in a threefold degeneracy. Perhaps this new insight has an interesting physical 'portée'. It draws attention to the fact that the JT distortions explore only a minute part of the interaction space. To appreciate the rich

structure of the degeneracy, other symmetry-breaking processes should be investigated. Such processes could be external to the system, e.g. by switching on a magnetic field, or intrinsic, i.e. related to the dynamics of the system itself. Interestingly Moore and Stedman (1990b) have pointed to the possibility of the actual observation of such intrinsic time-odd electron–phonon coupling mechanisms in ligand field systems.

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