Study of the Aharonov-Anandan Quantum Phase by NMR Interferometry

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Aharonov and Anandan have recently reformulated and generalized Berry's phase by showing that a quantum system which evolves through a circuit $C$ in projective Hilbert space acquires a geometrical phase $\beta(C)$ related to the topology of the space and the geometry of the circuit. We present NMR interferometry experiments in a three-level system which demonstrate the Aharonov-Anandan phase and its topological invariance for different circuits.

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It was shown by Berry that a nondegenerate quantum state $|\psi(t)\rangle$ of a Hamiltonian $\mathcal{H}(t)$ which varies adiabatically through a circuit $C$ in parameter space acquires, in addition to the "normal" dynamical phase $\gamma_d$ given by

$$
\gamma_d = -\frac{i}{\hbar} \int \langle \psi(t)| \mathcal{H}(t) |\psi(t)\rangle dt
$$

(1)

the generalization of $\omega t$, a phase which is related to the geometry of the circuit. Simon explained that this geometrical phase could be viewed as a consequence of parallel transport in a curved space appropriate to the quantum system. Much experimental and theoretical work on "Berry's phase" and its connection to the early work of Aharonov and Bohm, and Mead and Truhlar, has since appeared. Recently, three important generalizations of this phase have appeared. In the first, Wilczek and Zee removed the constraint of nondegenerate states and related the evolution and phases of a degenerate manifold to a non-Abelian gauge. In the second, Berry, as well as Jackiw and co-workers, removed the constraint of adiabaticity in the Hamiltonian circuit and developed asymptotic expansions for the evolving (noncyclic) states and phases.

The third generalization, a fundamental one, which forms the subject of NMR experiments in this Letter and optical experiments in the accompanying Letters by Chiao and co-workers and Bhandari and Samuel, is due to Aharonov and Anandan. They cast the problem in terms of circuits of the quantum system itself, rather than circuits of the Hamiltonian in parameter space. It is clear from recent work that this is, in some sense, a continuous version of the phase discovered by Pancharatnam more than 30 years ago. A simple formulation of the Aharonov-Anandan (AA) phase is as follows: If the density operator $|\psi(\tau)\rangle$ for a pure state (generalized to mixed states by superposition) undergoes a cyclic evolution through a circuit $C$ in projective Hilbert (density operator) space $|\psi(\tau)\rangle \rightarrow_C |\psi(\tau)\rangle$, then the quantum state $|\psi\rangle$ acquires a geometrical phase $\beta(C)$ related to the (a) holonomy associated with parallel transport around the circuit. This phase appears in addition to the dynamical phase $\gamma_d$ given by (1), where $\mathcal{H}(t)$ may be noncyclic and nonadiabatic. Thus, we can write

$$
|\psi\rangle \rightarrow C \exp[i(\gamma_d + \beta(C))]|\psi\rangle.
$$

The importance of the AA formulation is that it applies whether or not the Hamiltonian $\mathcal{H}(t)$ is cyclic or adiabatic- the geometrical phase depends only on the cyclic evolution of the system itself. This establishes a simple connection of the geometrical phase to the Aharonov-Bohm effect which does not invoke adiabaticity of the circuit. The Berry phase thereby emerges as a manifestation of the AA geometry in the case of adiabatic evolution. Bouchiat and Gibbons have presented a thorough theoretical analysis of the AA phase for a three-level spin-1/2 system.

As a demonstration of the AA phase, consider the situation depicted in Fig. 1, a version of NMR interferometry related to neutron (say) interferometry. The two-level system (TLS) comprising states 2 and 3 can be treated as a fictitious spin-1/2 system. Time-dependent magnetic fields are applied to take the TLS through a circuit. The resultant phase factor associated with state 2 is detected by means of its effect on transition 1-2. Suppose the system begins in thermal equilibrium so that the initial density operator for the three-level system is diagonal in the eigenbase of the unperturbed Hamiltonian. The $\pi/2$ pulse applied to the 1-2 transition

![Diagram](attachment://fig1.png)

FIG. 1. Schematic representation of the NMR interferometry experiment to demonstrate the Aharonov-Anandan (AA) phase. The 2-3 two-level system (TLS) undergoes a circuit $C$ in projective Hilbert space and the phase is determined by means of its effect on the echo produced by the coherent superposition of levels 1 and 2.

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produces a coherent superposition of the two states whose phase serves as the reference for the subsequent measurement of the geometrical phase. The π pulse refocusses the 1-2 superposition as an echo\(^{13}\) of transverse magnetization. This 1-2 echo corresponds to the element \(\rho_{prz}\) of the density matrix,\(^{31}\)

\[
\rho_{\text{echo}} = \begin{pmatrix}
    \rho_{11} & \rho_{12} & 0 \\
    \rho_{21} & \rho_{22} & 0 \\
    0 & \rho_{23} & 0 \\
\end{pmatrix},
\]

and can be observed by means of a phase-sensitive detector at frequency \(\omega_{12}\). The submatrix \(\rho_{23}\) is proportional to \(1 + \mathbf{p}^2 3 \cdot \sigma_z\), the projected density matrix for the 2-3 TLS, where \(\mathbf{p}^2 3\) is the TLS polarization vector (initially along \(z\)) and \(\sigma_z\) is the vector \((\sigma_x, \sigma_y, \sigma_z)\) of Pauli matrices. Imagine now that \(\mathbf{p}^2 3\) is made to undergo a circuit, that is \(\mathbf{p}^2 3 \rightarrow c \mathbf{p}^2 3\) (and therefore \(\rho_{23} \rightarrow c \rho_{23}\)), by means of a perturbation applied selectively to the 2-3 transition. The phase factor acquired by the quantum states does not affect \(\rho_{23}\) and is therefore unobservable in the TLS.\(^{10}\) However, the phase factor acquired by state 2 does manifest itself in the phase of the 1-2 echo in the element \(\rho_{12} (c \rho_{12} \rightarrow \mathbb{I} \mathbb{I} + \beta(C)) \rho_{12}\) and is therefore detected by the phase-sensitive detector at frequency \(\omega_{12}\).

For the fictitious spin-\(\frac{1}{2}\) TLS, projective Hilbert space corresponds to a two-sphere and the geometrical phase becomes

\[
\beta(C) = m \Omega(C) = \pm \frac{1}{2} \Omega(C),
\]

where \(m\) is the magnetic quantum number and \(\Omega(C)\) is the solid angle subtended by the circuit \(C\) at the origin. We add that the π pulse and the echo in the 1-2 transition are not necessary in principle since the phases could be detected in the coherent signal following the π/2 pulse. In practice, however, the echo is a convenient experimental means of compensating for any extraneous dephasing due to nonuniform magnetic fields and other inhomogeneous broadening mechanisms. We also mention that \(\mathbf{p}^2 3\) need not begin and end along \(z\); it is necessary only that it go through a circuit.

Experiments were performed on the spin-1 manifold of two proton spins-\(\frac{1}{2}\) coupled by magnetic dipolar interactions in the molecule \(\text{CH}_2\text{Cl}_2\) oriented in a nematic liquid-crystal solvent.\(^{14}\) The static magnetic field was 8.4 T and the rotating magnetic fields at frequencies \(\omega_{12} = 362.023524\) MHz and \(\omega_{23} = 362.019675\) MHz had amplitudes of 1.7 and 1.3 \(\mu\text{T}\), respectively. The system was allowed to reach thermal equilibrium in the magnet and three types of circuits with a range of solid angles \(\Omega(C)\) were implemented for the TLS as shown in Fig. 2, by the application of time-dependent phase-shifted magnetic fields near \(\omega_{23}\).\(^{15}\) The cone circuits were induced by a magnetic field which was, in the rotating frame, tilted at an angle \(\theta\) with respect to the \(z\) axis. For the spherical triangles and the slices, rectangular magnetic field pulses were applied perpendicular to the polarization vector. For the spherical triangles the pulses were a π/2 pulse along \((0,1,0)\), a π pulse along \((0,0,1)\), and finally a π/2 pulse along \((\sin \theta, -\cos \theta, 0)\). Similarly, the slices were generated with a π pulse along \((0,1,0)\) followed by another π pulse along \((\sin \theta, -\cos \theta, 0)\). The geometrical phase was determined by measurement of the phase relative to a reference phase \(\gamma_d\) determined by pure dynamical evolution. For the spherical triangles

FIG. 2. Three types of circuits experienced by the polarization \(\mathbf{p}(\mathbf{p}^2 3\) in the text) for the TLS of Fig. 1 in the 2-3 frame of reference. The solid angles are \(\Omega(C) = 2\pi (1 - \cos \theta)\) for the cone, \(\Omega(C) = \theta\) for the triangle, and \(\Omega(C) = 2\theta\) for the slice.

FIG. 3. Oscilloscope traces of the 1-2 echoes detected in the two orthogonal \((x,y)\) channels of a phase-sensitive detector at frequency \(\omega_{12}\). In this case, the 2-3 TLS undergoes the slice circuits of Fig. 2, with solid angles \(\Omega(C)\) equal to 0, \(\pi/2, \pi,\) and \(2\pi\).
and the slice circuits the dynamical phase vanished, since in our experiment the applied field was always orthogonal to $p^{2-3}$, generating parallel transport. For the cone, the dynamical phase was determined via a reference experiment that included only the component of the field parallel to $p^{2-3}$.

Figure 3 shows some examples of the echoes observed, exhibiting the phase shifts induced by evolution around the slice circuits of Fig. 2, with various solid angles. The geometrical phase is given by $eta(C) = \arctan(S_y/S_x)$, where $S_x$ and $S_y$ are the integrated amplitudes of the signals in the two detector channels. Figure 4 shows a plot of $\beta(C)$ versus solid angle for all three circuits of Fig. 2. It is clear that the geometrical quantum phase is proportional to the solid angle subtended by the circuit. Our results illustrate the invariance of the AA phase to details of the circuit geometry and to the (perhaps nonadiabatic and noncyclic) Hamiltonian responsible for generating the circuit.

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