Brief Reports

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Non-Abelian effects in a quadrupole system rotating around two axes

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The ^{35}Cl nuclear quadrupole resonance spectrum of a single crystal of sodium chlorate [R. Tycko, Phys. Rev. Lett. **58**, 2281 (1987)] rotating about two axes reflects a non-Abelian gauge potential. This gauge potential is an example of Wilczek and Zee's [Phys. Rev. Lett. **52**, 2111 (1984)] generalization of Berry's phase to the adiabatic transport of degenerate states.

A large body of work beginning with the contributions of Berry and Simon[1] has demonstrated how geometric effects may arise in systems undergoing cyclic evolution.[2] In the simple and well-studied case of nondegenerate eigenstates adiabatically following a slowly varying Hamiltonian, the effect takes the form of a geometric phase, Berry's phase, acquired during evolution. This phase may be interpreted as being generated by a U(1) gauge potential. Experimental studies of Berry's phase have been performed in optics,[3] with neutron beams,[4] and in magnetic resonance.[5,6] Other invariance groups for the gauge potential are of course possible. For nondegenerate real wave functions, for example, only a Z_2 subgroup of U(1) is observed. This is relevant to molecules subject to Jahn-Teller distortions.[7] Following Berry and Simon's work, Wilczek and Zee[8] showed that for *n degenerate* levels the relevant gauge group is U(n). Such systems may exhibit much richer structure than the Abelian case, since eigenstates can exchange population in addition to acquiring phases. Examples that have been discussed include λ doubling in diatomic molecules[9] and isolated atoms in external fields.[10]

A particle with spin **I** in a slowly moving magnetic field **B**, represented by the Hamiltonian **B·I**, forms the standard example of the Abelian Berry phase. In order to study the non-Abelian generalization it is necessary to implement a Hamiltonian whose degeneracy structure is invariant with respect to changes in the parameters. A particularly appealing system with these characteristics is the quadrupole Hamiltonian $H = \mathbf{I} \cdot \mathbf{Q} \cdot \mathbf{I}$, for which the theory has been studied in detail by Avron *et al.*[11] Here **I** is taken to be half-odd-integral; time-reversal symmetry thereby ensures that the energy levels occur in degenerate pairs, with the degeneracy being maintained while the

five parameters of the real, symmetric, traceless 3×3 quadrupole matrix **Q** are varied. One convenient realization of this bilinear Hamiltonian arises from the interaction of a nuclear quadrupole moment with an electric field gradient (EFG), a situation which is routinely studied in condensed matter by nuclear quadrupole resonance (NQR) spectroscopy. The five parameters in this case are the strength and asymmetry of the interaction (which depend on the nucleus and the crystal structure), and three Euler angles, describing the orientation of the EFG tensor principal axis system with respect to a laboratory fixed frame of reference.[12] In fact, Tycko[6] studied just such a system, ^{35}Cl (spin $\frac{3}{2}$) in an oriented crystal of sodium chlorate (Fig. 1). The EFG tensor in this system has cylindrical symmetry, and thus two parameters (two of the Euler angles) can easily be varied by simply reorienting the crystal in the laboratory. In Tycko's experiment
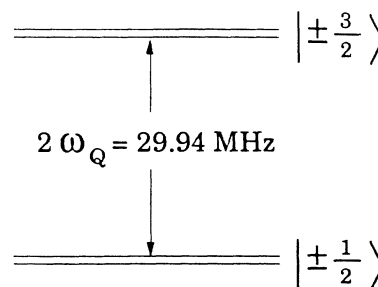

$$2\,\omega_Q = 29.94 \text{ MHz}$$

$$\left| \pm \frac{3}{2} \right\rangle$$

$$\left| \pm \frac{1}{2} \right\rangle$$

FIG. 1. Spin states of ^{35}Cl in sodium chlorate. Because this is a fermionic system with time-reversal symmetry (spin $\frac{3}{2}$) the levels appear in doublets, even as parameters in the Hamiltonian are varied. The quadrupole splitting is 29.94 MHz.

this was accomplished by rotating the crystal about a single axis, thus varying one angle and exploiting a one-parameter subgroup of the full non-Abelian structure. As pointed out by Tycko[6] and elaborated by Zee,[13] this gives a geometric effect which can be interpreted as a Berry phase.

In order to expose the full non-Abelian gauge structure it is necessary to use paths in parameter space for which both angles vary. We implement such a path by means of a double rotor,[14] in which a sample holder spins freely on an air bearing and is contained in a larger spinner, which itself spins about a different axis from the sample holder (see Fig. 2). We excite and detect the ^{35}Cl resonance by means of a radio-frequency coil around the double rotor; the symmetry axis of the coil coincides with the axis of rotation of the large spinner. A nonzero magnetization is prepared with a radio-frequency pulse and its time evolution is monitored as the crystal undergoes the double rotor trajectory. Our system is described by the following Hamiltonian:

$$H = \omega_Q e^{-i\varphi(t)I_z} e^{-i\theta(t)I_y} I_z^2 e^{i\theta(t)I_y} e^{i\varphi(t)I_z} , \qquad (1)$$

where ω_Q is the strength of the quadrupolar interaction and I_z is the z component of the nuclear spin operator in the EFG principal axis system. The angles $\varphi(t)$ and $\theta(t)$ describe the orientation of the crystal in the laboratory frame. Zee has shown[13] that the evolution of this system
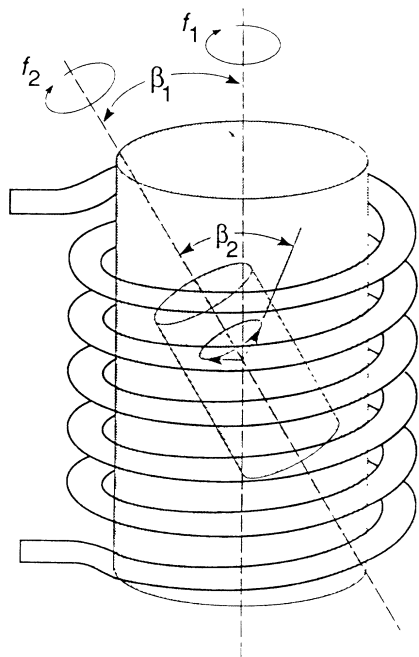

FIG. 2. Schematic diagram of the double rotor. An oriented single crystal of sodium chlorate ($NaClO_3$) is placed in the sample holder, which spins on an air bearing at a frequency f_2. The z axes of the ^{35}Cl EFG tensors (four per unit cell) make an angle $\beta_2 = 54.7°$ with the axis of rotation. The sample holder is contained in a large spinner, which rotates independently at a frequency f_1. The axes of rotation of the two spinners make an angle $\beta_1 = 30.6°$. A coil concentric with the large spinner is used to excite and detect the ^{35}Cl quadrupole resonance.

is governed by a gauge potential A, which is expressed in the I_z basis by

$$A_{\pm 1/2,\varphi} = \left[(-1 + \cos\theta)\frac{\sigma_3}{2} \right.$$
$$\left. - (I + 1/2)\sin\theta \left[\cos\varphi\frac{\sigma_1}{2} - \sin\varphi\frac{\sigma_2}{2} \right] \right] , \qquad (2)$$

$$A_{\pm 1/2,\theta} = (I + 1/2) \left[\cos\varphi\frac{\sigma_2}{2} + \sin\varphi\frac{\sigma_1}{2} \right] \qquad (3)$$

for the $m = \pm 1/2$ pair, using Pauli matrices, and for the $m \neq \pm 1/2$ pairs by

$$A_{mm',\varphi} = m(-1 + \cos\theta)\delta_{mm'} , \qquad (4)$$

$$A_{mm',\theta} = 0 . \qquad (5)$$

Off-diagonal matrix elements between nondegenerate basis states are neglected because of the adiabatic condition; the selection rules of the angular momentum operators further restrict the off-diagonal couplings to the $m = \pm 1/2$ pair of states.

The geometric part of the evolution is expressed in terms of A as

$$U(t) = \mathcal{P}\exp\left[-\int_C [A_\theta(t)d\theta(t) + A_\varphi(t)d\varphi(t)] \right] , \qquad (6)$$

where C denotes the path in the parameter space θ,φ. The path-ordering operator $\mathcal{P}$ is necessary, in general, because the components of A do not commute with themselves at different times. In the case where θ is constant there exists a single transformation, valid at all times, which diagonalizes A. In this case each eigenvector is multiplied by a diagonal element of U, and hence only a U(1) subgroup of U(2) is relevant.[6] Evolution of the density operator factors into a dynamic and a geometric part as

$$\rho(t) = U(t)\exp(-i\omega_Q I_z^2 t)\rho(0)\exp(i\omega_Q I_z^2 t)U^\dagger(t) , \qquad (7)$$

where under conditions of high temperature ($\hbar\omega_Q \ll kT$) the initial density operator $\rho(0)$ is obtained from a state of thermal equilibrium[15] and path ordering is implied in the time evolution. One sees that the nontrivial part of the evolution can be expressed in purely geometric terms. The observable in our experiment is the component of magnetization along the coil axis; the signal detected is then proportional to

$$\text{Tr}[I_z^{\text{lab}}\rho(t)] = \text{Tr}\{[I_z\cos\theta(t) - I_x\sin\theta(t)]\rho(t)\} . \qquad (8)$$

Note that an additional dependence on $\theta(t)$ is introduced by detecting the signal in the laboratory frame and not in the EFG principal axis system.

Figure 3 shows various trajectories of the ^{35}Cl EFG tensor z axis, which may be realized by different combinations of the rotor speeds and the angle between the rotors. Paths (a) and (b) correspond to rotations around a single axis and give Berry's phases, whereas (c) and (d) experience the full non-Abelian structure. A path subtending a net zero solid angle (for example, a figure eight) coupled with stroboscopic observation (once per cycle)
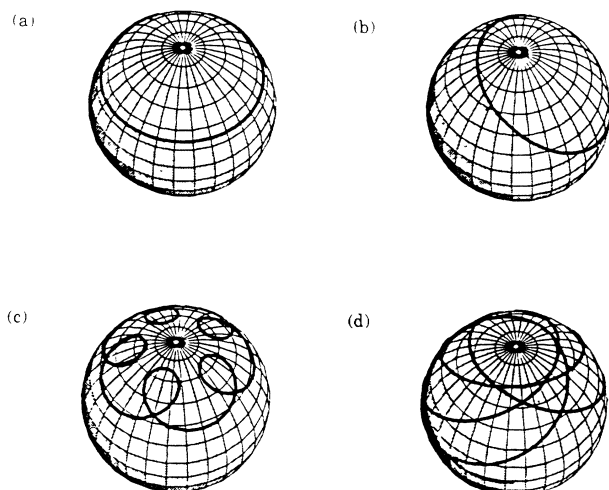
FIG. 3. Different possible paths followed by the z axis of the ^{35}Cl EFG tensor in the double rotor. For path (a), $\beta_1 = 0°$, $\beta_2 = 54.7°$, and $f_1/f_2 = 0$. This corresponds to the single-axis rotation of Tycko's experiment (Ref. 6), where φ varies, but θ is constant. For path (b), $\beta_1 = 30.6°$, $\beta_2 = 54.7°$, and $f_1/f_2 = 0$. This is still a single-axis cone, but tilted with respect to the coil axis. Both θ and φ in this case are time dependent, but there exists a basis, valid at all times, in which A is diagonal (Ref. 13) and thus commutes with itself at different times. Paths (c) and (d) are double rotation trajectories, with different angles and speed ratios. For (c), they are $\beta_1 = 34.4°$, $\beta_2 = 17.2°$, and $f_1/f_2 = 0.20$; for (d) they are $\beta_1 = 30.6°$, $\beta_2 = 54.7°$, and $f_1/f_2 = 0.18$. Path (d) represents the actual path of the experiment. Paths (c) and (d) show non-Abelian effects, since there exists no global transformation that diagonalizes A.

FIG. 4. Comparison of simulations and experiment. (a) shows the ^{35}Cl NQR spectrum of a static crystal of $NaClO_3$. (b) shows the same sample under double rotation with the trajectory of Fig. 3(d). This result is simulated in (c) using the parameters $\beta_1 = 30.6°$, $\beta_2 = 54.7°$, $f_1 = 0.36$ kHz, and $f_2 = 2.0$ kHz, and Gaussian broadening of full width at half-maximum, 1.0 kHz.

would naturally factor out any Abelian geometric phases, but is difficult to realize experimentally.

The ^{35}Cl NQR spectrum of $NaClO_3$, in the absence of sample rotation, consists of a single line at 29.94 MHz, with full width at half maximum of 1.0 kHz [Fig. 4(a)]. Under double rotation this spectrum breaks up into five resolved lines [Fig. 4(b)], which are simulated [Fig. 4(c)] by the geometric treatment of Eq. (7). For the simulation, the rotation frequencies of the two rotors were measured independently by means of a strip of piezoelectric foil. Rotation frequencies of 360 Hz for the large spinner and 2020 Hz for the sample holder were obtained. This shows that we are indeed in the adiabatic regime, as both frequencies are much lower than the transition frequency of 29.94 MHz. In contrast to the case of rotation about a single axis, simulation of our spectrum requires path-ordered integration of the full non-Abelian gauge potential [Eqs. (2)–(5)]; this integration cannot be simplified to a solid angle formula of the type which characterizes the (Abelian) Berry phase. Thus the spectrum is consistent with the non-Abelian geometric character of the double rotating quadrupole system.
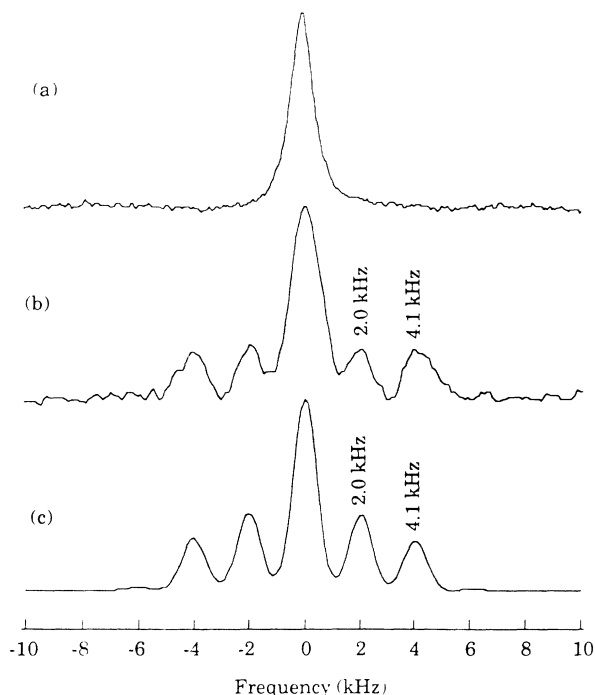
This work is a preliminary study of the consequences

of the Wilczek-Zee non-Abelian geometric phase in a quadrupole system. A complete experiment would involve transporting a degenerate pair of eigenstates along two *different, closed* paths, and showing that the resulting phase factors are elements of U(2) and do not commute with each other. Experimental efforts along these lines are currently under way in our laboratory. We close by noting that the mathematical structure of the system we have studied is that of a classical instanton.[11] To study this analogy in more detail, it would be necessary to vary all parameters in the quadrupole. It would also be interesting to extend the experiments to more general (nonadiabatic and perhaps nonunitary) evolutions of degenerate systems.[16]

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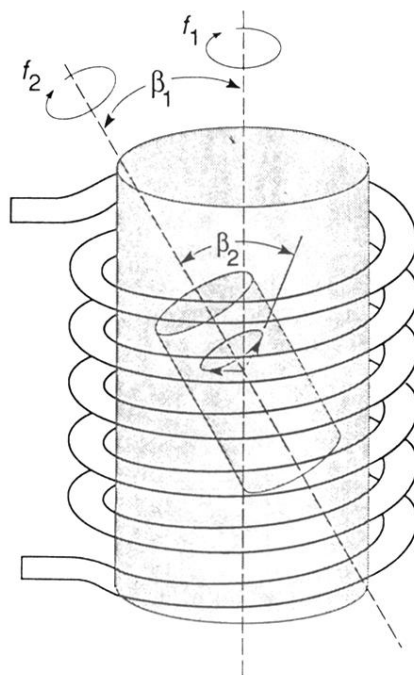[16]J. Anandan, Phys. Lett. A **133**, 171 (1988); J. Anandan and A. Pines, *ibid.* **141**, 335 (1989).

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