Non-Abelian Geometric Phase and Long-Range Atomic Forces

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Using fully quantal methods, we investigate various manifestations of the geometric phase in bound and free diatom systems. Adiabaticity and degeneracy are not invoked in the discussion.

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It is generally recognized that the fundamental constituents of matter interact through the exchange of gauge-field quanta. Recently, the concept of a gauge field, or a nonintegrable phase factor,[1,2] has found application in systems other than fundamental ones.[3-9] In an analysis of the quantum adiabatic theorem, Berry[4] illustrated how a quantum state can, under certain conditions, acquire a nonintegrable phase factor during an adiabatic evolution. This phase factor, now commonly referred to as Berry's geometric phase, has since been observed in diverse physical systems.[5] Wilczek and Zee[6] generalized Berry's analysis to include degenerate states undergoing adiabatic evolution. They showed that such systems can exhibit a non-Abelian geometric phase and suggested, using quasiclassical arguments, that geometric phases alter energy splittings in simple quantum systems. Diatomic systems exhibiting non-Abelian gauge-field structure have been explored previously;[7-9] in this Letter we illustrate how gauge fields, or geometric phases,[10] manifest as observable effects in both bound and free diatom systems. We show that, in addition to altering energy splittings in bound systems, geometric phases induce transitions in levels separated by a finite-energy gap. An example is given where the non-Abelian gauge field couples *nondegenerate* electronic levels in a diatom. This gauge-field coupling gives rise to an observable effect (Λ doubling). We show that when the diatom is "pulled apart" the non-Abelian geometric phase manifests as a long-range atomic force. Below we present final results; the details will be published elsewhere.[11] Atomic units are used throughout.

The Hamiltonian for a diatomic system (after factoring out the c.m. motion) is given by

$$H=-\frac{1}{2\mu}\nabla_R^2+H_{\rm ad}(\mathbf{R},\mathbf{r}), \tag{1}$$

where $\mathbf{R}$ is the internuclear radius vector between the two atomic centers, μ is the reduced mass of the system, and all coordinates are taken with respect to a space-fixed frame. $H_{\rm ad}(\mathbf{R},\mathbf{r})$ is the adiabatic Hamiltonian; it includes the kinetic energy of the electrons and all electrostatic interactions. Its eigenstates, $\phi_n(\mathbf{R},\mathbf{r})$, are complete with respect to the electronic or "fast" coordinates for all values of the parameter $\mathbf{R}$, and they form a representation for the system. The total wave function can be expressed by

$$\Psi(\mathbf{R},\mathbf{r},t)=\sum_n \phi_n(\mathbf{R},\mathbf{r})F_n(\mathbf{R},t), \tag{2}$$

where $F_n(\mathbf{R},t)$ are expansion coefficients and are effective amplitudes for the nuclear motion. In applications only a finite, incomplete, set $\{M\}$ of adiabatic states is used. This approximation is sometimes called the Born-Huang approximation, or the method of perturbed stationary states.[12] It is particularly well suited for systems where the distinction between "slow" and "fast" degrees of freedom is unambiguous. Treating the nuclear amplitudes as variational parameters and Ψ as a variational wave function we get[9,11]

$$-\frac{1}{2\mu}[\nabla_R\tilde{I}-i\tilde{\mathbf{A}}(\mathbf{R})]^2\tilde{F}+\tilde{V}(\mathbf{R})\tilde{F}=i\frac{\partial\tilde{F}}{\partial t}, \tag{3}$$

where $\tilde{F}=\{F_n\}$ is a column vector and $\tilde{\mathbf{A}},\tilde{V}$ are matrices. Their components are

$$\mathbf{A}_{ij}(\mathbf{R})=i\langle\phi_i|U(\hat{\mathbf{R}})\nabla_R U^{-1}(\hat{\mathbf{R}})|\phi_j\rangle, \tag{4a}$$

$$V_{ij}(\mathbf{R})=\langle\phi_i|H_{\rm BO}|\phi_j\rangle+\frac{1}{2\mu}\sum_{k\neq ij}\mathbf{A}_{ik}\cdot\mathbf{A}_{kj}, \tag{4b}$$

where $H_{\rm BO}\equiv H_{\rm ad}(\mathbf{r},|\mathbf{R}|\hat{\mathbf{z}})$, $\hat{\mathbf{z}}$ is the unit vector along the z axis of the space-fixed frame, and $|\phi_m\rangle$ are the eigenstates of $H_{\rm BO}$. We call them Born-Oppenheimer states. They are specified, among other quantum numbers, by the value of the electronic angular momentum, Λ (throughout, we assume integer values for Λ, the more general case is discussed in Ref. 11), along the $\hat{\mathbf{z}}$ axis. The adiabatic states are related to the BO states by a rotation $U(\hat{\mathbf{R}})$ so that $\phi_n(\mathbf{R},\mathbf{r})=U^{-1}(\hat{\mathbf{R}})|\phi_n\rangle$, and $U(\hat{\mathbf{R}})H_{\rm ad}U^{-1}(\hat{\mathbf{R}})=H_{\rm BO}$. Equation (3) possesses unitary gauge symmetry [U(M) group], and $\tilde{\mathbf{A}},\tilde{V}$ are the $3+1$ components of the gauge field.[9] For a complete set the second term in (4b) and the spatial components of the field strength tensor[9] vanish identically.[11] In this case one can always find a gauge (pure gauge) where the vector potential also vanishes. However, for an incomplete set, $\{M\}$, nontrivial gauge fields are induced; i.e., the Wilson-loop integral is not unity for an arbitrary closed circuit in $\mathbf{R}$ space. We illustrate this below.

In the one-state approximation (Born-Oppenheimer approximation) (3) becomes

$$-\frac{1}{2\mu}[\nabla_R-i\mathbf{A}(\mathbf{R})]^2F(\mathbf{R},t)+V(R)F(\mathbf{R},t)=i\frac{\partial F(\mathbf{R},t)}{\partial t} \tag{5}$$

where $F(\mathbf{R})$ is the scalar wave function. $V(R)$ is given by

$$V(R)=\epsilon(R)+\frac{\Omega^2(R)}{2\mu R^2}+\frac{B(R)}{2\mu}; \tag{6}$$

$\epsilon(R)$ is the Born-Oppenheimer eigenvalue, $\Omega^2(R) = \langle\phi|L_x^2+L_y^2|\phi\rangle$, where L_x, L_y are the angular momentum operators of the electrons, and $B(R)$ is the radial component of the nonadiabatic correction

$$B(R) = \sum_{n'} |\langle\phi|d/dR|\phi_{n'}\rangle|^2 ,$$

where the sum is over all states excluding $|\phi\rangle$. $B(R)/2\mu$ is usually small and we absorb it into the definition of $\epsilon(R)$. We assume that $\epsilon(R)$ describes a potential curve containing a deep attractive well. Bound vibrational states exist for such a potential and we can approximate the well by a harmonic-oscillator potential near, R^*, the equilibrium value of the internuclear separation. To calculate the rotational structure of the bound diatom we evaluate the vector potential (4a). In order to obtain a nonsingular vector potential in the fixed coordinate frame we divide the region into two overlapping hemispheres (sections) [13] R_a, R_b corresponding to the northern and southern polar regions, respectively. We choose

$$U = \exp(\mp i\phi L_z)\exp(i\theta L_y)\exp(i\phi L_z) ,$$

where $\mp$ identifies regions R_a and R_b, respectively, and θ, ϕ are the polar and azimuthal angles of $\mathbf{R}$ in the space-fixed coordinate system. Using (4a) we get

$$\mathbf{A} = i\langle\phi_\Lambda|U(\hat{\mathbf{R}})\nabla_R U^{-1}(\hat{\mathbf{R}})|\phi_\Lambda\rangle = \Lambda\hat{\boldsymbol{\phi}}\frac{\cos\theta\mp 1}{\sin\theta} . \quad (7)$$

The vector potential (7) is that of a "magnetic" monopole with charge Λ located at the origin. In the field of such a monopole the mechanical angular momentum of the rotating nuclei is no longer conserved, but the total angular momentum

$$\mathbf{J} = \mathbf{N} + \mathbf{\Sigma} ,$$

where $\mathbf{N} = \mathbf{R}\times -i\nabla_R$ and

$$\Sigma_x = \Lambda\cos\phi\frac{1\mp\cos\theta}{\sin\theta} ,$$
$$\Sigma_y = \Lambda\sin\phi\frac{1\mp\cos\theta}{\sin\theta} , \quad \Sigma_z = \pm\Lambda , \quad (8)$$

is conserved. Because $\mathbf{J}$ is a constant of the motion the nuclear amplitude can be expanded in terms of the eigensections, $Y^{\Lambda}_{JM}(\theta\phi)$, of $\mathbf{J}^2$ and J_z. These eigensections are the monopole harmonics described in detail by Wu and Yang.[13] To obtain a stationary state of (5) with energy E we let

$$F(\mathbf{R}) = \sum_{JM} Y^{\Lambda}_{JM}(\theta\phi)\frac{F^{\Lambda}_{JM}(R)}{R} , \quad (9a)$$

where

$$\mathbf{J}^2 Y^{\Lambda}_{JM} = J(J+1)Y^{\Lambda}_{JM} , \quad J_z Y^{\Lambda}_{JM} = MY^{\Lambda}_{JM} , \quad (9b)$$

to get

$$-\frac{1}{2\mu}\frac{d^2F^{\Lambda}_{JM}}{dR^2} + \left[\frac{J(J+1)-\Lambda^2+\Omega^2(R)}{2\mu R^2}\right]F^{\Lambda}_{MJ} + \epsilon(R)F^{\Lambda}_{MJ} = EF^{\Lambda}_{MJ} . \quad (9c)$$

Using (9c), and the clamped-nuclei approximation, we obtain an expression for the rotational eigenvalues ϵ^{Λ}_{JM}. For a given vibrational level, v, we get

$$\epsilon^{\Lambda}_{JM} = B\left[J(J+1)-\Lambda^2\left(1-\frac{1}{I_C}\right)\right] ,$$
$$B \equiv \frac{1}{2\mu R^{*2}} , \quad I_C = \frac{\Lambda^2}{\Omega^2(R^*)} , \quad (10)$$

where the allowed values of J are $|\Lambda|, |\Lambda|+1, \ldots$. These are the rotational eigenvalues of a symmetric top.[14] In the formalism presented here the coupling of the nuclear and electronic angular momenta, that give rise to the rotational structure (10), is mediated by a "magnetic" monopole gauge field. In addition to the monopole contribution to spectrum (10), there is also a contribution resulting from an induced scalar potential given by the second term on the right-hand side of Eq. (4b). Spectrum (10) is an observable consequence of the presence of the background Abelian gauge potentials coupling to the rotating nuclei.

We assume that $\epsilon(R)$ represents the potential curve of a π ($|\Lambda| = 1$) electronic state of a diatom that dissociates to an ion and degenerate p-state atom. At large internuclear separations this potential curve is degenerate with the potential curve of a Σ ($\Lambda = 0$) state. For high vibrational quantum numbers, v, and in the continuum region, the single-channel *Ansatz* is no longer adequate, and we must include the degenerate π and Σ states in *Ansatz* (2). Now the effective nuclear amplitude is expressed as a vector in an abstract "isospin" space,

$$\tilde{F}(\mathbf{R}) \equiv \begin{pmatrix} F_\Sigma(\mathbf{R}) \\ F_\pi(\mathbf{R}) \\ F_{-\pi}(\mathbf{R}) \end{pmatrix} , \quad (11)$$

where the subscripts are the isospin components and identify the amplitude for the system to be in the corresponding electronic state. The gauge potentials are operators in isospin space and are given by[11] (we use the notation of Ref. 9)

$$\tilde{A}_\theta = \kappa(R)[\tilde{\sigma}_y\cos\phi \mp \tilde{\sigma}_x\sin\phi] ,$$
$$\tilde{A}_\phi = \tilde{\sigma}_z(\cos\theta\mp 1) - \kappa(R)\sin\theta[\tilde{\sigma}_x\cos\phi \pm \tilde{\sigma}_y\sin\phi] , \quad (12a)$$

where

$$\tilde{\sigma}_x = \frac{1}{\sqrt{2}}\begin{pmatrix} 0 & 1 & 1 \\ 1 & 0 & 0 \\ 1 & 0 & 0 \end{pmatrix} , \quad \tilde{\sigma}_y = \frac{1}{\sqrt{2}}\begin{pmatrix} 0 & i & -i \\ -i & 0 & 0 \\ i & 0 & 0 \end{pmatrix} ,$$
$$\tilde{\sigma}_z = \begin{pmatrix} 0 & 0 & 0 \\ 0 & 1 & 0 \\ 0 & 0 & -1 \end{pmatrix} , \quad (12b)$$

are spin-1 matrices,

$$\kappa(R) \equiv \frac{1}{\sqrt{2}}|\langle\Sigma|L_x - iL_y|\pi\rangle| , \quad (12c)$$

and the $\mp$ notation refers to sections R_a and R_b, respectively. Using (12) we obtain the effective Hamiltonian for $\tilde{F}(\mathbf{R})$,

$$\tilde{H}=\tilde{H}_0+\kappa\tilde{H}_1 .$$

$$\tilde{H}_0=-\frac{\tilde{I}}{2\mu}\left\{\frac{1}{R^2}\frac{\partial}{\partial R}\left[R^2\frac{\partial}{\partial R}\right]\right\}$$

$$+\frac{\tilde{J}^2-\tilde{\sigma}_z^2+\tilde{\Omega}^2}{2\mu R^2}+\tilde{W}(R) ; \tag{13}$$

$\tilde{\mathbf{J}}=\mathbf{N}+\tilde{\mathbf{\Sigma}}$, where now $\tilde{\mathbf{\Sigma}}$ is given by (8) with Λ replaced by the $\tilde{\sigma}_z$ matrix, $\tilde{\Omega}$ is a diagonal matrix with entries $\Omega_{ii}=\langle\phi_i|L_x^2+L_y^2|\phi_i\rangle$, and $\tilde{W}(R)$ is diagonal with the entries $\epsilon_i(R)$ corresponding to the Born-Oppenheimer eigenvalue for the ith electronic state. The interaction term, $\tilde{H}_1$, is given by

$$\tilde{H}_1=\frac{\tilde{\sigma}_-\tilde{L}_--\tilde{\sigma}_+\tilde{L}_+}{\sqrt{2}\mu R^2} , \tag{14}$$

where $\tilde{\sigma}_+,\tilde{\sigma}_-$ are the raising and lowering operators in isospin space, and

$$\tilde{L}_+=e^{i\mp\phi}\left[\frac{\partial}{\partial\theta}-\frac{1}{\sin\theta}\left(i\frac{\partial}{\partial\phi}+(\cos\theta\mp1)\tilde{\sigma}_z\right)\right] ,$$

$$\tilde{L}_-=e^{i\pm\phi}\left[-\frac{\partial}{\partial\theta}-\frac{1}{\sin\theta}\left(i\frac{\partial}{\partial\phi}+(\cos\theta\mp1)\tilde{\sigma}_z\right)\right] . \tag{15}$$

We introduce isospin monopole harmonics, $\tilde{Y}^{\Lambda}_{JM}(\theta\phi)$, with the properties

$$\tilde{\mathbf{J}}^2\tilde{Y}^{\Lambda}_{JM}=J(J+1)\tilde{Y}^{\Lambda}_{JM} , \quad \tilde{J}_z\tilde{Y}^{\Lambda}_{JM}=M\tilde{Y}^{\Lambda}_{JM} ,$$

$$\tilde{\sigma}_z\tilde{Y}^{\Lambda}_{JM}=\Lambda\tilde{Y}^{\Lambda}_{JM} , \quad \tilde{\sigma}^2\tilde{Y}^{\Lambda}_{JM}=2\tilde{Y}^{\Lambda}_{JM} . \tag{16}$$

We also have[11]

$$\tilde{\sigma}_+\tilde{L}_+\tilde{Y}^{\Lambda}_{JM}=[(J-\Lambda)(1+J+\Lambda)]^{1/2}\tilde{Y}^{1+\Lambda}_{JM}$$

and

$$\tilde{\sigma}_-\tilde{L}_-\tilde{Y}^{\Lambda}_{JM}=-[(J+\Lambda)(1+J-\Lambda)]^{1/2}\tilde{Y}^{\Lambda-1}_{JM}$$

provided that the triangle relations for J, M, and Λ are satisfied. Because $\tilde{H}$ commutes with $\tilde{\mathbf{J}}^2$ and $\tilde{J}_z$,[11] we obtain an effective Hamiltonian for the radial nuclear motion by expanding

$$\tilde{F}(\mathbf{R})=\sum_{JM\Lambda}\frac{F^{\Lambda}_{JM}(R)}{R}\tilde{Y}^{\Lambda}_{JM}(\theta\phi)$$

to get

$$-\frac{1}{2\mu}\frac{d^2\tilde{F}_{JM}}{dR^2}+\left[\frac{J(J+1)\tilde{I}-\tilde{\sigma}_z^2+\tilde{\Omega}^2}{2\mu R^2}+\tilde{W}(R)-\frac{\kappa(R)[J(J+1)]^{1/2}}{\mu R^2}\tilde{\sigma}_x\right]\tilde{F}_{JM}=E\tilde{F}_{JM} , \tag{17}$$

where $\tilde{F}_{JM}(R)$ is now a radial isospin vector, its isospin components corresponding to the ones in definition (11). Using the clamped-nuclei approximation and neglecting the term proportional to $\kappa(R)$, we obtain the spectrum (10) for E well below the dissociation limit $\epsilon_\Sigma(\infty)$. Because of the degeneracy of the π electronic states the bound rovibrational states are doubly degenerate. When the term proportional to κ is included as a perturbation the degeneracy is removed in second order, and the levels are split by the amount

$$\Delta E=4B^2\kappa^2J(J+1)\sum_{v'}\frac{|\langle JM,|\Lambda|=1,v|v',\Lambda=0,JM\rangle|^2}{\epsilon_{v'J\Lambda=0}-\epsilon_{vJ|\Lambda|=1}} \tag{18}$$

where $|v\Lambda JM\rangle$ is a vibrational eigenstate of H_0 and $\epsilon_{vJ\Lambda}$ its eigenvalue. This is the familiar example of Λ doubling.[14] In the formulation presented here it is a manifestation of a non-Abelian geometric phase; i.e. H_1 arises from the coupling of the non-Abelian gauge field (12a) with the nuclear motion. The non-Abelian nature of the $\pm\pi$-Σ state coupling is apparent when the field strength tensor component,[9] $\tilde{F}_{\theta\phi}$, is evaluated:

$$\tilde{F}_{\theta\phi}=[\kappa^2(R)-1]\sin\theta\sigma_z . \tag{19}$$

The electrostatic interactions of the electrons with the nuclei break the spherical symmetry of the electronic states, and the κ parameter (12c) is not unity. Thus, for finite R, the field strength tensor (19) does not vanish and, because κ is not quantized, gauge field (12a) is "truly non-Abelian."[7] In the asymptotic limit $R\rightarrow\infty$ the electronic states correlate to atomic states centered at one of the nuclei. The triply degenerate atomic states form a representation of the rotation group and, in this limit, the κ parameter tends to unity and $\tilde{F}_{\theta\phi}$ vanishes. Therefore, the field strength (19) describes a non-Abelian, monopolelike, object and κ acts as a monopole "charge" screening parameter. For the nondegenerate triplet discussed above we obtain a truly non-Abelian monopole for integer values of Λ ($|\Lambda|=1$).

We choose a new gauge so that

$$\tilde{\mathbf{A}}'=\tilde{U}^{-1}\tilde{\mathbf{A}}\tilde{U}+i\tilde{U}^{-1}\nabla_R\tilde{U} ,$$

where

$$\tilde{U}=\exp(\mp i\phi\tilde{\sigma}_z)\exp(i\theta\tilde{\sigma}_y)\exp(i\phi\tilde{\sigma}_z) ,$$

and $\mp$ refer to the sections R_a and R_b, respectively. In this gauge the vector potential is[11]

$$\tilde{A}'_\theta=[\kappa(R)-1](\tilde{\sigma}_x\sin\phi+\tilde{\sigma}_y\cos\phi) , \quad \tilde{A}'_\phi=[1-\kappa(R)][-\tilde{\sigma}_z\sin^2\theta+\cos\theta\sin\theta(\tilde{\sigma}_x\cos\phi+\tilde{\sigma}_y\sin\phi)] . \tag{20}$$

We note that the components in (20) vanish as $R\rightarrow\infty$. In this gauge the conserved angular momentum is

$\tilde{\mathbf{J}}'=\tilde{U}^{-1}(\mathbf{N}+\tilde{\boldsymbol{\Sigma}})\tilde{U}=\mathbf{N}+\tilde{\boldsymbol{\sigma}}$, and the transformed Hamiltonian is

$$H'=-\frac{1}{2\mu}\left\{\frac{1}{R^2}\frac{\partial}{\partial R}\left[R^2\frac{\partial}{\partial R}\right]\right\}+\frac{\mathbf{N}^2}{2\mu R^2}+\frac{1-\kappa(R)}{\mu R^2}\tilde{\boldsymbol{\sigma}}\cdot\mathbf{N}+\frac{1-\kappa(R)}{2\mu R^2}(\tilde{\boldsymbol{\sigma}}\cdot\hat{\mathbf{R}})(\tilde{\boldsymbol{\sigma}}\cdot\hat{\mathbf{R}})+\frac{[1-\kappa(R)]^2}{\mu R^2}+H_{\text{atomic}}\,,$$

$$H_{\text{atomic}}=\frac{\epsilon_\Sigma(R)}{2}\tilde{\boldsymbol{\sigma}}\cdot\tilde{\boldsymbol{\sigma}}-\Delta(R)(\tilde{\boldsymbol{\sigma}}\cdot\hat{\mathbf{R}})(\tilde{\boldsymbol{\sigma}}\cdot\hat{\mathbf{R}})\,,\quad \Delta(R)\equiv\frac{\epsilon_\Sigma(R)-\epsilon_\pi(R)}{2}\,. \tag{21}$$

Expression (21) is valid for all R but, here, we use it to study the asymptotic limit of the ion-atom interaction. For large R, $\Delta(R)\sim Q/R^3$, where Q is proportional to the quadrupole moment of the atom.[12] Thus, in the asymptotic limit, H_{atomic} represents the well-known quadrupole interaction between an ion and atom.[12] However, we also get higher-order terms proportional to the non-Abelian parameter κ. Using perturbation theory we get the asymptotic expansion[11]

$$\kappa(R)\sim 1+\frac{C}{R^4}+\cdots\,, \tag{22}$$

where C is a constant and is on the order of an atomic dipole polarizability.[15]

Using (22) in (21) we obtain a spin-orbit-type potential

$$V\sim\frac{C}{\mu}\frac{\tilde{\boldsymbol{\sigma}}\cdot\mathbf{N}}{R^6} \tag{23}$$

valid at large internuclear separations. In addition to (23), we also get higher-order corrections to the quadrupole potential. The effects of this potential should be seen in the profiles of collision-broadened lines found in a gas at low temperatures. The above analysis is also valid for a treatment of Rydberg systems.[11] For a Rydberg atom with a p-state core, the Rydberg electron interacts with the core via a term analogous to (23) (where now $\mu=1$). In addition to the quadrupole electron-core interaction,[16] this interaction influences the rotational structure of the Rydberg levels.

The "spin-orbit" interaction (23) is a direct consequence of the coupling of the relative nuclear motion with gauge field (20). We can use the effective Hamiltonian (21) to analyze the slow collision of an ion with a p-state atom. At finite collision velocities, the dynamic phase[12] induces transitions among the sublevels of the atom. However, because of the gauge-field coupling (20) connecting the different sublevels, transition probabilities also depend on the geometric phase.[10]

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