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# Geometric (Berry) Phases in Neutron Molecular Spectroscopy

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# GEOMETRIC (BERRY) PHASES IN NEUTRON MOLECULAR SPECTROSCOPY

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## *Abstract*

A theory of neutron scattering by nuclei in a molecule, accompanied by an electronic transition, is formulated with attention to gauge potentials and geometric phases in the Born-Oppenheimer scheme. Non-degenerate and nearly degenerate electronic levels are considered. For nearly degenerate levels it is shown that, the cross-section is free of the singular structure which characterizes the corresponding gauge potential for the phase, and much larger than for well separated electronic states.

## 1. INTRODUCTION

The occurrence of non-trivial phase factors in wave functions of particles responding to a slow change in parameters was recognized in molecular science some time ago; see [1] and references therein. In this context, the generic model system is a few electrons and nuclei described by the Born-Oppenheimer scheme [2]. Even though the dynamics of this model is a well researched problem, covered to some extent in most texts on quantum mechanics, it displays subtle features which are today interpreted in terms of a geometric phase factor. An example of the latter in classical mechanics appears in transport of a vector around a circuit on a sphere; when parallel transported, and not twisted, the vector taken on an excursion round a closed circuit is turned through an angle, called the holonomy (or sometimes anholonomy). A spate of work during the past few years, by mathematicians, chemists and physicists, has shown that geometric phases are present in a wide variety of models. Much of the recent work has been motivated by an intimate relation between geometric phases and gauge (vector) potentials which are a key ingredient in preferred models of elementary particles. For one thing, it is simply intriguing to find gauge potentials in a model, such as a molecule described by the *B*-O scheme, when no such potentials are initially required to specify it.

Here, we formulate the theory of neutron scattering by a molecule, accompanied by an electronic transition, using a language that displays the rôle of gauge potentials and geometric phases. We will draw on earlier work [3], adding to it these recent developments and the behaviour of the cross-section in the vicinity of a level crossing where it is very large.

In §§2,3 we briefly survey, following Berry [4] and Aitchison [5], the occurrence of gauge potentials and geometric phases in a molecular system described by the *B*-O scheme. This material is deemed necessary to fully define notation and concepts, bearing in mind that treatment of the *B*-O scheme in standard texts is incomplete. The bulk of the paper is taken up with the calculation of the neutron cross-section for a molecule when scattering by the nuclei induces an electronic transition. This process does not occur within the standard *B*-O approximation, since it completely decouples nuclear and electronic degrees of freedom. The next approximation to the total molecular wave function, obtained by an expansion in terms of the electron-nucleon mass ratio, is given in §4 together with a compact expression for the appropriate matrix element. Features of the matrix element in the vicinity of a level crossing are discussed in §5 using a model discussed by Berry [4]. Neutron cross-sections for non-degenerate and nearly degenerate electronic energy surfaces are given in §§6,7, and §8 contains a short discussion.

## 2. GEOMETRIC PHASE FACTOR

Because the ratio of the electron and nuclear masses is very small, dynamical properties of a molecule can be calculated to a good approximation by treating the electrons as enslaved to the nuclear degrees of freedom. The latter are relatively very slow, with a fine spacing between energy levels, whereas electronic degrees of freedom are fast. In the **adiabatic approximation** the nuclear spatial coordinates, $\boldsymbol{R}$, are assumed to be constant. If $\mathcal{H}_{el}(\boldsymbol{R})$ is the sum of the electronic kinetic energy, and the potential and interaction energies of the electrons and nuclei, the stationary state Schrödinger equation for the electrons (coordinates $\boldsymbol{r}$) is simply,

$$\mathcal{H}_{el}(\boldsymbol{R})\varphi_j(\boldsymbol{r},\boldsymbol{R}) = E_j(\boldsymbol{R})\varphi_j(\boldsymbol{r},\boldsymbol{R}) \ . \tag{2.1}$$

The complete set of electronic eigenfunctions $\{\varphi_j\}$ and eigenvalues $\{E_j\}$ are labelled by an index $j$. These quantities depend on $\boldsymbol{R}$ as a parameter, whose actual time dependence is so slow that it does not induce transitions from one $E_j$ level to another.

However, the time evolution of $\boldsymbol{R} = \boldsymbol{R}(t)$ influences the phase of the electronic wave function, $\phi(t)$. The instantaneous eigenstates determined by (2.1) depend parametrically on the slow coordinate $\boldsymbol{R}(t)$, and in the following working we employ the notation $|j(\boldsymbol{R}(t))\rangle$ for these eigenstates. Adiabatically, a molecule in one of these states $|j(\boldsymbol{R}(t=0))\rangle$, say, will evolve with $\mathcal{H}_{el}(\boldsymbol{R}(t))$ and be in the state $|j(\boldsymbol{R}(t_o))\rangle$ at some later time $t_o$. The wave function $\phi(t_o)$ differs from $|j(\boldsymbol{R}(t_o))\rangle$ by a phase factor, part of which depends on the circuit made by $\boldsymbol{R}(t)$ on the excursion between $t = 0$ and $t = t_o$, and this part of the phase $\gamma_j$ cannot be written as a function just of the end points, i.e. it depends on the geometry of the path connecting them as well. In the formulation given below, due to Berry [4], the circuit is introduced as part of the argument, whereas in alternative formulations, using path-integral techniques, it naturally emerges. For the adiabatic approximation to apply, $t_o$ must be large.

Writing,

$$|\phi(t_o)\rangle = |j(\boldsymbol{R}(t_o))\rangle \, exp\left\{i\gamma_j(t_o) - i\int_0^{t_o} dt'\, E_j(\boldsymbol{R}(t'))\right\} \tag{2.2}$$

the time-dependent Schrödinger equation,

$$\mathcal{H}_{el}(\boldsymbol{R}(t))|\phi(t)\rangle = i(d/dt)|\phi(t)\rangle \ , \tag{2.3}$$

together with (2.2), yields the following equation for the geometric phase,

$$\gamma_j(t) = i\int_0^t dt' \left\langle j(\mathbf{R}(t'))\right|(d/dt')\left|j(\mathbf{R}(t'))\right\rangle = \int_0^t d\mathbf{R} \, . \, \mathbf{A}_j(\mathbf{R}(t)) \ , \tag{2.4}$$

where we have defined

$$\mathbf{A}_j(\mathbf{R}) = i\left\langle j(\mathbf{R})\right|\nabla_{\mathbf{R}} j(\mathbf{R})\rangle \equiv i\int d\mathbf{r} \, \varphi_j^*(\mathbf{r},\mathbf{R})\nabla_{\mathbf{R}}\varphi_j(\mathbf{r},\mathbf{R}) \tag{2.5}$$

for later convenience. The Berry phase $\gamma_j(t)$ is non-integrable, and cannot be written as a function of $\mathbf{R}$. In particular, it is not single-valued under continuation around a circuit, so $\gamma_j(t_o) \neq \gamma_j(0)$ for $\mathbf{R}(0) = \mathbf{R}(t_o)$ . Given that $|j(\mathbf{R})\rangle$ is normalizable, $\mathbf{A}_j(\mathbf{R})$ vanishes if $|j(\mathbf{R})\rangle$ is purely real, otherwise $\mathbf{A}_j(\mathbf{R})$ is real. A specific example is provided in §5.

The notation introduced in (2.5) is in line with other workers; it is intended to convey the concept that the geometric phase is related to a gauge (vector) potential, $\mathbf{A}_j$. Physical effects of a gauge potential depend only on the gauge (magnetic) field proportional to curl $\mathbf{A}_j$. Hence, we are led to expect that the equation of motion for the nuclear coordinates contains such a gauge field, and this is indeed revealed in the Born-Oppenheimer scheme. By and large though, it is at first sight a surprise to find a gauge field in a manifestly non-magnetic, and seemingly innocuous, physical model.

## 3. BORN-OPPENHEIMER SCHEME

The equation of motion for the (slow) nuclear coordinates is obtained, in the Born-Oppenheimer scheme, by integrating out the (fast) electronic degrees of freedom [2]. As mentioned in the previous section, it contains non-trivial structure, left behind by the adiabatic decoupling of the electronic degrees of freedom, which might be viewed as a reaction of the light particles on the heavy particles.

In the $B$-0 scheme, the total wave function $\Psi(\mathbf{r},\mathbf{R})$ is taken to be a single product of a nuclear wave function, $\chi(\mathbf{R})$, to be determined, and an electronic wave function, $\varphi(\mathbf{r},\mathbf{R})$, which is a solution of the Schrödinger equation (2.1); so,

$$\Psi_j(\mathbf{r},\mathbf{R}) = \chi_j(\mathbf{R}) \, \varphi_j(\mathbf{r},\mathbf{R}) \ . \tag{3.1}$$

To obtain an equation for $\chi_j$ apply to $\Psi$ the total Hamiltonian that describes the molecule, which is the sum of $\mathcal{H}_{el}$ and the nuclear kinetic energy. After multiplying the Schrödinger equation by $\varphi^*_{j'}(\boldsymbol{r},\boldsymbol{R})$ and integrating over the electronic coordinates $\boldsymbol{r}$ one finds for $\{\chi_j\}$ the set of equations,

$$(1/2M)\sum_{jj'}\left(\delta_{j_oj}\boldsymbol{P}-\mathcal{A}_{j_oj}\right)\cdot\left(\delta_{jj'}\boldsymbol{P}-\mathcal{A}_{jj'}\right)\chi_{j'}(\boldsymbol{R})=\left(E-E_{j_o}(\boldsymbol{R})\right)\chi_{j_o}(\boldsymbol{R})\ . \tag{3.2}$$

In this equation, $\boldsymbol{P}$ is the momentum conjugate to $\boldsymbol{R}$, $E$ is the total energy, $M$ the nuclear mass, and

$$\mathcal{A}_{jj'}=i\langle j(\boldsymbol{R})|\nabla_{\boldsymbol{R}}j'(\boldsymbol{R})\rangle\ . \tag{3.3}$$

The standard B-0 approximation for $\chi$ is obtained from (3.2) by (a) use of real wave functions to give $\mathcal{A}_{jj}=0$ and (b) neglect of off-diagonal matrix elements. (For the special case when $\{\varphi_j\}$ are real the only possible phase factors associated with a circuit are ±1.) If we relax (a) and keep (b) the approximate equation for the nuclear wave function is ($\mathcal{A}_{jj}=\boldsymbol{A}_j$)

$$(1/2M)\left(\boldsymbol{P}-\boldsymbol{A}_j\right)^2\chi_j(\boldsymbol{R})=\left(E-E_j(\boldsymbol{R})\right)\chi_j(\boldsymbol{R})\ . \tag{3.4}$$

Equation (3.4) shows that the fast system induces into the slow system a potential energy $E_j(\boldsymbol{R})$, and a velocity dependent interaction arising from the gauge potential $\boldsymbol{A}_j$ which enters dynamics only through the gauge field = curl $\boldsymbol{A}_j$. The gauge potential is also associated with gauge invariance. It is found that, when the phase of the electronic eigenfunction is changed,

$$\varphi(\boldsymbol{r},\boldsymbol{R})\rightarrow\varphi(\boldsymbol{r},\boldsymbol{R})\,exp\,\{i\mu(\boldsymbol{R})\}$$

the potential transforms as,

$$\boldsymbol{A}\rightarrow\boldsymbol{A}-\nabla\mu(\boldsymbol{R})\ .$$

The total product wave function $\Psi=\chi\varphi$ is gauge independent. In consequence, a change of phase of $\varphi$ $(\boldsymbol{r},\boldsymbol{R})$ must be compensated for by a change of phase in $\chi$ $(\boldsymbol{R})$ which then inherits a phase dependence from the integrated out electrons.

## 4. PERTURBATION OF THE NUCLEI

The previous section has addressed the reaction of the electrons on the nuclei brought about by the adiabatic decoupling employed in the $B$-0 scheme. Here, we consider the matrix element of a (perturbation) operator $B = B\ (\boldsymbol{R})$ that acts only on the nuclear coordinates. Intuitive reasoning suggests that these two topics are closely related, and this is borne out by a simple calculation.

With the standard $B$-0 total wave function (3.1),

$$\langle \Psi_j | B | \Psi_{j'} \rangle \equiv \int d\boldsymbol{r} \int d\boldsymbol{R}\, \chi_j^*(\boldsymbol{R})\, \varphi_j^*(\boldsymbol{r},\boldsymbol{R}) B(\boldsymbol{R}) \chi_{j'}(\boldsymbol{R}) \varphi_{j'}(\boldsymbol{r},\boldsymbol{R}) = \delta_{jj'} \langle \chi_j | B | \chi_j \rangle \ . \tag{4.1}$$

Non-diagonal matrix elements arise from corrections to the standard $B$-0 scheme in part determined by the gauge potentials introduced in §3.

Employing the electron-nucleon mass ratio as a small parameter in a perturbation expansion, the next approximation to the total wave function is found to be,

$$\Psi_j(\boldsymbol{r},\boldsymbol{R}) = \chi_j(\boldsymbol{R}) \varphi_j(\boldsymbol{r},\boldsymbol{R}_o) + \left(i\hbar^2 / M\right) \nabla_{\boldsymbol{R}} \chi_j(\boldsymbol{R}) . \sum_{j'}{}' \varphi_{j'}(\boldsymbol{r},\boldsymbol{R}_o)\, \mathcal{A}_{jj'}(\boldsymbol{R}_o) / \Delta_{jj'} \ .$$

Here we use (3.3), $\Delta_{jj'} = \{E_j(\boldsymbol{R}_o) - E_{j'}(\boldsymbol{R}_o)\}$, $\boldsymbol{R}_o$ denotes the equilibrium nuclear coordinates, and the prime on the sum means that the term $j = j'$ is omitted. The matrix element of $B(\boldsymbol{R})$ obtained with (4.2) is,

$$\langle \Psi_j | B | \Psi_{j'} \rangle = \delta_{jj'} \langle \chi_j | B | \chi_j \rangle + \left(i\hbar^2 / M\right) \left(\mathcal{A}_{jj'} / \Delta_{jj'}\right) . \langle \chi_j | \nabla_{\boldsymbol{R}} B | \chi_{j'} \rangle \ . \tag{4.3}$$

The compact expression for the matrix element is obtained by use of the identity

$$\int d\boldsymbol{R}\, \nabla \{\chi_j^* B \chi_{j'}\} = 0 \ .$$

In an actual calculation of (4.3) it is useful to note the relation,

$$\langle j(\boldsymbol{R}) | \nabla_{\boldsymbol{R}} \mathcal{H}_{e\ell} | j'(\boldsymbol{R}) \rangle = i \mathcal{A}_{jj'}\, \Delta_{jj'} \ ; \ j \neq j' \tag{4.4}$$

which follows directly from (2.1). Values of $\mathcal{A}_{jj'}$ with $j \neq j'$ for a particular model are given in §5, and compared with the corresponding $\mathbf{A}_j$.

The results (4.3) and (4.4) are used in §§6,7 to calculate the cross-section for neutron scattering by a molecule accompanied by an electronic transition. Before reporting this work we discuss the behaviour of the matrix element (4.3) in the vicinity of a degeneracy.

## 5. NEAR-DEGENERACY

Our discussion of the matrix element (4.3) in the vicinity of a degeneracy of two electronic levels $E_j(\boldsymbol{R})$ and $E_{j'}(\boldsymbol{R})$ is couched in terms of a model described by a 2 x 2 Hamiltonian matrix ($R^2 = x^2 + y^2 + z^2$, with $\boldsymbol{R}$ relative to the location of the crossing point),

$$\mathcal{H}_{el}(\boldsymbol{R}) = \xi \begin{pmatrix} -z & x+iy \\ x-iy & z \end{pmatrix} , \tag{5.1}$$

in which the parameter $\xi$ sets the energy scale. This model has the property that the eigenvalues $\varepsilon_{\pm} = \pm \xi R$ cross at $R = 0$. The gauge potentials for (5.1) required in (4.3) have been calculated via (4.4). When $y = 0$ the model is purely real, and the only type of phase change for a circuit is just a change of sign. Cases that correspond to $y \neq 0$ include (*i*) application of a magnet field (*ii*) diatomic molecules not in a $^1\Sigma$ state and (*iii*) molecules that possess a Kramers degeneracy. Here, the model serves to illustrate the expected structure of the cross-section in the vicinity of a degeneracy , and the differences between $\mathcal{A}_{jj'}$ $(j \neq j')$ and $\mathbf{A}_j = \mathcal{A}_{jj}$

If we choose $j = (+)$ and $j' = (-)$, and adopt the notation

$$\langle \chi_+ | \nabla_{\boldsymbol{R}} B | \chi_- \rangle = \boldsymbol{J} \ , \tag{5.2}$$

we obtain the result,

$$(i\hbar^2/M)(\mathcal{A}_{+-}/\Delta_{+-})\cdot\langle\chi_+|\nabla_{\boldsymbol{R}}B|\chi_-\rangle = \left(\frac{\hbar^2}{4\xi MR^3}\right)\left(x^2+y^2\right)^{-\frac{1}{2}}$$

$$\left\{iR\left(yJ_x - xJ_y\right) + z\left(xJ_x + yJ_y\right) - \left(x^2+y^2\right)J_z\right\} . \tag{5.3}$$

It is instructive to compare the components $\mathcal{A}_{+-}$ used in deriving (5.3) with $\boldsymbol{A}_{\pm}$, the gauge potentials for the two energy surfaces used to calculate geometric phases from the relation (2.4). To this end, let us write $\boldsymbol{R}$ in spherical components, e.g. $x = \boldsymbol{R}\, sin\alpha\, cos\beta$, and choose to compare $\mathcal{A}_{+-}$ with $\boldsymbol{A}_{-}$, derived from (2.5). We find,

$$\mathcal{A}_{+-} = (1/2R)\begin{cases} -i\, cos\alpha\, cos\beta - sin\beta \\ -i\, cos\alpha\, sin\beta + cos\beta \\ -i\, sin\alpha \end{cases}$$

and

$$A_{-} = (1/2R)\begin{cases} sin\beta\, tan(\alpha/2) \\ -\, cos\beta\, tan(\alpha/2) \\ 0\ . \end{cases}$$

These results illustrate the general feature that $\boldsymbol{A}$ is purely real, while there is no such condition for $\mathcal{A}$. Also significant is the fact that $\boldsymbol{A}$ is not defined on the z-axis ($\alpha=\pi$), unlike $\mathcal{A}$ which is smooth. However, the geometric phase, a line integral of $\boldsymbol{A}$, is finite for a circuit around the degeneracy (the singularity is reminiscent of a Dirac pole); the phase is proportional to the solid angle that the circuit subtends at the degeneracy. Additional properties of $\boldsymbol{A}$ are found in reference [1] and [5].

## 6. NEUTRON CROSS-SECTION : NON-DEGENERATE LEVELS

For neutron scattering by nuclei the interaction operator,

$$B = b\, exp\,(i\boldsymbol{Q}.\boldsymbol{R}) \tag{6.1}$$

where $\boldsymbol{Q}$ is the change in wave vector on scattering, and $b$ is the scattering length. The cross-section for scattering by a single molecule is proportional to,

$$I = \left|\langle\Psi_j|b\, exp\,(i\boldsymbol{Q}.\boldsymbol{R})|\Psi_{j'}\rangle\right|^2 \tag{6.2}$$

and it has the dimension of area. For scattering accompanied by an electronic transition $(j' \rightarrow j)$ we find, using (4.3),

$$I = \left(b\hbar^2 / M\right)^2 \left| \frac{\mathbf{Q} \cdot \mathcal{A}_{jj'}}{\Delta_{jj'}} \langle \chi_j | exp(i\mathbf{Q}.\mathbf{R}) | \chi_{j'} \rangle \right|^2 \tag{6.3}$$

The cross-section is seen to be proportional to the square of the scalar produce of the scattering vector and gauge potential.

To complete the connection with previous calculations of the cross-section [3] note that $(j' = 1, j = 2)$,

$$\left( \frac{\hbar^2}{m\Delta_{21}} \right) \mathbf{Q}.\mathcal{A}_{21} \cong i\mathbf{Q}.\mathbf{R} \int d\mathbf{r} \; \varphi_2(\mathbf{r},\mathbf{R})\varphi_1(\mathbf{r},\mathbf{R}+g\mathbf{R}) \; . \tag{6.4}$$

Here the right-hand side is evaluated for $\mathbf{R} = \mathbf{R}_o$ the equilibrium nuclear coordinates, and $g$ is the ratio of the rotational energy constant and the difference in the electronic energies of the states of interests, viz.

$$g = \left(\hbar^2 / MR_o^2\right) / \left(E_2 - E_1\right) \; . \tag{6.5}$$

The approximation involved in obtaining the result (6.4) is good when $g << 1$. This situation is likely to be realized in the majority of cases. In consequence, the cross-section (6.3) is small: an explicit example is discussed in [3]. For nearly degenerate electronic states $g$ need not be small. In this case, the cross-section is calculated with (6.3), and it is no longer so small.

## 7. CROSS-SECTION : NEAR-DEGENERACY

The structure of the cross-section in the vicinity of a degeneracy of two states is derived from the material gathered in §5 for the model Hamiltonian (5.1).

Using (6.1) for the neutron beam interaction operator $B$, one has

$$J = iQ \langle \chi_+ | exp(iQ.R) | \chi_- \rangle . \tag{7.1}$$

Since the two levels in the model cross at $\boldsymbol{R} = 0$ it is not unreasonable to use the approximate value,

$$J \cong iQ \langle \chi_+ | \chi_- \rangle . \tag{7.2}$$

The (Franck-Condon) matrix element is finite because the nuclear wave functions $\chi_+$, $\chi_-$ are derived for two distinct electronic energy surfaces. If the level crossing occurs at a location such that $\boldsymbol{Q}.\boldsymbol{R} \sim 1$, then the $\boldsymbol{Q}$-dependence of the vibrational-rotational matrix element should be explicitly included. However, in this more general case an estimate of the magnitude of the cross-section can be obtained by summing over all final nuclear states; use of closure reduces the square of the nuclear matrix element to $\langle \chi_+ | \chi_+ \rangle = 1$, and so the following expression remains a meaningful estimate.

The following result for the cross-section (6.3) is obtained with considerable algebra directly from (5.3);

$$I = \left\{ \frac{Qb\hbar^2 \langle \chi_+ | \chi_- \rangle sin\, \alpha}{4MR^2 \xi} \right\}^2 , \tag{7.3}$$

where $\alpha$ is the angle between $\boldsymbol{Q}$ and $\boldsymbol{R}$. Observe that the cross-section (7.3) is not singular on the $z$-axis, unlike the gauge potential from which the geometric phase is derived. For scattering by a free molecule $sin^2\, \alpha$ is replaced by $({}^2/_3)$. The presence of a factor $({}^1/_{R}{}^4)$ in the cross-section shows that it is large in the vicinity of a level crossing. Since the cross-section is obtained from the correction to the standard $B$-0 wave function, consistency in the level of approximation requires in the extreme limit $R \to 0$ consideration of higher-order terms in (4.2).

## 8. DISCUSSION

Our formulation of neutron molecular spectroscopy, involving an electronic transition, places the theory in a wider context, demonstrating as it does the rôle in this problem of gauge potentials and geometric phases that are indigenous to the Born-Oppenheimer scheme. The experimental feasibility of making measurements has been established by Johnson and Bowman [6] since when neutron techniques have dramatically improved [7].

Predicted cross-sections are relatively small if the spacing of the electronic levels is large compared to the rotational energy constant which is the general rule for transitions from the ground state of a stable molecule. In view of this, the case of nearly degenerate electronic levels is of considerable practical interest. A simple model of a two-level system is used to show that, the cross-section is much enhanced in the vicinity of a level crossing, and it has a remarkably simple structure. One feature which merits comment is that, in the vicinity of a degeneracy the gauge potential for the geometric (Berry) phase is singular (reminiscent of a Dirac pole) while the corresponding cross-section is free of the singularity, in accord with physical intuition.

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