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# Origin Invariance of the Fully Retarded Rotational Strength

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The gauge invariance of the rotational strength is examined. It is shown that the invariance of the rotational strength derived in the quantized field formalism follows only when all terms (to order $e^2$) are retained in the $T$-matrix from which the rotational strength is derived. The gauge invariance of the semiclassical theory of natural circular dichroism is discussed. The origin invariance of the Schrödinger equation is considered and the difficulties associated with making truncated expansions of the retardation factors for the rotational strength are outlined.

The origin invariance of the rotational strength has been discussed for about forty years.[1-5] The deliberations of these workers have centred around the two different expressions for calculating the rotational strength, namely, the dipole velocity and the dipole length forms. The choice of the appropriate expression is of considerable importance for the numerical calculation of rotational strengths. Since exact eigenstates are generally unavailable, it is highly desirable to employ that form for the dipole operator which minimizes inaccuracies due to the use of inexact wavefunctions. For inexact wavefunctions, the usual transformation between the matrix element of the electronic momentum operator $\boldsymbol{p}_j$ and the matrix element of the electronic coordinate operator $\boldsymbol{r}_j$

$$\langle\psi_a|\boldsymbol{p}_j|\psi_b\rangle = (\mathrm{i}m/\hbar)(E_a-E_b)\langle\psi_a|\boldsymbol{r}_j|\psi_b\rangle \tag{1}$$

is no longer justified.

Since origin invariance in the present context may be related to the more general requirement that a theory be gauge invariant, it is then the latter which will be examined in some detail. We will consider the rotational strength in its fully retarded form, which has not been the case previously. Practical considerations of calculations are set aside. The reasoning for this is due to the fact that the operators themselves are not gauge invariant; it is only the various matrix elements and their products which are invariant. This requires us to restrict the discussion to cases involving only exact eigenstates. The purpose of this paper is twofold. First, we wish to establish to order $e^2$, that gauge invariance of the $T$-matrix is satisfied. Secondly, to emphasize that some careful consideration of phase factors, is necessary, when expansion of the retardation factors is carried out to obtain the conventional Rosenfeld expression for the rotational strength.

## GAUGE INVARIANCE OF THE $T$-MATRIX

In this section, it is shown that the $T$-matrix[6] (to order $e^2$) which contains the fully retarded rotational strength, is invariant under the transformation of the quantized potentials

$$\boldsymbol{A}' = \boldsymbol{A}+\nabla\chi \tag{2}$$

$$\phi' = \phi-\frac{1}{c}\frac{\partial\chi}{\partial t}. \tag{3}$$

For convenience we assume the $T$-matrix is initially described in the coulomb gauge. It is then required to show that

$$T^{(2)}(\chi)-T^{(2)}(\chi=0)=0. \tag{4}$$

A method used to demonstrate the invariance of the second order corrections to the energy [7] in calculations subject to the change given by eqn (2) is modified to prove eqn (4). The problem is more involved than the energy calculation, since we need to consider specifically the quantized nature of the radiation field.

The system under consideration is described by a Hamiltonian

$$H = H_R + H_M + V \tag{5}$$

where $H_R$ is the Hamiltonian of the radiation field, $H_M$ the molecular Hamiltonian and $V$ describes the interaction between the molecule and the radiation field. An eigenfunction of $H_R+H_M$ will be described by the notation $|\boldsymbol{k},a\rangle$. The following results may then be written

$$\langle \boldsymbol{k}_a, a|[H_R+H_M, \chi]|b, \boldsymbol{k}_b\rangle = (E_{ab}+\hbar c k_{ab})\langle \boldsymbol{k}_a, a|\chi|b, \boldsymbol{k}_b\rangle \tag{6}$$

$$\langle \boldsymbol{k}_a, a|[H_M, \chi]|b, \boldsymbol{k}_b\rangle = (2m)^{-1}\langle \boldsymbol{k}_a, a|2(\boldsymbol{p}\chi)\,.\,\boldsymbol{p}+(p^2\chi)|b, \boldsymbol{k}_b\rangle \tag{7}$$

$$\langle \boldsymbol{k}_a, a|[H_R, \chi]|b, \boldsymbol{k}_b\rangle = 0 \tag{8}$$

and hence the result

$$\left\langle \boldsymbol{k}_a, a\left|\frac{e}{2mc}[\boldsymbol{p}\,.\,\nabla\chi+\nabla\chi\,.\,\boldsymbol{p}]\right|b, \boldsymbol{k}_b\right\rangle = (ie/\hbar c)(E_{ab}+\hbar c k_{ab})\langle \boldsymbol{k}_a, a|\chi|b, \boldsymbol{k}_b\rangle. \tag{9}$$

Introducing the substitutions $|\boldsymbol{k}_a\rangle = |\boldsymbol{k}_b\rangle = |\boldsymbol{k}\rangle$, $|a\rangle = |b\rangle$ and $\chi\to\chi^2$ into eqn (9), then the following result is obtained

$$\left\langle \boldsymbol{k}, a\left|\chi\nabla\chi\,.\,\nabla+\tfrac{1}{2}\nabla\chi\,.\,\nabla\chi+\tfrac{1}{2}\chi\nabla^2\chi\right|a, \boldsymbol{k}\right\rangle = 0. \tag{10}$$

Eqn (9) and (10) are the two principal results required to establish eqn (4).

The rotational strength is derived from the second order $T$-matrix. To see how gauge invariance of the rotational strength is maintained, it is necessary to carry all terms of order $e^2$.

Now the $T$-matrix is given as

$$T^{(2)}(\chi=0) = \frac{e^2}{2mc^2}\langle \boldsymbol{k}_a, a|A^2|b, \boldsymbol{k}_b\rangle + \frac{e^2}{m^2c^2}\sum_n\left\{\frac{\langle \boldsymbol{k}_a, a|\boldsymbol{A}\,.\,\boldsymbol{p}|n\rangle\langle n|\boldsymbol{A}\,.\,\boldsymbol{p}|b, \boldsymbol{k}_b\rangle}{E_b-E_n+\hbar c k_b} + \frac{\langle \boldsymbol{k}_a, a|\boldsymbol{A}\,.\,\boldsymbol{p}|n, \boldsymbol{k}_a\boldsymbol{k}_b\rangle\langle \boldsymbol{k}_b\boldsymbol{k}_a, n|\boldsymbol{A}\,.\,\boldsymbol{p}|b, \boldsymbol{k}_b\rangle}{E_b-E_n-\hbar c k_a}\right\}. \tag{11a}$$

For a nonzero $\chi$, the $T$-matrix is given by

$$\begin{aligned}T^{(2)}(\chi) = {}& \frac{e^2}{2mc^2}\langle \boldsymbol{k}_a, a|(\boldsymbol{A}+\nabla\chi)^2|b, \boldsymbol{k}_b\rangle + \\ & \frac{e^2}{m^2c^2}\sum_n\sum_{k_1,k_2,\ldots}\Big\{\langle \boldsymbol{k}_a, a\,|\,\tfrac{1}{2}[\boldsymbol{p}\,.\,(\nabla\chi+\boldsymbol{A})+(\boldsymbol{A}+\nabla\chi)\,.\,\boldsymbol{p}]\,|\,n, \boldsymbol{k}_1\boldsymbol{k}_2\boldsymbol{k}_3\ldots\rangle\times \\ & \langle \ldots \boldsymbol{k}_3\boldsymbol{k}_2\boldsymbol{k}_1, n\,|\,\tfrac{1}{2}[\boldsymbol{p}\,.\,(\nabla\chi+\boldsymbol{A})+(\nabla\chi+\boldsymbol{A})\,.\,\boldsymbol{p}]\,|\,b, \boldsymbol{k}_b\rangle\times \\ & [E_b+\hbar c k_b-(E_n+\hbar c k_1+\hbar c k_2+\ldots)]^{-1}\Big\}.\end{aligned} \tag{11b}$$

For a consideration of the rotational strength, $|\boldsymbol{k}_a\rangle = |\boldsymbol{k}_b\rangle = |\boldsymbol{k}\rangle$ and $|a\rangle = |b\rangle$. For the case where $\chi = 0$, eqn (11$b$) simplifies to give eqn (11$a$). From eqn (11$b$) with $|a\rangle = |b\rangle$ and $|\boldsymbol{k}_a\rangle = |\boldsymbol{k}_b\rangle$, we can see that there are only nonvanishing matrix elements in the summation which involve the interaction $\boldsymbol{A}\cdot\boldsymbol{p}$ for single photon processes. The only nonvanishing matrix elements involving the $c$-number $\chi$ are those for which no change occurs in the photon field. Then from eqn (11$a$) and (11$b$) we have

$$\begin{aligned} &T^{(2)}(\chi)-T^{(2)}(\chi=0)\\ &=\frac{e^2}{2mc^2}\langle \boldsymbol{k}, a|\nabla\chi\,.\,\nabla\chi|a, \boldsymbol{k}\rangle+\\ &\frac{e^2}{4m^2c^2}\sum_n\Big\{\langle \boldsymbol{k}, a\,|\,\boldsymbol{p}\,.\,\nabla\chi+\nabla\chi\,.\,\boldsymbol{p}\,|\,n, \boldsymbol{k}\rangle\times\\ &\langle \boldsymbol{k}, n\,|\,\boldsymbol{p}\,.\,\nabla\chi+\nabla\chi\,.\,\boldsymbol{p}\,|\,a, \boldsymbol{k}\rangle(E_a-E_n)^{-1}\Big\}. \end{aligned} \tag{11c}$$

Employing eqn (9), we have

$$\begin{aligned} T^{(2)}(\chi)-T^{(2)}(\chi=0) &= \frac{e^2}{mc^2}\langle \boldsymbol{k}, a\,|\,\tfrac{1}{2}\nabla\chi\cdot\nabla\chi+\chi\{\nabla\chi\cdot\nabla+\tfrac{1}{2}\nabla^2\chi\}\,|\,a, \boldsymbol{k}\rangle \\ &= 0. \end{aligned} \tag{12}$$

on using eqn (10). This completes the proof of eqn (4) when $\chi$ is taken as a $c$-number.[8]

A few comments can be made when $\chi$ is allowed to be a $q$-number, although we do not consider this case in detail. For $\chi$ a $q$-number, eqn (6) and (7) will remain unchanged, although eqn (8) will be altered. If, for example, $\chi$ is chosen to be of the form

$$\chi = \sum_j\{c_j(r)\exp(-\mathrm{i}\omega_j t)a_j+c_j^*(r)\exp(\mathrm{i}\omega_j t)a_j^\dagger\},$$

then eqn (8) becomes

$$\langle \boldsymbol{k}, a|[H_{\mathbf{R}}, \chi]|b, k_b\rangle = -\mathrm{i}\hbar\langle \boldsymbol{k}_a, a\left|\frac{\partial\chi}{\partial t}\right|b, \boldsymbol{k}_b\rangle.$$

Eqn (9) is then modified by the appearance of a term $(e/c)(\partial\chi/\partial t)$ in the matrix element on the left hand side. In addition, a result similar to eqn (10) may be derived if $[\chi, \nabla\chi]$, $[\chi, \nabla^2\chi]$ and $[\chi, \partial\chi/\partial t]$ all equal zero. Eqn (11$c$) will be modified by the appearance of time derivatives of $\chi$ in the appropriate matrix elements, and there is no longer a direct simplification of eqn (11$c$) as was the case when $\chi$ was a $c$-number.

Eqn (11$c$) can be treated by considering terms linear in $\chi$ and those quadratic in $\chi$ separately. The quadratic terms can be shown to be zero with the use of the modified eqn (10). The terms linear in $\chi$ can be shown to be zero with the assumptions $[\chi, \boldsymbol{A}] = 0$ and $[\nabla\chi, \boldsymbol{A}] = 0$. The unrealistic demands of requiring these five comutators to vanish makes the treatment of $\chi$ as a $q$-number rather unsatisfactory.

The case of origin invariance for the rotational strength as conventionally employed has been discussed fully by Buckingham and Dunn.[9] These authors have shown the need for adding an electric dipole–electric quadrupole term to the usual electric dipole–magnetic dipole expression when anisotropic media are considered.

Higher asymmetry effects in optical rotation, which are still second-order optical processes, are possible.[10, 11] Since the rotational strength can be derived in a quantum electrodynamical formulation,[10] which yields the rotational strength directly in terms of the $T$-matrix elements, this then appears to be a logical starting point for discussions of gauge invariance which encompass all the higher asymmetry effects.

Since only physical observables are required to be gauge invariant, it is the total sum of all contributions to the rotational strength which must be gauge invariant. This sum will include higher asymmetry corrections; although numerically small, they must nevertheless satisfy the requirements of gauge invariance. This seems to dictate an examination of the general $T$-matrix elements.

## SEMICLASSICAL ARGUMENT FOR CIRCULAR DICHROISM

Natural circular dichroism based on a semiclassical treatment of the radiation field is determined directly from the matrix element [12]

$$\langle n|V^{(1)}|a\rangle = \sum_j \left\langle n\left|\frac{e}{mc}\boldsymbol{A}_j \cdot \boldsymbol{p}_j\right|a\right\rangle \tag{13}$$

and $\boldsymbol{A}$ now denotes the vector potential of the field in unquantized form and the sum is over all particles. In the presence of a gauge transformation, eqn (13) becomes

$$\langle n|V^{(1)}(\chi)|a\rangle = \sum_j \left\langle n\left|\frac{e}{mc}\boldsymbol{A}_j \cdot \boldsymbol{p}_j + \frac{e}{2mc}[2\nabla_j\chi_j \cdot \boldsymbol{p}_j + (\boldsymbol{p}_j \cdot \nabla_j\chi_j)] + \frac{e}{c}\frac{\partial\chi_j}{\partial t}\right|a\right\rangle. \tag{14}$$

Employing the following result

$$(E_n - E_a)\langle n|\chi_j|a\rangle = (-\mathrm{i}\hbar/2m)\langle n|2\nabla_j\chi_j \cdot \boldsymbol{p}_j + (\boldsymbol{p}_j \cdot \nabla_j\chi_j)|a\rangle \tag{15}$$

allows us to write eqn (14) as

$$\langle n|V^{(1)}(\chi)|a\rangle = \sum_j \left\langle n\left|\frac{e}{mc}\boldsymbol{A}_j \cdot \boldsymbol{p}_j + \frac{\mathrm{i}e}{c}(\omega_{na}+\omega)\theta_j \exp(\mathrm{i}\omega t) + \frac{\mathrm{i}e}{c}(\omega_{na}-\omega)\theta_j^* \exp(-\mathrm{i}\omega t)\right|a\right\rangle \tag{16}$$

where we have introduced a gauge of the form

$$\chi_j = \theta_j \exp(\mathrm{i}\omega t) + \theta_j^* \exp(-\mathrm{i}\omega t). \tag{17}$$

With the standard assumption that circular dichroism is only appreciable at a resonance, i.e., $\omega = \omega_{na}$, then the ellipticity per unit path length, which is proportional to $|\langle n|V^{(1)}|a\rangle|^2$, is independent of $\chi$ as can be seen from the form of $\langle n|V^{(1)}(\chi)|a\rangle$. This simple argument is limited to gauge transformations of the form given by eqn (17), and is further restricted by the constraint that the non-resonant components be vanishingly small.

To effect a displacement of the coordinate system we take $\chi$ to be of the following form

$$\chi = \alpha(x+\mathrm{i}gy)\exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{r}-\mathrm{i}\omega t)\{\exp(\mathrm{i}\boldsymbol{k}\cdot\boldsymbol{R})-1\}+\text{complex conjugate (c.c.)} \tag{18}$$

where $\boldsymbol{R}$ is a constant vector displacement, $\alpha$ an amplitude factor, $g = -1$ or $+1$ for left and right circularly polarized waves respectively and we assume for simplicity that the wave is propagating in the $z$-direction. $\chi$ satisfies

$$\nabla^2\chi - c^{-2}\frac{\partial^2\chi}{\partial t^2} = 0 \tag{19}$$

and is of the form given by eqn (17). The transformed vector potential

$$\boldsymbol{A}' = \boldsymbol{A} + \nabla\chi \tag{20}$$

becomes

$$A' = [A_0\exp(ik\,.\,[r+R]-i\omega t)+\text{c.c.}] + [\alpha k(ix-gy)\{\exp(ik\,.\,R)-1\}\exp(ik\,.\,r-i\omega t)+\text{c.c.}] \tag{21}$$

where $A$ has been taken to be of the form

$$A = \alpha(\mathbf{i}+ig\mathbf{j})\exp(ik\,.\,r-i\omega t)+\text{c.c.}$$

and $A_0 = \alpha(\mathbf{i}+ig\mathbf{j})$ is an amplitude factor.

Introducing $A'$ into the matrix element for the ellipticity per unit path length leads to a result which is independent of $R$ based on the conclusion drawn from eqn (16). However if we neglect the non-transverse component of $A'$ and expand the retardation factor $\exp\{ik\cdot(r+R)\}$ we obtain the rotational strength of the form

$$R_{an} \approx Im(\langle a|r|n\rangle\,.\,\langle n|r\;x\;p|a\rangle+\langle a|r|n\rangle\,.\,R\;x\;\langle n|p|a\rangle). \tag{22}$$

The assumption of neglecting the non-transverse component and the approximation involved in expanding the factor $\exp(ik\,.\,R)$ leads to a result now dependent upon $R$ in contradiction to the previous conclusion. It therefore appears essential to retain $\exp(ik\,.\,R)$ as a phase factor and to retain the non-transverse component. As a result of eqn (16), we may deduce that $R_{an}$ is gauge invariant, independent of any properties of the matrix elements $r_{an}$ and $p_{an}$.

## ORIGIN INVARIANCE OF THE SCHRÖDINGER EQUATION

We now take up the question of how the origin invariance of the Schrödinger equation can be used to examine the truncated expansions of the retardation factors in the formula for the rotational strength. The operator that generates finite displacements is

$$\mathcal{O}(R) = \exp\left(-\frac{i}{\hbar}R\,.\,\sum_j p_j\right) \tag{23}$$

and hence the time dependent Schrödinger equation

$$i\hbar\,\partial\Phi/\partial t = H\Phi$$

becomes

$$i\hbar\,\partial\Phi'/\partial t = \mathcal{O}(R)H\mathcal{O}(R)^{-1}\Phi' \tag{24}$$

and

$$\begin{aligned}\mathcal{O}(R)H\mathcal{O}(R)^{-1} &= \mathcal{O}(R)H_M\mathcal{O}(R)^{-1}+\mathcal{O}(R)V\mathcal{O}(R)^{-1}\\ &= H_M+\mathcal{O}(R)\left\{\sum_j\frac{e}{mc}A(r_j,t)\,.\,p_j+\frac{e^2}{2mc^2}A^2(r_j,t)\right\}\mathcal{O}(R)^{-1}\\ &= H_M+\sum_j\left\{\frac{e}{mc}A(r_j+R,t)\,.\,p_j+\frac{e^2}{2mc^2}A^2(r_j+R,t)\right\}\end{aligned} \tag{25}$$

where the expansion

$$e^{\beta G}F\,e^{-\beta G} = F+\beta[G,F]+\frac{\beta^2}{2}[G,[G,F]]+\ldots \tag{26}$$

has been employed to obtain eqn (25). The above modification of $A(r,t)$ leads to no experimentally observable change, since the electric and magnetic fields remain unchanged.

Evaluation of the appropriate matrix element for determining the rotational strength, based on eqn (25), leads to the following result

$$\langle a|V^{(1)}|g\rangle = \exp(ik\,.\,R)\left\langle a\left|\sum_j\frac{e}{mc}A_0\,.\,p_j+\frac{ie}{2mc}kxA_0\,.\,r_jxp_j\right|g\right\rangle \tag{27}$$

where well known approximations have been employed.[12] Thus $|\langle a|V^{(1)}|g\rangle|^2$ is invariant with respect to the origin displacement $\boldsymbol{R}$. Alternatively, the matrix element $V_{ag}$ may be written as

$$\langle a|V^{(1)}|g\rangle = \left\langle a\left|\sum_j\left\{\frac{e}{mc}\boldsymbol{A}_0\,.\,\boldsymbol{p}_j+\frac{\mathrm{i}e}{2mc}\boldsymbol{k}x\boldsymbol{A}_0\,.\,\boldsymbol{r}_jx\boldsymbol{p}_j+\frac{\mathrm{i}e}{2mc}\boldsymbol{k}x\boldsymbol{A}_0\,.\,\boldsymbol{R}x\boldsymbol{p}_j\right\}\right|g\right\rangle \tag{28}$$

which appears to lead to an additional contribution to $R_{ag}$ of the form

$$R_{ag} \approx Im(\sum_j\langle g|\boldsymbol{A}_0\,.\,\boldsymbol{p}_j|a\rangle\langle a|\boldsymbol{k}x\boldsymbol{A}_0\,.\,\boldsymbol{R}x\boldsymbol{p}_j|g\rangle). \tag{29}$$

However, eqn (28) is equivalent to writing

$$\langle a|V^{(1)}|g\rangle = \sum_j\left\{\frac{e}{mc}\boldsymbol{A}_0\,.\,\langle a|\boldsymbol{p}_j|g\rangle(1+\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{R})+\frac{e}{2mc}(\mathrm{i}\boldsymbol{k}x\boldsymbol{A}_0)\,.\,\langle a|\boldsymbol{r}_jx\boldsymbol{p}_j|g\rangle\right\}. \tag{30}$$

Eqn (30) then to the approximation considered may be written

$$\langle a|V^{(1)}|g\rangle = \sum_j\left\{\frac{e}{mc}\boldsymbol{A}_0\,.\,\langle a|\boldsymbol{p}_j|g\rangle\exp(\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{R})+\frac{e}{2mc}(\mathrm{i}\boldsymbol{k}x\boldsymbol{A}_0)\,.\,\langle a|\boldsymbol{r}_jx\boldsymbol{p}_j|g\rangle\exp(\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{R})\right\}. \tag{31}$$

The exponential dependence of the second term becomes clearer if eqn (30) is written to higher powers of $(\boldsymbol{k}\,.\,[\boldsymbol{r}+\boldsymbol{R}])$, then eqn (30) would be

$$\langle a|V^{(1)}|g\rangle = \sum_j\left\{\frac{e}{mc}\boldsymbol{A}_0\,.\,\langle a|\boldsymbol{p}_j|g\rangle\left[1+\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{R}+\frac{i^2}{2}(\boldsymbol{k}\,.\,\boldsymbol{R})^2+\ldots\right]+\frac{e}{2mc}(\mathrm{i}\boldsymbol{k}x\boldsymbol{A}_0)\,.\,\langle a|\boldsymbol{r}_jx\boldsymbol{p}_j|g\rangle[1+\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{R}+\ldots]\right. \tag{32}$$

Eqn (32) expresses the fact that the factor $(\boldsymbol{k}\ x\ \boldsymbol{A}_0)\,.\,(\boldsymbol{R}\ x\ \boldsymbol{p}_j)$ may be regrouped with the term $\boldsymbol{A}_0\,.\,\boldsymbol{p}_j$ and replaced by a phase factor $\exp(i\boldsymbol{k}.\boldsymbol{R})$. The important point is that the arbitrary phase factor dependence must be expanded to all orders, even though the $\exp(\mathrm{i}\boldsymbol{k}\,.\,\boldsymbol{r})$ term is truncated after the second term. Thus the rotational strength under origin displacement is correctly defined in terms of the matrix element of eqn (27) and not eqn (28). This choice is dictated by the fact that since the Hamiltonian is invariant under the operation $\mathcal{O}(\boldsymbol{R})$, the function $\mathcal{O}(\boldsymbol{R})\Phi$ is also a solution, so that $\Phi' = \mathcal{O}(\boldsymbol{R})\Phi$ must be of the form $e^{\mathrm{i}\gamma}\Phi$ with $\gamma$ real. So we may expand $\Phi'$ as $e^{\mathrm{i}\gamma}\phi_a^{(0)}+\lambda e^{\mathrm{i}\gamma}\phi_a^{(1)}+\ldots$ which leads to matrix elements $\langle e^{\mathrm{i}\gamma}\phi_a^{(0)}|\boldsymbol{r}|e^{\mathrm{i}\gamma}\phi_a^{(0)}\rangle$ etc., which yields an invariant expression for the rotational strength independent of the form of the operators employed. So if the action of $\mathcal{O}(\boldsymbol{R})$ on $H$ and $\Phi$ is to lead to consistent results, eqn (27) must define the matrix element from which the rotational strength is derived.

## DISCUSSION

The term rotational strength is often loosely associated with just the first order contribution to the rotational strength. The first-order contribution is clearly not related to a physical observable and need not be rigorously gauge invariant. Only the total sum of all order contributions represents a gauge invariant quantity. However it is most logical to define the rotational strength in terms of contributions which are each gauge invariant, in an analogous manner in which higher order corrections in energy calculations are gauge invariant. The advantage of the scheme is clearly that an infinite order perturbation calculation is no longer required to obtain a gauge invariant quantity.

The question as to which form of the rotational strength, dipole velocity versus dipole length, should be employed, when exact eigenfunctions are unavailable, cannot be inferred from the requirements of gauge invariance as set forth in this paper. This is due to the requirement that we have been employing exact eigenstates throughout.

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