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**LETTER TO THE EDITOR**

# Interpreting Stone's model of Berry phases

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**Abstract**

We show that a simple quantum-mechanical model, put forward by Stone some time ago, affords a description of site magnetoelectricity, a phenomenon which takes place in crystals (and molecular systems) when space inversion is locally broken and coexistence of electric and magnetic moments is permitted by the site point group. We demonstrate this by identifying a local order parameter, which is odd under both space inversion and time reversal. This order parameter (a magnetic quadrupole) characterizes Stone's ground state. Our results indicate that the model, extended to a lattice of sites, could be relevant to the study of electronic properties of transition-metal oxides. A generalization of Stone's Hamiltonian to cover cases of different symmetry is also discussed.

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In 1986 Stone reported a study of the Hamiltonian

$$H = \frac{1}{2I}\boldsymbol{L}^2 - \mu \boldsymbol{n}\cdot\boldsymbol{\sigma} \tag{1}$$

which he performed by path-integral and conventional quantum-mechanical techniques (Stone 1986). The conventional method considers the basis set of spinor spherical harmonics

$$\left|j \pm \tfrac{1}{2}, jm\right\rangle = \sum_{m',\xi} C^{j,m}_{j\pm\frac{1}{2},m';\frac{1}{2},\xi} \left|j \pm \tfrac{1}{2}, m'\right\rangle \left|\tfrac{1}{2}, \xi\right\rangle \tag{2}$$

as suggested by $[H, \boldsymbol{J}]_- = 0$, with $\boldsymbol{J} = \boldsymbol{L} + \frac{1}{2}\boldsymbol{\sigma}$; $\boldsymbol{L}$ stands for the angular momentum operator that generates rotations of $\boldsymbol{n} = \mathbf{r}/r$; $\boldsymbol{\sigma}$ are Pauli matrices and $I$ is the rotor moment of inertia; $[\,,\,]_-$ denotes a commutator and $C^{j,m}_{j_1,m_1;j_2,m_2}$ is a Clebsch–Gordan coefficient. On account of the property (Stone 1986, Varshalovich *et al* 1988)

$$\boldsymbol{n}\cdot\boldsymbol{\sigma}\left|j \pm \tfrac{1}{2}, jm\right\rangle = -\left|j \mp \tfrac{1}{2}, jm\right\rangle \tag{3}$$

 

the Hamiltonian $H$ is readily diagonalized in this basis. In the large-$\mu$ limit, the ground state is found to be

$$|g\rangle_- = \tfrac{1}{\sqrt{2}}\left(\left|j+\tfrac{1}{2}, jm\right\rangle - \left|j-\tfrac{1}{2}, jm\right\rangle\right) \tag{4}$$

with energy $E = -\mu$.

Stone's motivation was to provide a simple quantum-mechanical example in which the Berry phase gives rise to Wess–Zumino terms. Indeed, for large $\mu$, equation (1) describes the motion of a constrained spin, which is equivalent to motion of a charged particle about a magnetic monopole (Leinaas 1978).

Equation (1) has been interpreted as a model for a solenoid, which is rotating about its centre of mass where a spin-$\frac{1}{2}$ particle is placed. When $\mu$ is small, the solenoid and the particle would spin independently. As $\mu$ becomes large, the spin will become slaved to the direction of the solenoid (Stone 1986, Aitchinson 1987). Note that this physical picture leads to the coupling $\boldsymbol{B}\cdot\boldsymbol{\sigma}$, which is space and time even. ($\boldsymbol{B}$ stands for the magnetic field generated by the solenoid.) Such a symmetry should be contrasted with that of $\boldsymbol{n}\cdot\boldsymbol{\sigma}$, which is space and time odd.

The current letter will demonstrate that the model defined by equation (1) affords a description of a different effect: *site magnetoelectricity*. Such a phenomenon occurs in crystalline and molecular systems when space-inversion symmetry is locally broken and coexistence of electric and magnetic moments is permitted by the pertinent site point group. An effective magnetoelectric interaction between these two moments would be described by equation (1) provided we identify $\boldsymbol{n}$ with a unitary electric-dipole moment. (The electric charge $e$ is merged into $\mu$.) This new interpretation of the model does not affect its dynamical regimes (Stone 1986), which remain those of the rotating solenoid with $\boldsymbol{B}$ replaced by $\boldsymbol{n}$.

As shown by Goulon and his collaborators (Goulon *et al* 2000, 2002), microscopic magnetoelectric behaviour of crystals can be investigated using near-edge absorption of x-rays, which implies excitations of inner-shell electrons to empty valence states. As is known, this experimental technique is site selective, a feature resulting from the tuning of x-ray energy at a given inner-shell threshold. Sensitivity to the long-range order of local magnetoelectric order parameters is obtained by recording dichroic signals which stem from an interference between electric-dipole and electric-quadrupole transitions. As a consequence, scalars (e.g. $\boldsymbol{n}\cdot\boldsymbol{\sigma}$) are not probed by these experiments, which detect the long-range order of local (on-site) magnetoelectric order parameters represented by one-particle irreducible tensors of ranks 1, 2 and 3. One set of these order parameters specifically serves our purposes: the *magnetic quadrupoles* (rank-2 tensors)[1]. In the *LS*-coupling scheme, they read

$$\begin{aligned} \mathcal{M}_L^{(2)} &\equiv [\boldsymbol{n}, \boldsymbol{L}]^{(2)} & \mathcal{M}_S^{(2)} &\equiv [\boldsymbol{n}, \boldsymbol{S}]^{(2)} \\ \mathcal{M}_T^{(2)} &\equiv \tfrac{\sqrt{3}}{\sqrt{2}}[\mathrm{i}[\boldsymbol{\Omega}_L, \boldsymbol{L}]^{(2)}, \boldsymbol{S}]^{(2)} & \mathcal{M}_F^{(2)} &\equiv \tfrac{\sqrt{35}}{2}[[\boldsymbol{n}, \mathcal{Q}^{(2)}]^{(3)}, \boldsymbol{S}]^{(2)} \end{aligned} \tag{5}$$

as shown by recent theoretical work on x-ray dichroism and resonant scattering in noncentrosymmetric crystals (Carra *et al* 2003, Marri and Carra 2004). (The symbol $[\,,\,]^{(k)}$ denotes Clebsch–Gordan coupling of irreducible tensors; the spin operator is defined by $\boldsymbol{S} = \frac{1}{2}\boldsymbol{\sigma}$; $\mathcal{Q}^{(2)} = [\boldsymbol{L}, \boldsymbol{L}]^{(2)}$ identifies an orbital quadrupole.) Inspection of magnetic nature identifies orbital, spin and spin-orbital elements in the set. All tensors in (5) are space and time odd, being thereby invariant under the combined action of these transformations.

A set of vector order parameters will also be considered in connection with Stone's model. Its elements are defined by (Marri and Carra 2004)

[1] A simple mechanical model of a magnetic quadrupole is provided by two parallel coils run through by opposite currents.

$$\boldsymbol{n} \qquad \boldsymbol{P}_S \equiv \boldsymbol{\Omega}_L \times \boldsymbol{S} \qquad \text{and} \qquad \boldsymbol{P}_T \equiv -\tfrac{2\sqrt{5}}{\sqrt{3}}[[\boldsymbol{n}, \boldsymbol{L}]^{(2)}, \boldsymbol{S}]^{(1)} \tag{6}$$

in *LS* coupling. These irreducible tensors have *polar* (electric) symmetry, i.e. they are space odd and time even[2].

The order parameters (5), (6) are defined in the second quantization formalism. For example,

$$\big(\mathcal{M}_L^{(2)}\big)_q = \sum_{\substack{l,l'=l\pm1\\ m,m',\sigma,\sigma'}} \frac{1}{2}\big[\langle\sigma'|\langle l'm'|\big(\mathcal{M}_L^{(2)}\big)_q|lm\rangle|\sigma\rangle c^\dagger_{l'm'\sigma'}c_{lm\sigma} + \text{h.c.}\big] \tag{7}$$

and similarly for the others. Here, $c^\dagger_{lm\sigma}$ and $c_{lm\sigma}$ denote fermionic creation and annihilation operators, respectively.

As stated, the main purpose of the current work is to discuss magnetoelectric properties of Stone's Hamiltonian in the large-$\mu$ limit. To this end, we will show that the symmetry property displayed by the scalar $\boldsymbol{n}\cdot\boldsymbol{\sigma}$, when acting on the spinor spherical harmonics $\big|j \pm \frac{1}{2}, jm\big\rangle$ (equation (3)), extends to the irreducible tensors defined by expressions (5), (6), leading to relations whereby the magnetoelectric behaviour of $|g\rangle_-$ is readily inferred.

The basis set $\big|j \pm \frac{1}{2}, jm\big\rangle$ provides a convenient framework for describing *parity-breaking electron hybridization* (e.g. pd mixing in transition-metal oxides), in the *jj* coupling scheme[3]. It is thus clear that to generalize equation (3) to higher-rank order parameters we must first determine the form of the corresponding tensors in *jj* coupling. This is accomplished by resorting to the theory of *LS*→ *jj* transformations (Edmonds 1974). In the case of space-odd irreducible tensors, such transformations contain matrix elements between the states $|jm\rangle$ and $|j'm'\rangle$, with $j = l \pm \frac{1}{2}$ and $j' = l' \pm \frac{1}{2}$. Solving the corresponding equations for $j' = j$, as demanded by $[H, \boldsymbol{J}]_- = 0$, provides the required *jj*-coupled order parameters, which will appear as linear combinations of space-odd *LS*-coupled irreducible tensors. (Note that $\boldsymbol{n}\cdot\boldsymbol{\sigma}/2 = \boldsymbol{n}\cdot\boldsymbol{J}$, as $\boldsymbol{n}\cdot\boldsymbol{L} = 0$; no transformation is needed in this case.) Technically, the derivation is laborious as it implements angular-momentum recoupling methods (Racah tensor calculus). Thus, for the convenience of the reader, we will first state our results, then illustrate their physical content and, at the end, discuss mathematical aspects of the formulation.

The required *jj*-coupled magnetic quadrupole is found to be

$$\begin{aligned}\widetilde{\mathcal{M}}_J^{(2)}(l', l) = {} & \frac{1}{5}\left(\frac{l + l' - 1}{2}\right)\left(\frac{l + l' + 3}{2}\right)\mathcal{M}_S^{(2)}(l', l) + \frac{2}{3}\mathcal{M}_T^{(2)}(l', l) \\ & + \frac{1}{5}\mathcal{M}_F^{(2)}(l', l) - \frac{1}{2}\mathcal{M}_L^{(2)}(l', l)\end{aligned} \tag{8}$$

yielding, after some algebra,

$$\sum_{l,l'=l\pm1} \widetilde{\mathcal{M}}_J^{(2)}(l', l)_z \big|j \pm \tfrac{1}{2}, jm\big\rangle = -\frac{3m^2 - j(j+1)}{\sqrt{6}} \big|j \mp \tfrac{1}{2}, jm\big\rangle \tag{9}$$

with $\hat{\boldsymbol{z}}$ the quantization axis. This result generalizes equation (3) and shows that $|g\rangle_-$ is an eigenstate of the *jj*-coupled magnetic quadrupole operator; in other words, the large-$\mu$ ground

[2] The *LS* tensors (5), (6) are given in compact forms. In this representation irrational prefactors appear. They can be removed by recoupling transformations. We find $[\boldsymbol{S}, [\boldsymbol{L}, \boldsymbol{n}]^{(2)}]^{(1)} = -\frac{\sqrt{3}}{2\sqrt{5}}(\boldsymbol{S}\cdot\boldsymbol{L}\boldsymbol{n} + \boldsymbol{L}\boldsymbol{S}\cdot\boldsymbol{n})$ and similarly for $[\boldsymbol{S}, [\boldsymbol{\Omega}_L, \boldsymbol{L}]^{(2)}]^{(1)}$; furthermore, $[\boldsymbol{S}, -\mathrm{i}[\boldsymbol{\Omega}_L, \boldsymbol{L}]^{(2)}]^{(2)} = \frac{\sqrt{2}}{\sqrt{3}}([\boldsymbol{S}\times\boldsymbol{\Omega}_L, \boldsymbol{L}]^{(2)} + \frac{1}{2}[\boldsymbol{S}, \boldsymbol{\Omega}_L \times \boldsymbol{L}]^{(2)})$ and $[\boldsymbol{S}, [\boldsymbol{n}, \mathcal{Q}^{(2)}]^{(3)}]^{(2)} = (3[\boldsymbol{S}, [\boldsymbol{n}, \mathcal{Q}^{(2)}]^{(2)}]^{(2)} + 6[[\boldsymbol{S}, \mathcal{Q}^{(2)}]^{(2)}, \boldsymbol{n}]^{(2)} - 3[\boldsymbol{S}\cdot\boldsymbol{n}, \mathcal{Q}^{(2)}]^{(2)} - 5[\boldsymbol{S}, [\mathcal{Q}^{(2)}, \boldsymbol{n}]^{(2)}]^{(2)})/\sqrt{35}$.

[3] As shown below, in this basis, parity breaking hybridization displays rotational symmetry ($SU(n)$, in the general case). Rather than by ordinary $|jm\rangle$, irreducible representations are spanned by $\frac{1}{\sqrt{2}}(|j + \frac{1}{2}, jm\rangle \pm |j - \frac{1}{2}, jm\rangle)$ for ranks 0 and 2, and by $\frac{1}{\sqrt{2}}(\mathrm{e}^{\mathrm{i}\pi/4}|j + \frac{1}{2}, jm\rangle \pm \mathrm{e}^{-\mathrm{i}\pi/4}|j - \frac{1}{2}, jm\rangle)$ for rank 1.

state of Stone's Hamiltonian is magnetoelectric. Our conclusion is further supported by what follows. Consider the *jj*-coupled operator

$$\widetilde{\boldsymbol{P}}_J(l',l) = \boldsymbol{n}(l',l) + \boldsymbol{P}_S(l',l) - 2\boldsymbol{P}_T(l',l). \tag{10}$$

We have

$$\sum_{l,l'=l\pm1} \widetilde{\boldsymbol{P}}_J(l',l)_0 \left| j \pm \frac{1}{2}, jm \right\rangle = -m \left| j \mp \frac{1}{2}, jm \right\rangle \tag{11}$$

showing that $|g\rangle_-$ is an eigenstate of the *jj*-coupled unitary electric-dipole moment, with eigenvalue $m$. $|g\rangle_-$ is thus characterized by the simultaneous presence of an electric and a magnetic moment in a parallel (as expected) configuration. (Reversing the sign of the coupling constant in Stone's model ($\mu \to -\mu$, large $\mu$) would change the ground state to $|g\rangle_+ = \frac{1}{\sqrt{2}}\big(\big|j+\frac{1}{2}, jm\big\rangle + \big|j-\frac{1}{2}, jm\big\rangle\big)$ which is characterized by an antiparallel alignment of the moments and by a magnetic quadrupole with opposite sign.)

Thus, we are led to the conclusion that, in the strong coupling limit, the magnetoelectric interaction can be viewed as a problem of a magnetic moment constrained by an electric field. In turn, this is equivalent to the problem of a charged particle moving in the field of a magnetic monopole, both classically and quantum mechanically (Leinaas 1978). This seems to tally with recent work on magnetic monopoles in crystal momentum space (Fang *et al* 2003).

According to our findings, Stone's model provides a good starting point in the study of interactions between (local) electric and magnetic moments in crystals. For this purpose, an extension of the model to a lattice of sites is now needed. Such a model, characterized by an order parameter which violates space inversion and time reversal, could be relevant in the analysis of electronic properties of transition-metal oxides.

In certain site point groups (Cracknell 1975), magnetoelectric symmetry results in configurations where electric and magnetic moments are mutually perpendicular. Magnetoelectricity arises from toroidal distributions of currents and is described by anapole moments, in these cases. In *LS* coupling, the set of anapolar order parameters reads (Carra *et al* 2003, Marri and Carra 2004)

$$\begin{aligned} &\boldsymbol{\Omega}_L \equiv \tfrac{1}{2}(\boldsymbol{n}\times\boldsymbol{L} - \boldsymbol{L}\times\boldsymbol{n}) \qquad \boldsymbol{\Omega}_S \equiv \boldsymbol{n}\times\boldsymbol{S} \\ &\boldsymbol{\Omega}_T \equiv -\tfrac{2\sqrt{5}}{\sqrt{3}}[[\boldsymbol{\Omega}_L, \boldsymbol{L}]^{(2)}, \boldsymbol{S}]^{(1)}. \end{aligned} \tag{12}$$

The required *jj*-coupled anapole takes the form

$$\widetilde{\boldsymbol{\Omega}}_J(l',l) = \frac{2}{l+l'+1}\boldsymbol{\Omega}_L(l',l) - (l+l'+1)\boldsymbol{\Omega}_S(l',l) + \frac{4}{l+l'+1}\boldsymbol{\Omega}_T(l',l) \tag{13}$$

giving

$$(\widetilde{\boldsymbol{\Omega}}_J)_z \left| j \pm \tfrac{1}{2}, jm \right\rangle = \sum_{l,l'=l\pm1} \widetilde{\boldsymbol{\Omega}}_J(l',l)_z \left| j \pm \tfrac{1}{2}, jm \right\rangle = \mp \mathrm{i}m \left| j \mp \tfrac{1}{2}, jm \right\rangle. \tag{14}$$

$|\tilde{g}\rangle_- = \frac{1}{\sqrt{2}}\big(\mathrm{e}^{\mathrm{i}\pi/4}\big|j+\frac{1}{2}, jm\big\rangle - \mathrm{e}^{-\mathrm{i}\pi/4}\big|j-\frac{1}{2}, jm\big\rangle\big)$ is therefore an eigenstate of the *jj*-coupled anapole operator. It is readily shown that $|\tilde{g}\rangle_-$ is the large-$\mu$ limit ground state of the Hamiltonian obtained by replacing $\boldsymbol{n}\cdot\boldsymbol{\sigma}$ with $\hat{\boldsymbol{z}}\cdot\widetilde{\boldsymbol{\Omega}}_J$ in equation (1). Note that $|\tilde{g}\rangle_-$ is also an eigenstate of a *jj*-coupled *pseudodeviator*, a polar rank-2 tensor, with eigenvalue $[3m^2 - j(j+1)]/\sqrt{6}$. This new Hamiltonian thus displays x-ray natural circular dichroism. (The derivation of this result will not be given for lack of space. *LS* pseudodeviators have been discussed by Marri and Carra (2004).)

We conclude with some remarks concerning technical aspects of our derivation. Generalizing the concept of coupled double tensor (Judd 1967), we define

$$w_\zeta^{(xy)z}(l',l) = \sum_{\xi,\eta,\lambda,\lambda',\sigma,\sigma'} C^{z\zeta}_{x\xi;y\eta} C^{y\eta}_{\frac{1}{2}\sigma';\frac{1}{2}\sigma} C^{x\xi}_{l'\lambda';l\lambda} c^\dagger_{l'\lambda'\sigma'} \tilde{c}_{l\lambda\sigma} + \text{h.c.} \tag{15}$$

and

$$v_{\zeta}^{(j'j)z}(l',l)=\sum_{m,m'}C^{z\zeta}_{j'm';jm}c^{\dagger}_{l',j'm'}\tilde{c}_{l,jm}+\text{h.c.} \tag{16}$$

where $\tilde{c}_{l\lambda\sigma}=(-1)^{l-\lambda+\frac{1}{2}-\sigma}c_{l-\lambda-\sigma}$ and $\tilde{c}_{l,jm}=(-1)^{j-m}c_{l,j-m}$, so that creation and annihilation operators transform as the components of irreducible tensors. (Note that $w^{(x0)x}(l',l)$ are spinless, whereas $w^{(x1)z}(l',l)$ depend on spin.) The importance of $w^{(xy)z}(l',l)$ lies in the fact that all one-electron *LS* order parameters defined by (5), (6) can be expressed as multiples of them (Wigner–Eckart theorem). For example

$$w^{(20)2}(l',l)=-\frac{\sqrt{2}}{\sqrt{l(l+1)}C^{l'0}_{l0;10}\left\{\begin{smallmatrix}1&1&2\\l'&l&l\end{smallmatrix}\right\}}\mathcal{M}^{(2)}_{L}(l',l). \tag{17}$$

In a similar way, one-electron *jj* order parameters can be expressed as multiples of $v^{(j'j)z}(l',l)$. The tensors $w^{(xy)z}(l',l)$ and $v^{(j'j)z}(l',l)$ are related by a standard $LS\rightarrow jj$ transformation (Edmonds 1974), which reads

$$w^{(xy)z}(l',l)=\sum_{j,j'}(-1)^{x+y+z}[x,y,j,j']^{\frac{1}{2}}\begin{Bmatrix}l&l'&x\\\frac{1}{2}&\frac{1}{2}&y\\j&j'&z\end{Bmatrix}v^{(j'j)z}(l',l) \tag{18}$$

with $[a,\ldots,b]=(2a+1)\cdots(2b+1)$. In the case of magnetic-quadrupole order parameters, equation (18) yields a system of four equations. By solving this system for $j'=j$ and $l'=l\pm1$, we find

$$\begin{aligned}\widetilde{\mathcal{M}}^{(2)}_{J}(l',l)&=\text{rhs of equation (8)}\\&=-\tfrac{3}{2}(2l+1)(2l'+1)\left\{[\boldsymbol{n},\boldsymbol{J}]^{(l'+\frac{1}{2},l-\frac{1}{2})2}\delta_{l',l-1}+[\boldsymbol{n},\boldsymbol{J}]^{(l'-\frac{1}{2},l+\frac{1}{2})2}\delta_{l',l+1}\right\}.\end{aligned}$$

Vector order parameters lead to systems of three equations. Their solutions for $j'=j$ and $l'=l\pm1$ read

$$\widetilde{\boldsymbol{P}}_{J}(l',l)=-\frac{3(l+l'+1)}{2}\left[\boldsymbol{n}_{J}^{l'+\frac{1}{2},l-\frac{1}{2}}\delta_{l',l-1}+\boldsymbol{n}_{J}^{l'-\frac{1}{2},l+\frac{1}{2}}\delta_{l',l+1}\right] \tag{19}$$

and

$$\widetilde{\boldsymbol{\Omega}}_{J}(l',l)=-\frac{4(2l+1)(2l'+1)}{l+l'+1}\left[\boldsymbol{\Omega}_{J}^{l'+\frac{1}{2},l-\frac{1}{2}}\delta_{l',l-1}+\boldsymbol{\Omega}_{J}^{l'-\frac{1}{2},l+\frac{1}{2}}\delta_{l',l+1}\right] \tag{20}$$

providing a full definition of equations (10) and (13). In equation (20),

$$\boldsymbol{\Omega}^{j'j}_{J}(l',l)=\sum_{m,m'}\langle l',j'm'|\left(\boldsymbol{\nabla}_{\Omega}-\boldsymbol{\nabla}^{\dagger}_{\Omega}\right)\times\boldsymbol{J}+\boldsymbol{J}\times\left(\boldsymbol{\nabla}_{\Omega}-\boldsymbol{\nabla}^{\dagger}_{\Omega}\right)|l,jm\rangle c^{\dagger}_{l',j'm'}\tilde{c}_{l,jm} \tag{21}$$

where $\boldsymbol{\nabla}_{\Omega}=-\mathrm{i}\boldsymbol{n}\times\boldsymbol{L}$.[4]

In the case of anapolar order parameters, *LS* Judd's tensors are defined as

$$\overline{w}^{(xy)z}(l',l)=\mathrm{i}\sum_{\xi,\eta,\lambda,\lambda',\sigma,\sigma'}C^{z\zeta}_{x\xi;y\eta}C^{y\eta}_{\frac{1}{2}\sigma';\frac{1}{2}\sigma}C^{x\xi}_{l'\lambda';l\lambda}c^{\dagger}_{l'\lambda'\sigma'}\tilde{c}_{l\lambda\sigma}+\text{h.c.} \tag{22}$$

and similarly in *jj* coupling.

[4] A natural choice for the anapole in *jj* coupling would be $(\boldsymbol{n}\times\boldsymbol{J}-\boldsymbol{J}\times\boldsymbol{n})/2$ (Dothan and Ne'eman 1966). However, this operator cannot be employed in our case as its matrix element vanishes for $j'=j$. The vector appearing in equation (21) removes this drawback (Jerez 2003).

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