X-ray Dichroism and Orbital Anapoles

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We present a theoretical analysis of photo-absorption spectroscopies in noncentrosymmetric systems, covering both x-ray and optical regions. Integrated dichroic spectra are interpreted using microscopic effective operators, which are obtained by coupling the orbital angular momentum to the orbital anapole. Symmetry arguments afford a classification of valence-electron states in the presence of parity nonconserving hybridisation. Enantiomorphism is identified by a two-particle chiral operator.

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The current paper aims at providing a theoretical interpretation of two recent experiments:

**X-ray natural circular dichroism** (XNCD), probed in α-LiIO$_3$ and in Na$_3$Nd(digly)$_3$·2NaBF$_4$·6H$_2$O. (The effect was observed at the iodine L edges and at the Nd L$_3$ edge.)

**X-ray nonreciprocal linear dichroism** (XNRLD), detected at vanadium K edge in the low-temperature insulating phase of a Cr-doped V$_2$O$_3$ crystal.

XNCD measures the difference in absorption between right and left circularly polarised radiations. XNRLD implies a difference in absorption in radiations with linear polarisation parallel or perpendicular to a local symmetry axis. Both phenomena stem from the interference between electric-dipole (E1) and electric-quadrupole (E2) transitions that raise an inner-shell electron to empty valence orbitals. Detecting a nonvanishing signal thus requires an ordered structure (crystal) and the breaking of space inversion.

As will be shown, Lie groups (or, alternatively, Lie algebras) furnish a powerful tool for interpreting x-ray dichroism in non-centrosymmetric crystals. In fact, effective microscopic operators, which express electronic properties revealed by the spectra, are readily deduced from the pertinent group generators: The **orbital angular momentum and orbital anapole**. Our approach hinges therefore on spectrum-generating algebras (SGAs), a concept originally introduced in nuclear and particle physics. We present a theoretical analysis of photo-absorption spectroscopies in noncentrosymmetric systems, over a finite energy range, corresponding to the two partners of a spin-orbit split inner-shell: $j_\pm = l_\pm \pm \frac{\hbar}{2}$.

We follow Carra and Benoist and consider the general framework of a de Sitter algebra: $so(3,2)$. A realisation of such an algebra is provided by the operators: $A^+ = i(A - A^\dagger)/2$, $A^− = (A + A^\dagger)/2$, $L$ and $N_0$, where $A = n f_1(N_0) + \nabla_{\Omega} f_2(N_0)$, with $n = r/r$ and $\nabla_{\Omega} = −i n \times L$: $L$ denotes the orbital angular momentum (in units of $\hbar$). Furthermore, $f_1(N_0) = (N_0 − 1/2)f_2(N_0)$ and $f_2(N_0) = \sqrt{(N_0 − 1)/N_0}$, where $N_0|lm⟩ = (l + \frac{1}{2})|lm⟩$, with $|lm⟩$ a spherical harmonic. $A$ and $A^\dagger$ are known as shift operators as their action on $|lm⟩$ changes $l$ into $l \pm 1$.

As will be seen, discussing x-ray dichroism in noncentrosymmetric systems only requires $L$ (rotations) and $A^−$ (boosts), which themselves generate a homogeneous Lorentz group: $SO(3,1)$. We observe that

$$L \cdot A^- = A^- \cdot L = 0,$$

and that a physical interpretation of $A^−$ is provided by the relation

$$Ω^- = (n \times L - L \times n)/2 = i [n, L^2]/2 = \frac{i}{2} (\nabla^\dagger_\Omega - \nabla_\Omega) = \frac{1}{2 \sqrt{N_0}} [N_0, A^-] + \frac{1}{\sqrt{N_0}},$$

where $[...]_+$ denotes an anticommutator. Eq. (2) defines the (purely angular) orbital anapole $Ω^-$. As pointed out by Dothan et al., $L$ and $Ω^−$ also generate $SO(3,1)$, $[L$ and $A^+}$ generate another $SO(3,1)$ subgroup of $SO(3,2)$. Being space-odd and time-even, $A^+$ does not enter our theory of dichroism in noncentrosymmetric systems.

Our formulation is based on a localised (atomic) model and exploits the Racah-Wigner calculus to relate integrated dichroic spectra to the ground-state expectation value of effective orbital operators. These operators are constructed by coupling the hermitean vector operators $L$ (space-even) and $Ω^−$ (space-odd) to obtain space-odd irreducible tensors. In such a framework, one-electron tensors are given by ($l$ is an electron label).
\[
\sum_{l} [L_{l} \otimes \Omega_{l}^{-}]_{q}^{(k)l+l'} = \sum_{l} [L_{l} \otimes \Omega_{l}^{-}]_{q}^{(k)l,l'} + \text{H.c.}
\]
\[
= \sum_{m,m'} \langle l'm'|[L \otimes \Omega_{l}^{-}]_{q}^{(k)l}|lm \rangle a_{l'}^{\dagger} a_{lm} + \text{H.c.} \tag{3}
\]

Here, \(l' = l \pm 1\); \(a_{l'}^{\dagger}\) and \(a_{lm}\) create and annihilate valence electrons. (As usual, tensor couplings are defined via Clebsch-Gordan coefficients: \([U^{(p)} \otimes V^{(\kappa)}]_{q}^{(k)} \equiv \sum_{\mu,\nu} C_{\mu\nu}^{pq} C_{\mu\nu}^{pq}^{(k)} \otimes \mu_{\nu}^{(p) \otimes \kappa_{\nu}^{(p)}}\).) Extending Eq. (3) to define two-electron space-odd irreducible tensors is straightforward.

\(E1-E2\) interference: XNCD \[1\] The effect is characterised by an orbital pseudodevator, i.e., a space-odd and time-even rank-two irreducible tensor \([14]\). In our formulation, its microscopically expression is given by a coupling of \(L\) to the orbital anapole; it reads: \([L \otimes \Omega_{l}^{-}]_{q}^{(2)l+l'}\). For the integrated XNCD spectrum, we thus find
\[
\int_{j_{-}+j_{+}} \frac{\sigma_{\text{XNCD}}(\omega)}{(\hbar \omega)^{2}} d(\hbar \omega) = \frac{8\pi^{2}a}{3h \epsilon_{c}} (2\epsilon_{c} + 1) \sum_{l,l'=l \pm 1} R_{l,l'}^{(1)}
\]
\[
\sum_{l,l'=l \pm 1} R_{l,l'}^{(1)} \langle \psi_{0}| \sum_{l} [L_{l} \otimes (L \times n - n \times L)]_{l}^{(2)l+l'} |\psi_{0}\rangle \tag{4}
\]

In the foregoing equation, \(\sigma_{\text{XNCD}}(\omega) = \sigma_{\text{X}}^{\text{a}}(\omega) - \sigma_{\text{X}}^{\text{b}}(\omega)\) denotes the cross section for \(E1-E2\) circular dichroism in the x-ray region (\(X\)), with
\[
\sigma_{\text{X}}^{\text{a}}(\omega) = 2\pi^{2}a\hbar \omega [i \sum_{f,f'} \langle g | e^{*} \cdot r_{i} | f \rangle \langle f | e \cdot r_{i} \cdot k \cdot r_{f'} | g \rangle + \text{c.c.}] \delta(E_{f} - E_{g} - \hbar \omega); \tag{5}
\]
\(\hbar \omega\), \(k\) and \(e^{\pm} = \mp (i/\sqrt{2})(\epsilon_{1} \pm i \epsilon_{2})\) represent energy, wave vector, and circular polarisations of the photon, respectively; \(\alpha = e^{2}/\hbar \epsilon\). The integral terms are given by \(R_{l,l'}^{(1)} = \int_{0}^{\infty} dt_{e} \varphi_{l}(r) r^{L+2} \varphi_{l}(r)\), where \(\varphi_{l}(r)\) and \(\varphi_{l}(r)\) denote inner-shell and valence radial wave functions, respectively [11,12].

A general expression can be given for the wave-vector and polarisation responses used throughout this work. It reads: \(i^{z}T_{q}^{(k)}(e^{*}, e, k)z\), where
\[
T_{q}^{(k)}(e^{*}, e, k)z = \sum_{y} \sqrt{(2z+1)(2y+1)}(-1)^{1+k} \tag{6}
\]
\[
\{\begin{array}{c}
1 \ k \ y \\
1 \ 1 \ z
\end{array}\} \sum_{\eta \beta} C_{\eta \beta}^{yq} C_{\eta \beta}^{qy} Y_{1 \eta}(e^{*}) Y_{1 \alpha}(e) Y_{1 \beta}(k),
\]
with \(z = 1, 2\) denoting \(E1-M1\) (imaginary) and \(E1-E2\) (real) responses, respectively; \(k = k/k\). Explicit forms are given in Table [1].

\(E1-E2\) interference: XNRLD \[3\] One effective operators is found in this case: a space and time odd tensor of rank three, i.e., an orbital septor. This unusual magnetic moment stems from a coupling of the orbital anapole with the quadrupolar moment of \(L\) and takes the form \([L \otimes L]_{q}^{(3)l+l'}\).

The integrated XNRLD spectrum can thus be written as
\[
\int_{j_{-}+j_{+}} \frac{\sigma_{\text{XNRLD}}(\omega)}{(\hbar \omega)^{2}} d(\hbar \omega) = \frac{8\pi^{2}a}{3h \epsilon_{c}} (2\epsilon_{c} + 1) \sum_{l,l'=l \pm 1} R_{l,l'}^{(2)}
\]
\[
R_{l,l'}^{(2)} \langle l',l \rangle \sum_{q} \left[T_{q}^{(3)l}(\epsilon^{*}, k)_{2} - T_{q}^{(3)l}(\epsilon, k)_{2}\right] \tag{7}
\]
\[
\langle \psi_{0}| \sum_{l} [L_{l} \otimes L_{l}]^{2} (L \times n - n \times L)_{l}^{(3)l+l'} |\psi_{0}\rangle,
\]

where \(\sigma_{\text{XNRLD}}(\omega) = \sigma_{\text{X}}^{b}(\omega) - \sigma_{\text{X}}^{a}(\omega)\) stands for the cross section for \(E1-E2\) linear dichroism; here, \(\parallel\) and \(\perp\) denote two orthogonal linear-polarisation states [12].

Notice that the \(E1-E2\) interference contains a further magnetic term, which vanishes in the geometry of Eq. (3). It is an orbital vector, with polarisation response \(T_{q}^{(1)l}(\epsilon, k)z\). In our formulation, it is given by \(\sum_{l} \langle [L_{l} \times n - n \times L_{l}]_{l}^{(1)l+l'} \rangle\), i.e., by the orbital anapole. This toroidal contribution \(\parallel\) to the orbital current (see below) could be detected by the following experiment. Absorb linearly polarised x rays at the 'magic angle' \(\vec{k}B \approx 39.23^{\circ}\) for parallel and antiparallel magnetoelectric annealing \([3]\) of the sample; \(B \equiv \text{external magnetic field}\). Subtract the two spectra.

Stemming from parity nonconserving hybridisation of the valence electrons, the effect should be easier to detect than its nuclear counterpart \([17]\), as the work of Ref. [3] appears to indicate.

\(E1-M1\) interference. As previously stated, our approach readily extends to cover the visible range, where the \(E1-M1\) interference is predominant. In this case (for optical natural circular dichroism (ONCD) takes the form: \(\sigma_{\text{ONCD}}(\omega) = \sigma_{\text{\Omega}}^{a}(\omega) - \sigma_{\text{\Omega}}^{b}(\omega)\), with
\[
\sigma_{\text{\Omega}}^{a}(\omega) = \frac{2\pi^{2}a}{m^{2}} \sum_{f,f'} \langle g | e^{*} \cdot r_{i} | f \rangle \langle f | e \cdot r_{i} \cdot k \cdot l_{f'} | g \rangle + \text{c.c.}] \delta(E_{f} - E_{g} - \hbar \omega), \tag{8}
\]
\(\langle f | e \times k \cdot l_{f'} | g \rangle + \text{c.c.}] \delta(E_{f} - E_{g} - \hbar \omega), \tag{9}
\)
\(l = h \mathbf{L}\).

For the integrated ONCD spectrum we obtain
\[
\int_{j_{-}+j_{+}} \frac{\sigma_{\text{ONCD}}(\omega)}{(\hbar \omega)^{2}} d(\hbar \omega) = \frac{4\pi^{2}a}{3mc} \sum_{l,l'=l \pm 1} R_{l,l'}^{(1)}
\]
\[
d(l,l') \langle \psi_{0}| \left\{ \sqrt{\frac{2}{3}} T_{0}^{(0)}(\epsilon^{*}, \epsilon, \hat{k}) + \hat{k}_{1} \right\} \right| \langle \psi_{0}| \tag{9}
\]}
\[
\sum_{l \neq l'} \left[ L_l \cdot (L \times n - n \times L)_{l'} \right]^{l+l'-1} - \sum_q \sqrt{2T_q} (e^+, e^-, \hat{k}),
\]

\[
\sum_{l, l'} \left[ L_l \otimes (L \times n - n \times L)_{l'} \right]^{l+l'-1}_{q}\}
\]

\[
\psi_0,
\]

with \(d(l, l+1) = 1/(l+1)\) and \(d(l, l-1) = -1/l\). Two irreducible tensors are thus associated to ONCD: a two-particle orbital pseudoscalar, which identifies chirality (see below), and a orbital pseudoelevator; the latter generalises the results of Eq. (1) to the two-particle case and will not be discussed any further. (There is an important difference between E1-E2 and E1-M1 involved dichroic spectra. E1-E2 implies excitations from inner shells, which are filled in the ground state and can therefore be 'integrated out'. As a result, one-electron contributions, and thus providing a physical basis to our consideration.

Interpretation of the results. The choice of \(L\) and \(\Omega^-\) as the building blocks of our analysis is physically motivated as follows. As both generators are magnetic, they are expected to contribute to the electron orbital current, a conjecture which is readily verified. To this purpose, consider Trammell's expansion of the Fourier transform of the atomic orbital magnetisation density [23]

\[
L(k) = \frac{1}{2} \langle \psi_0 | \sum_i [L_i f(k \cdot r_i) + f(k \cdot r_i) L_i] | \psi_0 \rangle,
\]

and perform a recoupling with use of [23]:

\[
f(k \cdot r) = 4\pi \sum_i g_i(kr) Y_{lm}(n) Y_{lm}^*(k),
\]

with \(g_i(x) = (2/x^2) \int_{0}^{\pi} d\xi j_i(\xi)\), where \(j_i(\xi)\) denotes a spherical Bessel function.

We have

\[
L(k) = \sum_i \langle \psi_0 | \left[ g_0(kr_i) L_i + 3 g_1(kr_i) \right] / 2
\]

\[
(\hat{k} \times (L_i \times n_i - n_i \times L_i) / 2 + \cdots ) | \psi_0 \rangle,
\]

displaying the orbital angular momentum and anapole contributions, and thus providing a physical basis to our chosen so(3,1) SGA, which is realised by \(L\) and \(\Omega^-\).

The representations of the homogeneous Lorentz group, which are identified by a pair of indexes, \(\nu\) and \(\rho\), afford a classification of electronic states in noncentrosymmetric systems. Unitary (unirreps) and nonunitary (nonunirreps) irreducible representations will be considered.

It is readily seen [24] that \(\Omega^-\) corresponds to having \(\nu = \rho = 0\), yielding the unirrep \(\mathcal{D}(\nu = 0, \rho = 0) = \sum_{l=0,1,\ldots} \mathcal{D}^l\) ("supplementary" series), with basis \(\sum_{l=0,1,\ldots} |lm\rangle\), thus confirming that we are dealing with a spinless case. (Here, \(\mathcal{D}^l\) identifies the representations of the rotation group.) Notice that the spherical harmonics are eigenstates not only of the so(3) invariant (Casimir) \(\mathcal{L}^2\), but also of the corresponding so(3,1) invariants \(\mathcal{L}^2 - (\Omega^-)^2\) and \(\mathcal{L} \cdot \Omega^-\). The latter, with eigenvalues \(\nu \rho\), identifies the chiral operator and evinces the deep interweaving of the Lorentz group with chirality. As \(\mathcal{L} \cdot \Omega^- = 0\) [Eq. (1)], orbital chirality cannot manifest itself at the one-particle level.

We can build a two-particle basis using the same so(3,1) SGA. The generators are given by \(L_T = L_1 + L_2\) and \(\Omega_T = \Omega_1^- + \Omega_2^-\), in this case. In such a basis, the two-particle states are eigenstates of the so(3,1) invariants; in particular of \(\mathcal{L} \cdot \Omega_T\), the chiral operator that emerges from the E1-M1 processes [Eq. (1)]. The Clebsch-Gordon series for the direct product of two \(\mathcal{D}(0,0)\) representation takes the form [22]:

\[
\mathcal{D}(0,0) \otimes \mathcal{D}(0,0) = \sum_{\nu=0}^{\infty} \int\mathcal{D}(\nu,\rho) d\mu(\nu,\rho).
\]

The direct product thus decomposes into a direct sum of representations some of which characterised by \(\nu \rho \neq 0\), i.e. orbital chirality appears naturally when considering two-particle properties.

From the relations between the so(3,1) Clebsch-Gordon coefficients [24], it is readily seen that states with \(\nu \rho \neq 0\) have mixed parity. For a given value of \(\nu|\rho| \neq 0\), there are two inequivalent representations with basis states of the form \([\mathcal{D}(0,0)]\), with eigenvalues \(j + 1\) and \(j\) (\(j\)'s values), respectively; \(j\)'s take integer or half-integers values. These nonunirreps are denoted by \(\mathcal{D}(J^J)\). The representation space is spanned by the \((2j+1)(2j'+1)\) basis vectors \([jm]\)\([j'm']\) (canonical basis). [All nonunirreps for which \(j + j'\) is an integer are true representations of SO(3,1), while those for which \(j + j'\) is half-integer are double-valued.] Observe that \(\mathcal{N}\) and \(\mathcal{N}^\dagger\) are not independent as they can be interchanged by space inversion. This is equivalent to swapping \(j \leftrightarrow j'\). Notice that \(\mathcal{L} \cdot \Omega^- = 0\) implies \(j = j'\). Again, orbital chirality does not appear at the one-particle level.

To illustrate the use of nonunirreps in constructing two-electron states, we resort to the simple case \(\mathcal{D}(\mathcal{J}^\mathcal{J}) \otimes \mathcal{D}(\mathcal{J}^\mathcal{J})\). We have

\[
\Psi_{MM'}^{LL'}(1,2) = \sum_{\alpha \sigma} C^{LM}_{\alpha1,\sigma1} C^{LM}_{\alpha2,\sigma2} \psi_{\sigma1,\sigma1'}(1) \psi_{\sigma2,\sigma2'}(2),
\]

with \(\psi_{\sigma,\sigma'} = |\frac{1}{2}\sigma\rangle \langle \frac{1}{2}\sigma'|\). In this basis, the two-particle chiral operator takes the form: \(\mathcal{L}_T \cdot \mathcal{\Omega}_T = \mathcal{L} \cdot \mathcal{\Omega} = \mathcal{L} \cdot \mathcal{\Omega}_T = 0\).
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A symmetry analysis based on homogeneous Lorentz
group has indicated that, in the presence of inversion-
symmetry-breaking hybridisation, valence electrons de-
velop an orbital anapole moment. The coupling of such a
moment to the orbital angular momentum yields a set of
microscopic effective operators, which identify the elec-
tronic properties probed by x-ray and optical dichroism
(E1-E2 and E1-M1 interferences).

It has also been shown that orbital chirality (enantio-
morphism) is described by a two-electron so(3, 1) in-
variant (pseudoscalar), which is measured by optical nat-
ural circular dichroism.

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with
b_{l}(l_{c}, l) = \frac{2(l + 1)(l’ + 1)}{h(l, l’)(l_{c} - 3l’ + 2l)(l_{c} + 3l’ - 2l + 1)h(l_{c}, l)},

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TABLE I. Polarisation responses of E1-E2 and E1-M1 in-
terferences.

| E1-E2 | E1-M1 |
|-------|-------|
| iT_0^{(0)}(\epsilon^*, \epsilon, \hat{k})_1 = -iT_0^{(0)}(\epsilon, \epsilon^*, \hat{k})_1 = -i/\sqrt{6}, |
| iT_q^{(2)}(\epsilon^*, \epsilon, \hat{k})_1 = -iT_q^{(2)}(\epsilon, \epsilon^*, \hat{k})_1 = -i/2[|\epsilon^* \otimes \epsilon|^{(1)} \otimes \hat{k}]^{(2)}, |
| T_q^{(1)}(\epsilon, \hat{k})_2 = T_q^{(1)}(\epsilon^*, \hat{k})_2 = -1/2\sqrt{2}\hat{k}q, |
| T_q^{(2)}(\epsilon^*, \epsilon, \hat{k})_2 = -T_q^{(2)}(\epsilon, \epsilon^*, \hat{k})_2 = \frac{\sqrt{5}}{2}[|\epsilon^* \otimes \epsilon|^{(1)} \otimes \hat{k}]^{(2)}, |
| T_q^{(3)}(\epsilon, \hat{k})_2 = [[|\epsilon^* \otimes \epsilon|^{(1)} \otimes \hat{k}]^{(2)} \otimes \hat{k}]^{(3)} \neq T_q^{(3)}(\epsilon^*, \hat{k})_2 = [[|\epsilon^* \otimes \epsilon|^{(1)} \otimes \hat{k}]^{(2)} \otimes \hat{k}]^{(3)} |