

Geometry of chiral temporal structures. II. The formalism

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We develop a mathematical formalism underlying the emergence of enantiosensitive molecular orientation due to photoionization or photoexcitation of chiral molecules. We consider geometric quantities such as the Berry connection and Berry curvature in light-driven chiral electronic states in the space of complex light polarization vectors. The parametric dependence of the light-driven electronic wave function on such vectors is based on various possible mutual orientations between the laser field and a chiral molecule. Using the tools of differential geometry, we show how the enantiosensitive observables arise from the geometry of the molecular response in such spaces.

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I. INTRODUCTION

Light-induced ultrafast electronic currents in chiral molecules [1,2] can trace temporal chiral structures even when the driving laser field itself is achiral. A chiral temporal structure is a trajectory that cannot be superimposed on its mirror image [3]. Inspired by the emergence of geometric properties in the photoionization of chiral molecules [4,5], we develop a unified approach to understanding and controlling enantiosensitive geometric observables based on the analysis of temporal shapes of chiral electron currents. Our framework is naturally geometric, grounded in the formalism of fiber bundle theory [6,7] over complex polarization spaces [8]. These complex vector spaces describe the polarization of the driving laser field and allow a differential-geometric description of enantiosensitive phenomena. In this setting, the wave function defines a section of a fiber bundle whose Berry connection and Berry curvature yield physical observables.

Our generalized approach reveals strategies for controlling geometric quantities such as the Berry curvature and Berry connection through multiphoton processes driven by polarization-shaped pulses targeting either geometries of bound states via photoexcitation or continuum states via photoionization or both. These opportunities are described in our companion paper [9]. Here, we focus on using the tools of

differential geometry [10,11] to derive the Berry connection and the Berry curvature in complex vector spaces to predict and analyze new enantiosensitive observables of geometric origin.

Choosing the electric field vector space as the parameter space emphasizes physical intuition, even though the orientation of the molecule remains the fundamental control parameter. This choice enables a clear geometric formulation of enantiosensitive responses and allows us to define a Berry connection and Berry curvature in the space of light polarization vectors. The orientation dependence of the wave function originates entirely from the dipole interaction. In the molecular frame, the electric field polarization vector can be seen as a tangent vector being parallel transported over the space of molecular orientations as the molecule rotates. For circular polarization, this space is reduced to the sphere S^2 . The polarization vector can be naturally viewed as an element of the complexified tangent space to the sphere, and thinking of the Berry connection and Berry curvature in terms of this complex tangent vector makes the geometric nature of the phase explicit. The geometric phase of the wave function can thus be related to the phase acquired by a complex vector undergoing parallel transport along the sphere. The $U(1)$ phase reflects the rotation of this complex tangent vector within the tangent plane.

In the following sections, we develop this geometric framework explicitly and show how key physical observables—such as molecular orientation or circular dichroism—are encoded in the Berry curvature and Berry connection defined over these complex vector spaces.

II. KEY GEOMETRIC QUANTITIES RELEVANT FOR ENANTIOSENSITIVE OBSERVABLES

The investigation of the wave-function dynamics in terms of the geometric details of polarization vector dynamics

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connects to general photoinduced processes independent of a specific molecular setup or field amplitude and is thus applicable to a broad bandwidth of phenomena coupling to polarization vector dynamics, such as molecular rotations (in the laboratory frame of reference) or three-dimensionally structured light-fields (synthetic chiral light). For such setups, it does not matter whether the light-matter coupling takes place in the strong- or weak-coupling regime, or is of pulsed or periodic character. As introduced in the companion paper [9], our analysis of the geometric structure of the manifold of temporal shapes in the space of electric field polarization vectors $\mathbf{e}$ reveals two key quantities. The first one is the geometric phase [12]:

$$S = \oint i \langle \psi(\mathbf{e}) | \nabla_{\mathbf{e}} \psi(\mathbf{e}) \rangle \cdot d\mathbf{e} = \oint \mathbf{A}(\mathbf{e}) \cdot d\mathbf{e}, \quad (1)$$

revealing the Berry connection vector defined as

$$\mathbf{A}(\mathbf{e}) = i \langle \psi(\mathbf{e}) | \nabla_{\mathbf{e}} \psi(\mathbf{e}) \rangle. \quad (2)$$

This vector quantifies the sensitivity of the electronic wave function $\psi[t, \mathbf{e}(\rho), \mathbf{r}]$ to changes in the orientation of the laser field polarization $\mathbf{e}(\rho)$ in the molecular frame, and ρ describes Euler angles quantifying the orientation of the molecular frame with respect to the laboratory frame. The wave function $\psi[t, \mathbf{e}(\rho), \mathbf{r}]$, expressed in the molecular frame, is the solution of the time-dependent Schrödinger equation for a fixed relative orientation ρ between the molecule and the laser field:

$$i \frac{\partial}{\partial t} \psi(\mathbf{r}, \rho, t) = [H_{el} + \mathbf{r} \cdot \mathbf{E}(\rho, t)] \psi(\mathbf{r}, \rho, t). \quad (3)$$

The vector $\mathbf{A}(\mathbf{e})$ “connects” wave functions at $\psi[\mathbf{r}, \mathbf{e}(\rho), t]$ and $\psi[\mathbf{r}, \mathbf{e}(\rho + d\rho), t]$. The second relevant geometric quantity is a pseudovector, defined as the curl of the Berry connection in the space of electric field polarization vectors:

$$\mathbf{\Omega}_a = \nabla_{\mathbf{e}} \times \mathbf{A} = i \langle \nabla_{\mathbf{e}} \psi(\mathbf{e}) | \times | \nabla_{\mathbf{e}} \psi(\mathbf{e}) \rangle. \quad (4)$$

Equations (2) and (4) highlight the dynamical, laser-driven origin of these geometric quantities.

The pseudovector $\mathbf{\Omega}_a$ identifies a well-defined direction in the molecular frame. In our companion paper [9] we show that the direction of this vector determines the orientation of a molecular cation upon two-photon ionization of a randomly oriented ensemble of molecules and thus it is a directly accessible experimental observable. As such, it deserves a dedicated analysis that we perform in this paper (Secs. III–V). In a real-valued, three-dimensional parameter space the curl of the vector potential, i.e., the pseudovector $\mathbf{\Omega}$, corresponds to the Berry curvature. In the following, we show that in complex vector spaces the Berry curvature tensor gets additional components.

III. THE EMERGENCE OF THE BERRY CURVATURE IN ONE-PHOTON IONIZATION

In order to demonstrate the concepts introduced in the previous section, in this section we limit the discussion to a specific excitation scheme. Let us consider one-photon ionization from an initial state ψ_0 by circularly polarized light to follow the emergence of the Berry curvature. We can use

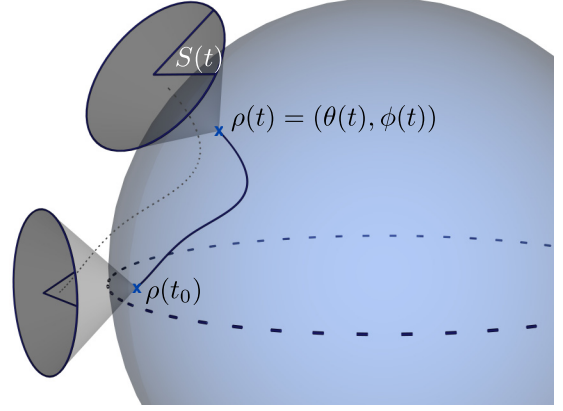


FIG. 1. The fiber bundle $\mathcal{H} \xrightarrow{\pi} S^2$. Each point in the projective base manifold (sphere) S^2 is associated with a fiber (drawn as circles) of the Hilbert space $\mathcal{H}$, which is determined by wave function $e^{iS}|\psi\rangle$. The angle S parametrizes the fiber, as can be seen for the two points $\rho(t_0)$ and $\rho(t)$ indicated by blue markers. Throughout a time evolution in the base space (solid line), a horizontal [13] motion is given by the phase $S(t)$ Eq. (19) (dotted line).

perturbation theory to calculate $|\psi(\mathbf{e})\rangle$:

$$|\psi\rangle = |\psi_0\rangle + \int d\Theta_k a_k(\mathbf{e}) |\psi_k\rangle \quad (5)$$

and thus evaluate the Berry connection explicitly. Here, $d\Theta_k = \sin\theta_k d\theta_k d\phi_k$ and θ_k, ϕ_k are angles characterizing the direction of the photoelectron momentum $\mathbf{k}$ for fixed $k = |\mathbf{k}|$, while $a_k = -\tilde{E}_{\omega_k}(\mathbf{D} \cdot \hat{\mathbf{e}})$ are the amplitudes of the continuum states and $\tilde{E}_{\omega_k}$ is the Fourier transform of the electric field amplitude at the transition frequency. The polarization of the laser field in the molecular frame is characterized by a complex unit vector $\hat{\mathbf{e}} = \frac{1}{\sqrt{2}}(\hat{\boldsymbol{\theta}} + i\sigma\hat{\boldsymbol{\phi}})$ that encodes the direction of field rotation $\sigma = \pm 1$. Each cartesian component e^j is a function defined on the 2-sphere (see Fig. 1). The Berry connection becomes

$$\mathcal{A} = i \langle \psi | d\psi \rangle = i \int d\Theta_k (\mathbf{D} \cdot \hat{\mathbf{e}})^* (\mathbf{D} \cdot d\hat{\mathbf{e}}). \quad (6)$$

We can expand the exterior derivative of the vector $\hat{\mathbf{e}}$ in terms of the unit cartesian basis vectors $\hat{\mathbf{j}}$ in the molecular frame,

$$d\hat{\mathbf{e}} = (de^j)\hat{\mathbf{j}}. \quad (7)$$

In this framework, the Berry curvature is the exterior derivative of the Berry connection $\Omega = d\mathcal{A}$:

$$\Omega = i |\tilde{E}_{\omega_k}|^2 \int d\Theta_k D_i^* D_j de^{i*} \wedge de^j. \quad (8)$$

Note that in Eq. (8) the Berry curvature is the full contraction of a molecular tensor—the dipole tensor $\mathbf{\Omega}^{ij} = i \int d\Theta_k D_i^* D_j$ —and the light polarization 2-form $de^{i*} \wedge de^j$. Since the dipole tensor is anti-Hermitian it may be partitioned into a symmetric (imaginary) and an antisymmetric (real) matrix,

$$\mathbf{\Omega}_s^{ij} \equiv \frac{i}{2} |\tilde{E}_{\omega_k}|^2 \int d\Theta_k (D_i^* D_j + D_j^* D_i), \quad (9)$$

$$\mathbf{\Omega}_a^{ij} \equiv \frac{i}{2} |\tilde{E}_{\omega_k}|^2 \int d\Theta_k (D_i^* D_j - D_j^* D_i), \quad (10)$$

such that $\Omega^{ij} = \Omega_s^{ij} + \Omega_a^{ij}$. The real part of the dipole tensor is vectorizable and can be expressed in terms of a cross product,

$$\Omega_a^{ij} = \frac{i}{2} |\tilde{E}_{\omega_k}|^2 \epsilon^{lij} \int d\Theta_k (\mathbf{D}^* \times \mathbf{D})_l, \quad (11)$$

where ϵ^{lij} is the Levi-Civita symbol.

On the basis of Eq. (11), we obtain the expression for the antisymmetric part of the Berry curvature:

$$\begin{aligned} \Omega_a &= \Omega_a^{ij} de^{i*} \wedge de^j \\ &= \frac{i}{2} |\tilde{E}_{\omega_k}|^2 \epsilon^{lij} \int d\Theta_k (\mathbf{D}^* \times \mathbf{D})_l de^{i*} \wedge de^j. \end{aligned} \quad (12)$$

It is now natural to define the *polarization Berry curvature 2-forms*,

$$d\xi^l \equiv \frac{1}{2} \epsilon^{lij} de^{i*} \wedge de^j, \quad (13)$$

and their molecular counterparts,

$$\Omega_a \equiv i |\tilde{E}_{\omega_k}|^2 \int d\Theta_k \mathbf{D}^* \times \mathbf{D}, \quad (14)$$

such that the antisymmetric contribution to the Berry curvature can be expressed as a scalar product,

$$\Omega_a = \Omega_a \cdot d\xi, \quad (15)$$

We deduce a very important result: the antisymmetric component of the Berry curvature tensor Ω reduces, when paired with the polarization differential $de^{i*} \wedge de^j$, to the scalar product of a molecular pseudovector and a surface element $d\xi$. This implies that Ω_a specifies a well-defined and molecular-specific direction in the molecular frame. Its physical meaning has been identified in our earlier work [5]. Specifically, Ω_a is equivalent to the net propensity field [4,5,14–16]. Its projection on photoelectron spin quantifies circular dichroism in one-photon ionization [5]. Its direction quantifies enantiosensitive orientation of molecular cations upon photoionization from a current-carrying state [5]. This pseudovector also plays an important role in spin-orientation locking in chiral molecules. However, the symmetric counterpart of the Berry curvature cannot be reduced to a cross product,

$$\Omega_s = \Omega_s^{ij} de^{i*} \wedge de^j. \quad (16)$$

Note that the symmetric and antisymmetric parts of the Berry curvature arise from the following components of the molecular dipole tensor:

$$\Omega_s^{ij} = |\tilde{E}_{\omega_k}|^2 \int d\Theta_k \text{Im}\{iD_i^* D_j\} = \text{Re}\langle \partial_{e^i} \psi | \partial_{e^j} \psi \rangle \quad (17)$$

$$\Omega_a^{ij} = |\tilde{E}_{\omega_k}|^2 \int d\Theta_k \text{Re}\{iD_i^* D_j\} = -\text{Im}\langle \partial_{e^i} \psi | \partial_{e^j} \psi \rangle. \quad (18)$$

Usually only the imaginary part $\text{Im}\langle \partial_{e^i} \psi | \partial_{e^j} \psi \rangle$ of a tensor $\langle \partial_{e^i} \psi | \partial_{e^j} \psi \rangle$ contributes to the Berry curvature (see, e.g., Ref. [17]), however in our case $\text{Re}\langle \partial_{e^i} \psi | \partial_{e^j} \psi \rangle$ also contributes because our base space involves complex polarization vectors. Thus, using a simple example of one-photon ionization we established that Eq. (4) is associated with the antisymmetric part of the molecular Berry curvature Eq. (18). In the next section, we extend the discussion to a general wave function to confirm our conclusions.

IV. GEOMETRIC QUANTITIES ASSOCIATED WITH CIRCULAR POLARIZATION VECTORS

The concepts we introduced in the previous sections find their place in the comprehensive research field of geometric phases in quantum systems. Thus, the understanding of the introduced observables may be deepened by a mathematical classification. A generalized geometric picture arising from the dynamics of circular polarization vectors is given within the framework of fiber bundle theory. Such frameworks are often used in order to investigate quantum systems that either depend on an adiabatic parameter set [10,18,19] or undergo a cyclic evolution [7,20,21]. We partition the Hilbert space $\mathcal{H}$ of pure-state, normalized wave functions $|\psi(\mathbf{e})\rangle$ into a set of fibers $\mathcal{H}_\rho = \{e^{iS} |\psi(\mathbf{e}(\rho))\rangle | S \in (-\pi, \pi]\}$ that consist of wave functions which are identical up to a $U(1)$ -phase factor e^{iS} . By further defining the projection map $\pi(e^{iS} |\psi(\mathbf{e}(\rho))\rangle) = \frac{1}{2i} \mathbf{e}^* \times \mathbf{e}$, we obtain the unit sphere S^2 parametrized by the angles $\rho = (\theta, \phi)$ as projective space (or base manifold). The construction of a total space, here the Hilbert space, endowed with a projective base manifold (S^2) and a projection map π , is referred to as the fiber bundle $\mathcal{H} \xrightarrow{\pi} S^2$. A schematic visualization is given in Fig. 1. Here, for two points on the sphere S^2 the corresponding fibers $U(1)$ are visualized as circles. The wave function $|\psi(t)\rangle$, the parameters $\rho(t)$, and the phase $S(t)$ evolve in time. The temporal evolutions of $\rho(t)$ and $S(t)$ are illustrated by the solid and dotted lines in Fig. 1, respectively. Thus, we distinguish between two kinds of dynamics in the total Hilbert space $\mathcal{H}$. A movement within one fiber, i.e., a change of phase, is referred to as vertical. This motion is fully determined by the Lie group action of the unitary group $U(1)$ on the Hilbert space and, thus, extends the introduced bundle into a principal fiber bundle. A definition of horizontal motion can be obtained using the Berry connection form $\mathcal{A}$ [10]. The horizontal motion is given by the phase

$$S(t) = S(t_0) + i \int_{\rho(t_0)}^{\rho(t)} \mathcal{A}. \quad (19)$$

The Berry connection form can be expressed in terms of the complex electric field polarization vector. Let the complex Wirtinger derivatives $\partial_{e_i} = \frac{1}{2}(\frac{\partial}{\partial \text{Re} e_i} - i \frac{\partial}{\partial \text{Im} e_i})$ and $\partial_{e_i^*} = \frac{1}{2}(\frac{\partial}{\partial \text{Re} e_i} + i \frac{\partial}{\partial \text{Im} e_i})$ be the components of ∇_e and ∇_{e^*} , respectively. In terms of the polarization vector, the Berry connection form reads [22,23]

$$\mathcal{A} = i \langle \psi | d\psi \rangle = i \langle \psi | \nabla_e \psi \rangle \cdot d\mathbf{e}, \quad (20)$$

where the differentials of the polarization vector are $de_i = d\boldsymbol{\rho} \cdot \nabla_{\boldsymbol{\rho}} e_i = d\theta \frac{\partial}{\partial \theta} e_i + d\phi \frac{\partial}{\partial \phi} e_i$. Here, we implicitly assumed the wave function to be a holomorphic section, i.e., $|\nabla_{e^*} \psi\rangle = 0$ [24]. This is in accordance with the perturbative approach of Sec. III. The Berry curvature form Ω is the exterior derivative of the Berry connection

$$\Omega = d\mathcal{A} = i \langle \partial_{e_i} \psi | \partial_{e_j} \psi \rangle de_i^* \wedge de_j. \quad (21)$$

Indeed, this expression is vectorizable in such a way that it nicely connects to physical observables such as the photoionization yield [9]. To this end, we define two bilinear vector

products,

$$\begin{aligned} (\mathbf{a} \diamond \mathbf{b})_i &\equiv |\varepsilon_{ilm}| a_l b_m, \\ (\mathbf{a} \odot \mathbf{b})_i &\equiv \delta_{il} a_l b_l. \end{aligned} \quad (22)$$

The full Berry curvature, then, is

$$\begin{aligned} d\mathcal{A} &= i d\boldsymbol{\xi} \cdot \langle \nabla_e \psi | \times | \nabla_e \psi \rangle + i d\boldsymbol{\zeta} \cdot \langle \nabla_e \psi | \odot | \nabla_e \psi \rangle \\ &+ i d\boldsymbol{\chi} \cdot \langle \nabla_e \psi | \odot | \nabla_e \psi \rangle, \end{aligned} \quad (23)$$

where we defined the polarization differentials

$$d\zeta^l \equiv \frac{1}{2} |\varepsilon^{lij}| d\mathbf{e}^{i*} \wedge d\mathbf{e}^j, \quad d\chi^l \equiv \delta_{li} d\mathbf{e}^{i*} \wedge d\mathbf{e}^i. \quad (24)$$

All the above differentials have natural representations on the unit sphere in terms of the Euler angles ρ ,

$$d\boldsymbol{\xi} = d\theta \wedge d\phi \operatorname{Re} \left[\frac{\partial \mathbf{e}^*}{\partial \theta} \times \frac{\partial \mathbf{e}}{\partial \phi} \right], \quad (25)$$

$$d\boldsymbol{\zeta} = d\theta \wedge d\phi i \operatorname{Im} \left[\frac{\partial \mathbf{e}^*}{\partial \theta} \diamond \frac{\partial \mathbf{e}}{\partial \phi} \right], \quad (26)$$

$$d\boldsymbol{\chi} = d\theta \wedge d\phi 2i \operatorname{Im} \left[\frac{\partial \mathbf{e}^*}{\partial \theta} \odot \frac{\partial \mathbf{e}}{\partial \phi} \right]. \quad (27)$$

A detailed derivation of the Berry curvature is given in Appendix A. Conclusively, the Berry curvature derived above quantifies the impact of the polarization vector dynamics on the phase of a pure-state wave function. Physically, it dictates the accumulation of the Berry phase and its antisymmetric component, and when applied to ionization experiments, it further determines the photoionization yield.

V. BERRY PHASE AND ANTISYMMETRIC BERRY CURVATURE IN TWO-FIELD PUMP-PROBE EXPERIMENTS

The formalism introduced in Sec. IV is extendable to pump-probe experiments, where a primary (pump) light field excites the target molecule and a secondary (probe) field subsequently ionizes it. The crucial difference for such experiments is that the two laser fields can have different polarization vectors. We summarize the geometric ingredients that arise when a pump-probe scheme is driven by two laser fields with circular $\mathbf{e} = \frac{1}{\sqrt{2}}(\hat{\boldsymbol{\theta}} + i\sigma\hat{\boldsymbol{\phi}})$ and a linear $\boldsymbol{\varepsilon} = \cos\alpha\hat{\boldsymbol{\theta}} + \sin\alpha\hat{\boldsymbol{\phi}}$ polarization vector. The angle α implies a phase shift between the fields. All derivational details are collected in Appendix B.

The Berry connection 1-form on the joint field manifold $(\mathbf{e}, \boldsymbol{\varepsilon})$ is

$$\mathcal{A} = i \langle \psi | \nabla_{(\mathbf{e}, \boldsymbol{\varepsilon})} \psi \rangle \cdot d(\mathbf{e}, \boldsymbol{\varepsilon}), \quad (28)$$

which splits naturally into the circular- and linear-field contributions

$$\mathcal{A} = \mathcal{A}_e + \mathcal{A}_\varepsilon \quad (29)$$

and yields a geometric phase

$$S = \oint (\mathcal{A}_e + \mathcal{A}_\varepsilon). \quad (30)$$

Taking an exterior derivative gives the Berry curvature 2-form $\Omega = d\mathcal{A}$. Its antisymmetric part decomposes into

$$\Omega_a = \Omega_a^e + \Omega_a^\varepsilon + \Omega_a^{\text{cross}}, \quad (31)$$

with the compact vector formula

$$\Omega_a^e = (\nabla_{\mathbf{e}^*} \times \mathbf{A}_e) \cdot d\boldsymbol{\xi}_e = \boldsymbol{\Omega}_a^e \cdot d\boldsymbol{\xi}_e, \quad (32)$$

which coincides with the expression for the one-field antisymmetric Berry curvature. The linear field polarization vector is real valued and thus its Berry curvature is fully antisymmetric:

$$\Omega^\varepsilon = \Omega_a^\varepsilon = (\nabla_\varepsilon \times \mathbf{A}_\varepsilon) \cdot d\boldsymbol{\xi}_\varepsilon = \boldsymbol{\Omega}_a^\varepsilon \cdot d\boldsymbol{\xi}_\varepsilon. \quad (33)$$

The mixed Berry curvature can be written as

$$\Omega_a^{\text{cross}} = -\operatorname{Im}\{\langle \nabla_\varepsilon \psi | \times | \nabla_e \psi \rangle \cdot d\boldsymbol{\xi}_{\text{cross}}\}. \quad (34)$$

Thus, by taking into account the geometric interplay of two polarization vectors, the geometric description of temporal polarization vector dynamics introduced in Sec. IV is extendable to setups incorporating two independent electric fields, such as pump-probe experiments. Neither the linear-field Berry curvature nor the mixed Berry curvature are relevant to isotropic signals (i.e., independent of the third Euler angle α distinguishing directions in polarization plane and breaking cylindrical symmetry) considered in our companion paper [9], and will be analyzed in a separate publication, where all three Euler angles will be taken into account.

VI. CONCLUSION

We have developed a unified geometric framework that highlights the interplay of geometric light-polarization dynamics and the photon-induced processes in chiral molecules. By treating the electronic wave function as a section of a $U(1)$ bundle over the space of molecular orientations, we can identify the fiber bundle connection and the antisymmetric Berry curvature as the fundamental geometric objects governing enantiosensitive observables. By applying the formalism to one-photon ionization, we have shown that the antisymmetric part of the Berry curvature reduces to the net molecular propensity field, whose direction dictates the net orientation of the ionized ensemble [9]. In our companion paper we exploit the geometric framework of polarization vectors, where the Berry curvature describes a photoionization-driven enantiosensitive orientation of the molecular cations and the Berry connection governs the accumulation of geometric phases during rotational time evolutions. Taken together, the two studies demonstrate that the bundle curvature and connection provide a common language for describing both the enantiosensitive yield [5] and the orientation of the cation. We have shown that the present formalism is readily generalized to multiphoton processes and pump-probe schemes. Since all observables follow from geometric quantities, the framework is conceptually transparent, making it a versatile tool for designing experiments and interpreting ultrafast chiral phenomena. We anticipate that further exploration of polarization-space geometry will provide control over singularities or nontrivial topologies that can be harnessed to amplify enantioselective signals.

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DATA AVAILABILITY

No data were created or analyzed in this study.

APPENDIX A: BERRY CURVATURE IN POLARIZATION VECTOR SPACE

1. The symmetrized Levi-Civita symbol

Lemma A1. Let ε be the Levi-Civita symbol in three dimensions. Then,

$$|\varepsilon_{nij}||\varepsilon_{nlm}| = (1 - \delta_{ij})(\delta_{il}\delta_{jm} + \delta_{im}\delta_{jl}). \quad (\text{A1})$$

Proof. Trivially, the equation holds true for $i = j$ and both sides of the equation equal zero. For the case $i \neq j$ we first consider $l = m$. The left-hand side of the equation equals zero and on the right-hand side we find

$$\delta_{il}\delta_{jm} + \delta_{im}\delta_{jl} = \delta_{il}\delta_{jl} + \delta_{il}\delta_{jl} \quad (\text{A2})$$

$$= 2\delta_{ij}\delta_{jl} \quad (\text{A3})$$

$$= 0. \quad (\text{A4})$$

The last case to prove is thus $i \neq j \cap l \neq m$. We rewrite $|\varepsilon_{nij}| = (1 - \delta_{ki})(1 - \delta_{ij})(1 - \delta_{kj})$ such that

$$|\varepsilon_{nij}||\varepsilon_{nlm}| = \sum_k (1 - \delta_{ki})(1 - \delta_{ij})(1 - \delta_{kj})(1 - \delta_{kl})(1 - \delta_{lm})(1 - \delta_{km}) \quad (\text{A5})$$

$$= \sum_k (1 - \delta_{ki})(1 - \delta_{kj})(1 - \delta_{kl})(1 - \delta_{km}) \quad (\text{A6})$$

$$= \sum_k (1 - \delta_{ki} - \delta_{kj} + \delta_{ki}\delta_{kj})(1 - \delta_{kl} - \delta_{km} + \delta_{kl}\delta_{km}) \quad (\text{A7})$$

$$= \sum_k (1 - \delta_{ki} - \delta_{kj} + \delta_{ki}\delta_{kj} - \delta_{kl} + \delta_{kl}\delta_{ki} + \delta_{kl}\delta_{kj} - \delta_{kl}\delta_{ki}\delta_{kj} - \delta_{km} + \delta_{km}\delta_{ki} \quad (\text{A8})$$

$$+ \delta_{km}\delta_{kj} - \delta_{km}\delta_{ki}\delta_{kj} + \delta_{kl}\delta_{km} - \delta_{kl}\delta_{km}\delta_{ki} - \delta_{kl}\delta_{km}\delta_{kj} + \delta_{kl}\delta_{km}\delta_{ki}\delta_{kj}) \quad (\text{A9})$$

$$= -1 + \delta_{ij} + \delta_{il} + \delta_{jl} - \delta_{il}\delta_{ij} + \delta_{im} \quad (\text{A10})$$

$$+ \delta_{jm} - \delta_{im}\delta_{ij} + \delta_{lm} - \delta_{il}\delta_{lm} - \delta_{jl}\delta_{lm} + \delta_{ij}\delta_{jl}\delta_{lm} \quad (\text{A11})$$

$$= -1 + \delta_{il} + \delta_{jl} + \delta_{im} + \delta_{jm}. \quad (\text{A12})$$

$$(\text{A13})$$

Thus, we need to prove

$$\delta_{il}\delta_{jm} + \delta_{im}\delta_{jl} = -1 + \delta_{il} + \delta_{jl} + \delta_{im} + \delta_{jm} \quad (\text{A14})$$

for $i \neq j \cap l \neq m$. Since $i, j, l, m \in \{1, 2, 3\}$, we have again two distinct cases: (1) $|\{i, j\} \cap \{l, m\}| = 1$ and (2) $|\{i, j\} \cap \{l, m\}| = 2$.

(1) Without loss of generality we may assume that $i = l$ and $i, j, l \neq m$. Then, the equation amounts to $0 = 0$.

(2) Without loss of generality $i = l$ and $j = m$. Then, we have $1 = 1$.

2. Vectorial expression for the Berry curvature

The full Berry curvature is

$$\Omega = i\langle \partial_{e_i} \psi | \partial_{e_j} \psi \rangle de_i^* \wedge de_j. \quad (\text{A15})$$

We partition the tensor $i\langle \partial_{e_i} \psi | \partial_{e_j} \psi \rangle$ into its symmetric and antisymmetric components. For the symmetric components we separate the diagonal from the off-diagonal matrix elements,

$$\Omega = \frac{i}{2}(\langle \partial_{e_i} \psi | \partial_{e_j} \psi \rangle - \langle \partial_{e_j} \psi | \partial_{e_i} \psi \rangle) de_i^* \wedge de_j + \frac{i}{2}(\langle \partial_{e_i} \psi | \partial_{e_j} \psi \rangle + \langle \partial_{e_j} \psi | \partial_{e_i} \psi \rangle) de_i^* \wedge de_j \quad (\text{A16})$$

$$= \frac{i}{2}(\langle \partial_{e_i} \psi | \partial_{e_j} \psi \rangle - \langle \partial_{e_j} \psi | \partial_{e_i} \psi \rangle) de_i^* \wedge de_j + \frac{i}{2}(1 - \delta_{ij})(\langle \partial_{e_i} \psi | \partial_{e_j} \psi \rangle + \langle \partial_{e_j} \psi | \partial_{e_i} \psi \rangle) de_i^* \wedge de_j \quad (\text{A17})$$

$$+ i\delta_{ij}\langle \partial_{e_i} \psi | \partial_{e_j} \psi \rangle de_i^* \wedge de_j \quad (\text{A18})$$

$$= \frac{i}{2}(\delta_{li}\delta_{mj} - \delta_{lj}\delta_{mi})\langle \partial_{e_l} \psi | \partial_{e_m} \psi \rangle de_i^* \wedge de_j + \frac{i}{2}(1 - \delta_{ij})(\delta_{li}\delta_{mj} + \delta_{lj}\delta_{mi})\langle \partial_{e_l} \psi | \partial_{e_m} \psi \rangle de_i^* \wedge de_j \quad (\text{A19})$$

$$+ i\delta_{ij}\langle \partial_{e_i} \psi | \partial_{e_j} \psi \rangle de_j^* \wedge de_j. \quad (\text{A20})$$

We employ the identity $\varepsilon_{nij}\varepsilon_{nlm} = \delta_{il}\delta_{jm} + \delta_{im}\delta_{jl}$ and Lemma A 1, such that we may write

$$\Omega = \frac{i}{2}\varepsilon_{nij}\varepsilon_{nlm}\langle\partial_{e_l}\psi|\partial_{e_m}\psi\rangle de_i^* \wedge de_j + \frac{i}{2}|\varepsilon_{nij}||\varepsilon_{nlm}|\langle\partial_{e_l}\psi|\partial_{e_m}\psi\rangle de_i^* \wedge de_j \quad (\text{A21})$$

$$+ i\delta_{ij}\langle\partial_{e_i}\psi|\partial_{e_i}\psi\rangle de_j^* \wedge de_j. \quad (\text{A22})$$

We use the definitions of the standard cross product and the bilinear forms Eq. (22),

$$\Omega = i\langle\nabla_e\psi|\times|\nabla_e\psi\rangle\cdot d\boldsymbol{\xi} + i\langle\nabla_e\psi|\diamond|\nabla_e\psi\rangle\cdot d\boldsymbol{\zeta} + i\langle\nabla_e\psi|\odot|\nabla_e\psi\rangle\cdot d\boldsymbol{\chi}. \quad (\text{A23})$$

3. Euler-angle representation of the polarization differentials

For each of the differential 2-forms $d\boldsymbol{\xi}$, $d\boldsymbol{\zeta}$, and $d\boldsymbol{\chi}$ an explicit expression in terms of the Euler angles $\rho = (\theta, \phi)$ is given by the exterior derivative of a 1-form ω : $d\omega = d\rho_i \frac{\partial}{\partial \rho_i} \omega$,

$$d\xi^l = \frac{1}{2}\epsilon^{lij} de^{*i} \wedge de^j \quad (\text{A24})$$

$$= \frac{1}{2}\epsilon^{lij}\left(d\theta\frac{\partial}{\partial\theta} + d\phi\frac{\partial}{\partial\phi}\right)e^{*i} \wedge \left(d\theta\frac{\partial}{\partial\theta} + d\phi\frac{\partial}{\partial\phi}\right)e^j. \quad (\text{A25})$$

Due to the antisymmetry of the wedge product, we have $d\theta \wedge d\theta = d\phi \wedge d\phi = 0$ and $d\phi \wedge d\theta = -d\theta \wedge d\phi$,

$$d\xi^l = \frac{1}{2}d\theta \wedge d\phi \epsilon^{lij}\left(\frac{\partial e^{*i}}{\partial\theta}\frac{\partial e^j}{\partial\phi} - \frac{\partial e^j}{\partial\theta}\frac{\partial e^{*i}}{\partial\phi}\right) \quad (\text{A26})$$

$$= \frac{1}{2}d\theta \wedge d\phi\left(\frac{\partial e^*}{\partial\theta} \times \frac{\partial e}{\partial\phi} + \frac{\partial e}{\partial\theta} \times \frac{\partial e^*}{\partial\phi}\right)^l \quad (\text{A27})$$

$$= d\theta \wedge d\phi \text{Re}\left(\frac{\partial e^*}{\partial\theta} \times \frac{\partial e}{\partial\phi}\right)^l. \quad (\text{A28})$$

Similarly, we get

$$d\zeta^l = \frac{1}{2}|\varepsilon^{lij}|de^{*i} \wedge de^j \quad (\text{A29})$$

$$= \frac{1}{2}d\theta \wedge d\phi|\varepsilon^{lij}|\left(\frac{\partial e^{*i}}{\partial\theta}\frac{\partial e^j}{\partial\phi} - \frac{\partial e^j}{\partial\theta}\frac{\partial e^{*i}}{\partial\phi}\right) \quad (\text{A30})$$

$$= \frac{1}{2}d\theta \wedge d\phi\left(\frac{\partial e^*}{\partial\theta} \diamond \frac{\partial e}{\partial\phi} - \frac{\partial e}{\partial\theta} \diamond \frac{\partial e^*}{\partial\phi}\right)^l \quad (\text{A31})$$

$$= d\theta \wedge d\phi i\text{Im}\left(\frac{\partial e^*}{\partial\theta} \diamond \frac{\partial e}{\partial\phi}\right)^l \quad (\text{A32})$$

and

$$d\chi^l = \delta_{li}de^{*i} \wedge de^i \quad (\text{A33})$$

$$= \delta_{li}d\theta \wedge d\phi\left(\frac{\partial e^{*i}}{\partial\theta}\frac{\partial e^i}{\partial\phi} - \frac{\partial e^i}{\partial\theta}\frac{\partial e^{*i}}{\partial\phi}\right) \quad (\text{A34})$$

$$= d\theta \wedge d\phi\left(\frac{\partial e^*}{\partial\theta} \odot \frac{\partial e}{\partial\phi} - \frac{\partial e}{\partial\theta} \odot \frac{\partial e^*}{\partial\phi}\right)^l \quad (\text{A35})$$

$$= d\theta \wedge d\phi 2i\text{Im}\left(\frac{\partial e^*}{\partial\theta} \odot \frac{\partial e}{\partial\phi}\right)^l. \quad (\text{A36})$$

4. Berry curvature for one-photon ionization

One-photon ionization processes are fully described by the first-order perturbative wave function

$$|\psi\rangle = |0\rangle + \int d\Theta_k a_k(\mathbf{e})|\mathbf{k}\rangle. \quad (\text{A37})$$

The real, antisymmetric part of the Berry curvature tensor Ω_a is determined by the vector

$$\Omega_a = \langle\nabla_e\psi|\times|\nabla_e\psi\rangle \quad (\text{A38})$$

$$= \int d\Theta_k \int d\Theta_{k'} (\nabla_{e^*} a_k^*) \times (\nabla_e a_{k'}) \langle\mathbf{k}|\mathbf{k}'\rangle \quad (\text{A39})$$

$$= \int d\Theta_k (\nabla_e a_k)^* \times (\nabla_e a_k). \quad (\text{A40})$$

The coefficient a_k may be expressed in terms of the ionization dipole $\mathbf{D} = \langle\mathbf{k}|\hat{\mathbf{r}}|0\rangle$ and the circular field $\mathbf{E}(t) = E(t)\mathbf{e}$: $a_k = -\tilde{E}_{\omega_k} \mathbf{D} \cdot \mathbf{e}$ such that

$$\Omega_a = |\tilde{E}_{\omega_k}|^2 \int d\Theta_k (\nabla_e \mathbf{D} \cdot \mathbf{e})^* \times (\nabla_e \mathbf{D} \cdot \mathbf{e}) \quad (\text{A41})$$

$$= |\tilde{E}_{\omega_k}|^2 \int d\Theta_k \mathbf{D}^* \times \mathbf{D}. \quad (\text{A42})$$

Analogously, we obtain

$$\langle\nabla_e\psi|\diamond|\nabla_e\psi\rangle = |\tilde{E}_{\omega_k}|^2 \int d\Theta_k \mathbf{D}^* \diamond \mathbf{D} \quad (\text{A43})$$

and

$$\langle\nabla_e\psi|\odot|\nabla_e\psi\rangle = |\tilde{E}_{\omega_k}|^2 \int d\Theta_k \mathbf{D}^* \odot \mathbf{D}. \quad (\text{A44})$$

Conclusively, the full Berry curvature is

$$\Omega = i|\tilde{E}_{\omega_k}|^2 \int d\Theta_k (\mathbf{D}^* \times \mathbf{D} \cdot d\boldsymbol{\xi} + \mathbf{D}^* \diamond \mathbf{D} \cdot d\boldsymbol{\zeta} + \mathbf{D}^* \odot \mathbf{D} \cdot d\boldsymbol{\chi}) \quad (\text{A45})$$

$$= id\theta \wedge d\phi |\tilde{E}_{\omega_k}|^2 \int d\Theta_k \quad (\text{A46})$$

$$\left[\mathbf{D}^* \times \mathbf{D} \cdot \text{Re}\left(\frac{\partial e^*}{\partial\theta} \times \frac{\partial e}{\partial\phi}\right) + \mathbf{D}^* \diamond \mathbf{D} \cdot i\text{Im}\left(\frac{\partial e^*}{\partial\theta} \diamond \frac{\partial e}{\partial\phi}\right) + \mathbf{D}^* \odot \mathbf{D} \cdot 2i\text{Im}\left(\frac{\partial e^*}{\partial\theta} \odot \frac{\partial e}{\partial\phi}\right) \right]. \quad (\text{A47})$$

APPENDIX B: BERRY PHASE AND ANTISYMMETRIC BERRY CURVATURE IN TWO-FIELD PUMP-PROBE EXPERIMENTS

Here, we derive the Berry connection and antisymmetric Berry curvature in the case of a two-field pump-probe experiment.

1. Berry connection and phase

The Berry connection 1-form on the parameter manifold is

$$\mathcal{A} = i\langle\psi|\nabla_\rho\psi\rangle \cdot d\rho. \quad (\text{B1})$$

Since the wave function only depends on ρ via the electric fields, we can express the gradient with respect to ρ using the

chain rule:

$$\nabla_\rho = \frac{\partial \mathbf{e}}{\partial \rho} \nabla_{\mathbf{e}} + \frac{\partial \boldsymbol{\varepsilon}}{\partial \rho} \nabla_{\boldsymbol{\varepsilon}}, \quad (\text{B2})$$

giving

$$A = A_{e_i} de_i + A_{e_j} de_j, \quad (\text{B3})$$

with

$$A_{e_i} = i \langle \psi | \partial_{e_i} \psi \rangle, \quad A_{e_j} = i \langle \psi | \partial_{e_j} \psi \rangle. \quad (\text{B4})$$

The geometric (Berry) phase is defined as a loop integral in the parameter space (ρ):

$$\begin{aligned} S &= i \oint \langle \psi | \nabla_\rho | \psi \rangle \cdot d\rho \\ &= i \oint \langle \psi | \nabla_{\boldsymbol{\varepsilon}} | \psi \rangle \cdot d\boldsymbol{\varepsilon} + i \oint \langle \psi | \nabla_{\mathbf{e}} | \psi \rangle \cdot d\mathbf{e}. \end{aligned} \quad (\text{B5})$$

Hence, the Berry connection and phase are naturally split into two contributions, one for each field.

2. Berry curvature

The Berry curvature 2-form is

$$\Omega = dA \quad (\text{B6})$$

$$\begin{aligned} &= \partial_{e_i}^* A_{e_j} de_i^* \wedge de_j + \partial_{e_i} A_{e_j} de_i \wedge de_j + \partial_{e_i}^* A_{e_j} de_i^* \wedge de_j \\ &\quad + \partial_{e_i} A_{e_j} de_i \wedge de_j + \partial_{e_i} A_{e_j} de_i \wedge de_j, \end{aligned} \quad (\text{B7})$$

where we assume that ψ is holomorphic in $\mathbf{e}$ and that $\boldsymbol{\varepsilon}$ is real. Let us consider the antisymmetric Berry curvature and identify three contributions:

$$\Omega_a = \Omega_a^e + \Omega_a^\varepsilon + \Omega_a^{\text{cross}} \quad (\text{B8})$$

with

$$\Omega_a^e = \frac{1}{2} (\partial_{e_i}^* A_{e_j} - \partial_{e_j}^* A_{e_i}) de_i^* \wedge de_j \quad (\text{B9})$$

$$\Omega_a^\varepsilon = \frac{1}{2} (\partial_{\varepsilon_i} A_{\varepsilon_j} - \partial_{\varepsilon_j} A_{\varepsilon_i}) d\varepsilon_i \wedge d\varepsilon_j \quad (\text{B10})$$

$$\begin{aligned} \Omega_a^{\text{cross}} &= \frac{1}{2} (\partial_{\varepsilon_i} A_{e_j} - \partial_{e_j} A_{\varepsilon_i}) d\varepsilon_i \wedge de_j \\ &\quad + \frac{1}{2} (\partial_{e_i}^* A_{\varepsilon_j} - \partial_{\varepsilon_j}^* A_{e_i}) de_i^* \wedge d\varepsilon_j \\ &\quad + \frac{1}{2} (\partial_{e_i} A_{\varepsilon_j} - \partial_{\varepsilon_j} A_{e_i}) de_i \wedge d\varepsilon_j. \end{aligned} \quad (\text{B11})$$

Next, we use the following identity for the contraction of a fully antisymmetric tensor T_{ij} with some arbitrary tensor K ,

$$T_{ij} K_{ij} = \frac{1}{2} \epsilon_{nij} \epsilon_{nlm} T_{ij} K_{lm}, \quad (\text{B12})$$

to rewrite the antisymmetric Berry curvature:

$$\Omega_a^e = \frac{1}{2} (\partial_{e_i}^* A_{e_j} - \partial_{e_j}^* A_{e_i}) de_i^* \wedge de_j \quad (\text{B13})$$

$$= \frac{1}{2} \epsilon_{nij} (\partial_{e_i}^* A_{e_j} - \partial_{e_j}^* A_{e_i}) \frac{1}{2} \epsilon_{nlm} de_l^* \wedge de_m. \quad (\text{B14})$$

Identifying $\mathbf{A}_e = (A_{e_1}, A_{e_2}, A_{e_3})$ and $d\xi_k^e = \frac{1}{2} \epsilon_{nlm} de_l^* \wedge de_m$ gives

$$\Omega_a^e = (\nabla_{\mathbf{e}^*} \times \mathbf{A}_e) \cdot d\xi_e = \Omega_a^e \cdot d\xi_e \quad (\text{B15})$$

with

$$\Omega_a^e = \nabla_{\mathbf{e}^*} \times \mathbf{A}_e = i \langle \nabla_{\mathbf{e}} \psi | \times | \nabla_{\mathbf{e}} \psi \rangle, \quad (\text{B16})$$

which coincides with the expression for the one-field antisymmetric Berry curvature. Analogously, the linear field Berry curvature is fully antisymmetric:

$$\Omega^\varepsilon = \Omega_a^\varepsilon = (\nabla_{\boldsymbol{\varepsilon}} \times \mathbf{A}_\varepsilon) \cdot d\xi_\varepsilon = \Omega_a^\varepsilon \cdot d\xi_\varepsilon, \quad (\text{B17})$$

with

$$\Omega_a^\varepsilon = \nabla_{\boldsymbol{\varepsilon}} \times \mathbf{A}_\varepsilon = i \langle \nabla_{\boldsymbol{\varepsilon}} \psi | \times | \nabla_{\boldsymbol{\varepsilon}} \psi \rangle. \quad (\text{B18})$$

The linear Berry curvature is equal to zero in one photon ionization.

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