

Enhancing the Hyperpolarizability of Crystals with Quantum Geometry

Wojciech J. Jankowski^{1,*}, Robert-Jan Slager^{2,1}, and Michele Pizzochero^{3,4,†}

¹*TCM Group, Cavendish Laboratory, University of Cambridge, J. J. Thomson Avenue, Cambridge CB3 0HE, United Kingdom*

²*Department of Physics and Astronomy, University of Manchester, Oxford Road, Manchester M13 9PL, United Kingdom*

³*Department of Physics, University of Bath, Claverton Down, Bath BA2 7AY, United Kingdom*

⁴*School of Engineering and Applied Sciences, Harvard University, Cambridge, Massachusetts 02138, USA*



(Received 10 February 2025; revised 26 June 2025; accepted 19 August 2025; published 16 September 2025)

We demonstrate that higher-order electric susceptibilities in crystals can be enhanced and understood through nontrivial topological invariants and quantum geometry, using one-dimensional π -conjugated chains as representative model systems. First, we show that the crystalline-symmetry-protected topology of these chains imposes a lower bound on their quantum metric and hyperpolarizabilities. Second, we employ numerical simulations to reveal the tunability of nonlinear, quantum geometry-driven optical responses in various one-dimensional crystals in which band topology can be externally controlled. Third, we develop a semiclassical picture to deliver an intuitive understanding of these effects. Our findings offer a firm interpretation of otherwise elusive experimental observations of colossal hyperpolarizabilities and establish guidelines for designing topological materials of any dimensionality with enhanced nonlinear optical properties.

DOI: [10.1103/z7lp-pqp6](https://doi.org/10.1103/z7lp-pqp6)

Introduction—Nonlinear optics offers a rich platform to explore a broad range of intriguing phenomena, with implications ranging from fundamental science to engineering [1]. One-dimensional crystals, most notably π -conjugated linear chains [2–4], constitute prime examples to probe enhanced optical responses at nonlinear orders. Because these crystals are centrosymmetric, i.e., they feature an inversion symmetry, their optical responses occur at odd orders in optical fields. The first- and third-order optical responses of one-dimensional π -conjugated chains have long been investigated, both experimentally [4–9] and computationally [10–20]. Of central interest in this context is the third-order hyperpolarizability, i.e., the third-order electric susceptibility $\chi^{(3)}$ in the static limit of vanishing optical field frequency, $\omega \rightarrow 0$, which quantifies the nonlinear polarization response, $P^{(3)} = \chi^{(3)} E_x^3$, to electric field E_x [21]. In π -conjugated chains, $\chi^{(3)}$ is known to acquire colossal values. For example, $\chi^{(3)}$ was inferred to be as high as $10^{-18} \text{ m}^2/\text{V}^2$ in polyacetylene [1], $10^{-19} \text{ m}^2/\text{V}^2$ in polydiacetylene, and $10^{-20} \text{ m}^2/\text{V}^2$ in polyarylenes [4]. This unusually large hyperpolarizability has been heuristically ascribed to the responses of p_z

electrons forming the conjugated π bonds [1]. However, a general theoretical understanding of the phenomenological origin of these nonlinear optical properties remains yet to be achieved [22–32].

In this Letter, we bridge this gap by deploying the modern notions of band topology and quantum geometry. We show that the crystalline-symmetry-protected topology of π -conjugated chains induces a lower bound on the quantum metric, and thus on the hyperpolarizability. Then, we use numerical simulations to demonstrate the tunability of nonlinear, quantum geometry-driven optical responses of a variety of one-dimensional crystals in which the band topology is controllable by an external parameter, such as lattice strain. Finally, we propose a general semiclassical picture to explain these remarkable optical properties, based on the scaling of quantum metric and band gaps with external parameters. Importantly, our theory offers a topological explanation for the distinct hyperpolarizabilities of the *cis* and *trans* isomers of π -conjugated linear polymers shown in Fig. 1(d) [7]. Unlike the *cis* configuration, the topological windings of the Bloch eigenvector in the *trans* configuration support the geometric enhancements of the optical matrix elements associated with the nonlinear optical responses. Overall, our findings provide a novel interpretation of otherwise elusive experimental observations, along with guiding principles for identifying and engineering materials for nonlinear optics.

Quantum geometry and topology in one dimension—Over the past several years, crystalline topologies have been thoroughly investigated [34–39], culminating in a rather uniform view upon comparing momentum space

*Contact author: wjj25@cam.ac.uk

†Contact author: mp2834@bath.ac.uk

Published by the American Physical Society under the terms of the [Creative Commons Attribution 4.0 International](https://creativecommons.org/licenses/by/4.0/) license. Further distribution of this work must maintain attribution to the author(s) and the published article's title, journal citation, and DOI.

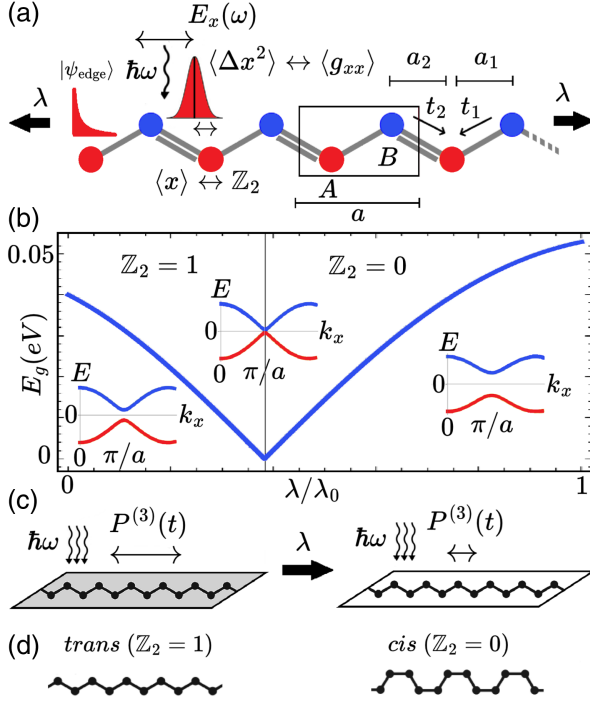


FIG. 1. Crystalline topology and quantum geometrically enhanced hyperpolarizability in one-dimensional crystals. (a) Schematic illustration of a one-dimensional inversion-symmetric model system with a topological invariant $\mathbb{Z}_2$ reflected by a Wannier charge center shift $\langle x \rangle$ and by the presence of a symmetry-protected exponentially localized edge state, $|\psi_{\text{edge}}\rangle$. The model system with unit cell parameter a realizes a bipartite basis A, B , and effective hoppings t_1, t_2 across bonds with lengths a_1, a_2 , and is coupled to an optical field $E_x(\omega)$ introduced by photons with energy $\hbar\omega$. The minimal spread of the Wannier function $\langle \Delta x^2 \rangle$ is topologically bounded, and evaluates to the band-averaged quantum metric $\langle g_{xx} \rangle$ [33], which determines the nonlinear polarizability. (b) Control of the band gap and band topology with lattice strain (λ), across a topological phase transition (TPT), where λ_0 is a unit strain. (c) Discontinuous control of the nonlinear optical properties, such as the time-dependent third-order polarization $P^{(3)}(t)$ from a three-photon coupling, across a strain-induced TPT. (d) Molecular structure of the *trans* and *cis* isomers of polyacetylene realizing different $\mathbb{Z}_2$ invariants, resulting in distinct hyperpolarizabilities.

constraints [37] and real space (Wannierizability) conditions [38,39]. Within this framework, taking into account the fact that an exponentially localized Wannier basis [40] can be always constructed [41] in one spatial dimension, it is directly inferred that the only form of nontrivial electronic topology is that of an obstructed limit. The topology [42,43] arises from the interplay of band wave function ($|\psi_{nk}\rangle = e^{ik \cdot r} |u_{nk}\rangle$) windings with additional symmetries, such as chiral or inversion ($\mathcal{I}$) symmetry [44]. Under $\mathcal{I}$ symmetry, in nonmagnetic systems, the topological invariant is determined by the band parity eigenvalues, $\delta_k = \pm 1$ at inversion-symmetric momenta, $k = 0, \pi/a$, where a is the lattice constant, which coincides with the Fu-Kane

formula [44]: $\nu_{\text{FK}} = (1 - \delta_0 \delta_{\pi/a})/2 \in \mathbb{Z}_2$. At the same time, the band topology of obstructed insulators provides for the minimal spread $\langle \Delta x^2 \rangle$ of the exponentially localized Wannier functions [33], as schematically presented in Fig. 1(a). Furthermore, this can be understood with the notions of quantum geometry [45], and can be related to topological bounds [46].

We now elucidate how, in one-dimensional (1D) crystals, the crystalline topology gives rise to quantum geometry and, as a direct consequence, enhances their optical responses. We define a non-Abelian multiband quantum geometric tensor, $[\mathbf{Q}_{ij}^{(m)}]_{np} = \langle \partial_{k_i} u_{nk} | u_{mk} \rangle \langle u_{mk} | \partial_{k_j} u_{pk} \rangle$ over momentum space, with its real part, commonly known as the non-Abelian quantum metric, $[\mathbf{g}_{ij}^{(m)}]_{np} = \text{Re}[\mathbf{Q}_{ij}^{(m)}]_{np}$ [45,47,48]. The multiband quantum metric encodes the optical transition dipole matrix elements, $[\mathbf{g}_{xx}^{(m)}]_{nm} = |\langle \psi_{nk} | \hat{x} | \psi_{mk} \rangle|^2$ [49,50], which is at the heart of both linear and nonlinear responses. The multiband quantum metric of electrons realizes lower bounds due to 1D crystalline topologies that translate into minimal values of (higher-order) optical susceptibilities,

$$\langle g_{xx} \rangle \equiv \sum_m \sum_k^{\text{unocc}} \text{Tr} \mathbf{g}_{xx}^{(m)} \geq \frac{a^2}{4} \nu_{\text{FK}}, \quad (1)$$

where $\langle g_{xx} \rangle$ is band-averaged quantum metric and $\nu_{\text{FK}} \in \mathbb{Z}_2$ the Fu-Kane parity invariant protected by the $\mathcal{I}$ symmetry [44]. The bound can be derived from (i) nontrivial inversion symmetry indicators pinpointing how the irreducible representations of the obstructed bands transform [37–39,44], (ii) translating these into real-space projectors [46], and (iii) using an optimization argument of Ref. [46]. The technical details of the derivation are given in Sec. I of the Supplemental Material [51]. In the context of the maximally localized Wannier functions, $\langle g_{xx} \rangle = \langle \Delta x^2 \rangle$ [33]. Hence, the invariant imposes a lower-bound constraint on the spread of the wave function in the real-space basis. In topological crystalline phases, any topologically nontrivial band realizes $\nu_{\text{FK}} = 1$. In the following, we demonstrate that this bound yields a topological enhancement of the multiband geometry-dependent response functions within the first- and third-order optical responses.

First- and third-order optical responses—The linear optical conductivity of a band insulator in the first-order response to an electric field of frequency ω reads as [50],

$$\sigma_{xx}(\omega) = \frac{e^2}{2\hbar^2} \int_{\text{BZ}} \frac{dk}{2\pi} \sum_{n,m} f_{nm} E_{mn} \text{Tr} \mathbf{g}_{xx}^{(m)} \delta(\omega - E_{mn}/\hbar), \quad (2)$$

where $f_{nm} = f_{nk} - f_{mk}$ are the thermal Fermi-Dirac occupation factors, momentum k runs over the first Brillouin zone (BZ), and E_{mn} is the difference in energy between

valence and conduction bands n and m , respectively. For crystals possessing the same band structure, the distinct first-order optical conductivity is purely determined by the multiband quantum metric. This allows one to uniquely distinguish band topologies by inducing the corresponding band geometries. Importantly, changes in band topology, as realized through external parameters such as lattice strain, as shown in Figs. 1(a) and 1(b), result in changes in the optical response. As we shall discuss in the representative case of π -conjugated chains and related one-dimensional crystals, lattice strain can be leveraged to engineer the opacity and optical nonlinearities, as illustrated in Fig. 1(c).

While the second-order optical responses vanish under the $\mathcal{T}$ symmetry in centrosymmetric crystals, third-order responses remain finite and can be controlled with band geometry. In the static limit, the third-order susceptibility can be enhanced quadratically with the considered multiband quantum metric:

$$\chi^{(3)} = \frac{e^4}{\epsilon_0} \int_{\text{BZ}} \frac{dk}{2\pi} \sum_{n,m,p,q} \frac{[\mathbf{g}_{xx}^{(p)}]_{nm} [\mathbf{g}_{xx}^{(q)}]_{mn}}{E_{mn} E_{pn} E_{qn}}, \quad (3)$$

where ϵ_0 is the vacuum permittivity. The derivation of Eq. (3) is provided in Sec. II of the Supplemental Material [51].

While the enhancement of $\chi^{(3)}$ can be induced by nontrivial quantum geometry in the numerator, it can be further achieved through the tunable band gap magnitudes in the denominator. Notably, materials with identical band gaps, yet with distinct crystalline topologies, realize distinguishable numerators, with topological phase experiencing a quadratic enhancement. By combining this enhancement with Eq. (1) and sum rules, we derive a fundamental topological lower bound on the hyperpolarizability of a nontrivial insulator,

$$\chi^{(3)} \geq \alpha^{(3)} \times \frac{a^{10}}{n_e^3}, \quad (4)$$

where $\alpha^{(3)} = \frac{1}{2} \pi^3 e^4 m_e^3 \epsilon_0^{-1} \hbar^{-6} = 6 \times 10^{50} \text{ m}^{-5} / \text{V}^2$, m_e is the electron mass, and n_e is the number density of electrons in the crystal. The derivation of Eq. (4) employs (i) the energy gap bound of Ref. [54] due to quantum metric (ii) combined with the sum rules of Ref. [55], as employed in the linear susceptibility bound of Ref. [56]. The technical details of the derivation are given in Sec. IV of the Supplemental Material [51].

From models to real materials—We next employ an effective, two-band Hamiltonian to capture the geometry induced by the $\mathbb{Z}_2$ obstructed topology in the presence of lattice strain. This model is schematically depicted in Fig. 1(a). The Hamiltonian that minimally realizes such general phenomenology on a quasi-one-dimensional chain takes the form of a strained Su-Schrieffer-Heeger (SSH) model [57,58],

$$H(k_x, \lambda) = \begin{pmatrix} 0 & t_1(\lambda) + t_2(\lambda) e^{-ik_x a(\lambda)} \\ t_1(\lambda) + t_2(\lambda) e^{ik_x a(\lambda)} & 0 \end{pmatrix}, \quad (5)$$

where $\lambda > 0$ is an external tensile stress parameter, $a(\lambda) \equiv a_1(\lambda) + a_2(\lambda)$ is the strain-renormalized lattice parameter with bond lengths $a_j(\lambda) = a_j(0)[1 + \lambda]$, and $t_j(\lambda) = t_j(0)e^{-\gamma_j[a_j(\lambda) - a_j(0)]}$ are the strain-dependent hopping amplitudes, where γ_j 's parametrize bond stiffness [59]. We validate our strain-dependent models with hopping parameters obtained from first-principles calculations [60]. We correspondingly present our numerical results in Fig. 2. In Fig. 2(a), we show how the profile of quantum metric g_{xx} over k space changes with the strain λ . The metric supporting the topological lower bound is enhanced at the band edge $k = \pi/a$, as the dominant contributions arise at the band inversion region, where the local derivatives of the Bloch eigenvectors winding across the BZ are maximized. Upon crossing the critical strain $\lambda = \lambda_c$, the band edge metric contributions provide for an average $\langle g_{xx} \rangle$ that satisfies the bound in the topological phase ($\nu_{\text{FK}} = 1$). Figure 2(b) presents a dependence of the optical weight, i.e., weighted integrated optical conductivity, or equivalently, integrated interband contribution to the imaginary part of the dielectric constant $W_{xx} = \int_0^\infty d\omega [\sigma_{xx}(\omega)/\omega] = (\pi e^2 / \hbar a) \langle g_{xx} \rangle$, which is also lower-bounded by the topological invariant. In Fig. 2(c), we benchmark the first-order optical conductivity and show how its contributions to the bounded optical weights gradually arise. Figure 2(d) shows how the colossal values of $\chi^{(3)}$ arise with strain λ , before vanishing sharply upon transitioning to the trivial phase across the critical point. We remark that the value of $\chi^{(3)}$ at vanishing λ is in excellent agreement with experimental observations. Importantly, the bound of the quantum metric, as manifested in Figs. 2(c) and 2(d), provides for the robustness of lower-bounded optical responses of topological phases, especially away from the singular critical points that correspond to the strain-induced phase transitions. Further details on the model-dependent parametrization and controllability of quantum geometry within π -conjugated chains are given in Sec. III of the Supplemental Material [51].

We stress that this model Hamiltonian and accompanying results are effectively realized by the low-energy bands of an ample variety of one-dimensional π -conjugated systems, such as polyacetylene [57,58], polyacenes [61], polyphenylenes, and graphene nanoribbons [59], as overviewed in Fig. 3. Their low-energy valence and conduction bands are universally governed by an effective physics of hopping between the Wannier orbitals A, B that are predominantly constituted by the weighted combinations of the atomic orbitals with p_z character [57,60,61]. The physical implication of our findings is that the materials shown in Fig. 3 are expected to exhibit quantum-geometrically enhanced hyperpolarizabilities that originate from their nontrivial, inversion symmetry-protected band topology. Indeed, the introduction of additional bands

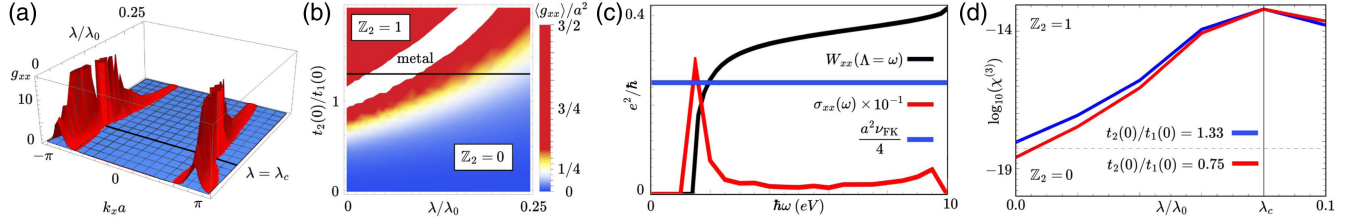


FIG. 2. Quantum geometry and (non)linear optics in one-dimensional crystals. (a) Quantum metric (red) over momentum space as a function of the lattice strain parameter (λ) realized in the effective low-energy Hamiltonian, Eq. (5), with hoppings $t_1(0) = 2.15$ eV, $t_2(0) = 2.85$ eV [$t_2(0)/t_1(0) = 1.33$]. The critical strain ($\lambda = \lambda_c$) induces a topological phase transition across which the metric scales up at the band edge at $k = \pm\pi/a(\lambda)$, overcoming the minimal average metric value per BZ (blue) necessary for satisfying the topological bound. (b) Band-averaged metric ($\langle g_{xx} \rangle$) as a function of strain and hopping amplitude ratio $t_2(0)/t_1(0)$. The horizontal line corresponds to the chain with $t_2(0)/t_1(0) = 1.33$. The corresponding optical absorptivity is manifestly larger and topologically lower-bounded in the topologically nontrivial phase. (c) Spectral resolution of strain-dependent optical conductivity $\sigma_{xx}(\omega)$ and cutoff (Λ) dependent optical weights $W_{xx}(\Lambda) = \int_0^\Lambda d\omega [\sigma_{xx}(\omega)/\omega]$ (black), as a function of the bounding value for $\Lambda \rightarrow \infty$ (blue) in a topological chain with $t_2/t_1 = 1.33$. (d) Lattice strain control of the topologically bounded hyperpolarizability $\chi^{(3)}$. In the subcritical strain limit, we find that due to distinct realized quantum metrics, the $\chi^{(3)}$ of a trivial phase [$\mathbb{Z}_2 = 0$] (red) is multiple times smaller than the $\chi^{(3)}$ of a topological phase [$\mathbb{Z}_2 = 1$] (blue) with identical band energies and band gaps, which determine equal λ_c for transitions to metallic states. The dashed line denotes the topological lower bound on $\chi^{(3)}$. Under the relatively high values of strain λ/λ_0 , where λ_0 is a unit strain, the anomalous hyperpolarizability of the $\mathbb{Z}_2$ -topological phase can be quenched; see also the Supplemental Material [51]. At vanishing strain, we note that the effective model fitted to the t_2/t_1 of polyacetylene yields $\chi^{(3)} = 8 \times 10^{-19} \text{ m}^2/\text{V}^2$, in excellent agreement with the corresponding experimental value, $\chi^{(3)} \approx 10^{-18} \text{ m}^2/\text{V}^2$ [3–5].

does not alter our conclusions, as we demonstrate in Sec. V of the Supplemental Material [51], where we also provide an extended discussion of multiband models. The reason for this traces back to the fact that the additional bands will experience higher energy gaps E_{mn} , suppressing the $\chi^{(3)}$ contributions as $1/E_{mn}^3$. Furthermore, the metric similarly scales with $1/E_{mn}^2$ [cf. Fig. 2(a)], which for topologically trivial additional bands provides only a marginal contribution to the numerator of $\chi^{(3)}$.

From the quantum to semiclassical picture—We now present an intuitive picture of the topologically induced quantum geometric phenomenology. Semiclassically, electrons in band gap insulators can be modeled with an effective nonlinear anharmonic oscillator Hamiltonian, $H = (p^2/2m_e) + U(x)$ with a $\mathcal{I}$ -symmetric confining nonlinear potential $U(x) = \frac{1}{2}\kappa x^2 + \frac{1}{4}bx^4$, where κ is an effective spring constant, $\kappa = m_e\omega_0^2$, in which ω_0 is a resonant frequency and b quantifies the degree of anharmonicity. Notably, $\kappa \propto 1/\langle g_{xx} \rangle = 1/\langle \Delta x^2 \rangle$ [56], where $\langle \Delta x^2 \rangle$ is a spatial spread of a localized state wave function, as representable in terms of Wannier functions [33]. From the theory of the nonlinear anharmonic oscillator discussed in Sec. VI of the Supplemental Material [51], one expects $\chi^{(3)} \propto b/\kappa^4$, by recognizing that, typically, $b \propto \kappa/\langle g_{xx} \rangle$, explaining the effective scaling of the $\chi^{(3)} \propto \langle g_{xx} \rangle^2$ within a full quantum-mechanical treatment. The semiclassical picture, in connection to quantum geometry, further supports the interpretation of our key result. Namely, the topological symmetry-indicated winding of the Bloch states in a one-dimensional insulator (i) induces a geometric spread in the localization of the bounded electrons, which

(ii) semiclassically reflects an effective spring constant and its interplay with anharmonicity. The latter determine the nonlinear optical response of the driven localized electrons within an anharmonic oscillator picture.

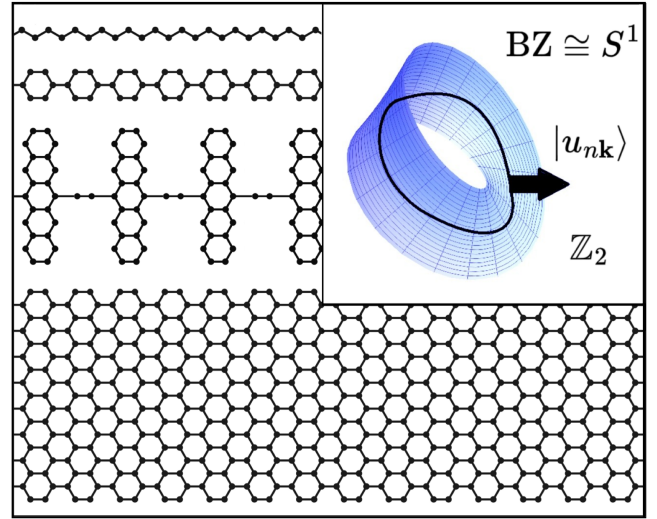


FIG. 3. Examples of materials for crystalline topology-induced, quantum-geometrically enhanced nonlinear optics. From top to bottom: polyacetylene, poly(*p*-phenylene), polypentacene, and graphene nanoribbons. Under external control, such as lattice strains, these materials can realize $\mathcal{I}$ -symmetry protected nontrivial $\mathbb{Z}_2$ topology and topologically enhanced hyperpolarizability $\chi^{(3)}$, which culminates in nonlinear polarization $P^{(3)}$. Further chemical functionalizations of such $\mathbb{Z}_2$ -topological materials, e.g., through perhalogenations, can provide a further knob to tune the quantum metric and higher-order susceptibilities.

Discussion and conclusions—In summary, we have shown that the colossal linear and higher-order optical responses observed in one-dimensional π -conjugated chains can be understood by means of the modern notions of band topology and quantum geometry, where the latter is bounded from below by the former. We have established a transparent connection of the multiband quantum geometry with the third-order optical responses, demonstrating that the crystalline-symmetry-protected topology induces a lower bound on the hyperpolarizability. Our quantum-geometric perspective sheds a new light on the long-standing controversy of the discrepancy between $\chi^{(3)}$ of the *cis* and *trans* geometric isomers of polyacetylene [4,7,62]. The distinction between *cis*- and *trans*-polyacetylene arises from the different spatial arrangements of atoms with identical chemical connectivity, which induces inequivalent band topology in the two isomers. The band topology classified by the winding of the Bloch eigenvectors provides for a lower bound in the quantum geometric tensor and in the associated enhanced nonlinear optical response matrix elements in the *trans* isomer. While being manifestly larger in the topological polymer, the metric band average lies above the topological lower bound and does not saturate, unless the band dispersion is forced to approach the flat band limit. The minimization of the lower-bounded quantum metric in such a limiting case is consistent with the other known flatness conditions for the metric bound saturations [63–65]. In the provided picture, *cis*-polyacetylene is topologically trivial ($\nu_{\text{FK}} = 0$), hence, realizes no bound on geometry with the average quantum metric $\langle g_{xx} \rangle$ that induces an enhancement of the $\chi^{(3)}$, unlike in the topological *trans*-polyacetylene ($\nu_{\text{FK}} = 1$).

The theory presented in this Letter offers a step forward by revealing that the origin of the p_z -orbital band polarizability is topologically induced, as associated with the lower-bounded quantum metric providing for an enhanced spread of the electronic wave function in the topological phase. To the best of our knowledge, the importance of band topology and geometry for nonlinear responses in the static limits—especially concerning hyperpolarizabilities—was not addressed in previous studies. Given that any band topology is fully encoded in the non-Abelian Berry connections, which are fully captured by the quantum geometry determining nonlinear optical responses [50], our conceptual framework consistently and comprehensively captures the role of band topology for hyperpolarizabilities in any class of materials. In addition to offering an understanding of the observed difference in the hyperpolarizabilities between distinct isomers through the notion of quantum geometric bound, the phenomenological picture underlying our findings translates into topologically induced quantum lattice fluctuations [22,24,27] in the form of virtual soliton-antisoliton pairs [22]. Following identifications of positional charge correlators with electronic localization lengths and quantum geometry of Refs. [54,55,66], such quantum charge density fluctuations are driven by a quantum metric.

To conclude, our Letter demonstrates that topologically lower-bounded higher-order geometric optical responses can be achieved in materials possessing nontrivial crystal-line topology, thus opening a pathway to identify and engineer novel platforms for enhanced nonlinear phenomena within the growing family of topologically classified materials.

Acknowledgments—W. J. J. and R.-J. S. acknowledge Bartomeu Monserrat and Joshua J. P. Thompson for stimulating discussions. W. J. J. acknowledges funding from the Rod Smallwood Studentship at Trinity College, Cambridge. R.-J. S. acknowledges funding from an EPSRC ERC underwrite Grant No. EP/X025829/1 and a Royal Society exchange Grant No. IES/R1/221060 as well as Trinity College, Cambridge.

Data availability—The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

-
- [1] R. Boyd, *Nonlinear Optics*, 4th ed. (Elsevier, Amsterdam, 2019).
 - [2] G. P. Agrawal, C. Cojan, and C. Flytzanis, Nonlinear optical properties of one-dimensional semiconductors and conjugated polymers, *Phys. Rev. B* **17**, 776 (1978).
 - [3] L. R. Dalton, J. Thomson, and H. S. Nalwa, The role of extensively delocalized π -electrons in electrical conductivity, non-linear optical properties and physical properties of polymers, *Polymer* **28**, 543 (1987).
 - [4] J. L. Bredas, C. Adant, P. Tackx, A. Persoons, and B. M. Pierce, Third-order nonlinear optical response in organic materials: Theoretical and experimental aspects, *Chem. Rev.* **94** (1994).
 - [5] C. R. Fincher, M. Ozaki, M. Tanaka, D. Peebles, L. Lauchlan, A. J. Heeger, and A. G. MacDiarmid, Electronic structure of polyacetylene: Optical and infrared studies of undoped semiconducting $(\text{CH})_x$ and heavily doped metallic $(\text{CH})_x$, *Phys. Rev. B* **20**, 1589 (1979).
 - [6] D. N. Beratan, J. N. Onuchic, and J. W. Perry, Nonlinear susceptibilities of finite conjugated organic polymers, *J. Phys. Chem.* **91**, 2696 (1987).
 - [7] M. Sinclair, D. Moses, K. Akagi, and A. J. Heeger, Anisotropy of the third-order nonlinear-optical susceptibility in a degenerate-ground-state conjugated polymer: Trans- $(\text{CH})_x$, *Phys. Rev. B* **38**, 10724 (1988).
 - [8] I. D. W. Samuel, I. Ledoux, C. Dhenaut, J. Zyss, H. H. Fox, R. R. Schrock, and R. J. Silbey, Saturation of cubic optical nonlinearity in long-chain polyene oligomers, *Science* **265**, 1070 (1994).
 - [9] I. Ledoux, I. D. W. Samuel, J. Zyss, S. N. Yaliraki, F. J. Schattenmann, R. R. Schrock, and R. J. Silbey, Third-order microscopic nonlinearities of very long chain polyenes: Saturation phenomena and conformational effects, *Chem. Phys.* **245**, 1 (1999).
 - [10] J. P. Hermann and J. Ducuing, Third-order polarizabilities of long-chain molecules, *J. Appl. Phys.* **45**, 5100 (1974).

- [11] K. C. Rustagi and J. Ducuing, Third-order optical polarizability of conjugated organic molecules, *Opt. Commun.* **10**, 258 (1974).
- [12] Z. G. Soos and G. W. Hayden, Static polarizability of interacting π electrons in conjugated polymers, *Phys. Rev. B* **40**, 3081 (1989).
- [13] A. Villesuzanne, J. Hoarau, L. Ducasse, L. Olmedo, and P. Hourquebie, π -electron calculations of polarizabilities in conjugated systems, *J. Chem. Phys.* **96**, 495 (1992).
- [14] Z. Huang, D. A. Jelski, R. Wang, D. Xie, C. Zhao, X. Xia, and T. F. George, Polarizabilities of trans and cis polyacetylene and interactions among chains in crystalline polyacetylene, *Can. J. Chem.* **70**, 372 (1992).
- [15] Z. Shuai and J. L. Brédas, Static and dynamic third-order susceptibilities in conjugated polymers: Hückel theory and VEH approach, *Synth. Met.* **49**, 37 (1992).
- [16] Z. Shuai and J. L. Brédas, Static and dynamic optical nonlinearities in conjugated polymers: Third-harmonic generation and the dc Kerr effect in polyacetylene, polyparaphenylene vinylene, and polythienylene vinylene, *Phys. Rev. B* **46**, 4395 (1992).
- [17] K. Schmidt and M. Springborg, Static (hyper-)polarizabilities of infinite, conjugated polymers in the framework of density-functional theory, *Phys. Chem. Chem. Phys.* **1**, 1743 (1999).
- [18] W. Hu, H. Ma, C. Liu, and Y. Jiang, Static polarizability and second hyperpolarizability of closed- and open-shell π -conjugated polymers, *J. Chem. Phys.* **126**, 044903 (2007).
- [19] V. Lacivita, M. Rérat, R. Orlando, M. Ferrero, and R. Dovesi, Calculation of longitudinal polarizability and second hyperpolarizability of polyacetylene with the coupled perturbed Hartree-Fock/Kohn-Sham scheme: Where it is shown how finite oligomer chains tend to the infinite periodic polymer, *J. Chem. Phys.* **136**, 114101 (2012).
- [20] A. V. Luzanov, Approximate computations of linear and nonlinear polarizabilities for conjugated systems within a configuration interaction approach, *Funct. Mater.* **30**, 268 (2023).
- [21] K. A. Pronin, R. H. Friend, and D. D. C. Bradley, Nonlinear optical susceptibility of conjugated polymers: d.c. Kerr effect, *Synth. Met.* **71**, 1689–1690 (1995).
- [22] J. Yu, B. Friedman, and W. P. Su, Quantum lattice fluctuations and nonlinear optics of conducting polymers, *MRS Online Proc. Libr.* **173**, 671 (1989).
- [23] T. W. Hagler and A. J. Heeger, Soliton-antisoliton configurations and the linear and nonlinear optical response of degenerate-ground-state conjugated polymers, *Phys. Rev. B* **49**, 7313 (1994).
- [24] A. Takahashi, Significant enhancement of optical nonlinearity by quantum lattice fluctuations in π -conjugated polymers, *Phys. Rev. B* **51**, 16479 (1995).
- [25] D. Yaron, Nonlinear optical response of conjugated polymers: Essential excitations and scattering, *Phys. Rev. B* **54**, 4609 (1996).
- [26] A. Takahashi, Nonlinear optical response of trans-polyacetylene to the strong static electric field, *Synth. Met.* **85**, 1087 (1997).
- [27] A. Takahashi, Effects of the quantum fluctuations on the nonlinear optical response of trans-polyacetylene to the static external electric field, *Phys. Rev. B* **56**, 3792 (1997).
- [28] Y. Matsuzaki, K. Tanaka, and T. Yamabe, Influence of backbone conformation on the third-order optical nonlinearity in polyacetylene, *Chem. Phys. Lett.* **284**, 261 (1998).
- [29] V. I. Margulis and E. A. Gaiduk, The effect of side chain substituents on third-order optical nonlinearity of conjugated polymers: A theoretical study, *Synth. Met.* **97**, 175 (1998).
- [30] S. Jiang and M. Xu, Hyperpolarizabilities for the one-dimensional infinite single-electron periodic systems. I. Analytical solutions under dipole-dipole correlations, *J. Chem. Phys.* **123**, 064901 (2005).
- [31] M. Xu and S. Jiang, Hyperpolarizabilities for the one-dimensional infinite single-electron periodic systems. II. Dipole-dipole versus current-current correlations, *J. Chem. Phys.* **123**, 064902 (2005).
- [32] J. A. Díaz-Ponce, Contribution of the transition moments, form of the absorption band, exciton, and the correlation effects in the linear and nonlinear optical properties of conjugated polymers, *Opt. Mater.* **66**, 595 (2017).
- [33] N. Marzari and D. Vanderbilt, Maximally localized generalized Wannier functions for composite energy bands, *Phys. Rev. B* **56**, 12847 (1997).
- [34] L. Fu, Topological crystalline insulators, *Phys. Rev. Lett.* **106**, 106802 (2011).
- [35] R.-J. Slager, The translational side of topological band insulators, *J. Phys. Chem. Solids* **128**, 24 (2019).
- [36] R.-J. Slager, A. Mesaros, V. Juričić, and J. Zaanen, The space group classification of topological band-insulators, *Nat. Phys.* **9**, 98 (2012).
- [37] J. Kruthoff, J. de Boer, J. van Wezel, C. L. Kane, and R.-J. Slager, Topological classification of crystalline insulators through band structure combinatorics, *Phys. Rev. X* **7**, 041069 (2017).
- [38] H. C. Po, A. Vishwanath, and H. Watanabe, Symmetry-based indicators of band topology in the 230 space groups, *Nat. Commun.* **8**, 50 (2017).
- [39] B. Bradlyn, L. Elcoro, J. Cano, M. G. Vergniory, Z. Wang, C. Felser, M. I. Aroyo, and B. Andrei Bernevig, Topological quantum chemistry, *Nature (London)* **547**, 298 (2017).
- [40] N. Marzari, A. A. Mostofi, J. R. Yates, I. Souza, and D. Vanderbilt, Maximally localized Wannier functions: Theory and applications, *Rev. Mod. Phys.* **84**, 1419 (2012).
- [41] W. Kohn, Analytic properties of Bloch waves and Wannier functions, *Phys. Rev.* **115**, 809 (1959).
- [42] A. P. Schnyder, S. Ryu, A. Furusaki, and A. W. W. Ludwig, Classification of topological insulators and superconductors in three spatial dimensions, *Phys. Rev. B* **78**, 195125 (2008).
- [43] A. Kitaev, V. Lebedev, and M. Feigel'man, Periodic table for topological insulators and superconductors, *AIP Conf. Proc.* **1134**, 22 (2009).
- [44] L. Fu and C. L. Kane, Topological insulators with inversion symmetry, *Phys. Rev. B* **76**, 045302 (2007).
- [45] J. P. Provost and G. Vallee, Riemannian structure on manifolds of quantum states, *Commun. Math. Phys.* **76**, 289 (1980).
- [46] J. Herzog-Arbeitman, V. Peri, F. Schindler, S. D. Huber, and B. A. Bernevig, Superfluid weight bounds from symmetry and quantum geometry in flat bands, *Phys. Rev. Lett.* **128**, 087002 (2022).

- [47] Y.-Q. Ma, S. Chen, H. Fan, and W.-M. Liu, Abelian and non-Abelian quantum geometric tensor, *Phys. Rev. B* **81**, 245129 (2010).
- [48] A. Bouhon, A. Timmel, and R.-J. Slager, Quantum geometry beyond projective single bands, [arXiv:2303.02180](https://arxiv.org/abs/2303.02180).
- [49] J. Ahn, G.-Y. Guo, and N. Nagaosa, Low-frequency divergence and quantum geometry of the bulk photovoltaic effect in topological semimetals, *Phys. Rev. X* **10**, 041041 (2020).
- [50] J. Ahn, G.-Y. Guo, N. Nagaosa, and A. Vishwanath, Riemannian geometry of resonant optical responses, *Nat. Phys.* **18**, 290 (2021).
- [51] See Supplemental Material at <http://link.aps.org/supplemental/10.1103/z7lp-pqp6> for more details on multi-band quantum geometry and bounds (Sec. I), derivation of quantum geometry-dependent hyperpolarizability (Sec. II), control of quantum geometry with external fields (Sec. III), topological bound on hyperpolarizability (Sec. IV), multi-band generalizations of the model for different π -conjugated polymers (Sec. V), and anharmonic oscillator (Sec. VI). The Supplemental Material also contains Refs. [52,53].
- [52] T. L. Hughes, Emil Prodan, and B. A. Bernevig, Inversion-symmetric topological insulators, *Phys. Rev. B* **83**, 245132 (2011).
- [53] J. E. Sipe and A. I. Shkrebtii, Second-order optical response in semiconductors, *Phys. Rev. B* **61**, 5337 (2000).
- [54] S. Kivelson, Wannier functions in one-dimensional disordered systems: Application to fractionally charged solitons, *Phys. Rev. B* **26**, 4269 (1982).
- [55] I. Souza, T. Wilkens, and R. M. Martin, Polarization and localization in insulators: Generating function approach, *Phys. Rev. B* **62**, 1666 (2000).
- [56] Y. Onishi and L. Fu, Universal relation between energy gap and dielectric constant, *Phys. Rev. B* **110**, 155107 (2024).
- [57] W. P. Su, J. R. Schrieffer, and A. J. Heeger, Solitons in polyacetylene, *Phys. Rev. Lett.* **42**, 1698 (1979).
- [58] W. P. Su, J. R. Schrieffer, and A. J. Heeger, Soliton excitations in polyacetylene, *Phys. Rev. B* **22**, 2099 (1980).
- [59] N. V. Tepliakov, J. Lischner, E. Kaxiras, A. A. Mostofi, and M. Pizzochero, Unveiling and manipulating hidden symmetries in graphene nanoribbons, *Phys. Rev. Lett.* **130**, 026401 (2023).
- [60] W. J. Jankowski, J. J. P. Thompson, B. Monserrat, and R.-J. Slager, Excitonic topology and quantum geometry in organic semiconductors, *Nat. Commun.* **16**, 4661 (2025).
- [61] B. C  rera, A. S  nchez-Grande, B. de la Torre, J. Santos, S. Edalatmanesh, E. Rodr  guez-S  nchez, K. Lauwaet, B. Mallada, R. Zbořil, R. Miranda, O. Gr  ning, P. Jel  nek, N. Mart  n, and D. Eci  ja, Tailoring topological order and π -conjugation to engineer quasi-metallic polymers, *Nat. Nanotechnol.* **15**, 437 (2020).
- [62] J. Yu, B. Friedman, P. R. Baldwin, and W. P. Su, Hyperpolarizabilities of conjugated polymers, *Phys. Rev. B* **39**, 12814 (1989).
- [63] R. Roy, Band geometry of fractional topological insulators, *Phys. Rev. B* **90**, 165139 (2014).
- [64] S. Kwon and B.-J. Yang, Quantum geometric bound and ideal condition for Euler band topology, *Phys. Rev. B* **109**, L161111 (2024).
- [65] T. B. Wahl, W. J. Jankowski, A. Bouhon, G. Chaudhary, and R.-J. Slager, Exact projected entangled pair ground states with topological Euler invariant, *Nat. Commun.* **16**, 284 (2025).
- [66] P. M. Tam, J. Herzog-Arbeitman, and J. Yu, Corner charge fluctuation as an observable for quantum geometry and entanglement in two-dimensional insulators, *Phys. Rev. Lett.* **133**, 246603 (2024).