Structural chirality measurements and computation of handedness in periodic solids

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We compare the various chirality measures most widely used in the literature to quantify chiral symmetry in extended solids, i.e., the continuous chirality measure and the Hausdorff distance. By studying these functions in an algebraically tractable case, we can evaluate their strengths and weaknesses when applied to more complex crystals. Going beyond those classical calculations, we propose a different method to quantify the handedness of a crystal based on a pseudoscalar function, i.e., the helicity during a soft phonon mode driven displacive phase transition from an achiral structure. This quantity, borrowed from hydrodynamics, can be computed from the eigenvector carrying the system from the high-symmetry nonchiral phase to the low-symmetry chiral phase. Different model systems like K_3NiO_2 , $CsCuCl_3$, and $MgTi_2O_4$ are used as test cases where we show the superior interest of using helicity to quantify chirality in displacive chiral transitions together with the handedness distinction.

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I. INTRODUCTION

A recent revival of chirality studies in materials has occurred due to its applicability in high-impact fields like topological insulators or chiral phonons in two-dimensional (2D) materials [1]. Despite this revival, relatively little attention has been paid to the structural chirality in crystalline phases. From a symmetry standpoint, the criteria for a crystal to exhibit chirality are straightforward: it must lack improper rotation symmetry elements within its symmetry group (i.e., no "operations of the second kind" which can convert righthanded coordinate systems into left-handed ones) [2]. This condition leads to a comprehensive classification of the 230 crystallographic space groups [1,3]. The first category encompasses 165 space groups which include improper symmetry operations, rendering them achiral. The second category comprises the remaining 65 groups, called Sohncke space groups, which are exclusively characterized by containing orientationpreserving operations (i.e., only "operations of the first kind") [2]. Among the Sohncke groups, we can further distinguish two different subcategories: 11 enantiomorphic pairs (22 chiral groups) characterized by containing screw axis of opposite handedness and 43 nonenantiomorphic space groups that will only preserve the chirality of the motif. The former subset of space groups will be the focus of this work.

However, this binary (chiral vs achiral) classification sometimes proves insufficient, particularly when we aim to identify materials with optimal chiral responses for specific functionalities such as optoelectronics [4]. A more nuanced approach involves categorizing crystals based on their varying degrees of chirality, offering a richer classification of materials [5,6]. To address this challenge, significant efforts have been dedicated in the literature to continuously quantify chirality over the past few years despite its inherent difficulty [7-9]. Evidence of such attempts can be found in various measures, including the continuous chirality measure [5,6], the Hausdorff distance [1,10], or the phonon pseudoangular momentum of the eigendisplacements [11,12]. Chirality measures are commonly divided into two primary categories based on their mathematical behavior under symmetry transformations: scalar and pseudoscalar functions. Scalar functions retain their value when subjected to mirror symmetry, making them invariant under such transformations. In contrast, pseudoscalar functions are sensitive to these symmetries and undergo a sign change when reflected. Intriguingly, in structural chirality determination, scalar functions are predominantly utilized in most established measures. This prevalence overshadows pseudoscalar functions, which remain relatively unexplored and elusive to the best of our knowledge.

In this study, we delve into the strengths and weaknesses of the previously announced chirality measures, e.g., the continuous chirality measure and the Hausdorff distance. Going further, we propose a fresh methodology to determine the handedness of crystalline solids that undergo displacive chiral transitions [13] by utilizing a pseudoscalar function inspired by the concept of helicity in hydrodynamics [14,15]. This innovative approach quantifies the degree of chirality in a crystal and distinguishes between right- and left-handed configurations.

However, it is worth noting that while all handed objects are inherently chiral, not all chiral objects possess unambiguous handedness, even if we can distinguish their enantiomers [1]. Consequently, our proposed method effectively assesses

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FIG. 1. Schematic view of the unit cell in the prototypical cubic ABO_3 perovskite structure together with their dominant instabilities without periodic boundary conditions. (a) Nonpolar cubic centrosymmetric phase. (b) Nonpolar rotation of the oxygen octahedra. (c) The off-centering motion of the *B* atom. Gray, blue, and red balls represent *A*, *B*, and O atoms, respectively. Yellow arrows indicate the direction for the nonzero displacements.

the chiral response of crystals undergoing a displacive chiral transition within the 11 pairs of enantiomorphic space groups [16], albeit it poses problems to those within chiral nonenan-tiomorphic space groups, limiting its applicability.

The paper is organized as follows. In the following section, we will inquire into the highly tractable scenario of a unit perovskite ABO_3 cell, where all the various chiral measurements can be computed algebraically, providing a clear understanding of their limitations. Moreover, we shall introduce the concept of helicity as a new chiral measure to quantify the handedness of a crystalline structure that undergoes a displacive chiral transition. After identifying the strengths and weaknesses of the aforementioned quantities, we shall apply them to different cases of interest. Subsequent sections will therefore be dedicated to the particular study of K₃NiO₂, Na₃AuO₂, CsCuCl₃, and MgTi₂O₄ that have been selected as paradigmatic examples.

II. CASE OF A UNIT PEROVSKITE CELL

In this section, we present the continuous chirality measure and the Hausdorff distance through a simple toy model formed by an isolated ABO_3 perovskite unit cell. This examination aims to delineate the applicability range of these approaches. Additionally, we shall introduce a different pseudoscalar measure to quantify the handedness of a given structure precisely.

A. Problems with continuous chirality measurements

We begin by examining a simplified model system: a prototypical ABO_3 perovskite unit cell, without considering its periodic replicas. In its centrosymmetric cubic phase, corresponding to the $Pm\bar{3}m$ space group under hypothetical periodic boundary conditions, the crystal is inherently nonchiral, as illustrated in Fig. 1(a). Next, we introduce structural distortions to this system and analyze their impact on different chirality measurements. According to Hlinka, chirality can arise from the interaction of a vector V with a rotational mode R, forming an axial vector. Although these distortions, when considered separately, might be achiral, their coupling can lead to the emergence of chiral distortions [17].

These vector and rotation modes can be achieved in our model by the distortions schematized in Fig. 1. In particular, a local rotation in a perovskite can be obtained by a rotation of the oxygen octahedra [Fig. 1(b)], and a polar vector can be

simplified by an off centering of the *B* cation [Fig. 1(c)]. Such transformations are easy to characterize algebraically, and the values of the new positions of the distorted atoms can be found in the following Eq. (1):

$$O_{x1} = \left(\frac{1}{2} - \frac{1}{2}\cos\theta, \frac{1}{2} - \frac{1}{2}\sin\theta, \frac{1}{2}\right),$$

$$O_{x2} = \left(\frac{1}{2} + \frac{1}{2}\cos\theta, \frac{1}{2} + \frac{1}{2}\sin\theta, \frac{1}{2}\right),$$

$$O_{y1} = \left(\frac{1}{2} + \frac{1}{2}\sin\theta, \frac{1}{2} - \frac{1}{2}\cos\theta, \frac{1}{2}\right),$$

$$O_{y2} = \left(\frac{1}{2} - \frac{1}{2}\sin\theta, \frac{1}{2} + \frac{1}{2}\cos\theta, \frac{1}{2}\right),$$

$$B = \left(\frac{1}{2}, \frac{1}{2}, \frac{1}{2} + \xi\right),$$
(1)

where θ is the rotation angle of the oxygen octahedra with respect to the z axis and ξ is the off centering of the B atom. We have, therefore, three different structures: (i) the high-symmetry cubic phase (that would correspond to a $Pm\bar{3}m$ space group with periodic boundary conditions), (ii) the lower-symmetry phase corresponding to the rotation of the oxygen octahedra (represented by a I4/mcm space group with periodic boundary conditions), and (iii) the phase corresponding to the off centering of the *B* cation (ascribed to a *I4/mcm* space group with periodic boundary conditions). Clearly, the two distortions are nonchiral individually, as they present mirror symmetry planes along z and x, respectively. However, when the octahedral rotation is combined with the off-centering motion of the B cation, a chiral structure emerges at the unit-cell level as it combines a vector (polar distortion) and an axial vector (octahedra rotation). It is important to highlight that, in the periodic crystal, the antiferrodistortive coupling of the oxygen octahedra causes octahedra to rotate cooperatively in opposite directions from unit cell to unit cell. In contrast, the off-centering direction is preserved, leading to opposite chirality and an overall achiral system. The system would be antichiral unit-cell wise as the chirality from one unit cell will reverse sign but keep the same absolute value concerning its neighboring cells. Nevertheless, for the sake of simplicity in our analysis, we will restrict our examination to the chiral nature observed at the unit-cell level in this straightforward scenario.

1. Continuous chirality measure

The continuous chirality measure (CCM) is a scalar measure introduced by Avnir that quantifies the structure's distance to its closest achiral reference [6]. Mathematically, it can be expressed as

$$CCM = \frac{1}{N} \sum_{1}^{N} ||\vec{x}_i - \vec{x}'_i||^2, \qquad (2)$$

where N is the number of atoms of the structure and \vec{x} , \vec{x}' are, respectively, the positions of the structure under study and its closest achiral reference. Although in later works, Avnir added a normalization factor by the root-mean-square size of the original centered structure [18], this will not be considered here for simplicity.

One inherent difficulty of the CCM [5,6] is that it requires selecting the closest nonchiral structure. One may think that a reliable choice would be the nondistorted high-symmetry phase, arguing that for small enough distortions, the closest nonchiral configuration is the original reference structure. This simple toy model demonstrates that such an assumption is generally incorrect. In the general case, we can follow a symmetry-adapted modes (SAMs) argument. Given a nonchiral high-symmetry phase (R) and a chiral low-symmetry phase (G), one can generally write the total distortion δ as the sum over SAMs, namely, as $a\delta_1 + b\delta_2$ where δ_1 and δ_2 are the modes associated with the irreducible representations brought up by the $R \rightarrow G$ transition and a and b are real projection coefficients. We can take δ_2 to be the symmetry-breaking mode, while δ_1 is the mode associated with an isotropy representation of the parent achiral group. Thus, the CCM is given by the norm of δ_1 rather than by the norm of δ that would be greater. In fact, the closest nonchiral structure in this example corresponds to the configuration with the octahedral rotation but without the B atom's off-center motion. This problem manifests when computing the CCM with the cubic centrosymmetric structure as a reference. Applying Eq. (2) to our perovskite toy model system with such a reference results in the following value:

$$\text{CCM} = \frac{1}{15} \sum_{i \in \text{atoms}} ||\vec{x}_i - \vec{x}_i^{Pm\bar{3}m}||^2 = \frac{1}{15} \left(\xi^2 + 4\sin^2\frac{\theta}{2}\right).$$
(3)

We can see that the CCM decouples the off-center motion of the *B* atom and the octahedra rotation. Therefore, we would obtain a nonzero value if θ is nonzero and ξ is zero or vice versa, even if we know that we need both distortions simultaneously to have a chiral structure. This is a consequence of the wrong selection of the reference structure. However, if we take as a reference the configuration containing the octahedral rotations, which is the closest nonchiral structure, the CCM would then have a value of

$$\text{CCM} = \frac{1}{15} \sum_{i \in \text{atoms}} \left| \left| \vec{x}_i - \vec{x}_i^{14/mcm} \right| \right|^2 = \frac{1}{15} \xi^2, \qquad (4)$$

which is not satisfactory either, as much important information is encoded in the selection of the reference structure, and the value of the final order parameter only relies on the off-centering value of the *B* atom. Consequently, the chirality contribution measure of two different distortions, D_1 and D_2 , will be higher for the distortion with greater off centering, regardless of the extent of their octahedral rotations. This discrepancy arises because the reference structures for both distortions differ, potentially leading to misconceptions.

These challenges become more pronounced as the crystal structure becomes more complicated. Identifying the closest nonchiral reference structure can be difficult. While recent numerical approximations, available at [19,20], have improved computational efficiency to N^2 (compared with the *N*! scaling of analytical methods [21]) with an error margin of 2% for cyclic symmetry groups [18], they may disrupt the connectivity map of the structure, resulting in unphysical references [22]. Furthermore, chiral distortions often involve multiple coupled modes in complex systems with many atoms in the unit cell. These modes can become decoupled, as seen in the case where octahedral rotation and the off-centering motion of the *B* cation are examined separately.

Aside from these problem-specific issues, other general problems and considerations about the difficulties of whether

continuous chiral measures can be well defined have been addressed in Refs. [8,9,23–25].

2. Hausdorff distances

The Hausdorff distance is defined as the supremum of the minimum distances between the structure and its closest nonchiral reference [1,10]. Therefore, it also relies on the preassumption of a reference structure concerning which computes the supremum of the infimum of the distances (i.e., the supremum for the case of maximal overlap between the structure and the reference). Consequently, the same types of problems discussed above for the CCM are at play too for the Hausdorff distance. When we compute the Hausdorff distance concerning the centrosymmetric structure, we obtain the following value:

$$\mathbb{H}(\operatorname{chir}, Pm\bar{3}m) = \sup\left\{|\xi|, \sin\frac{|\theta|}{2}\right\},\tag{5}$$

depending on the values of the off-centering motion and the octahedral rotation, the Hausdorff distance would vary. However, similar to the case of the CCM, since we are computing positive-definite distances, no distinction can be made between enantiomers if we reverse either the direction of the rotation or the direction of the off-centering motion. This problem is common to all scalar measures as mirror symmetries will leave the value unaffected.

Another problem arises with the Hausdorff distance when considering the distances' supremum. For small distortions, the distance between the *B* atoms is ξ , and the distance between the equatorial oxygens is $\sin \frac{\theta}{2} \sim \frac{\theta}{2}$. Let us imagine that the value of the off centering and the rotation is the same. In such a case, the Hausdorff distance would be ξ . However, the multiplicity of the oxygen atoms is not taken into account. Therefore, even if the overall contribution of the oxygen sites is larger than the one coming from the *B* atom, as the individual inputs are smaller, they are not considered in the chiral measure. This is an added problem to the ones highlighted above for the case of the CCM.

B. Computation of handedness

Handedness is intrinsically linked to rotational direction in physics. For instance, in chemistry, helicity refers to the sense of rotation of helical structures, with right-handed helices assigned a positive helicity value and left-handed helices assigned a negative helicity value [26]. Similarly, in hydrodynamics, the handedness of a flow is determined by the sign of its helicity, which measures the degree of linkage of the streamlines [15]. This quantity can be computed from the velocity lines of the flow as the following integral [14,15]:

$$\mathcal{H} = \int d^3 \vec{r} \, \vec{v} \cdot [\vec{\nabla} \times \vec{v}]. \tag{6}$$

The integrand of Eq. (6), the helicity density, is a pseudoscalar quantity that changes its sign under a mirror-symmetry operation. Thus, a nonzero helicity is associated with a lack of mirror symmetry: right (respectively left) handedness can be associated with positive (respectively negative) values of \mathcal{H} . Accordingly, the helicity modulus $|\mathcal{H}|$ quantifies the

strength of the handedness [15]. Contrary to scalar measures (like CCM or Hausdorff distances), the helicity measure can discriminate between enantiomers. However, other problems arise due to the chiral connectedness property [27-30]. This property refers to the fact that two enantiomorphs can be transformed into one another while remaining chiral throughout the transformation process. As elucidated in Refs. [27-29], this inevitably leads to what is known as the false-zeros problem: any pseudoscalar function will, in general, fail to determine the handedness of some objects assigning zero values to chiral entities. A notable exception to the falsezeros problem occurs in the set of helices with variable pitch, as discussed in Ref. [31]. In these cases, pseudoscalar functions exclusively yield zero values for achiral objects, making them suitable measures of chirality. Consequently, enantiomorphic space groups are unaffected by this issue and can be classified according to their handedness, indicating that pseudoscalar functions are more convenient for such classifications. Conversely, nonenantiomorphic space groups showing chirality like the Pb₅Ge₃O₁₁ compound [32] may exhibit chiral connectedness, making the presence of chiral nonhanded configurations inevitable.

The definition of helicity from fluid dynamics has been extended to the case of discrete fields defined in periodic crystals to compute the handedness of topological polar textures and is now widely accepted [33,34]. Borrowing this knowledge, we shall try to apply it to our particular problem where a velocity field can be established linking the achiral and chiral phases, i.e., going from continuous to discrete vector fields. In such a case, the velocity field can be defined, by the individual atomic displacements from the high-symmetry phase to the low-symmetry chiral phase (the difference in positions can be related with a velocity in an arbitrary units of time system following a finite difference approach). Indeed, we will choose the high-symmetry reference as the nonexcited phase and follow the eigenvector direction towards the nonexcited chiral state in an arbitrary time length. Therefore, in our perovskite toy model system, the velocities of the atoms give the following values:

$$\begin{aligned} \dot{\vec{x}}_{O_{x1}} &= \left(\frac{1}{2} - \frac{1}{2}\cos\theta, -\frac{1}{2}\sin\theta, 0\right), \\ \dot{\vec{x}}_{O_{x2}} &= \left(-\frac{1}{2} + \frac{1}{2}\cos\theta, \frac{1}{2}\sin\theta, 0\right), \\ \dot{\vec{x}}_{O_{y1}} &= \left(\frac{1}{2}\sin\theta, \frac{1}{2} - \frac{1}{2}\cos\theta, 0\right), \\ \dot{\vec{x}}_{O_{y2}} &= \left(-\frac{1}{2}\sin\theta, -\frac{1}{2} + \frac{1}{2}\cos\theta, 0\right), \\ \dot{\vec{x}}_{B} &= (0, 0, \xi). \end{aligned}$$
(7)

The curl $\vec{\nabla} \times \dot{\vec{x}}_i$, is then computed following a finitedifference approach and the integral is substituted by a regular sum at each atomic site in the unit cell analogous to the approach employed in polar textures at the supercell level [33,34] where the polarization vector field is also discrete. Note that whenever this field cannot be established as it is the case, for instance, of reconstructive phase transition this method cannot be applied. Furthermore, in the case of a sequence of displacive phase transitions $A \rightarrow$ $B \rightarrow C$, where both A and B are achiral and C is chiral, a stepwise approach should be adopted. This approach relies on well-defined eigenvectors governing each individual transition. Consequently, the helicity between the achiral states $A \rightarrow B$ would be zero, and the helicity would only be nonzero for the transition $B \rightarrow C$, where chirality emerges. With this definition and the set of velocities described in Eq. (7), we obtain a value of the helicity of

$$\mathcal{H} = \sin \theta \xi. \tag{8}$$

Therefore, to have a nonzero helicity, we need the coupling of the oxygen rotation and the polar displacement, as desired, to characterize a chiral structure. Moreover, suppose we reverse the sense of rotation or the sign of the polar distortion inducing a change of handedness in the structure. In that case, the helicity changes sign accordingly in contrast to the scenario observed with the CCM or Hausdorff distances.

In the following sections, we shall apply this method (available at [35]) to compute the handedness of different crystalline structures that undergo such continuous chiral transitions.

III. APPLICATION TO K₃NiO₂-LIKE CHIRAL STRUCTURE

One interesting application case is found in the K₃NiO₂ compound [36]. At 423 K, this crystal exhibits a first-order chiral phase transition from a high-symmetry achiral phase (P4₂/mnm space group) to a low-symmetry enantiomorphic phase (either $P4_12_12$ or $P4_32_12$). The transition is continuous and has been identified to come from a zone boundary soft phonon mode that explains the cell doubling during the transition [13]. Such a phenomenon could be present in all crystals that can be stabilized into the same high-symmetry $P4_2/mnm$ crystal structure. For example, although Na₃AuO₂ does not crystallize in this structure, a hypothetical Na₃AuO₂ phase isostructural to K₃NiO₂ is a useful nonmagnetic and closed shell toy model system for computational studies. In this section, we shall compute the CCM, Hausdorff distances, and helicity for K₃NiO₂ and Na₃AuO₂. The latter is much simpler from a computational perspective due to its nonmagnetic nature. It will be used to systematically analyze how the different chirality measures behave when connecting the different possible phases. In Fig. 2, we show a schematic representation of the K₃NiO₂ crystal. Atomic positions correspond to the high-symmetry $P4_2/mnm$ phase, and the arrows indicate how the atoms move when transitioning to the chiral $P4_12_12$ phase. In that way, we have defined a vector field for every atomic site in the reference structure and, therefore, we can compute all the quantities discussed in previous sections. We can see from the arrows plotted in Fig. 2 that a handed helical distortion pattern along the *c* direction is present.

As we can see in Table I for the case of Na_3AuO_2 , the values of the CCM and Hausdorff distances calculations give each the same value for both enantiomers, which is in line with what we discussed before in Sec. II A, i.e., that these measures cannot distinguish two enantiomers as distances are always positive definite.

Finally, the sign of helicity is sensitive to the system's change of handedness while its modulus is constant. Hence, and as anticipated in Sec. II B, the helicity modulus can measure the chiral distortion amplitude while its sign encodes the handedness "sign" of the structure.

To compare those different measures, we plot in Fig. 3 the evolution of the CCM, Hausdorff, and helicity



FIG. 2. Schematic representation of the K_3NiO_2 crystal structure where atoms occupy the $P4_2/mnm$ high-symmetry positions. Arrows indicate the direction of the atom displacements that bring the system to the $P4_32_12$ chiral phase. Purple, gray, and red balls represent K, Ni, and O atoms.

measures as a function of the distortion amplitude when going from the high-symmetry $P4_2/mnm$ phase to both the $P4_12_12$ and $P4_32_12$ phases. Those two phases are degenerate in energy and correspond to the two energy minima of the leftand right-handed enantiomers that can be formed from the $P4_2/mnm$ phase. The difference in the distortion pattern of those two enantiomeric phases is simply a change of sign, i.e., a change between right- and left-handed helical distortion. As expected from the different definitions, the module of the Hausdorff distance shows a linear dependence with η . In contrast, the module of the CCM and the helicity measurements have a quadratic behavior, as already reported in Ref. [1]. Moreover, as anticipated, the helicity sign is reversed for the different enantiomers, whereas the CCM and Hausdorff distances are not. Note that the CCM and Hausdorff distances could be multiplied by a sign representing the sense of rotation of the screw axis to differentiate between enantiomers.

TABLE I. Values of the different chirality measures discussed before for the Na₃AuO₂ compound taking the $P4_2/mnm$ phase as a reference. The + and – superscripts represent the enantiomorphic $P4_12_12$ and $P4_32_12$ phases, respectively. Fractional units and normalization by the number of atoms in the unit cell have been used to compute the CCM and helicity.

	$Na_3AuO_2^+$	$Na_3AuO_2^-$
ССМ	3.30×10^{-3}	3.30×10^{-3}
Hausdorff	1.18×10^{-1}	1.18×10^{-1}
Helicity	7.75×10^{-3}	-7.75×10^{-3}



FIG. 3. Comparison of the evolution of the different chiral measures as a function of the amplitude of the chiral distortion η in Na₃AuO₂. Positive (negative) values of η correspond to the condensation of the modes towards the $P4_12_12$ ($P4_32_12$) phase. Red dots, blue squares, and green triangles correspond to CCM, Hausdorff, and helicity measures. The values of the different measures have been normalized to display a value of 1 at the optimal amplitude of the chiral distortion ($\eta = 1$).

However, this measure will not serve to differentiate the degree of chirality of two different compounds due to the reference choice problem highlighted in Sec. II A 1.

Furthermore, when assessing the same calculations between the $P4_2/mnm$ phase and the *Cmcm* intermediate achiral phase, which is found to be an intermediate achiral structure before the enantiomorphic groups [13], we obtain a zero value from the helicity calculation. Therefore, the helicity calculation appears robust in determining the chirality of helical structures and can be applied straightforwardly. Moreover, as one can see in Table II, when we apply this procedure to the case of the K₃NiO₂ and compare the results with the ones obtained for the Na₃AuO₂, we observe that the ratio between of the CCM's and the ratio of the helicities is constant, suggesting the same helical structure for both compounds.

TABLE II. Values of the different chirality measures for the distinct compounds discussed in the work. Fractional units and normalization by the number of atoms in the unit cell have been performed to compute the CCM and helicity.

	Na ₃ AuO ₂	K ₃ NiO ₂	CsCuCl ₃	MgTi ₂ O ₄
CCM Hausdorff Helicity	3.30×10^{-3} 1.18×10^{-1} 7.75×10^{-3}	$\begin{array}{c} 1.24 \times 10^{-3} \\ 7.35 \times 10^{-2} \\ 2.75 \times 10^{-3} \end{array}$	$5.66 \times 10^{-3} \\ 1.07 \times 10^{-1} \\ 2.08 \times 10^{-2}$	1.03×10^{-4} 1.19×10^{-2} 3.08×10^{-4}



FIG. 4. Schematic representation of the CsCuCl₃ where atoms occupy the high-symmetry positions. Arrows indicate the direction of the displacements into the $P6_122$ chiral phase. Gray, blue, and green balls represent Cs, Cu, and Cl atoms.

IV. CASE OF CsCuCl₃

Next, we will discuss the chiral transition in CsCuCl₃. This material undergoes a transition from a high-temperature, high-symmetry $P6_3/mmc$ phase to one of the enantiomorphic groups $P6_122$ or $P6_522$ [37,38]. This transition is driven by a cooperative Jahn-Teller distortion as reported in Refs. [39,40]. Figure 4 illustrates a schematic representation of the CsCuCl₃ crystal, similar to our previous example. It shows the chiral crystal distortions, depicted as differences in atomic positions between the high-symmetry phase and one of the chiral phases. The displacement vector field demonstrates the righthanded helical structure. In Table II, the values of the different chirality measures can be encountered compared to other compounds. As we can see from the data, if we compare the values of the different measures for the Na₃AuO₂ and the CsCuCl₃ cases, we can see that the ratio between the CCM is half of the ratio between the helicities, indicating that the CsCuCl₃ adopts a more pronounced helical structure at equal distortion.

V. CASE OF MgTi₂O₄

A crucial prerequisite for computing the helicity of a given structure is the ability to establish a one-to-one mapping between atomic positions in the high-symmetry phase and those in the low-symmetry phase, thereby defining the displacement vector field. In previous examples, establishing this mapping has been straightforward. However, in the case of $MgTi_2O_4$, this is not so. The high-temperature phase exhibits a structure



FIG. 5. Schematic representation of the $MgTi_2O_4$. Atoms occupy the high-symmetry undistorted positions in a (a) $P4_12_12$ or (b) $P4_32_12$ representation. Arrows indicate the direction of the displacements into their respective chiral phases. Orange, blue, and red balls represent Mg, Ti, and O atoms, respectively.

of Fd-3m symmetry with 56 atoms in the conventional unit cell. In contrast, the chiral phase exhibits a $P4_12_12$ or $P4_32_12$ space-group symmetry with 28 atoms in the unit cell [41,42]. To establish such a mapping, we used the ISODISTORT software [43,44]. Beginning with the high-symmetry phase and setting the distortions that bring the system to the $P4_12_12$ or $P4_32_12$ phase to zero, we could derive a set of undistorted atomic positions that adhere to the desired symmetry. Afterward, we provided to AMPLIMODES [45,46] the undistorted and distorted atomic coordinates to obtain the set of displacements. In Fig. 5, we present the atomic displacements obtained using this method.

Similar to previous examples, the CCM and Hausdorff distances are equivalent for both enantiomers, while the helicity exhibits the same magnitude but opposite signs between them. The numerical values of the different chirality quantification methods can be found in Table II. In comparison to the Na₃AuO₂ case, the ratio between the CCM values roughly equals the ratio between the helicities, suggesting that the two compounds present approximately the same chiral strength. In contrast to the previously discussed cases where a clear helical structure was observable along the *c* direction, this compound exhibits a more evenly distributed helicity due to the lattice's transition from a face-centered-cubic to a tetragonal structure. The corresponding distortion results in the absence of a preferred direction, making it more difficult to identify the helical structure of the compound visually.

VI. CONCLUSIONS

In this work, we have delved into various continuous chirality measures proposed in the literature. Utilizing the simple model system of a unit perovskite cell, we have examined the challenges these measures pose when applied to extended solids. For the case of the CCM or Hausdorff distances, in addition to the difficulties associated with employing a position-based operator in periodic systems [47],¹ not discussed in the text, we have identified the critical issue of selecting an appropriate reference, which can often be nontrivial. Even in the case of small chiral distortions, relying on the high-symmetry phase as an accurate reference may not suffice, as the deformation could involve multiple nonchiral modes that only become chiral when coupled together as exemplified by the rotation (axial-like) and off-centering (vector) distortions. Consequently, much important information about the distortion can remain implicitly hidden even with the right reference selection. Moreover, because these methods rely on computing distances, which are positive-definite quantities, they cannot discriminate between the different enantiomorphic structures of a given compound. An interesting venue to explore would be to create tuples of chiral measures and increasing the number of degrees of freedom in order to have a better description of the distortion. However, there is strong mathematical evidence to believe that it will be insufficient to solve the chiral contentedness and consequent false-zeros problem [28,30].

Aside from the analysis of the various chirality measures in the literature [8], we have proposed using a unique pseudoscalar function, the helicity, to quantify the handedness of solids that undergo a continuous transition from an achiral to a chiral enantiomorphic space group. Borrowing the definition from hydrodinamics [14,15], and its extension to discrete fields in periodic solids [33,34], this quantity can be directly computed from the eigenvector that brings the system from the high-symmetry nonchiral phase to the low-symmetry chiral one. We have demonstrated the method's robustness in measuring the handedness of a given distortion in enantiomorphic groups that do not suffer the false-zeros problem. In such groups, helicity not only yields zero values for nonchiral transformations but also provides finite values for chiral structures, with equal magnitude and opposite signs for each enantiomorph. Such approach has unveiled the handedness of different compounds like Na₃AuO₂, CsCuCl₃, or MgTi₂O₄. The use of helicity as defined in the paper could therefore be incorporated as a potential descriptor for the high-throughput characterization of materials, similar to how the CCM has been utilized for organic molecules [48] as well as interesting to characterize chiral phonons [49]. While effective in measuring the handedness of crystals with enantiomorphic space groups (the 11 enantiomorphic pairs), it is important to note that chiral crystals that are within the 43 nonenantiomorphic space groups present chiral connectedness such that this approach may be inadequate in those cases enhancing the well-known difference between chirality and handedness [1]. We note that the method is limited to measure the handedness of systems that exhibit a displacive transition from an achiral reference to a chiral state. In order to extend this mechanism to a broader range of compounds, a minimal supergroup search can be conducted, analogous to the approach used for identifying displacive ferroelectrics [50]. A comprehensive study detailing the workflow and introducing new candidates for soft phonon mode-driven displacive chiral phase transitions is underway [51]. Additionally, the Appendix presents

a perspective on a potential reciprocal-space formulation of the helicity operator. Although not implemented in this work, this approach could inspire further advancements in the study of chiral distortions, potentially enabling its integration into density functional perturbation theory and finite-wavelength techniques [52].

Even though the helicity described in this paper remains reliant on a position-based operator and thus susceptible to common challenges in dealing with periodic systems [47], we hope that our proposed approach will facilitate a more systematic quantification of crystal handedness of enantiomorphic space groups. We believe that the helicity measurement described here shares some similarities to the polarization quantification problem in ferroelectrics. The most evident example is that we aim to measure to what extent a chiral crystal deviates from its achiral high-symmetry phase. Besides, some of the issues discussed in the paper, as the reference problem, also existed (and were solved by the modern theory of polarization [53]) in the case of the polarization and periodic boundary conditions. We hope that this paper will stimulate further work towards refining the quantification of structural chirality. For instance, it would be interesting to establish a relation between helical structures' helicity from enantiomorphic groups and their optical activity, e.g., from density functional theory as the optical activity calculation has been recently implemented [54].

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APPENDIX: RECIPROCAL SPACE FORMULATION OF THE HELICITY

This brief Appendix explores a potential alternative formulation of the helicity operator in reciprocal space. Notably, the value of the Fourier transform of the helicity density operator

¹The position operator does not commute with the Hamiltonian.

at the Γ point directly corresponds to the helicity

$$\hat{H}(\vec{k}=0) = \int \vec{v} \cdot (\nabla \times \vec{v}) \cdot e^{i\vec{0}\vec{r}} d^{3}\vec{r}$$
$$= \int \vec{v} \cdot (\nabla \times \vec{v}) \cdot 1 d^{3}\vec{r} = \mathcal{H}.$$

Now, we only have to compute the Fourier transform of the helicity density operator $\hat{H}(\vec{r}) = \vec{v} \cdot (\nabla \times \vec{v})$. Applying the convolution theorem [55], the Fourier transform of the product of two vectorial functions corresponds to the componentwise

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convolution of their respective Fourier transforms. Therefore, the following equality holds:

$$\mathcal{F}[\vec{v} \cdot (\nabla \times \vec{v})] = \mathcal{F}[\vec{v}] \dot{*} \mathcal{F}[\nabla \times \vec{v}]. \tag{A1}$$

Moreover, the Fourier transform of the curl of a function can be rewritten in the following way [55]:

$$\mathcal{F}[\nabla \times \vec{v}] = i\vec{k} \times \mathcal{F}[\vec{v}],\tag{A2}$$

which offers a concise expression for the reciprocal-space formulation of the operator discussed in the main text.

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