

Letter

On the concept of chirality

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In the literal sense of Kelvin's classical definition, chirality is a dichotomous concept. In this letter, we report on theoretical results which tend to alter profoundly this conception of chirality in a class of spaces of chiral systems. The example space considered here is the space of 2D square-integrable complex fields. Our results show that, in such spaces, chirality can be considered as a continuous, extensive and local geometrical phenomenon. The presented analysis, based on a theory of symmetry groups structure, provides a rigorous description of "the way", "the place where", and "the extent to which" an element of such spaces lacks indirect symmetries. Kelvin's definition is shown to describe the exterior signs of this phenomenon. A major interest of this theory is that all results can be applied to molecular wavefunctions and orbitals. Then there is hope that such results provide a renewed insight in basic stereochemical issues related to chirality.

Chirality and related notions such as prochirality are central concepts of stereochemistry [5] and tend to pervade all fields in chemistry and biochemistry. The name "chirality" itself was coined by Lord Kelvin. Now to this name Kelvin associated his celebrated definition: "I call any geometrical figure, or group of points, chiral, and say it has chirality, if its image in a plane mirror, ideally realized, cannot be brought to coincide with itself" [12]. Kelvin's definition universally dominates the conception of chirality in chemistry and biochemistry. An alternative statement of that definition is that a system is said achiral if its symmetry group G contains an indirect isometry (rotation–reflection axis, etc.) and that, if no such element can be found, it is said chiral. In the framework of this dichotomous definition, the property which is positively defined – by the fact that G contains some special type of elements – is achirality, and not chirality. As a corollary, the only stated property of chiral systems is that they fail to be achiral. Therefore, if specialized to the molecular case, Kelvin's framework indiscriminately sets chiral molecules into a single broad negatively defined category. As a consequence, *the ways* different molecules are chiral are neither distinguished nor described. *A fortiori*, in that framework, chirality is clearly and definitely not conceived as an extensive phenomenon.

In this letter, we report on recent theoretical results, of purely geometrical nature, which tend to alter profoundly this conception of chirality in a definite class of spaces

of potentially chiral systems. The considered spaces are the spaces of square-integrable complex fields. These results show that, in such spaces, there exists a definite geometrical phenomenon, to be precisely defined below, which determines the presence or the absence of indirect isometries in symmetry groups. Moreover, this phenomenon provides, in a unique way, a definition and a description of the way, the place where, and the extent to which an element of such spaces lacks indirect symmetries. This phenomenon, which is continuous, local, extensive and positively defined, therefore appears as the “hidden” geometrical mechanism at work behind the presence or the absence of indirect symmetries. From this point of view, indirect symmetries reduce to the dichotomous *exterior signs* of existence of this phenomenon. In other words, this phenomenon is similar to the continuous and extensive conception of the opening of a door, while the presence or the absence of indirect symmetries are similar to the door being dichotomously said “closed” or “open” (respectively, presence or absence of indirect symmetries). In that sense, our presently unnamed phenomenon appears as a *deeper* geometrical point of view than Kelvin’s dichotomous point of view in the considered spaces (figure 1). On the other hand, Kelvin’s definition is a symmetry statement. As will be shown below, the definition of the “unnamed phenomenon” relies exclusively on a group theoretical analysis. Therefore, both definitions equally are symmetry statements, the latter being much more elaborate than the former. These arguments suggest that the most adapted and relevant denomination of this phenomenon is the denomination of *chirality* itself. Therefore, in the considered spaces we propose to transfer to this phenomenon the denomination which is presently applied to Kelvin’s definition. Other general arguments in support of this proposition are dis-

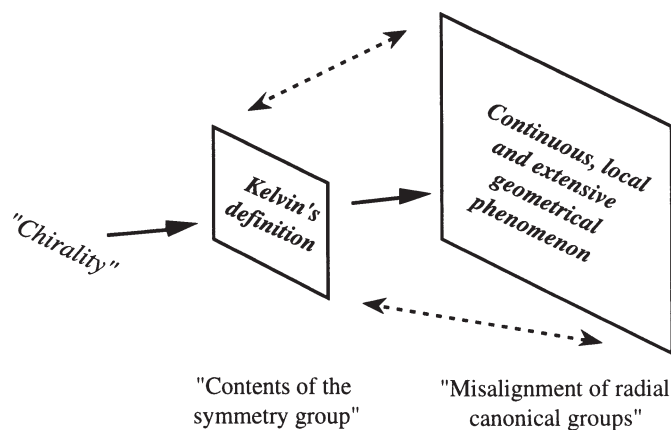


Figure 1. A representation of the relation between Kelvin’s definition and the continuous conception of chirality in H . First plain arrow: to the name “chirality” is associated Kelvin’s dichotomous definition. Second plain arrow: Kelvin’s definition is here shown to be the discrete expression of a continuous, local and extensive geometrical phenomenon. This result is only based on confronting this definition to the structure of H . Conversely, considering now a whole class of spaces like the Hilbert spaces $L^2(R^n, dm)$, this result shows that at a fixed level of space structure the two conceptions of chirality are logically equivalent (dashed arrows).

cussed in [16]. Then an expression of the central result of the presented theory is that *chirality can be considered as a continuous, extensive, local and positively defined geometrical phenomenon in a definite class of spaces.*

In order to avoid misinterpretations of the following theory, two important points will be discussed now. Firstly, the considered spaces are the Hilbert spaces of square-integrable spatial distributions of complex numbers (*fields*) [16]. The scope of this article will be limited to the first non-trivial example, that of two-dimensional (2D) fields

$$\iint_{R^2} |\psi(x, y)|^2 dx dy < +\infty. \quad (1)$$

This space would be denoted $L^2(R^2, dm)$ in analysis, where $dm = dx dy$ is the Lebesgue measure on the plane, and will be denoted H here. This space is the basic space of states of 2D non-relativistic quantum mechanics [16]. Then the major interest of the presented theory is that *all results can be applied to quantum mechanics.* However, it must be emphasized from the outset that our results are completely independent from the *interpretation* of H as a quantum space: the nature of the proposed theory is purely geometrical, so that it is firstly a mathematical theory. Nevertheless, it can be considered from the specialized point of view of quantum mechanics, and, if considered from this point of view, the following results practically provide physics and chemistry with a theory of the chirality of wavefunctions, that applies to all types of one-electron orbitals (HOMO, LUMO, etc.) or nuclear wavefunctions and also to molecular electronic densities,¹ in particular in the framework of the adiabatic approximation [2].² Secondly, note that the following theory should *not* be confused with the interesting and long-standing attempts at measuring or quantifying chirality by appropriate “degrees” or “indices” [1,3,4,7,9,11,13,14,17,19,20,22,23]. First of all, to the best of our knowledge, Kelvin’s definition has never been questioned in itself in such attempts, while this question is central to this work: we report here on a theory of the concept of chirality in itself, not on a theory of chirality degrees or indices, however fruitful and important such approaches prove to be. Moreover, the proposed degrees of chirality have been applied to very special objects – geometrical figures or finite sets of points in two and three dimensions [1,3,4,7,9,11,13,14,17,19,20,22,23]³ – while the results henceforth reported tend to emphasize the fundamental role of the space *structure* in the theory, as fully developed in [16]. To further insist on the basic

¹ Continuous integrable fields vanishing at infinity are square-integrable. Hence, molecular electronic densities, being continuous square moduli of wavefunctions, also belong to H .

² In particular, this theory can equally be applied to stationary and non-stationary quantum states [12]. If the Hamiltonian is invariant by parity, then stationary states can be chosen achiral, both in the sense of Kelvin’s framework and of the present conception. Then questions on the states (density operators) relevant to the description of statistical ensembles of chiral molecules appear largely distinct from questions on the notion of chirality itself. Former questions are firstly the province of ergodic theory and statistical mechanics.

³ An interesting distinction between physical and geometrical chiralities was recently introduced [6,8]. A thorough discussion of chirality in the physical context is found in [10].

theoretical and practical distinction between the concept of chirality and the notion of chirality degrees, note that no mention is made of “measuring” chirality in the scope of this article.

Salient features of the 2D theory will now be discussed. Complete proofs will appear elsewhere [16]. The theory starts with a simple remark, which can be grossly schematized in the following way: molecular theory is basically concerned with quantum wavefunctions. Then the discussion of molecular chirality should first be concerned with wavefunctions (see footnote 3 on the previous page). Now wavefunctions are very specific objects, in that quantum theory requires that they form vector spaces of well-defined structure called Hilbert spaces. The question is: what happens if we confront the structure of those spaces with Kelvin’s definition? Is it possible that, by embedding Kelvin’s general definition in a space of definite overall structure, chirality gets qualitatively new features originating in that structure? The advantage of considering the example of H is that H is the prototype of a category of spaces which are basic both to quantum mechanics and to analysis. Let us then analyse Kelvin’s definition in H . This definition can be expressed through symmetry groups, which are subgroups of the group of isometries [16]. Therefore the emphasis is placed on the properties of symmetry groups and isometries in H . Now the key argument relies on an original theory of the *structure* of these groups [16].⁴ This idea is best explained in a simplified context. Suppose that H could be decomposed as the direct sum of two linear subspaces H_1, H_2 globally invariant by isometries.⁵ Let ψ_1 (ψ_2) be the projection of $\psi \in H$ on H_1 (H_2) and G (G_1, G_2) be the symmetry group of ψ (ψ_1, ψ_2). Then it is shown [16] that $G = G_1 \cap G_2$. In plain words, G has a definite structure as the intersection of two more “elementary” symmetry groups G_1, G_2 . If G_1, G_2 are achiral, all questions concerning achirality of G are therefore changed into questions of *alignment* of the indirect symmetry elements of G_1, G_2 , since the indirect symmetry elements of G must be present in both groups. In the general case, the strategy is to iterate such decompositions down to achiral building blocks. A detailed structure of G is uncovered in this way. There appears a *single* preferred decomposition [16] whose 2D expression is

$$\begin{cases} G^{(r)} = \bigcap_{n \geq 1} G_n^{(r)}, \\ G_n = \bigcap_{r > 0} G_n^{(r)}, \end{cases} \quad G = \bigcap_{n \geq 1} G_n = \bigcap_{r > 0} G^{(r)} = \bigcap_{n \geq 1} \bigcap_{r > 0} G_n^{(r)}. \quad (2)$$

Let us briefly discuss the sense of this decomposition. This decomposition presupposes that a given origin O is selected. It is shown that this origin can be arbitrarily chosen in the plane (cf. the inversion center in figure 4 and the center of the double bond in figure 5). This result relies on an appropriate local generalization of Kelvin’s definition, called local chirality, which is argued to be of direct chemical significance in [16].

⁴ This argument is basically concerned with real fields. Complex fields are treated as two independent real fields [16].

⁵ This is equivalent to stating that the elements of H_1, H_2 have well-defined transformation laws under isometries.

The radial group $G^{(r)}$ is the symmetry group of the restriction $\psi^{(r)}$ of ψ to the circle of radius r and center O . The canonical group G_n is the symmetry group of the n th “canonical projection” ψ_n of ψ , i.e., the part of ψ having C_n rotational symmetry [16]. The radial canonical groups $G_n^{(r)}$ are the radial groups of canonical projections. Following (2), the $G_n^{(r)}$ are the universal *building blocks* of symmetry groups. All $G_n^{(r)}$ are shown to belong to the class D_n of 2D achiral groups [16]. Now, there are infinitely many groups $D_n[\alpha]$ in that class, which differ by the orientation α of the set of n indirect axes (n -star). It happens [16] that the orientation of the n -star of $G_n^{(r)}$ is related to the phase $\varphi_n(r)$ of the so-called absolute radial function (ARF) $\Delta_n(r)$,

$$\Delta_n(r) = \frac{1}{\sqrt{2\pi}} \int_0^{2\pi} \psi(r, \theta) e^{-in\theta} d\theta, \tag{3}$$

by the basic formula

$$G_n^{(r)} = D_n [-\varphi_n(r)/n]. \tag{4}$$

The relative orientation of $G_n^{(r)}, G_m^{(r)}$ depends similarly on the phase of the so-called relative radial function (RRF) $\Delta_{n,m}(r)$,

$$\Delta_{n,m}(r) = [\bar{\Delta}_n(r)]^M [\Delta_m(r)]^N \tag{5}$$

($M = m/p, N = n/p, p = \text{highest common factor of } n, m$) [16]. Radial functions are to play a crucial role in the following, so their basic representation is introduced now. In the complex plane (real part versus imaginary part), the complex functions $\Delta_n(r)$ and $\Delta_{n,m}(r)$ are represented by paths oriented in the direction of increasing r . These paths are shown to begin and to end at the origin [16]. Then they will respectively be called absolute and relative chiral loops (ACL, RCL) (figure 2).

What do we get by this analysis of symmetry groups? The fundamental result is that, following the decomposition (2), all questions on chirality (contents of G) are turned into questions on *misalignment* of the $G_n^{(r)}$ (n -stars). Two special forms of misalignment of the $G_n^{(r)}$ can be distinguished: (i) over r at fixed n , (ii) over n at fixed r . Following (2), case (i) deals with the chirality of the projections ψ_n , to be called *absolute* chirality, while case (ii) deals with the chirality of the restrictions $\psi^{(r)}$, to be called *relative* chirality. We shall concentrate now on absolute chirality. At fixed n , it is readily shown from (4) that two radial canonical groups $G_n^{(r_1)}, G_n^{(r_2)}$ are aligned – that is, have common indirect elements – if and only if (*iff*) $\Delta_n(r_1), \Delta_n(r_2)$ are aligned with the origin in the complex plane (figure 2). By straightforward generalization, a first result is that ψ_n is *achiral* iff the ACL is included in a *straight line* containing the origin.

However, the interpretation of ACLs is not confined to this extreme case. In the general case, relation (4) shows that the phase difference modulo π between $\Delta_n(r_1)$ and $\Delta_n(r_2)$ is a continuous measure of the relative orientation of $G_n^{(r_1)}, G_n^{(r_2)}$. Hence the ACL is an exact representation of the *distribution of orientations* among all groups $G_n^{(r)}$ in which G_n is decomposed. This is the basic property of ACLs. Let us now consider specific examples (figure 3). The ACL (a) is close to a line segment, hence the

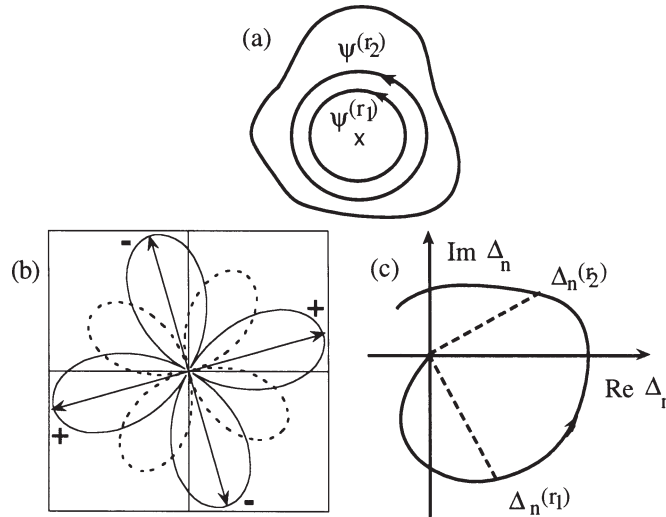


Figure 2. Relation between field, radial canonical groups and ACLs. (a) shows the restrictions $\psi^{(r_1)}, \psi^{(r_2)}$ of a field ψ to two circles of radii r_1, r_2 . The cross denotes an arbitrary origin. Arrows show the integration contours defining ARFs. (b) shows a polar plot in the case $n = 2$ of the component $\psi_n^{(r_1)}$ of $\psi^{(r_1)}$ having C_n rotational symmetry, which can be written [16] $\psi_n^{(r_1)}(\theta) = |\Delta_n(r_1)| \cos(n\theta + \varphi_n(r_1))$ (plain line). The sign reported beside every lobe is the sign of $\psi_n^{(r_1)}$. Arrowed lines show the n -star of $G_n^{(r_1)}$. The angle of the n -star to the Ox axis is $-\varphi_n(r_1)/n$. The dotted line is the polar plot of $\psi_n^{(r_2)}$ in the case of maximal misalignment (angular gap) of the n -stars. (c) is the translation of this situation in the language of ACLs. The phase difference modulo π between $\Delta_n(r_1)$ and $\Delta_n(r_2)$ measures the misalignment of n -stars. Whatever n , maximal misalignment corresponds to $\varphi_n(r_2) - \varphi_n(r_1) = \pm\pi/2$. The importance of the ACL is that it faithfully echoes all information on radial canonical groups misalignment.

decomposition of G_n results in almost aligned $G_n^{(r)}$. Accordingly, any of their n -stars is an approximate symmetry of ψ_n , and a small phase change $\delta\varphi_n(r)$ makes ψ_n achiral. This is the situation of weak misalignment. On the contrary, the ACL (b), wildly wandering in the plane, requires large phase changes in order to make ψ_n achiral so that no n -star can qualify as an approximate symmetry of ψ_n . This is the situation of large misalignment. While both ACLs correspond to chiral projections, since both ACLs differ from a line segment, these results suggest that (a) can properly be said “weakly chiral” and (b) “highly chiral”. These examples illustrate the basic result that the phenomenon of *radial misalignment* of the $G_n^{(r)}$, which is exhaustively echoed in the shape and the extent of ACLs, does provide a natural definition of “the way” and “the extent to which” a projection is chiral [16]. In other words, in H , and in the case of projections, this phenomenon indeed appears as the geometrical mechanism at work behind Kelvin’s conception of chirality. As previously stated, this phenomenon, to be called absolute chirality in keeping with the previous proposal, is continuous, extensive and positively defined. Achirality is now negatively defined as the nullity of misalignment. This phenomenon is also local in the sense that radial misalignment, just as the vibration of a string, can be unevenly distributed or localized in definite regions. As a final property, we get that this phenomenon is conveniently visualized

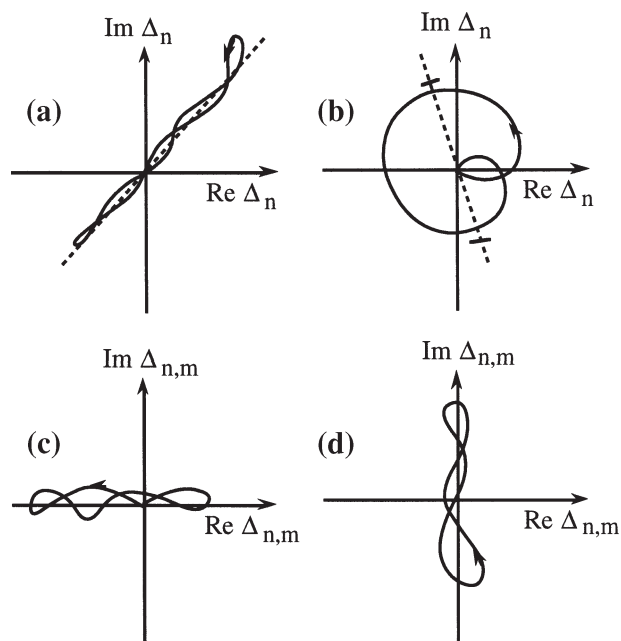


Figure 3. Example shapes of absolute ((a),(b)) and relative ((c),(d)) chiral loops. Orientations are emphasized by arrows. In case (a), the dashed line indicates the average direction of alignment. Radial canonical groups are approximately aligned, hence the projection whose ACL is (a) is to be considered as weakly chiral. In case (b), an arbitrary line was drawn (dashed line). The corresponding radial canonical group is a very poor symmetry of the projection except in the neighbourhood of the radii of its intersections with the ACL. The projection whose ACL is (b) is to be considered as highly chiral because of this large misalignment of radial canonical groups. In case (c), RRF values are close to the real axis, showing that the angular gap between radial canonical groups remains almost constantly small. Then the couple of projections whose RCL is (c) is to be considered as weakly relatively chiral. On the contrary, in case (d), the angular gap between radial canonical groups is almost constantly close to its maximum, so the couple of projections whose RCL is (d) is to be considered as highly relatively chiral [16].

through ACLs. An illustrative example of the shape of ACLs in the context of a 2D model of Walden inversion is shown in figure 4. Practical interpretation of the shape of ACLs follows from their basic properties and will be discussed in a forthcoming paper.

The analysis of relative chirality completely parallels that of absolute chirality [16], hence will not be detailed here. Relative chirality is a matter of misalignment between *sets* of projections (“over n ”). An important result is that this question can always be reduced to the case of *two* projections, in the sense that a set of projections is relatively achiral iff all couples formed from this set are relatively achiral [16]. The relative orientation of the radial canonical groups of a pair of projections is controlled by the phase of the RRF associated to this pair, so RRFs are to relative chirality what ARFs are to absolute chirality. However, the two forms of chirality differ by the criterium of achirality: a pair of projections is relatively achiral *at a given r* iff

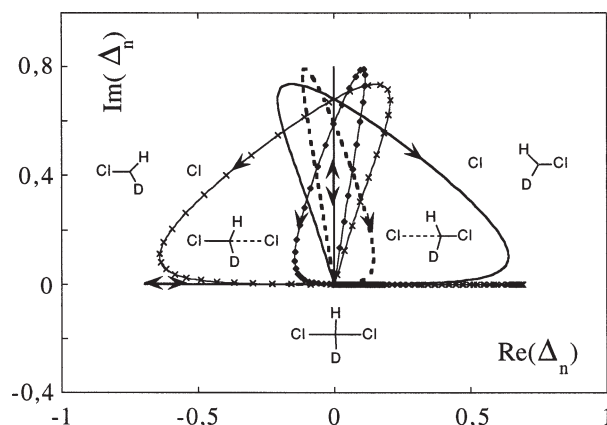


Figure 4. Evolution of absolute chirality in the S_N2 mechanism. We assumed definite geometrical changes in a 2D model simulating Walden inversion at a trivalent center with substituents H, D, Cl. Here the aim is to show the relation between given geometrical changes in a system and the subsequent evolution of chirality. The evolution of chirality during a chemical reaction is a special case of this relation. Atoms are modeled by Gaussian electronic densities of different widths (atomic radii) and weights. Analytically known ARFs of order $n = 1$ of the system as a whole are reported at various reduced times t (unit \AA^{-2}). From left to right: (i) $t = -1$ (crossed line), the ACL with a distant nucleophile, (ii) $t = -1/4$ (dotted line), the system becomes weakly chiral as both Cl are close to the center, (iii) $t = 0$ (vertical line), the ACL of the assumed achiral transition state is a straight line, (iv) $t = 1/4$ (dashed line) and (v) $t = 1$ (plain line), enantiomer systems generate symmetrical ACLs [16]. Such diagrams are typical of Walden inversion.

the RCL crosses the *real axis* at r . Then, by a straightforward generalization, two projections are relatively achiral (i.e., over all r) iff the RCL is included in the real axis (figures 3 and 5). As in the case of absolute chirality, the phenomenon at work behind the previous dichotomous definition (presence or absence of indirect symmetries in $G^{(r)}$) is the continuous, local, extensive and positively defined phenomenon of relative misalignment (i.e., over n at fixed r) of the $G_n^{(r)}$. According to our proposition, this phenomenon is to be called relative chirality. An illustrative example of the shape of RCLs in the context of 2D *cis-trans*-isomerism is shown in figure 5.

Gathering these results, we get a synthetic conception of chirality in H . There appears two and only two special theoretical forms of chirality – absolute and relative chirality. In that respect, a basic result is that a field is achiral iff it is both absolutely and relatively achiral [16]. Then previous results on the special cases of absolute and relative chirality show that the presence/absence of indirect symmetries in G is determined by the *phenomenon of misalignment of the radial canonical groups* $G_n^{(r)}$ (n -stars in 2D) over the *full domain* of indices n, r [16]. This phenomenon is exhaustively represented by the set of relevant ACLs and RCLs. It is, indeed, continuous, local, extensive and positively defined, as announced in introduction. In keeping with our proposal, this phenomenon is to be called “chirality”. Absolute and relative chirality appear as differing only by the two “independent directions” over which misalignment is considered.

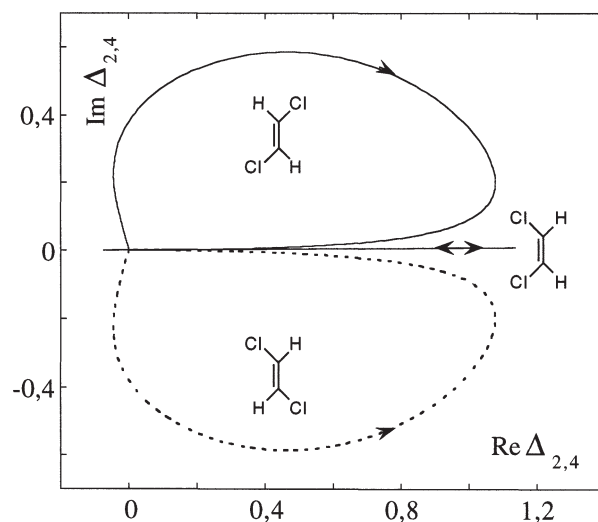


Figure 5. 2D *cis-trans*-isomerism. Atoms are modeled by Gaussian electronic densities whose parameters simulate 1,2-dichloroethylene [16]. In 2D, there are two *trans*- and one *cis*-isomers. *Trans*-isomers have C_2 symmetry at the double-bond center, which is the considered origin, so their odd order ARFs are zero. Non-zero RCLs of lowest order $n, m = 2, 4$ are reported for the two *trans*-isomers ($\Delta_{2,4} = [\overline{\Delta}_2]^2 \Delta_4$). The *cis*-RCL of the same order is also reported. Data are expressed in 10^{-5} \AA^{-6} . The *trans*-isomers are 2D enantiomers, so have symmetrical RCLs of opposite orientations [16]. The *cis*-isomer is achiral, so the *cis*-RCL lies on the real axis. *Cis*- and *trans*-isomers are diastereoisomers, hence have RCLs of different shapes. In the *cis* case, 2D prochirality could be evaluated by slightly varying the description of one model substituent. Enantiotopes are then shown to generate symmetrical loops. Note that the 2D *trans*-isomers correctly appear as a single achiral molecule in the 3D theory.

Let us briefly – and in an extremely preliminary way – comment on this conception of chirality in the light of experimental evidence. From the experimental point of view, all signatures of chirality, such as NMR chemical shift differences between enantiomers in chiral solvents, or circular dichroism, point at continuously varying quantitative differences between the pseudo-tensorial observables associated to different chiral molecules. Other types of signatures, such as enantiomeric excess or reaction rate measurements, point at the same conclusion. Such continuous variations are directly related to the various observables at stake in every peculiar experiment, so have nothing to do a priori with chirality being conceived either as a dichotomous or a continuous phenomenon. However, there are many molecules, such as 1-stearoyl-2,3-dipalmitoylglycerin or 7-methyl-[1- ^{14}C]-tridecane [3] for which *all* experimental signatures tend to be simultaneously weak. There exists a wide-spread intuitive tendency to consider such molecules as “weakly” chiral [1,3,4,6–11,13–15,17–23]. What is usually more or less explicitly understood by such a statement is, firstly, that the origin of such coherent experimental evidence is ascribed to the “geometry” of the molecule, and secondly, that it is assumed that, in some indetermined sense, this geometry is such that the molecule is “close” to being achiral. Such intuitive stereochemical interpretations are in direct conflict with Kelvin’s conception of chirality.

Now this paradoxical issue is naturally resolved in the continuous conception, which precisely provides room for weakly chiral electronic/nuclear distributions, in the above well-defined and geometrically profound sense. Similarly, we feel that many chemists would agree that L-Alanin is “more” chiral than L-[2-*d*]-Glycin, in the light of coherent reactivity evidence. Such comparisons are put on a firm and profound basis by this theory. Therefore, there is hope that these results provide renewed insight in the basic issues of chirality in chemistry and biochemistry. As exemplified by figures 4 and 5, practical applications of chiral loops can be expected both in structural studies, as a means to visualize and analyse the distribution of chirality in all types of (bio)molecules, and in dynamical studies, in particular in relation with asymmetric synthesis. Work on the 3D theory is in progress. However, the 2D approach is immediately relevant to the active field of chiral phenomena connected to interfaces (chiral monolayers, membranes, other types of interfaces) or to systems properly represented by 2D models (planar molecules, some host–guest systems, etc.). The chirality of such systems can already be studied in the proposed framework, as we showed in the case of *cis-trans*-isomerism and Walden inversion.

In conclusion, we showed that an extension of Kelvin’s conception of chirality as a continuous, extensive, local and positively defined geometrical phenomenon naturally follows from analysing the structure of symmetry groups in the space H . This concept, to which the denomination of chirality is naturally extended, tends to support intuitive experimental interpretations of chirality as a continuous phenomenon, and leads practically to a detailed representation of chirality by the two basic forms of chiral loops – absolute and relative chiral loops. This approach was exemplified on two model systems, Walden inversion and *cis-trans*-stereoisomerism.

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