

Handedness Selection and Hysteresis of Chiral Orders in Crystals

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A phase transition can drive the spontaneous emergence of chiral orders in crystals below a critical temperature. However, selecting either a right- or a left-handed phase with the aid of electromagnetic fields is challenging, particularly when intrinsic polar and axial moments are lacking. In this Letter, we show that purely chiral phases with opposite handedness, when both deriving from one degenerate instability, are linked by accessible transition states. While these states compete with the chirality under an electromagnetic field, a circularly polarized source can select the handedness of the system. This selection is mediated by a chiral monopole and may further result in a hysteresis process of the gyrotropic properties, namely, the optical activity, below the critical temperature. We suggest several materials, among which K_3NiO_2 , as candidates for possible experimental observation.

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Introduction—Many systems in nature are chiral and cannot be superimposed on their mirror images. While the structural chirality is often fixed by the crystallization process, it can emerge as a time-reversal even order parameter [1,2] (as an active degree of freedom) and also spontaneously appear below a finite critical temperature (T_c) [3–8]. Following Ref. [9], a chiral monopole G_0 , a time-reversal even polarization $\mathbf{P}$, and an electric-toroidal (axial, parity even) $\mathbf{G}$ order [10–13] couple via the $G_0 \cdot \mathbf{P} \cdot \mathbf{G}$ energy invariant. This suggests that a handedness (right, R and left, L) of choice could be selected in some crystals with the help of electromagnetic (EM) fields that control the sign of $\mathbf{P} \cdot \mathbf{G}$. In particular, a linearly polarized electric field may be used if either $\mathbf{P}$ or $\mathbf{G}$ are intrinsically present [14,15], as experimentally realized [16] in the case of spontaneous helical dipolar textures and photoinduced chiral distortions [17]. In particular, helical textures show a hysteresis of the handedness ($R \leftrightarrow L$ conversion below T_c) under a dc field [16]. However, chiral materials possess neither polar nor axial orders in general. In this case, a circularly polarized EM field is necessary to select a handedness [18], and heating-cooling cycles with respect to T_c seem to be required to convert it ($R \leftrightarrow L$) when a spontaneous chiral order is present [6,18], possibly due to a lack of a suitable transition path. It is hence questionable whether the handedness of a spontaneous chiral order below T_c can generally be controlled.

With the help of density functional theory (DFT) and Landau theory, we show that a way to overcome this hindrance is to consider a degenerate chiral instability that induces both right- and left-handed phases, so that a low-energy transition path connecting R and L is guaranteed. Therefore, we analyze the nonlinear interaction between the chirality and induced polar and electric-toroidal

moments as a way to realize handedness conversion. In particular, we distinguish between selective and nonselective effects that are simultaneously activated by an external source. First, we show that a field-mediated competition between chiral and transition states can take place. The latter are particularly favored if the external source is linearly polarized, which hinders the conversion process. Nevertheless, through static and dynamical calculations, we show that selecting a handedness and realizing a hysteresis of the chirality and associated properties (for instance, the optical activity) below T_c is feasible if a $\mathbf{P} \cdot \mathbf{G}$ monopole is switched on, for instance, by a chiral electromagnetic field. While evaluating our model on the K_3NiO_2 compound [19], this mechanism applies to several families of materials, as long as one degenerate instability generates both right- and left-handed chiral phases.

Chiral degenerate instability and transition paths—A degenerate instability is not a fundamental requirement for the spontaneous emergence of a chiral order; nevertheless, such an occurrence is not uncommon in nature. For instance, it can be expected if the resulting right- and left-handed low-symmetry phases are enantiomorphs; namely, their respective space groups are different, as is the case for many materials such as K_3NiO_2 [19], ABX_3 halide perovskites (e.g., CsCuCl_3) [20,21], AB_2X_4 spinels (e.g., MgTi_2O_4) [22–25], or SiO_2 cristobalite [26–29]. For symmetry reasons, the triggering chiral distortions $|R\rangle$ and $|L\rangle$ must be independent. As follows, we consider an insulator K_3NiO_2 (KNO), as a practical example. The experimental high-symmetry phase of KNO adopts an achiral tetragonal structure with $P4_2/mnm$ (no. 136) space group, and a first-order phase transition to low-temperature phases of either right-handed $P4_12_12$ (no. 92) or left-handed $P4_32_12$ (no. 96) symmetries has been measured on

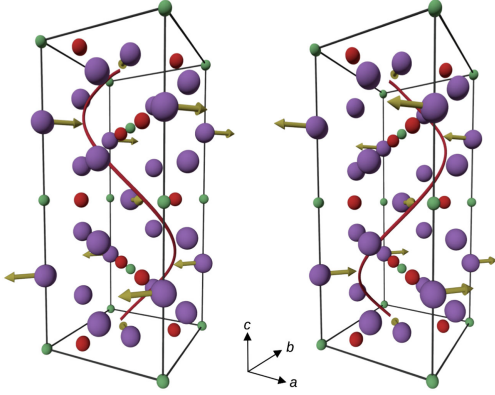


FIG. 1. Scheme of the nonequivalent handed helices spontaneously induced by a degenerate instability in K_3NiO_2 . The purple, green, and red spheres correspond to K, Ni, and O atoms. The ions are in the $P4_2/mnm$ configuration ($1 \times 1 \times 2$ supercell). At the same time, the golden arrows and the red helices represent the Z_4 chiral ($P4_12_12$ left, $P4_32_12$ right) displacement (for clarity, only the dominant potassium displacement is shown).

cooling below ~ 423 K [19]. A symmetry mode analysis [30] of the experimental structures shows that the transition is induced and chiefly described by an instability at the $Z = (0, 0, 1/2)$ point of the Brillouin zone, with the Z_4 doubly degenerate representation. This analysis is supported by our DFT + U calculations [31–35] ($U = 4.2$ eV [35]), highlighted in a separate work [36], of the phonons [37,38] and relaxed structures. Indeed, we identify a doubly degenerate unstable mode at the Z point, with frequency $\omega_Z = 1.35i$ THz and Z_4 irreducible representation that leads to the R and L low-symmetry ground states (see Fig. 1). As a consequence of the degeneracy, arbitrary $|\psi(\theta)\rangle \equiv \sin\theta|R\rangle + \cos\theta|L\rangle$ combinations of right- and left-handed eigenstates can be considered, where θ is a mixing angle. Thus, the Landau free-energy expression F reads [36,39]

$$F(\phi, \theta) = \alpha\phi^2 + \{\beta_1 + \beta_2\sin^2 2\theta\}\phi^4, \quad (1)$$

where ϕ is the amplitude of the distortion. While choosing $\theta = \{k\pi, k\pi + \pi/2\}$ to correspond to the $|R\rangle$ and $|L\rangle$ distortions, respectively, their $|R\rangle \pm |L\rangle$ combinations possess $Cmcm$ (no. 63) nonchiral symmetry. A generic $\theta \neq \{\pi/4 + k\pi/2, \pi/2 + k\pi, k\pi\}$ lowers the symmetry to $C222_1$ (no. 20) instead. The β_2 parameter breaks the rotational invariance of the energy surface in Eq. (1). As such, and upon full structural relaxation, we compute a $\Delta E = 17.9$ meV per formula unit (f.u.) energy gain for both the R and L ground states. This gain reduces to $\Delta E = 12.2$ meV f.u. in the (degenerate) $Cmcm$ phases, denoting possible competition with the chiral orders. Importantly, epitaxial constraints and hydrostatic pressure may tune these energy scales [36]. Also, Eq. (1) is additionally justified since secondary distortions are negligible [36].

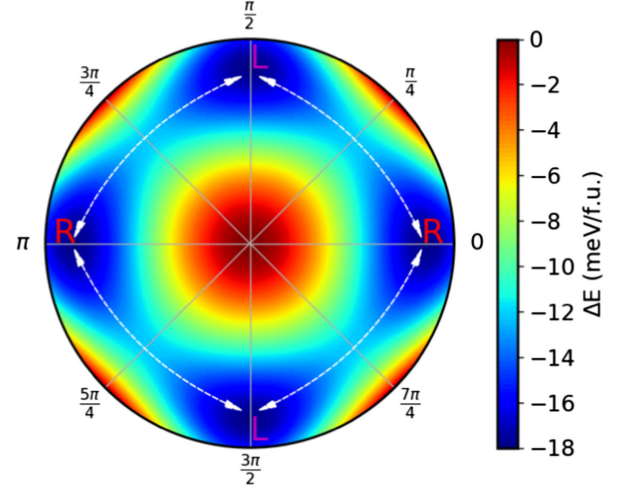


FIG. 2. Energy surface $F = \alpha\phi^2 + \{\beta_1 + \beta_2\sin^2 2\theta\}\phi^4$ of the Z mode instability of K_3NiO_2 (amplitude ϕ in arbitrary units). The origin corresponds to the high-symmetry $P4_2/mnm$ structure. The chiral $P4_12_12$ and $P4_32_12$ phases are located at $\theta = \{0, \pi\}$ and $\theta = \{\pi/2, 3\pi/2\}$, respectively, while the $Cmcm$ phases is at $\theta = \{\pi/4 + k\pi/2\}$. The white arrows indicate possible transition paths connecting R and L through low-energy $Cmcm$ transition states.

A free energy as in Eq. (1) is universal whenever $|R\rangle$ and $|L\rangle$ span a multidimensional space [39]. This suggests low-energy $L \leftrightarrow X \leftrightarrow R$ transition paths (absent if the instability is nondegenerate) to convert the handedness of a crystal below T_c , where the X orthorhombic transition state is energetically closer to the chiral structures compared to the reference configuration. In Fig. 2, we show these paths in the case of KNO ($X = Cmcm$ phases).

Chiral order and EM field interaction—To see if an external electromagnetic field can be used to trigger a permanent handedness conversion, we need to understand how the energy surface interacts with parity-odd polar $\mathbf{P}$ ($\Gamma_3^- \oplus \Gamma_5^-$ representation) and parity-even axial $\mathbf{G}$ ($\Gamma_3^+ \oplus \Gamma_5^+$ representation) distortions generated by such a field. At the lowest (fourth) order, this interaction is given by

$$\begin{aligned} V^{(4)} = & \phi^2 \sum_{i=x,y,z} (a_i P_i^2 + b_i G_i^2) + \\ & + \phi^2 \cos 2\theta \sum_{i=x,y,z} \lambda_i P_i G_i + \\ & + \phi^2 \sin 2\theta [\gamma_1 P_x P_y + \gamma_2 G_x G_y], \end{aligned} \quad (2)$$

and we can evaluate its effect on the energy surface as follows: The isotropic $V_{\text{biquad}} \equiv \phi^2 \sum_i (a_i P_i^2 + b_i G_i^2)$ term does not break any symmetry of Eq. (1). The second term $V_{\text{flip}} \equiv \phi^2 \cos 2\theta \sum_i \lambda_i P_i G_i$ instead crucially lifts the degeneracy of the right- and left-handed phases by coupling the intrinsic chiral order to $P_i G_i$ monopoles. The final

$V_{\text{ortho}} \equiv \phi^2 \sin 2\theta [\gamma_1 P_x P_y + \gamma_2 G_x G_y]$ term does not affect the R and L structures, but splits the energies of non-equivalent transition states (generated by $|R\rangle + |L\rangle$ and $|R\rangle - |L\rangle$, respectively). We show in Supplemental Material Sec. II [31] that an interaction such as Eq. (2) and a 2D chiral order parameter are present in various different materials, thus suggesting a common phenomenology for handedness selection. In the absence of intrinsic polar and electric-toroidal moments, a finite $\mathbf{P} \cdot \mathbf{G}$ monopole must be induced to generate a net chiral effect in the material [9]. While a static linearly polarized electric field $\mathcal{E}$ can trigger both polar $\mathbf{P}$ and electric axial distortions, the latter $\tilde{\mathbf{G}}_{xy} \sim (\mathbf{P}_{xy} \times \mathbf{P}_z)$ as a dipolar torque [39,40], one can easily see that the resulting $\mathbf{P} \cdot \tilde{\mathbf{G}}_{xy}$ monopole is zero, and thus, no handedness can be selected in tetragonal, hexagonal, and cubic systems [e.g., if $\lambda_x = \lambda_y$ in Eq. (2)]. On the other hand, a related but different electric-toroidal order $\mathbf{G} \equiv \sum_{i=1}^N \mathbf{u}_{xy,i} \times \mathbf{p}_{zi} / \Omega_{\text{u.c.}}$ [11,41] originates from the multipole expansion of the polarization density field $\mathbf{P}(\mathbf{r})$ [31,42]. Hence, the interaction between $\mathbf{P}$, $\mathbf{G}$, and an EM field can be expanded as

$$V = -\mathbf{P} \cdot \mathcal{E} - \mathbf{G} \cdot \nabla \times \mathcal{E} - \text{other multipoles}, \quad (3)$$

which results in $\mathbf{P} \sim \mathcal{E}$ and $\mathbf{G} \sim \nabla \times \mathcal{E}$ [11,41]. The R - L energy difference is thus given by $\sim \phi^2 \sum_i \lambda_i \mathcal{E}_i (\nabla \times \mathcal{E})_i$, which couples the dynamical chirality of an electromagnetic field $\rho_{\chi} \equiv (\epsilon_0/2)\mathcal{E} \cdot (\nabla \times \mathcal{E}) + (1/2\mu_0)\mathcal{B} \cdot (\nabla \times \mathcal{B})$ [43] to that of the material, as experimentally shown in Ref. [18].

Chiral and orthorhombic competition—Since an EM source is likely to activate both selective (V_{flip}) and nonselective effects (V_{biquad} , V_{ortho}), it is important to visualize how they compete with each other. To support our analysis, we develop a DFT [44,45], static, and

nonmagnetic model isostructural to K_3NiO_2 by replacing $\{\text{K}, \text{Ni}\} \rightarrow \{\text{Na}, \text{Au}\}$ (see Supplemental Material Sec. III A [31] and Ref. [46] for the details of its construction). We start with the nonselective processes.

First, we apply a constant linearly polarized electric field [47] $\mathcal{E} = 4.63 \times 10^{-2}$ V/nm along the $[1,1,1]$ direction of the high-symmetry phase and relax the unit cell. Then, we extract the polar and the torque-induced axial modes with polarization $\mathbf{P}_0 = (0.25, 0.25, 0.19)$ e/nm² and electric-toroidal moment $\mathbf{G}_0 = (-0.002, 0.002, 0.0)$ e/nm, which corresponds to Eq. (3) with $\nabla \times \mathcal{E} = 0$. Their effect on the energy landscape is shown in Fig. 3. An up-shift of all energies due to the overall positive isotropic term of Eq. (2) (V_{biquad}) and the splitting of the $Cmcm$ phases (V_{ortho}) along the $|R\rangle + |L\rangle$ and $|R\rangle - |L\rangle$ directions [cyan and green Fig. 3 (b) curves] are observed. Moreover, no R - L energy difference is detected, consistent with the symmetries of the unit cell. While an additional strain x deformation ϵ_{xx} breaks the tetragonal symmetry and introduces a $\mathbf{P} \cdot \mathbf{G}$ monopole, the calculated R - L splitting $\sim (\lambda_x - \lambda_y)\phi^2 P_x P_y P_z$ is found to be negligible (see Supplemental Material Sec. III B [31]). Clearly, and as shown in Fig. 3, a $Cmcm$ phase may be stabilized by a linearly polarized electric field (temporarily if $\mathcal{E} \rightarrow \mathcal{E}e^{-t/\tau}$), which prevents or strongly hinders the selection or conversion of a handedness unless a chiral source is used.

R - L splitting and handedness hysteresis—To probe a static net chiral effect, we can simulate a $\mathbf{P} \cdot \mathbf{G} \equiv P_{x,0}G_{x,0}$ monopole by applying the previously calculated polar and axial distortions corresponding to $\mathbf{P} \equiv (P_{x,0}, 0, 0)$ and $\mathbf{G} \equiv (G_{x,0}, 0, 0)$ to the energy surface, which eliminates the splitting of the transition states and downsizes the biquadratic terms. We also introduce scaling factors $P_x/P_{x,0}$ and $G_x/G_{x,0}$ as controllable parameters in our model. This is equivalent to employing a circularly polarized EM source

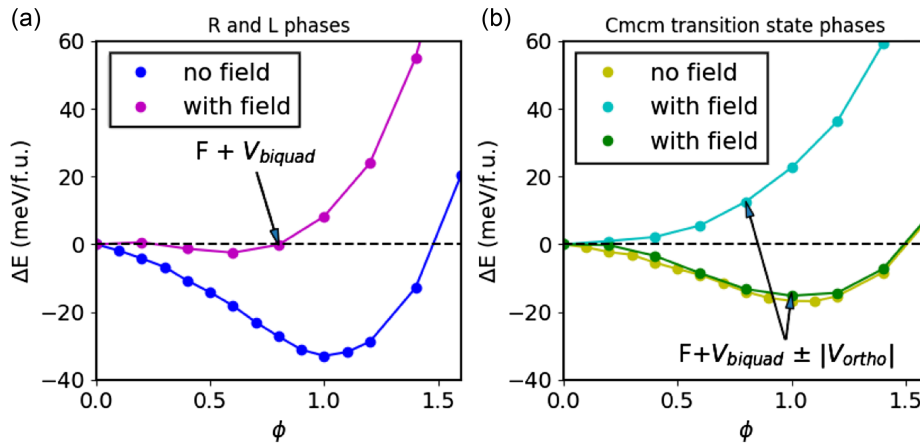


FIG. 3. Density functional theory energy landscape as a function of the ϕ amplitude (arbitrary units), with and without an electric field $[1, 1, 1] \times 4.63 \times 10^{-2}$ V/nm and no axial conjugate field. F , V_{biquad} , and V_{ortho} are defined in the main text. The induced chiral-polar-axial interaction cannot generate any R - L splitting but allows (a) an up-shift of the R and L energies and (b) the up-shift and splitting ($\pm V_{\text{ortho}}$) of the transition states $Cmcm$ energies.

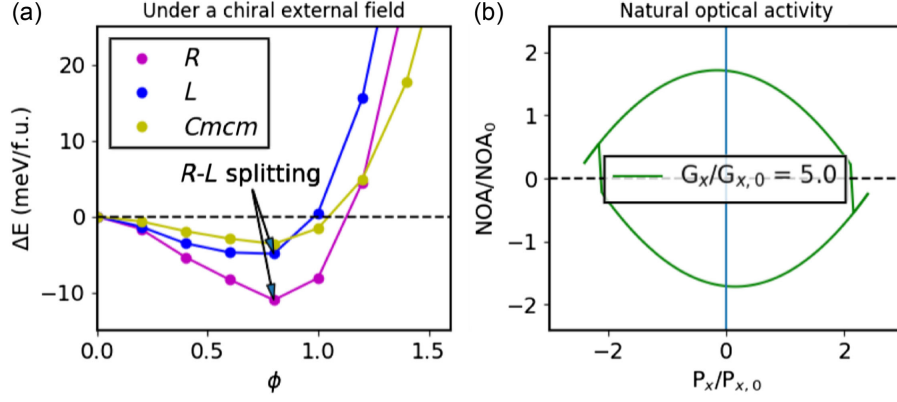


FIG. 4. (a) Density functional theory energy landscape as a function of the ϕ amplitude (arbitrary units) and in the presence of independent induced polar and toroidal moments along the x axis. $P_{x,0}$ and $G_{x,0}$ are the reference polar and electric-toroidal orders, and we fix $P_x/P_{x,0} = 1$ and $G_x/G_{x,0} = 5$, respectively. In this setting, the $|R\rangle \pm |L\rangle$ ($Cmcm$) degeneracy is unbroken. We observe the R - L splitting induced by $V_{\text{flip}} \equiv \lambda_x \phi^2 \cos 2\theta P_x G_x$, along with the onset of an $L \rightarrow R$ crossing via the $Cmcm$ transition state. (b) Hysteresis of the natural optical activity ($\phi^2 \cos 2\theta$ normalized by its zero-field value) as a function of $P_x/P_{x,0}$. The axial distortion is again fixed at $G_x/G_{x,0} = 5$.

with nonzero chirality [43], where $\nabla \times \mathcal{E}$ is the conjugate field of G_x .

Our DFT calculations are shown in Fig. 4(a), where $P_x/P_{x,0}$ and $G_x/G_{x,0}$ are chosen to be 1 and 5, respectively. Along with global shifts (V_{biquad}) that preserve the four-well structure of the free energy and clear R - L splitting, we notice that the L and $Cmcm$ phases are now saddle points in the energy landscape, suggesting that the aforementioned $L \rightarrow Cmcm \rightarrow R$ crossing below T_c is possible at rather modest values of $P_{x,0}$ and $G_{x,0}$. Clearly, the reciprocal $R \rightarrow L$ transition may be realized by changing the sign of $\mathbf{P} \cdot \mathbf{G} = P_x G_x$ (namely, the handedness of the chiral electromagnetic source). Thus, the scenario of Fig. 4(a), along with favoring the right-handed structure, corresponds to the application of an effective coercive field and to a still experimentally unobserved field-mediated hysteresis of the chiral order handedness below T_c , in the absence of intrinsic polar and electric-toroidal orders. An experimental signature of this hysteresis process would be given by quantities that probe the gyrotropic $\phi^2 \cos(2\theta)$ term of Eq. (2), namely, the natural optical activity (NOA) tensor in insulators [48] or the circular photogalvanic current in metals [18]. In Fig. 4(b), we show the hysteresis behavior of the normalized optical activity as a function of the polar and axial modes (the parameters of the model fitted from DFT are reported in Supplemental Material Sec. IV A [31]). Biquadratic fluctuations ($\phi^2 P_x^2, \phi^2 G_x^2$) constrain the magnitude of the fields and, if too large, can suppress the four-well structure of the energy surface [31]. Finally, we note that selecting the handedness of the system by applying a chiral external field from above T_c while quenching the temperature as in Ref. [18] is also compatible with this picture.

Dynamical effects—To better visualize the enantioselectivity process, we consider how the chirality of the system

evolves under a circularly polarized short EM pulse with a driving frequency ω in the THz range (see Supplemental Material [31], Sec. IV B). Treating the generated $\mathbf{P}(t)$ and $\mathbf{G}(t)$ orders as damped harmonic oscillators [11,15,17,31] (polar resonance at 3.26 THz) along the x and y directions, we find that the $\|\mathbf{P}(t)\|$ and $\|\mathbf{G}(t)\|$ amplitudes alongside the resulting $\mathbf{P}(t) \cdot \mathbf{G}(t)$ monopole only depend on time through an $\exp(-t/\tau)$ factor. Hence, the chiral-polar-axial interaction is almost equivalent to that of Fig. 4 in the $\tau \rightarrow \infty$ limit, with the difference that $V_{\text{ortho}} \propto e^{-t/\tau} \sin(2\omega t)$ is not zero. To gain additional insight, we solve the equations of motion of the system:

$$M\ddot{\phi} + \gamma\dot{\phi} + \nabla(F + V^{(4)}) = 0 \quad (4)$$

using DFT-obtained parameters. We show in Fig. 5 that starting from the zero-field R and L phases at $t = 0$, once $\mathbf{P} \cdot \mathbf{G} < 0$ is activated we can observe the stabilization of a net right-handed system ($R \rightarrow R, L \rightarrow R$). Analogously, inducing $\mathbf{P} \cdot \mathbf{G} > 0$ stabilizes an overall left-handed phase ($L \rightarrow L, R \rightarrow L$) [31]. Therefore, these conversion processes mediated by a circularly polarized EM field are consistent with the emergence of a net handedness, which may be controlled in a hysteresis fashion below T_c . Using the $\phi^2 \sin 2\theta$ quantity to measure the orthorhombic character of the distortion over time, we further confirm that the final equilibrium state is enantiomorphic [$\phi^2 \sin(2\theta) = 0$ with $\phi \neq 0$].

Application to other materials—While suggesting $K_3\text{NiO}_2$ or analogous $A_3\text{BO}_2$ oxides [36,49] for the experimental exploration of the handedness hysteresis process, other systems may be evaluated. As previously discussed (see Supplemental Material Sec. II [31]), a degenerate instability at the energy surface generating chirality below a critical temperature can be found in

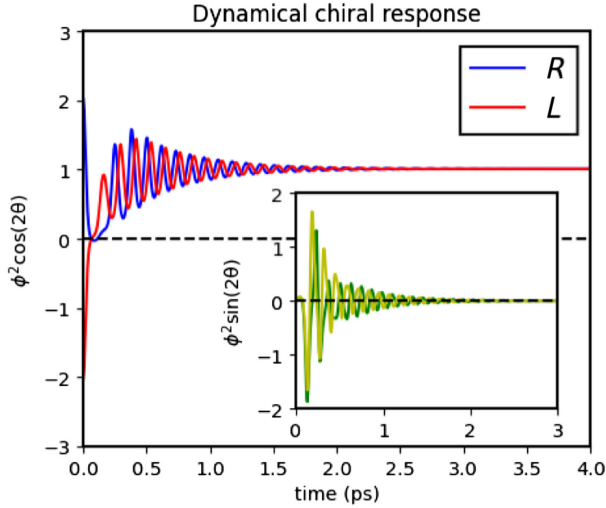


FIG. 5. Dynamics of the handedness conversion process. Starting from a system where the R and L phases are equally probable, an external chiral EM plane wave having frequency $\omega = 4.89$ THz and lifetime $\tau = 0.163$ ps triggers a $\mathbf{P} \cdot \mathbf{G} < 0$ monopole (the field is selected so that $\|\mathbf{P}\|/\|\mathbf{P}_0\| = 1$ and $\|\mathbf{G}\|/\|\mathbf{G}_0\| = 0.8$). This induces a stable $L \rightarrow R$ conversion over time, while the handedness of the R phase is instead preserved at the end of the evolution, hence resulting in a net stable right-handed system. Inset: damped oscillations of $\phi^2 \sin(2\theta)$, which project the order parameters on the orthorhombic states, starting from R (green) or L (yellow).

ABX_3 halide perovskites, AB_2X_4 spinels, and SiO_2 cristobalite. Additionally, several $A(B, B')_2X_4$ spinels may adopt cation-ordered structures of $P4_{1,3}22$ symmetry [50] (e.g., LiZnNbO_4 [51–53], B/B' ratio = 1:1), a process suitable to be influenced by Eq. (2) due to the chiral and multi-dimensional nature of the occupational order parameter. Consequently, the here-proposed strategy to use low-energy transition paths for $R \leftrightarrow L$ interconversion has a rather general character and could be extended to many materials.

Conclusions—We have reported how spontaneous right- and left-handed orders in crystals are connected by low-energy transition states when generated by one degenerate instability. Relying on this condition, we have developed a model to understand how a handedness may be selected by external fields and in the absence of intrinsic polar and axial moments. Importantly, we have shown that competing nonselective effects, for instance, triggered by a linearly polarized electric field, are present. Nevertheless, an external electromagnetic source generating a $\mathbf{P} \cdot \mathbf{G}$ monopole has a gyrotropic effect, and, depending on its sign, can stabilize a handedness of choice below the critical temperature. We argue that the hysteresis of chiral properties such as the natural optical activity is a signature of this phenomenon. Additionally, the interaction of Eq. (2) could be explored at the phonon level by replacing $\mathbf{P}$ and $\mathbf{G}$ with infrared and Raman active phonon modes [16]. The present

Letter highlights new perspectives on the chirality as a dynamical degree of freedom rather than a static symmetry property of materials.

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Data availability—The data that support the findings of this article are openly available [54].

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