I. COORDINATE SYSTEMS

Figure S1: 2D Brillouin zone in \( k_x - k_y \) plane \((k_z = 0)\) containing the \( \Gamma, M \) and \( K \) points. \( a \) and \( b \) are the lattice vectors, while \( a^* \) and \( b^* \) are the reciprocal lattice vectors. Here, \( x \) and \( y \) directions show the orthogonal reciprocal cartesian vectors \( k_x \) and \( k_y \), respectively.

The orientation of the lattice vectors, reciprocal lattice vectors, wave vectors and high symmetry path are shown in Fig. S1. For simplicity, we have added only the \( k_x - k_y \) plane containing \( k \)-points \( M, \Gamma \) and \( K \), which are used for computing spin textures and spin splitting. \( a \) and \( b \) are the lattice vectors, while \( a^* \) and \( b^* \) are the reciprocal lattice vectors. The angles between lattice vectors and reciprocal lattice vectors are 120° and 60°, respectively. Here, \( x \) and \( y \) directions show the orthogonal wave vectors \( k_x \) and \( k_y \), respectively. The \( \Gamma - K \) direction is symmetrically same as \( k_x \), which leads to band structures computed along \( \Gamma - K \) coincides with \( k_x \). Similarly, \( \Gamma - M \) direction is same as \( k_y \). We have used reciprocal cartesian basis i.e., \( k_x \) and \( k_y \) to compute spin textures and spin splitting. The lattice vector \( c \) is perpendicular to \( a \) and \( b \), pointing out of the page. The wave vector \( k_z \) is perpendicular to \( k_x \) and \( k_y \). The reciprocal lattice vector \( c^* \), \( k_z \) and \( \Gamma - \Lambda \) point along the same direction.

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II. HEXAGONAL DISTORTION UNDER STRAIN

Increasing the out-of-plane lattice parameter from 7.7 Å to 8.5 Å decreases the in-plane lattice parameters (i.e. \(a\) and \(b\)) and accordingly, the \(c/a\) ratio also increases. Figure S2 shows the variation of \(a\) and \(c/a\) as a function of \(c\).

![Figure S2: Calculated in-plane lattice parameter \((a)\) and the \(c/a\) ratio as a function of out-of-plane lattice parameter \((c)\).](image)

III. COMPARISON BETWEEN PBE+SOC AND HSE06+SOC FUNCTIONALS

We have computed the band structures of \(R3m\) phase of KIO\(_3\) using semi-local Perdew-Burke-Ernzerhof (PBE) and non-local Heyd-Scuseria-Ernzerhof (HSE06) exchange-correlation (\(\epsilon_{xc}\)) functional with spin-orbit coupling (SOC). Fig. S3a and S3b show the band structures calculated using PBE+SOC and HSE06+SOC, respectively. A slightly indirect band gap of 2.24 and 3.27 eV is observed using PBE+SOC and HSE06+SOC, respectively, at \(k\)-point A. Rashba spin splitting energy (\(\delta E\)), offset momentum (\(\delta k\)) along A-H and A-L directions, Rashba coefficient (\(\alpha_R\)) and Dresselhaus coefficient (\(\alpha_D\)) at conduction band minimum (CBM) are compared using both functionals in Table S1. Since Rashba parameters are comparable using both functionals, we have used PBE+SOC for further calculations being more cost effective. In addition, we have also compared the spin textures computed using PBE+SOC with HSE06+SOC (see Fig. S4). We have used uniformly space same dense \(k\)-grid for the comparison. The spin textures computed using both functionals are in close agreement, However, there is small inconsistencies near the center. This can be removed by taking more dense \(k\)-grid near the center.
Figure S3: Band structure for $R3m$ phase of KIO$_3$ calculated using (a) PBE+SOC and (b) HSE06+SOC. The Fermi energy is set to VBM.

Figure S4: (a)-(b) Spin textures of two lowest conduction bands and (c)-(d) two uppermost of valence bands around the $k$-point A obtained using PBE+SOC. Similarly, [(e)-(h)] are the counterparts of [(a)-(d)] obtained using HSE06+SOC.

### Table S1: Rashba parameters for conduction band-splitting at $k$-point A for $R3m$ phase.

| $\epsilon_{xc}$ functional | $\delta E$ (meV) | $\delta k$ (Å$^{-1}$) | $\alpha_R$ (eVÅ) |
|-----------------------------|------------------|----------------------|------------------|
| PBE+SOC                     | 27.3             | 0.047                | 1.16             |
| HSE06+SOC                   | 28.1             | 0.048                | 1.17             |
IV. TWO-BAND $k.p$ HAMILTONIAN

We aim at deriving the symmetry adapted two-band effective model Hamiltonian around $k$-point A. All those terms are included which are invariant under symmetry group operation, i.e., $O^†H(k)O = H(k)$. Here, $O$ represents the symmetry group operations belonging to the group of wave vectors and time-reversal operations. It is noteworthy that the considered $k.p$ Hamiltonian includes only spin degrees of freedom and does not take into account the orbital degrees of freedom. We have incorporated all the terms of the form $k_α^νσ_β$, where $k_α$ and $σ_β$ are the crystal momenta and Pauli spin matrices, respectively, along with the free particle Hamiltonian $H_o(k)$. Since the time-reversal operator transforms $k_α$ to $-k_α$ and $σ_β$ to $-σ_β$, the terms which are odd in momentum space, are only allowed to hold the time-reversal symmetry. Thus, the general expression of two-band $k.p$ model can be written as

$$H(k) = H_o(k) + \sum k_α^νσ_β$$  \hspace{1cm} (1)

where $α, β = x, y, z$ and $n$ takes only odd positive integers. In this model, for the specific case of $k$-point A, we have included up to cubic terms in crystal momentum, i.e., $k_ασ_z, k_ασ_y, k_ασ_z, k_ασ_yσ_z, k_ασ_y, k_α^2σ_z$. Point group symmetry at $k$-point A is $C_{3v}$, which can be generated by trivial identity operation (E), three fold rotation about z-axis ($C_{3z}=e^{-iπ/3σ_z}$) and mirror plane reflection in y-z plane ($M_y=iσ_z$) [1]. Transformation rules for $σ_β$ and $k_α$ under $C_{3v}$ point group and time-reversal operations are summarized in Table S2. Thus, the constructed Hamiltonian

TABLE S2: The transformations of $(σ_α, σ_γ, σ_δ)$ and $(k_x, k_y, k_z)$ with respect to the generators of the $C_{3v}$ point group and time-reversal operator ($T$). Note that the generators $C_3$ and $M_γ$ are enough to form the whole group of $C_{3v}$. Hence, only these generators with time-reversal $T=iσ_yK$ operation ($K$ is complex conjugation operator) are considered to construct the $k.p$ model for the $k$-point A.

| TABLE S2: The transformations of $(σ_α, σ_γ, σ_δ)$ and $(k_x, k_y, k_z)$ with respect to the generators of the $C_{3v}$ point group and time-reversal operator ($T$). Note that the generators $C_3$ and $M_γ$ are enough to form the whole group of $C_{3v}$. Hence, only these generators with time-reversal $T=iσ_yK$ operation ($K$ is complex conjugation operator) are considered to construct the $k.p$ model for the $k$-point A. |
|---|---|---|
| $C_3=e^{-iπ/3σ_z}$ | $M_y=iσ_z$ | $T=iσ_yK$ |
| $k_x$ | $-k_x/2+\sqrt{3}k_y/2$ | $-k_x$ |
| $k_y$ | $-\sqrt{3}k_x/2-k_y$ | $k_y$ |
| $k_z$ | $k_z$ | $-k_z$ |
| $σ_x$ | $-σ_x/2+\sqrt{3}σ_y/2$ | $σ_x$ |
| $σ_y$ | $-\sqrt{3}σ_x/2-σ_y/2$ | $-σ_y$ |
| $σ_z$ | $σ_z$ | $-σ_z$ |

can be written as

$$H_A(k) = H_o(k) + H_{SO}$$  \hspace{1cm} (2)

where,

$$H_{SO} = ασ_yk_x + βσ_zk_y + γσ_zf(k_x, k_y)$$  \hspace{1cm} (3)

and $H_o(k)$ is free particle Hamiltonian. Here, $α, β$ are the coefficients of linear terms and $γ$ is the coefficient of cubic term in SOC Hamiltonian. The linear Rashba and Dresselhaus Hamiltonian are given by $αR(σ_xk_y - σ_yk_x)$ and $αD(σ_xk_y + σ_yk_x)$, respectively [2]. Here, $αR$ and $αD$ are linear Rashba and Dresselhaus coefficients, respectively, which depend upon the properties of materials. The function $f(k_x, k_y)$, which has cubic dependence on crystal momentum, is given by

$$f(k_x, k_y) = (k_x^2 + k_y^2) - 3(k_xk_y^2 + k_yk_x^2)$$  \hspace{1cm} (4)

Writing the Hamiltonian in matrix representation

$$H_A(k) = \begin{pmatrix} E_0(k) - γf & βk_y - iαk_x \\ βk_y - iαk_x & E_0(k) - γf \end{pmatrix}$$  \hspace{1cm} (5)

where the $E_0(k) = \frac{ℏ^2k_x^2}{2m_x} = \frac{ℏ^2k_y^2}{2m_y}$ is the energy eigenvalue of free particle Hamiltonian. On diagonalizing the Hamiltonian, i.e., matrix in Eq. 5, gives
\[ E(\mathbf{k})^\pm = E_0(\mathbf{k}) \pm \sqrt{\alpha^2 k_x^2 + \beta^2 k_y^2 + \gamma^2 f^2(k_x, k_y)} \]

and the corresponding spinor eigenfunctions are given by

\[ \Psi^\pm_k = \frac{e^{i k x}}{\sqrt{2\pi(\rho^2_k + 1)}} \left( \frac{i \alpha k_x - \beta k_y}{\gamma f(k_x, k_y) + E_{SO}} \right) \]

where \( \rho^2_k = (\gamma f(k_x, k_y) + E_{SO})^2 \) is normalization constant, \( E_{SO} = | E(\mathbf{k}) - E_0(\mathbf{k}) | \) is the absolute energy eigenvalue of spin-orbit coupling Hamiltonian. Spin textures can be computed using the expectation values of spin operators.

Using the following expression

\[ \{s_x, s_y, s_z\}^\pm = \frac{1}{2} \{\langle \sigma_x \rangle^\pm, \langle \sigma_y \rangle^\pm, \langle \sigma_z \rangle^\pm\} = \pm \frac{1}{E_{SO}} \{\beta k_y, \alpha k_x, \gamma f(k_x, k_y)\} \]

where \( \{\langle \sigma_i \rangle\} = \{\Psi^\dagger_k | \sigma_i | \Psi^\pm_k\} \) are the expectation values of the spin operators. Using Eq. 4 and 8, we can say that the three-fold degeneracy of out of plane spin texture \( S_z \) is a consequence of cubic term \( f(k_x, k_y) \). The out of plane spin component is zero, when \( S_z = 0 \) or \( f(k_x, k_y) = 0 \). The lines \( L_1: k_y = -k_x, L_2: k_y = 2x - \sqrt{3}x \) and \( L_3: k_y = 2x + \sqrt{3}x \) in the momentum space are the directions of out of plane spin component. Slope of the lines \( L_1, L_2 \) and \( L_3 \) are -1, 2-\( \sqrt{3} \) and 2+\( \sqrt{3} \), respectively. Angle between two lines of slopes \( m_1 \) and \( m_2 \) can be computed using \( \theta = \tan^{-1}\left(\frac{m_2 - m_1}{1 + m_1 m_2}\right) \). It is straightforward to see that smaller angle between any two lines is 60°, confirming the existence of three-fold degeneracy.

The linear Rashba (LR) Hamiltonian is given by \( \alpha_R(\sigma_z k_y - \sigma_y k_x) \). The energy dispersion curves of LR splitted bands are given by \( E_{\pm} = \hbar^2 k_x^2/2m \pm \alpha_R k_x \). This energy dispersion relation leads to shift in degenerate bands from high symmetry point by the offset momentum \( \delta k \). For \( k \)-points \( \Gamma \) and \( \Lambda \), \( k_x = k_y = 0 \) and \( k_x \) is constant within the \( k_x - k_y \) plane. Therefore, \( \hbar^2 k_x^2/2m + \alpha_R k_x \) is taken to be constant and neglected in further calculations. Suppose we are calculating \( \alpha_R \) in \( k_x \) direction and \( \delta k \) is offset momentum along \( k_x \) direction, then \( \alpha_R \) can be estimated as follows

\[ E_{\pm} = \hbar^2 k_x^2/2m \pm \alpha_R k_x \] (9)

\( E_{\pm} \) have minimum energy \( E_{\text{min}} \) at offset momentum \( \mp \delta k \). Rashba spin splitting energy \( (\delta E) \) is \( E_{\text{min}} - E_{\text{LR}} \). Thus, \( \alpha_R \) can be estimated by approximating \( \delta E \) and \( \delta k \) using DFT and given by

\[ \alpha_R = \frac{2\delta E}{\delta k} \] (10)

Eq. 10 is used when there is pure LR effect.

Our considered SOC Hamiltonian \( (H_{SO} = \alpha \sigma_y k_x + \beta \sigma_x k_y + \gamma \sigma_z (k_x^3 + k_y^3) - 3(k_x k_y^2 + k_y k_x^2)) \) differs from LR Hamiltonian. Thus, possibility of splitting coming from linear Dresselhaus (LD) and cubic terms (cubic in \( k \)) are also taken in account. Energy eigenvalues corresponding to \( H_{SO} \) are given by \( E_{SO}(k) = \sqrt{\alpha^2 k_x^2 + \beta^2 k_y^2 + \gamma^2 f^2(k_x, k_y)} \), where \( f(k_x, k_y) = (k_x^3 + k_y^3) - 3(k_x k_y^2 + k_y k_x^2) \). Initially, we have fitted the band structures along \( k_x \) direction using the relation \( E_{SO}(k) = \sqrt{\alpha^2 k_x^2 + \gamma^2 f^2(k_x, k_y)} \). Since our considered range for DFT band structure is \( |k_x| \leq 0.125 \AA^{-1} \), \( |\alpha k_x| \gg |\gamma k_x^2| \). Thus, we have estimated the values of \( \alpha \) and \( \beta \) using the parametric fitting of DFT band structures along \( k_x \) and \( k_y \) directions, respectively. \( \gamma \) is estimated using the \( z \)-component of spin texture since it comes from cubic terms of \( H_{SO} \). Moreover, overall band fitting including the cubic terms is also compared with only linear terms and are in well agreement. The \( H_{SO} \) can be written as linear combinations of LR and LD, which is given by the following expression

\[ H_{SO}(k) = H_R + H_D = \alpha \sigma_y k_x + \beta \sigma_x k_y = (\alpha_R + \alpha_D) \sigma_y k_x + (\alpha_D - \alpha_R) \sigma_x k_y \] (11)

Using the \( \alpha \) and \( \beta \), we can estimate the values of \( \alpha_R \) and \( \alpha_D \). Since \( \alpha_D \approx 0 \), the splitting mainly comes from the LR effect. Therefore, the \( \alpha_R \) calculated using the standard DFT \( (\frac{\alpha_R}{\alpha_D}) \) and band fitting \( (\frac{\alpha_R}{\alpha_D}) \) are found to be consistent with each other.
V. FOUR-BAND \( k.p \) HAMILTONIAN

The four-band \( k.p \) Hamiltonian including spin and orbital degrees of freedom is derived to understand the splitting around \( k \)-point \( A \) in \( R3c \) phase. The representation of point group appropriate to spin-1/2 particles is usually determined by a group of double the order of the point group under consideration, known as the double group. The point group of wave vector associated with the \( k \)-point \( A \) is \( C_{3v} \). The character table of double group corresponding to \( C_{3v} \) point group is given by Table S3. The number of conjugacy classes and irreducible representations (IR) (denoted by \( \Gamma_i \)) are 6. The lowest conduction band is two-fold degenerate at \( k \)-point \( A \) (see Fig. 4 upper panel in main manuscript) and corresponds to IR \( \Gamma_4 \) (representing the orbital degree of freedom). The inclusion of SOC leads to the splitted four bands around \( k \)-point \( A \) and degenerate at \( k \)-point \( A \) (see Fig. 4 lower panel in main manuscript). On including SOC, lowest conduction bands at \( k \)-point \( A \) correspond to direct product of \( \Gamma_3 \) and \( \Gamma_4 \). IR \( \Gamma_4 \) represents the spin degrees of freedom. Using this, four-band \( k.p \) model is constructed employing the basis (\( \{|x\uparrow\rangle, |x\downarrow\rangle, |y\uparrow\rangle, |y\downarrow\rangle\})\). The transformation rules of (\( \sigma_x, \sigma_y, \sigma_z \), \( \gamma_x, \gamma_y, \gamma_z \)) and (\( k_x, k_y, k_z \)) are given in Table S4 [3]. \( \sigma_i \) and \( \gamma_j \) are both Pauli matrices signifying the spin and orbital degrees of freedom, respectively. We have used the Hamiltonian, which is invariant under the operations \( C_{3v}, M_x \) and \( T \) and is given by

\[
H_A(k) = \alpha(k_x^2 + k_y^2) + \beta(k_x^2 + k_y^2)^2 + \delta(k_x^2 + k_y^2)^3 + \eta((k_x^2 - k_y^2)\gamma_z + 2k_xk_y\gamma_x) + [\Delta + \Lambda(k_x^2 + k_y^2) + K(k_x^2 + k_y^2)^2]\gamma_y\sigma_z + [\kappa + \zeta(k_x^2 + k_y^2)(k_x\gamma_x - k_y\gamma_y) + \lambda k_x(k_x^2 - 3k_y^2)\sigma_z + \xi k_x(k_x^2 - 3k_y^2)\gamma_z + \rho[(k_x\gamma_x - k_y\gamma_y)\sigma_x - ((k_y\gamma_x - k_x\gamma_y)\sigma_y)]
\]

(12)

Here, \( \alpha, \beta, \delta, \eta, \Delta, \Lambda, \kappa, \zeta, \Lambda, \xi \) and \( \rho \) are coefficients signifying the strength of splitting. The terms having coefficients \( \alpha, \beta \) and \( \delta \) are the effective mass terms. The term with coefficients \( \eta \) and \( \xi \) includes only orbital degrees of freedom. Spin degrees of freedom are taken into account with the terms containing \( \kappa, \zeta \) and \( \lambda \). The remaining terms with coefficients \( \Delta, \Lambda, K \) and \( \rho \) couple the orbital and spin degrees of freedom. The coupling terms are responsible for the novel spin textures observed around \( k \)-point \( A \) in \( R3c \) phase (see Fig. 4i-l in main manuscript). Alongside of full four-band \( k.p \) Hamiltonian, we have constructed a modified Hamiltonian to understand the importance of orbital degrees of freedom in four-band \( k.p \) Hamiltonian. The modified Hamiltonian excludes the terms containing the Pauli matrices, which signify the orbital degrees of freedom from four-band \( k.p \) Hamiltonian (\( \eta = \xi = \Delta = \Lambda = K = \rho = 0 \)). Fig. 5d in main manuscript shows lowest conduction bands around \( k \)-point \( A \) using DFT and modified Hamiltonian. The band structure (Fig. 5d in main manuscript) obtained using modified Hamiltonian deviates from the DFT band structure. This signifies the importance of the full four-band \( k.p \) Hamiltonian. The Hamiltonian without the terms containing orbital degrees of freedom cannot reproduce the band dispersion and spin textures observed around \( k \)-point \( A \).

| \( \Gamma \) | \( E \) | \( \bar{E} \) | \( 2C_3 \) | \( 2\bar{C}C_3 \) | \( 3\sigma_v \) | \( 3\bar{E} \) | \( \sigma_v \) | Basis functions |
|---|---|---|---|---|---|---|---|---|
| \( \Gamma_1 \) | 1 | 1 | 1 | 1 | 1 | 1 | \( \bar{z} \) | |
| \( \Gamma_2 \) | 1 | 1 | 1 | 1 | -1 | -1 | \( S_z \) | |
| \( \Gamma_3 \) | 2 | 2 | -1 | -1 | 0 | 0 | \( (x, y), (S_x, S_y) \) | |
| \( \Gamma_4 \) | 2 | -2 | 1 | -1 | 0 | 0 | \( (\uparrow, \downarrow) \) | |
| \( \Gamma_5 \) | 1 | -1 | -1 | 1 | -1 | -1 | --- | |
| \( \Gamma_6 \) | 1 | -1 | -1 | 1 | -1 | -1 | --- | |
TABLE S4: The transformations of \((\sigma_x, \sigma_y, \sigma_z), (\gamma_x, \gamma_y, \gamma_z)\) and \((k_x, k_y, k_z)\) with respect to generators of the \(C_{3v}\) point group and time-reversal operator \((T)\). Note that the generators \(C_{3z}\) and \(M_x\) are enough to form the whole group of \(C_{3v}\). Hence, only these generators with time-reversal operation \(T=i\sigma_yK\) \((K\) is complex conjugation operator) are considered to construct the \(k.p\) model for \(k\)-point \(A\).

| \((T=)\) | \((C_{3z}=)\) | \((M_x=)\) |
|---|---|---|
| \(+k_x/2+\sqrt{3}k_y/2\) | \(e^{-i\pi/3}\sigma_z\) | \(-k_x\) |
| \(-k_x\) | \(-k_x\) | \(k_y\) |
| \(k_x\) | \(k_x\) | \(-k_x\) |
| \(-\sigma_x/2+\sqrt{3}\sigma_y/2\) | \(\sigma_x\) | \(+\sigma_x\) |
| \(-\sigma_x\) | \(-\sigma_x\) | \(+\sigma_x\) |
| \(-\gamma_x/2+\sqrt{3}\gamma_y/2\) | \(-\gamma_x\) | \(+\gamma_x\) |
| \(-\gamma_y\) | \(+\gamma_y\) | \(+\gamma_y\) |
| \(-\sqrt{3}\gamma_x/2+\gamma_z/2\) | \(\gamma_x\) | \(+\gamma_x\) |

VI. RASHBA PARAMETERS FOR SELECTED BULK FERROELECTRIC MATERIALS

In the Table S5, we have compared the Rashba coefficients \((\alpha_R)\) of some well known bulk ferroelectric materials with the bulk KIO\(_3\). Space group symmetry is also included with the material. For hafnia \((\text{HfO}_2)\), \(\delta E\) and \(\delta k\) are not provided in the literature. All the values are reported upto respective significant Figures in the references and may not be consistent with each other.

TABLE S5: Rashba spin splitting energy \((\delta E)\), offset momentum \((\delta k)\) and Rashba coefficient \((\alpha_R)\) of some selected bulk ferroelectric materials.

| Material | Space group | \(\delta E\) (meV) | \(\delta k\) \((\AA^{-1})\) | \(\alpha_R\) (eV\(\AA\)) | Reference |
|---|---|---|---|---|---|
| KIO\(_3\) | \(R3m\) | 23.2 | 0.053 | 1.16 | This work |
| KIO\(_3\) | \(R3c\) | 14.8 | 0.047 | 0.78 | This work |
| BiAlO\(_3\) | \(R3c\) | 7.34 | 0.04 | 0.39 | [4] |
| BiAlO\(_3\) (along Z-R) | \(P4mm\) | 9.40 | 0.03 | 0.74 | [4] |
| BiAlO\(_3\) (along A-Z) | \(P4mm\) | 8.62 | 0.03 | 0.65 | [4] |
| LaWN\(_3\) | \(Pna2_1\) | 2.20 | 0.014 | 0.31 | [5] |
| LaWN\(_3\) | \(R3c\) | 3.49 | 0.051 | 0.18 | [5] |
| BiInO\(_3\) | \(Pna2_1\) | 130 | 0.19 | 1.91 | [3] |
| PbTiO\(_3\) | \(P4mm\) | 5.45 | 0.50 | 0.51 | [6] |
| HfO\(_2\) | \(Pca2_1\) | - | - | 0.06 | [7] |
| KMgSb | \(P6_3mc\) | 10 | 0.024 | 0.83 | [8] |
| LiZnSb | \(P6_3mc\) | 21 | 0.023 | 1.82 | [8] |
| \(\beta\)-(MA)PbI\(_3\) | \(P4mm\) | 12 | 0.015 | 1.5 | [9] |
| \(\beta\)-(MA)SnI\(_3\) | \(P4mm\) | 11 | 0.011 | 1.9 | [9] |
| GeTe | \(R3m\) | 227 | 0.09 | 4.8 | [10] |

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