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Uncovering and tailoring hidden Rashba spin–orbit splitting in centrosymmetric crystals

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Numerous physical effects and the technologies enabled by the conditional on the presence of conditional on the presence of certain symmetric order of certain symmetric \mathbf{a} $\frac{1}{2}$ in the material that $\frac{1}{2}$ is the material that $\frac{1}{2}$ is the material that $\frac{1}{2}$ is the material term of $\frac{1}{2}$ include effects predicated on the absence of inversion symmetry a inversion symmetry a $($ - centrosystems) such as the Dressell as $\frac{1}{2}$, the Rashba effect $\frac{1}{2}$ $\frac{1}{2}$ $\frac{1}{2}$, the Rashba effect in non-chiral molecules in $\frac{3}{2}$, $\frac{6a}{2}$ a a_a and its $6a_b$ $\begin{pmatrix} 4 & a \\ b & a \end{pmatrix}$ $\begin{pmatrix} 4 & a \\ b & a \end{pmatrix}$ $\begin{pmatrix} 4 & a \\ b & a \end{pmatrix}$ and valley a_a \mathbf{a} (2D) a \mathbf{a} \mathbf{s} and \mathbf{s} \mathbf{s} \mathbf{s} $\frac{1}{2}$ are supposed to lack the system are supposed to lack the suppose effects of $\frac{1}{2}$ and $\frac{1}{2}$ class of systems whose global crystal systems whose global crystal symmetry a is (C) centrosymmetric but they consist of individual sectors with $\frac{1}{2}$ centrosymmetric local symmetric local symmetric symmetric symmetric symmetric symmetric symmetry α metric site point groups). The term $\frac{m}{\alpha}$ refers to the term $\frac{m}{\alpha}$ compound compound is often confused compound, which is often confused confused by $\frac{20}{3}$, investigate the evolution of the R-1 spin splitting from a R-2 spin $\sum_{\mathbf{a}}$ (" -1 $\sum_{\mathbf{a}}$ -2") **by** using the first-principles calculated tions on R-2 compounds and placing on it a tiny electric field \mathfrak{g} as \mathfrak{a} inversion symmetry. A a -2 compounds is 2×10 , which is a fig. coordinated Ni(II) structure consisting \mathbf{a} is putting 2D at a structure consisting \mathbf{a} edge-sharing square pyramidal polyhedral and crystalizes in the tetragonal system, space group P4/nmm. Conductivity and **s** indicate $\frac{1}{22,23}$ $\frac{1}{22,23}$ $\frac{1}{22,23}$ $\frac{1}{22,23}$ $\frac{1}{22,23}$ indicate the indicate that it is a metallic that it is a metal **a** a a a **b** + a a a \bullet \bullet J = 0.95 α) also predicts a low-temperature and predicts a low-temperature and phase an \bullet a \bullet $0.7 \mu_{\text{B}}$ \bullet ($0.6 \mu_{\text{B}}$ a monolayer and anti-ferromagnetic phase is slightly more in the anti-ferromagnetic phase is slightly more in the a \mathbf{a}^2 \mathbf{a}^2 as \mathbf{a}^3 meV(f)⁻¹ **for a** and 28 $(\frac{1}{2})^{-1}$ for and **D** + a_1 a a_2 undergoes a phase a_3 undergoes a phase a_1 undergoes a phase a_2 undergoes a phase a_3 $\begin{array}{lllllllllll} \text{a} & \text{a$ increasing the used U-value from 2 to 3 eV. Given the difficulty **a** a_a **b** a_b **b** a_c **b** a_c and e^{2a} (e^{2a} e^{2a} e^{2a} e^{2a} \mathbf{a} and \mathbf{a} and \mathbf{a} in the neutralic vector \mathbf{a} a $\bullet a$ - a \bullet avoid Ba₂ \bullet a**void the unit band is a** $a_{\mathbf{a}}$ and $a_{\mathbf{a}}$ relaxed lattice con-relaxed lattice con-relaxed lattice con-relaxed lattice con- \mathbf{a} and interaction in the non-magnetic distances in the non-magnetic \mathbf{a} $a \rightarrow A$ as (A) agrees a since measured results and 1% ^{10,22}. In the non-magnetic model, $\frac{1}{2}$ Ba₂ **i** 6 **i** 3 **i** 6 **i** 8 **i** 6 **a**

sechial S β (here β -reformed S β -Rashba band). We therefore β -Rashba band iidentify the splitting of the splitting $\delta E_{\rm AB}({\bf k})$ as a consequence of the R-2 effect of **a** a finite a Rashba a Rashba a α (2) = 0.24 values and a electric fields function function function \int the R-2 splitting δ AB(k) of the Sa- and S β -Rashba and S \bullet $\stackrel{6}{\sim}$ \bullet $\stackrel{7}{\sim}$ $\stackrel{8}{\sim}$ $\stackrel{8}{\sim}$ $\stackrel{3a}{\sim}$ $\stackrel{3a}{\sim}$ $\stackrel{3a}{\sim}$ $\stackrel{3a}{\sim}$ $\stackrel{4}{\sim}$ a a a a $\frac{\alpha}{\epsilon} = \delta E_{AB}(\mathbf{k} - \frac{\epsilon}{2})(\mathbf{k} - \frac{\epsilon}{2})$ **a** a linear **a** \mathbf{E} \mathbf{F} a \mathbf{a} a a discrete to \mathbf{a} a discrete to \mathbf{a} increases and the S β -Rashba band decreases at rates of the same of the sam $\begin{pmatrix} 1 & B \end{pmatrix} \qquad D\left(\begin{pmatrix} \frac{1}{\alpha/\beta}(A,k^-{}_{-\bar\Gamma}) \end{pmatrix} = D\left(\begin{pmatrix} \frac{1}{\alpha/\beta}(B,k^-{}_{-\bar\Gamma}) \end{pmatrix} = 0\right)$ 2% - spin-down components of \bullet and \bullet A and B and \bullet . α , the α the spin-up component of the sp a A 43% find and a and b 57% find in sector \mathbf{D} such \mathbf{D} satisfy \mathbf{D} satisfy \mathbf{D} satisfy \mathbf{D} satisfy \mathbf{D} $D\left(\begin{array}{cc} \uparrow \\ \alpha/\beta\end{array}(A,k_{^- - \bar{\Gamma}})\right) = D\left(\begin{array}{cc} \downarrow \end{array}\right)$ $\frac{1}{\alpha/\beta}$ $(B, k^-$ _{$-\bar{\Gamma})$} 14% for spin-up \cdots spin-up components. Thus, the wavefunctions of the w $\overline{I} - \overline{I}$ a a sa a $\overline{6}$. $\overline{6}$ a_3 and α and β . Such a_4 a_1 a complete comple local internal dipole fields within S^α by that within Sβ, when each local dipole weight and by its wavefunction and \mathfrak{g} is a major specified by its analysis of \mathfrak{g} a_0 a a according to according to a_1 according to a_2 according to a_3 .

Uni 'cation of R-1 and R-2 into a single theoretical framework. $\frac{1}{2}$ from R-1 from R-2" evolution (Fig. 3a) subsets that $\frac{1}{2}$ and $\frac{1}{2}$ a as a field $\frac{1}{2}$ field $\frac{1}{2}$ and $\frac{1}{2}$ electric field $a_{\overline{r}}$ \therefore internal dipole internal local dipole i $E(r) = E(r) + E$ (4)

Thus, both R-1 and R-2 spin splitting have a common fundamental source being the dipole electric fields of the local source \mathbf{f}_1 sectors rather than from the global crystal asymmetry per se. Such local dipole electric field \int_a^b field \int_a^b \int_a^b \int_a^b individual local sectors. The fundamental difference between R-1 and R-2 effects is that in R-2 the spin splitting is hidden by the overlapping energy bands arrising from the $\frac{1}{2}$ supersion-partners in the R-1 sectors, whereas in the R-1 sectors, whereas in the R-1 a is forbidden by the global inversion asymmetry. $\frac{1}{2}$ $\frac{1}{2}$ $\frac{1}{2}$ also shows the applied electric field is a shown that the application of $\frac{1}{2}$ and $\frac{1}{2}$ an $\begin{array}{ccccc}\n\mathbf{a} & \mathbf{a} & \mathbf{a} & -\bar{\mathbf{F}} & \mathbf{a} & \mathbf{a} & \mathbf{a}\n\end{array}$ linearly from zero at Eext = 0 to saturation at |Eext = 10 mV Å−¹ a_a and a_a . The a_b is in striking contrast to the striking contrast of the striking contras

linear field dependence of the band \overline{M} and \overline{M} (see Fig. 3a). Such unusual field dependence of α confirms again that the R-2 spin splitting evolves smoothly to the R-1 spin splitting upon the breaking of the global inversion symmetry, a a a $-\bar{\Gamma}$ a above 2 a α (2) = 0 in the absence of an external field. a_a and f_b , f_b and a $\frac{1}{\epsilon}$ $-\bar{\Gamma}$ a band and $\frac{1}{\epsilon}$ become gradually segments on $\frac{1}{\epsilon}$ the inversion of α is a result of α results as a result of 2^{25} . $\overline{3}$ subsequently field f sas as **b** $\left(\cdot , (1) \right)$ $\left(\cdot , (1) \right)$ $\left(\cdot , (1) \right)$ of a 14% to $> 80\%$ as the magnitude of E_st increases in E the magnitude of E in E 0 **s** 50 $^{-1}$. 6 , D(φ_k) a a 0 **s** 50 $^{-1}$. **6** $D(\varphi_k)$ a a
E \rightarrow 50 $^{-1}$ (a**j** a_i **h** $^$ ₁). **s s s a**_i a_i \mathbf{D} of the corresponding spin-down components is not shown components in the components is not shown components in the compone \bullet a similar response to the application of the application of \bullet application straightforward to learn that the internal electric dipole fields as sated by as the bands become uncompeted as the a tions a single into single into segregation on a single sector, evoking the R-2 effect with its strength highly related to D(φk) according to \bullet [\(3\)](#page-4-0). a a fia $D(\varphi_k)$ by the application of a fiall $a_1 a_2$ for the α for α for α for α $a \overline{C} - \overline{\Gamma}$ and $a \overline{B}$ and $a \overline{B}$ $a_1 a_2 a_3 a_4 a_5 a_6 a_7 a_8 a_8 a_9 a_1 a_1 a_2 a_3 a_4 a_5 a_7 a_7 a_8 a_1 a_1 a_2 a_3 a_4 a_4 a_5 a_7 a_7 a_8 a_1 a_1 a_2 a_3 a_4 a_4 a_5 a_7 a_7 a_8 a_1 a_1 a_2 a_3 a_4 a_4 a_5 a_7 a_1 a_2 a_3 a_4 a_4 a_5 a_1 a_2 a$ $\frac{1}{2}$ fi.

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 E , a $\frac{25}{1}$, α s = a − energy doublets become linear field-dependent but in rates a_1 \overline{M} is in a signal dependence as the response of $\overline{3}$ signs, $\overline{1}$ as the response of D(φ k) $\overline{\bullet}$ $\overline{\bullet}$ a $\overline{\bullet}$ E_r, 6, a fi $\frac{1}{4}$ a fin, $\frac{1}{4}$ indicating the state states remain function $\frac{1}{4}$ is $\frac{1}{4}$ indicates on $\frac{1}{4}$ of two inversion-partner sectors. The linear change of ^α along M direction as shown in Fig. 3a thus arises entirely from the \therefore a finite induced as \therefore i.e., i.e., \therefore i.e., i.e $\begin{array}{ccc} \text{a} & \text{b} & \text{c} & \text{c} & \text{d} \\ \text{c} & \text{d} & \text{e} & \text{e} & \text{f} & \text{c} & \text{d} \\ \text{d} & \text{e} & \text{f} & \text{f} & \text{e} & \text{d} \end{array}$ parameter of the R-2 spin splitting can be explained regarding the model of the R-1 spin splitting (Eq. ([3](#page-4-0))), indicating a unified theoretical view for \mathbf{a} and \mathbf{a} and \mathbf{a} and \mathbf{a} and \mathbf{a} \int fia $\frac{1}{2}$ \int \int \int \int fi \int a a a a a a and -2 a a superposition of the application of t external electric field plus the internal local electric fields originating from the dipoles of the individual local sectors, $\frac{1}{2}$ and $\frac{1}{2}$ a sectors.

 a apply this uniform theoretical framework to a non-tensor a non-te a -1 a, $\frac{1}{2}$ a-GeTe a $\frac{1}{2}$ a $\$ \mathbf{a}^2 a compound predicted in $\mathbf{a} = \mathbf{a} - 1$ compound predicted in 2013^{[26](#page-6-0)} a and a firm $2016^{27,28}$, and can be cannot confirm to cannot confirm to cannot can be cannot can be cannot can be cannot confirm to a index two inversions and the corresponding α inversion-partner sectors and the corresponding α $a_1 a_2 a_3 a_4 a_5 a_6$ symmetry in the rhombohedral phase (details see Supplementary \bullet 1). According to the unit of the unit of the unit of \bullet ([3](#page-4-0)), \overline{a} a \overline{b} a \overline{a} \overline{b} a \overline{a} a residual field \overline{a} band states and thus give rise to a finite Rashba spin splitting, a $a_1 a_2$ -2 spin Ba₂. A as a_1 α_{\bullet} a 111] α_{\bullet} a a phase transition from non-centrosymmetric rhombohedral phase to centrosymmetric rocksalt phase. We demonstrate that in the contros are even a a_1 and a a_2 are even as a_3 and a_4 are even as a_1 and a_2 are even as a_1 \bullet \bullet inversion-partner and partners in a perfect of a perfec compensation of the local dipole fields and the local dipole fields and thus vanishing Rashbaum and thus vanish
Thus vanishing Rashbaum and thus vanishing Rashbaum and thus vanishing Rashbaum and thus vanishing Rashbaum an \ddot{i} in the centrosymmetric rocks and \ddot{j} . ([3](#page-4-0)).

Design principles for increasing the strength of the R-2 effect. $\overline{P_1^2}$ a defined by having global inversion symmetry and $\overline{P_1^2}$ $\frac{103.84.10}{2}$ and two recognizations are inversion-partners with a inversion-partners with a in a point group symmetries. Designing -2 as a contract and a hidden spin splitting and hence strong local spin polarization can $\hat{h}_{\hat{a}}$ additional design principles:

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