Supplementary Materials: Interplay between Relativistic Spin-Momentum Locking and Breaking of Inversion Symmetry: conditions for effective p-wave magnetism

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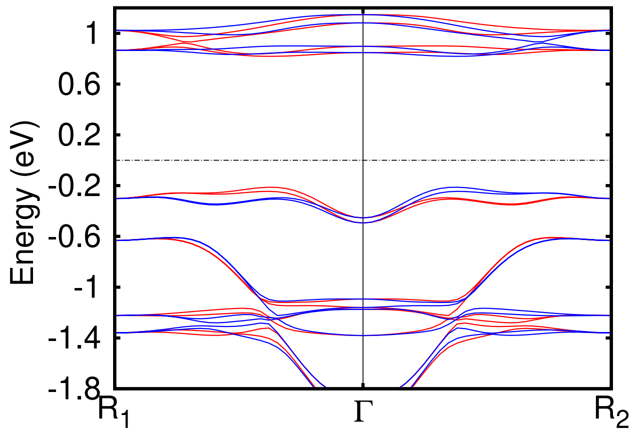

FIG. S1. Band structure of Ca_2RuO_4 in the B-centered magnetic phase without SOC along the path R_1-Γ-R_2. Blue and red represent the spin-up and spin-down channels, respectively. The Fermi level is set to zero energy. The band gap is 0.85 eV.

S1. COMPUTATIONAL DETAILS

All calculations were conducted without accounting for relativistic effects, using the Vienna Ab Initio Simulation Package (VASP)[1–3]. We have symmetrized our results and projected the charge on the atomic orbitals with version 6 of VASP. To include the correlations within the Ru-4d states, a Coulomb repulsion term was considered in the simulations. For the exchange correlation potential, we employed the generalized gradient approximation (GGA)[4]. Ca_2RuO_4 can have two orthorhombic phases named as long (L-Pbca) and short (S-Pbca) phases; all the density functional calculations reported in this paper were performed for the S-Pbca phase, characterized by space group number 61. A plane-wave energy cutoff of 480 eV was applied, ensuring total energy convergence to less than 1×10^{-5} eV. The computations were performed using an $11 \times 11 \times 4$ k-point grid, based on the experimental lattice constants $a = 5.3945$ Å, $b = 5.5999$ Å, and $c = 11.7653$ Å[5]. A Coulomb repulsion $U = 3$ eV was applied to the Ru-4d orbitals, along with a Hund's coupling $J_H = 0.15U$. Within this framework, the magnetic moment per Ru atom was calculated to be 1.4 μ_B, consistent with the typical magnetic configuration of Ca_2RuO_4.

The computational framework is the same one used in the literature for the Ca_2RuO_4[6? ,7] with an increased set of k-points. The electric field was simulated via the shift along the x-, y- or z-axis of the Ru atoms; such an approach was defined as the so-called "lattice-mediated" method, in which a polar displacement of the ions simulates the application of an electric field[8]. The displacements were set to 1% of the relative lattice constants along all directions.

S2. NON-RELATIVISTIC SPIN-SPLITTING OF THE B-CENTERED Ca_2RuO_4

We report the band structure of the B-centered magnetic phase for the centrosymmetric case and in the presence of a stripe. In the centrosymmetric case, the high-symmetry points of the BZ are the same as in the A-centered case, reported in the main text. Unlike the A-centered configuration, in the B-centered mode, the d_{xy} orbitals show non-relativistic spin-splitting, and the band structure is reported in Fig. S1. However, there is a different spin-momentum locking with respect to the A-centered phase as discussed in the main text.

In Fig. S2, we report how the band structure in the B-centered configuration changes when we consider a modulated electric field similar to the stripe with Ru shifted along the (a) x, (b) y, (c) z-axis. We can observe that, associated with the rise of the altermagnetism in the bands of d_{xy} orbitals, there is a non-relativistic spin-splitting in the bands of the γz orbitals that is much larger with respect to the A-centered magnetism. As in the A-centered case, the band gap gets reduced when Ru atoms are under ferroelectric distortion, due to an increase of the splitting in the bands of the d_{xy} orbitals.

S3. TIGHT BINDING MODEL WITH t_{2g} ORBITALS FOR THE A-CENTERED

To unveil the origin of the altermagnetism, we develop a tight-binding model without spin-orbit coupling composed of a 6×6 Hamiltonian for the spin-up and 6×6 Hamiltonian for the spin-down for the A-centered magnetic phase of

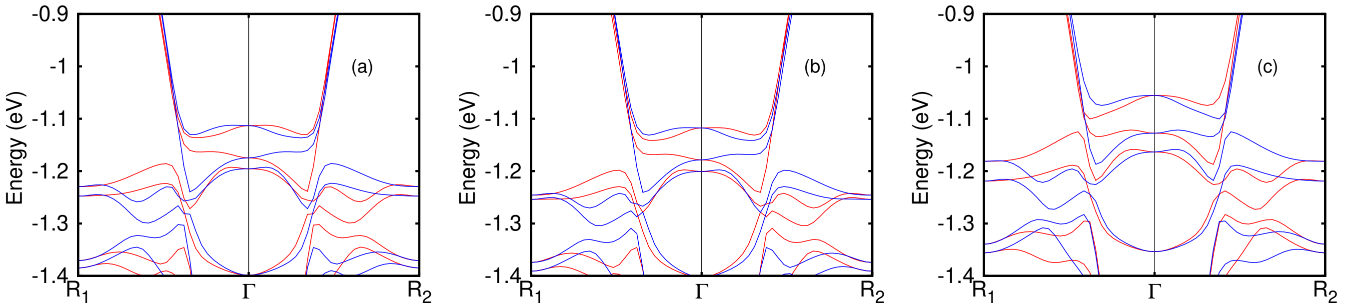

FIG. S2. Zoom of the band structure of Ca_2RuO_4 along the high-symmetry positions R_1-Γ-R_2 in the antiferromagnetic B-centered configuration and in the presence of the stripe for different layers with Ru shifted along: (a) the x-axis, (b) the y-axis and (c) the z-axis. The band gaps for these cases are 0.82 eV, 0.83 eV, and 0.87 eV, respectively.

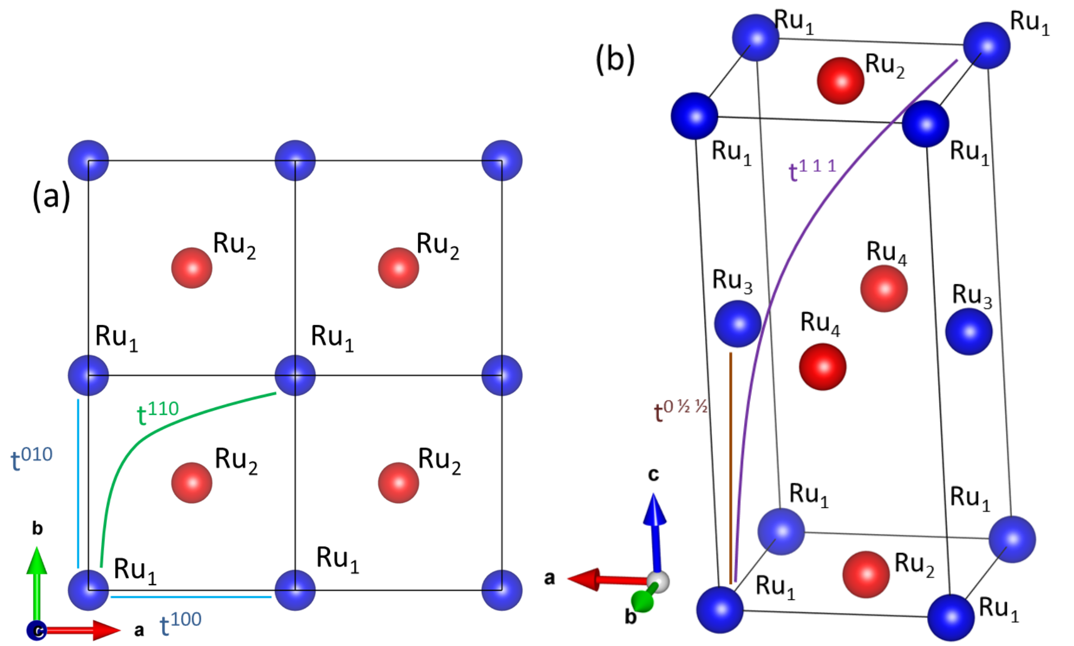

FIG. S3. Schematic representation of the position of Ru_1, Ru_2, Ru_3 and Ru_4 atoms. (a) Top view with a 2×2 repetition of the unit cell in the *ab* plane, with the in-plane hoppings represented, (b) primitive cell with the hoppings between the planes considered in the model. In the Figure, the hoppings in the ↑-subsector are shown; we have considered the same terms for the ↓-subsector. Ru atoms are blue (spin ↑ oriented) and red (spin ↓ oriented), while the oxygen and calcium atoms are not shown.

Ca_2RuO_4. The magnetism is given by the on-site term $+\Delta_z$ for the minority spin-channels and $-\Delta_z$ for the majority spin-channels. We performed the wannierization for the subsectors ↑ and ↓. In the unit cell we have Ru_1=(0,0,0), Ru_2=(0.5,0,0.5), Ru_3=(0,0.5,0.5) and Ru_4=(0.5,0.5,0). The tight-binding model is derived from the Wannier90 code[9], which includes two nearest-neighbor in-plane and two nearest-neighbor out-of-plane hoppings among Ru atoms. In the A-centered configuration, Ru_1 and Ru_3 are spin ↑ oriented, while Ru_2 and Ru_4 spin ↓. Therefore, for Ru_1 and Ru_3, the majority channel is the spin ↑ channel. The cell and the hoppings considered are shown in Fig. S3. As we can see from the Figure, we have considered two in-plane hoppings, namely t^{100} and t^{110}, and two hoppings among the planes, namely $t^{0\frac{1}{2}\frac{1}{2}}$ and t^{111}. Note that the hopping $t^{0\frac{1}{2}\frac{1}{2}}$ is the one named $t^{\frac{1}{2}\frac{1}{2}\frac{1}{2}}$ when we have a single Ru atom in the unit cell[10]. We start by writing the matrix elements only for the spin majority. The majority spins on Ru_1 and Ru_3 are spin up, while the majority spins on Ru_2 and Ru_4 are spin down; they will have the same Δ_z, which will be omitted in the next equations. The basis for the spin-up channel is (Ru_1 d_{xy}, Ru_1 d_{xz}, Ru_1 d_{yz}, Ru_3 d_{xy}, Ru_3 d_{xz}, Ru_3 d_{yz}). Therefore, the Hamiltonian for the matrix for the spin up is:

$$\hat{H}(k_x,k_y,k_z) = \begin{pmatrix} H_{11} & H_{13} \\ H_{31} & H_{33} \end{pmatrix}$$

with the explicit form of the Hamiltonian terms being:

$$H_{11}=\begin{pmatrix} \varepsilon^0_{1xy,1xy}+2t^{100}_{1xy,1xy}\cos k_x a+2t^{010}_{1xy,1xy}\cos k_y b.. & \varepsilon^0_{1xy,1xz}+2t^{100}_{1xy,1xz}\cos k_x a+2t^{010}_{1xy,1xz}\cos k_y b.. & \varepsilon^0_{1xy,1yz}+2t^{100}_{1xy,1yz}\cos k_x a+2t^{010}_{1xy,1yz}\cos k_y b.. \\ & \varepsilon^0_{xz,xz}+2t^{100}_{1xz,1xz}\cos k_x a+2t^{010}_{1xz,1xz}\cos k_y b.. & \varepsilon^0_{1xz,1yz}+2t^{100}_{1xz,1yz}\cos k_x a+2t^{010}_{1xz,1yz}\cos k_y b.. \\ & & \varepsilon^0_{yz,yz}+2t^{100}_{1yz,1yz}\cos k_x a+2t^{010}_{1yz,1yz}\cos k_y b.. \end{pmatrix}$$

and

$$H_{33}=\begin{pmatrix} \varepsilon^0_{3xy,3xy}+2t^{100}_{3xy,3xy}\cos k_x a+2t^{010}_{3xy,3xy}\cos k_y b.. & \varepsilon^0_{3xy,3xz}+2t^{100}_{3xy,3xz}\cos k_x a+2t^{010}_{3xy,3xz}\cos k_y b.. & \varepsilon^0_{3xy,3yz}+2t^{100}_{3xy,3yz}\cos k_x a+2t^{010}_{3xy,3yz}\cos k_y b.. \\ & \varepsilon^0_{xz,xz}+2t^{100}_{3xz,3xz}\cos k_x a+2t^{010}_{3xz,3xz}\cos k_y b.. & \varepsilon^0_{3xz,3yz}+2t^{100}_{3xz,3yz}\cos k_x a+2t^{010}_{3xz,3yz}\cos k_y b.. \\ & & \varepsilon^0_{yz,yz}+2t^{100}_{3yz,3yz}\cos k_x a+2t^{010}_{3yz,3yz}\cos k_y b.. \end{pmatrix}$$

We simplify the Hamiltonian using the following relations, which are derived by symmetry: where $\varepsilon^0_{1xy,1xy}$=$\varepsilon^0_{3xy,3xy}$=$\varepsilon^0_{xy,xy}$ and $\varepsilon^0_{1xz,1xz}$=$\varepsilon^0_{3yz,3yz}$=$\varepsilon^0_{xz,xz}$, while $\varepsilon^0_{1yz,1yz}$=$\varepsilon^0_{3xz,3xz}$=$\varepsilon^0_{yz,yz}$ and for the diagonal hopping parameters, we have: $t^{100}_{1xy,1xy}=t^{100}_{3xy,3xy}=t^{100}_{xy,xy}$; $t^{010}_{1xy,1xy}=t^{010}_{3xy,3xy}=t^{010}_{xy,xy}$; $t^{100}_{1xz,1xz}=t^{100}_{3yz,3yz}=t^{100}_{xz,xz}$; $t^{010}_{1xz,1xz}=t^{010}_{3yz,3yz}=t^{010}_{xz,xz}$, while for the non-diagonal terms we have $\varepsilon^0_{1xy,1xz}$=$-\varepsilon^0_{3xy,3yz}$=$\varepsilon^0_{xy,xz}$; $\varepsilon^0_{1xy,1yz}$=$-\varepsilon^0_{3xy,3xz}$=$\varepsilon^0_{xy,yz}$; $\varepsilon^0_{1xz,1yz}$=$\varepsilon^0_{3xz,3yz}$=$\varepsilon^0_{xz,yz}$. $t^{100}_{1xy,1xz}=-t^{100}_{3xy,3yz}=t^{100}_{1xy,1xz}$; $t^{100}_{1xy,1yz}=-t^{100}_{3xy,3xz}=t^{100}_{1xy,1yz}$; $t^{100}_{1xz,1yz}=t^{100}_{3xz,3yz}=t^{100}_{1xz,1yz}$. $t^{010}_{1xy,1xz}=-t^{010}_{3xy,3yz}=t^{010}_{1xy,1xz}$; $t^{010}_{1xy,1yz}=-t^{010}_{3xy,3xz}=t^{010}_{1xy,1yz}$; $t^{010}_{1xz,1yz}=t^{010}_{3xz,3yz}=t^{010}_{1xz,1yz}$.

By using these equalities and using a=b=c=1, we get a simpler form for H_{11}:

$$H_{11}=\begin{pmatrix} \varepsilon^0_{xy,xy}+2t^{100}_{xy,xy}\cos k_x+2t^{010}_{xy,xy}\cos k_y.. & \varepsilon^0_{xy,xz}+2t^{100}_{xy,xz}\cos k_x+2t^{010}_{xy,xz}\cos k_y.. & \varepsilon^0_{xy,yz}+2t^{100}_{xy,yz}\cos k_x+2t^{010}_{xy,yz}\cos k_y.. \\ & \varepsilon^0_{xz,xz}+2t^{100}_{xz,xz}\cos k_x+2t^{010}_{xz,xz}\cos k_y.. & \varepsilon^0_{xz,yz}+2t^{100}_{xz,yz}\cos k_x+2t^{010}_{xz,yz}\cos k_y.. \\ & & \varepsilon^0_{yz,yz}+2t^{100}_{yz,yz}\cos k_x+2t^{010}_{yz,yz}\cos k_y.. \end{pmatrix}$$

while the H_{33} matrix in the A-centered magnetic order can be obtained by exchanging x an y amd reads:

$$H_{33}=\begin{pmatrix} \varepsilon^0_{xy,xy}+2t^{100}_{xy,xy}\cos k_x+2t^{010}_{xy,xy}\cos k_y.. & -(\varepsilon^0_{xy,yz}+2t^{100}_{xy,yz}\cos k_x+2t^{010}_{xy,yz}\cos k_y).. & -(\varepsilon^0_{xy,xz}+2t^{100}_{xy,xz}\cos k_x+2t^{010}_{xy,xz}\cos k_y).. \\ & \varepsilon^0_{yz,yz}+2t^{100}_{yz,yz}\cos k_x+2t^{010}_{yz,yz}\cos k_y.. & \varepsilon^0_{xz,yz}+2t^{100}_{xz,yz}\cos k_x+2t^{010}_{xz,yz}\cos k_y.. \\ & & \varepsilon^0_{xz,xz}+2t^{100}_{xz,xz}\cos k_x a+2t^{010}_{xz,xz}\cos k_y.. \end{pmatrix}$$

The explicit expressions of spin-up inter-orbital terms are:

$$\begin{aligned}
H_{1xy,1xz} &= \varepsilon^0_{xy,xz} + 2t^{100}_{xy,xz}\cos k_x + 2t^{010}_{xy,xz}\cos k_y + 2(t^{110}_{Axy,xz} + t^{110}_{Bxy,xz})\cos k_x\cos k_y - 2(t^{110}_{Axy,xz} - t^{110}_{Bxy,xz})\sin k_x\sin k_y \\
&\quad +2(t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} + t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\cos k_x\cos k_y\cos k_z \\
&\quad -2(t^{111}_{Axy,xz} - t^{111}_{Bxy,xz} - t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\sin k_x\sin k_y\cos k_z \\
&\quad +2(-t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} - t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\sin k_x\cos k_y\sin k_z \\
&\quad +2(-t^{111}_{Axy,xz} - t^{111}_{Bxy,xz} + t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\cos k_x\sin k_y\sin k_z \\
H_{1xy,1yz} &= \varepsilon^0_{xy,yz} + 2t^{100}_{xy,yz}\cos k_x + 2t^{010}_{xy,yz}\cos k_y + 2(t^{110}_{Axy,yz} + t^{110}_{Bxy,yz})\cos k_x\cos k_y - 2(t^{110}_{Axy,yz} - t^{110}_{Bxy,yz})\sin k_x\sin k_y \\
&\quad +2(t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} + t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\cos k_x\cos k_y\cos k_z \\
&\quad -2(t^{111}_{Axy,yz} - t^{111}_{Bxy,yz} - t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\sin k_x\sin k_y\cos k_z \\
&\quad +2(-t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} - t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\sin k_x\cos k_y\sin k_z \\
&\quad +2(-t^{111}_{Axy,yz} - t^{111}_{Bxy,yz} + t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\cos k_x\sin k_y\sin k_z \\
H_{1xz,1yz} &= \varepsilon^0_{xz,yz} + 2t^{100}_{xz,yz}\cos k_x + 2t^{010}_{xz,yz}\cos k_y + 2(t^{110}_{Axz,yz} + t^{110}_{Bxz,yz})\cos k_x\cos k_y - 2(t^{110}_{Axz,yz} - t^{110}_{Bxz,yz})\sin k_x\sin k_y \\
&\quad +2(t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} + t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\cos k_x\cos k_y\cos k_z \\
&\quad -2(t^{111}_{Axz,yz} - t^{111}_{Bxz,yz} - t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\sin k_x\sin k_y\cos k_z \\
&\quad +2(-t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} - t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\sin k_x\cos k_y\sin k_z \\
&\quad +2(-t^{111}_{Axz,yz} - t^{111}_{Bxz,yz} + t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\cos k_x\sin k_y\sin k_z \\
H_{3xy,3xz} &= -(\varepsilon^0_{xy,yz} + 2t^{100}_{xy,yz}\cos k_x + 2t^{010}_{xy,yz}\cos k_y) - 2(t^{110}_{Axy,yz} + t^{110}_{Bxy,yz})\cos k_x\cos k_y - 2(t^{110}_{Axy,yz} - t^{110}_{Bxy,yz})\sin k_x\sin k_y \\
&\quad -2(t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} + t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\cos k_x\cos k_y\cos k_z \\
&\quad -2(t^{111}_{Axy,yz} - t^{111}_{Bxy,yz} - t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\sin k_x\sin k_y\cos k_z \\
&\quad -2(-t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} - t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\sin k_x\cos k_y\sin k_z \\
&\quad +2(-t^{111}_{Axy,yz} - t^{111}_{Bxy,yz} + t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\cos k_x\sin k_y\sin k_z \\
H_{3xy,3yz} &= -(\varepsilon^0_{xy,xz} + 2t^{100}_{xy,xz}\cos k_x + 2t^{010}_{xy,xz}\cos k_y) - 2(t^{110}_{Axy,xz} + t^{110}_{Bxy,xz})\cos k_x\cos k_y - 2(t^{110}_{Axy,xz} - t^{110}_{Bxy,xz})\sin k_x\sin k_y \\
&\quad -2(t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} + t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\cos k_x\cos k_y\cos k_z \\
&\quad -2(t^{111}_{Axy,xz} - t^{111}_{Bxy,xz} - t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\sin k_x\sin k_y\cos k_z \\
&\quad -2(-t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} - t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\sin k_x\cos k_y\sin k_z \\
&\quad +2(-t^{111}_{Axy,xz} - t^{111}_{Bxy,xz} + t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\cos k_x\sin k_y\sin k_z \\
H_{3xz,3yz} &= \varepsilon^0_{xz,yz} + 2t^{100}_{xz,yz}\cos k_x + 2t^{010}_{xz,yz}\cos k_y + 2(t^{110}_{Axz,yz} + t^{110}_{Bxz,yz})\cos k_x\cos k_y + 2(t^{110}_{Axz,yz} - t^{110}_{Bxz,yz})\sin k_x\sin k_y \\
&\quad +2(t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} + t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\cos k_x\cos k_y\cos k_z \\
&\quad +2(t^{111}_{Axz,yz} - t^{111}_{Bxz,yz} - t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\sin k_x\sin k_y\cos k_z \\
&\quad +2(-t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} - t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\sin k_x\cos k_y\sin k_z \\
&\quad -2(-t^{111}_{Axz,yz} - t^{111}_{Bxz,yz} + t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\cos k_x\sin k_y\sin k_z
\end{aligned}$$

where t_A, t_B, t_C and t_D are linear combinations of the hopping parameters obtained from the wannierization. The non-diagonal terms of the spin-down describing the hybridizations among different atoms are the following:

$$
\begin{aligned}
H_{1xy,3xy} &= 4t^{0\frac{1}{2}\frac{1}{2}}_{1xy,3xy}\cos k_yb/2\cos k_zc/2\\
H_{1xy,3xz} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{A1xy,3xz}+t^{0\frac{1}{2}\frac{1}{2}}_{B1xy,3xz})\cos k_yb/2\cos k_zc/2-2(t^{0\frac{1}{2}\frac{1}{2}}_{A1xy,3xz}-t^{0\frac{1}{2}\frac{1}{2}}_{B1xy,3xz})\sin k_yb/2\sin k_zc/2\\
H_{1xy,3yz} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{13Axy,yz}+t^{0\frac{1}{2}\frac{1}{2}}_{B1xy,3yz})\cos k_yb/2\cos k_zc/2-2(t^{0\frac{1}{2}\frac{1}{2}}_{13Axy,yz}-t^{0\frac{1}{2}\frac{1}{2}}_{B1xy,3yz})\sin k_yb/2\sin k_zc/2\\
H_{1xz,3xy} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{13Axz,xy}+t^{0\frac{1}{2}\frac{1}{2}}_{13Bxz,xy})\cos k_yb/2\cos k_zc/2-2(t^{0\frac{1}{2}\frac{1}{2}}_{13Axz,xy}-t^{0\frac{1}{2}\frac{1}{2}}_{13Bxz,xy})\sin k_yb/2\sin k_zc/2\\
H_{1xz,3xz} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{A1xz,3xz}+t^{0\frac{1}{2}\frac{1}{2}}_{B1xz,3xz})\cos k_yb/2\cos k_zc/2-2(t^{0\frac{1}{2}\frac{1}{2}}_{A1xz,3xz}-t^{0\frac{1}{2}\frac{1}{2}}_{B1xz,3xz})\sin k_yb/2\sin k_zc/2\\
H_{1xz,3yz} &= 4t^{0\frac{1}{2}\frac{1}{2}}_{13xz,yz}\cos k_yb/2\cos k_zc/2\\
H_{1yz,3xy} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{13Ayz,xy}+t^{0\frac{1}{2}\frac{1}{2}}_{13Byz,xy})\cos k_yb/2\cos k_zc/2-2(t^{0\frac{1}{2}\frac{1}{2}}_{13Ayz,xy}-t^{0\frac{1}{2}\frac{1}{2}}_{13Byz,xy})\sin k_yb/2\sin k_zc/2\\
H_{1yz,3xz} &= 4t^{0\frac{1}{2}\frac{1}{2}}_{13yz,xz}\cos k_yb/2\cos k_zc/2\\
H_{1yz,3yz} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{A1yz,3yz}+t^{0\frac{1}{2}\frac{1}{2}}_{B1yz,3yz})\cos k_yb/2\cos k_zc/2-2(t^{0\frac{1}{2}\frac{1}{2}}_{A1yz,3yz}-t^{0\frac{1}{2}\frac{1}{2}}_{B1yz,3yz})\sin k_yb/2\sin k_zc/2
\end{aligned}
$$

After simplifying the notation and using some equivalences among the hopping parameters, we obtain:

$$
\begin{aligned}
H_{1xy,3xy} &= 4t^{0\frac{1}{2}\frac{1}{2}}_{xy,xy}\cos\frac{k_y}{2}\cos\frac{k_z}{2}\\
H_{1xy,3xz} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,xz}+t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,xz})\cos\frac{k_y}{2}\cos\frac{k_z}{2}-2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,xz}-t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,xz})\sin\frac{k_y}{2}\sin\frac{k_z}{2}\\
H_{1xy,3yz} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,yz}+t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,yz})\cos\frac{k_y}{2}\cos\frac{k_z}{2}-2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,yz}-t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,yz})\sin\frac{k_y}{2}\sin\frac{k_z}{2}\\
H_{1xz,3xy} &= -2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,yz}+t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,yz})\cos\frac{k_y}{2}\cos\frac{k_z}{2}-2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,yz}-t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,yz})\sin\frac{k_y}{2}\sin\frac{k_z}{2}\\
H_{1xz,3xz} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axz,xz}+t^{0\frac{1}{2}\frac{1}{2}}_{Bxz,xz})\cos\frac{k_y}{2}\cos\frac{k_z}{2}-2(t^{0\frac{1}{2}\frac{1}{2}}_{Axz,xz}-t^{0\frac{1}{2}\frac{1}{2}}_{Bxz,xz})\sin\frac{k_y}{2}\sin\frac{k_z}{2}\\
H_{1xz,3yz} &= 4t^{0\frac{1}{2}\frac{1}{2}}_{xz,yz}\cos\frac{k_y}{2}\cos\frac{k_z}{2}\\
H_{1yz,3xy} &= -2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,xz}+t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,xz})\cos\frac{k_y}{2}\cos\frac{k_z}{2}-2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,xz}-t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,xz})\sin\frac{k_y}{2}\sin\frac{k_z}{2}\\
H_{1yz,3xz} &= 4t^{0\frac{1}{2}\frac{1}{2}}_{yz,xz}\cos\frac{k_y}{2}\cos\frac{k_z}{2}\\
H_{1yz,3yz} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axz,xz}+t^{0\frac{1}{2}\frac{1}{2}}_{Bxz,xz})\cos\frac{k_y}{2}\cos\frac{k_z}{2}+2(t^{0\frac{1}{2}\frac{1}{2}}_{Axz,xz}-t^{0\frac{1}{2}\frac{1}{2}}_{Bxz,xz})\sin\frac{k_y}{2}\sin\frac{k_z}{2}
\end{aligned}
$$

For the spin-down part, H_{22}=H_{11} and H_{44}=H_{33}; however, there is a sign change in the hopping of H_{24} with respect

TABLE S1. Hopping integrals $t^{nlm}_{\alpha,\beta}$ along the direction [lmn] between the Ru atoms intralayer extracted from the tight-binding Hamiltonian with Wannier function as basis, where α and β are different orbitals. The connecting vector **T** is expressed in terms of the integer set [lmn] and the lattice constants a, b and c as **T** = l a **x** + m b **y** + n c **z**. The unit is meV. In our notation, $t^{000}_{\alpha,\beta}=\varepsilon^0_{\alpha,\beta}$.

	on site	hoppings		
$t^{nlm}_{xy,xy}$	000	100	010	110
$t_{xy,xy}$	3926	−174	−181	
$t_{xz,xz}$	3673	12	19	
$t_{yz,yz}$	3724	3	0	
$t_{xy,xz}$	−50	−2	42	A=−17; B=13
$t_{xy,yz}$	94	9	−11	A=−19; B=−21
$t_{xz,yz}$	−7	−14	31	A=−3; B=7

to H_{13}. The spin-down part of the model is reported below:

$$H_{22} = \begin{pmatrix} \varepsilon^0_{xy,xy}+2t^{100}_{xy,xy}\cos k_x+2t^{010}_{xy,xy}\cos k_y+.. & -(\varepsilon^0_{xy,xz}+2t^{100}_{xy,xz}\cos k_x+2t^{010}_{xy,xz}\cos k_y)+.. & -(\varepsilon^0_{xy,yz}+2t^{100}_{xy,yz}\cos k_x+2t^{010}_{xy,yz}\cos k_y)+.. \\ & \varepsilon^0_{xz,xz}+2t^{100}_{xz,xz}\cos k_x+2t^{010}_{xz,xz}\cos k_y+.. & \varepsilon^0_{xz,yz}+2t^{100}_{xz,yz}\cos k_x+2t^{010}_{xz,yz}\cos k_y+.. \\ & & \varepsilon^0_{yz,yz}+2t^{100}_{yz,yz}\cos k_x+2t^{010}_{yz,yz}\cos k_y+.. \end{pmatrix}$$

and

$$H_{44} = \begin{pmatrix} \varepsilon^0_{xy,xy}+2t^{100}_{xy,xy}\cos k_x+2t^{010}_{xy,xy}\cos k_y+.. & \varepsilon^0_{xy,yz}+2t^{100}_{xy,yz}\cos k_x+2t^{010}_{xy,yz}\cos k_y+.. & \varepsilon^0_{xy,xz}+2t^{100}_{xy,xz}\cos k_x+2t^{010}_{xy,xz}\cos k_y+.. \\ & \varepsilon^0_{yz,yz}+2t^{100}_{yz,yz}\cos k_x+2t^{010}_{yz,yz}\cos k_y+.. & \varepsilon^0_{xz,yz}+2t^{100}_{xz,yz}\cos k_x+2t^{010}_{xz,yz}\cos k_y+.. \\ & & \varepsilon^0_{xz,xz}+2t^{100}_{xz,xz}\cos k_x+2t^{010}_{xz,xz}\cos k_y+.. \end{pmatrix}$$

The explicit form for H_{22} and H_{44} reads:

$$\begin{aligned}
H_{2xy,2xz} &= -(\varepsilon^0_{xy,xz} + 2t^{100}_{xy,xz}\cos k_x + 2t^{010}_{xy,xz}\cos k_y) - 2(t^{110}_{Axy,xz} + t^{110}_{Bxy,xz})\cos k_x\cos k_y + 2(t^{110}_{Axy,xz} - t^{110}_{Bxy,xz})\sin k_x\sin k_y \\
&\quad +2(t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} + t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\cos k_x\cos k_y\cos k_z \\
&\quad +2(t^{111}_{Axy,xz} - t^{111}_{Bxy,xz} - t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\sin k_x\sin k_y\cos k_z \\
&\quad +2(-t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} - t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\sin k_x\cos k_y\sin k_z \\
&\quad +2(-t^{111}_{Axy,xz} - t^{111}_{Bxy,xz} + t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\cos k_x\sin k_y\sin k_z \\
H_{2xy,2yz} &= -(\varepsilon^0_{xy,yz} + 2t^{100}_{xy,yz}\cos k_x + 2t^{010}_{xy,yz}\cos k_y) - 2(t^{110}_{Axy,yz} + t^{110}_{Bxy,yz})\cos k_x\cos k_y + 2(t^{110}_{Axy,yz} - t^{110}_{Bxy,yz})\sin k_x\sin k_y \\
&\quad -2(t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} + t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\cos k_x\cos k_y\cos k_z \\
&\quad +2(t^{111}_{Axy,yz} - t^{111}_{Bxy,yz} - t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\sin k_x\sin k_y\cos k_z \\
&\quad +2(-t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} - t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\sin k_x\cos k_y\sin k_z \\
&\quad +2(-t^{111}_{Axy,yz} - t^{111}_{Bxy,yz} + t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\cos k_x\sin k_y\sin k_z \\
H_{2xz,2yz} &= \varepsilon^0_{xz,yz} + 2t^{100}_{xz,yz}\cos k_x + 2t^{010}_{xz,yz}\cos k_y + 2(t^{110}_{Axz,yz} + t^{110}_{Bxz,yz})\cos k_x\cos k_y - 2(t^{110}_{Axz,yz} - t^{110}_{Bxz,yz})\sin k_x\sin k_y \\
&\quad +2(t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} + t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\cos k_x\cos k_y\cos k_z \\
&\quad -2(t^{111}_{Axz,yz} - t^{111}_{Bxz,yz} - t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\sin k_x\sin k_y\cos k_z \\
&\quad -2(-t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} - t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\sin k_x\cos k_y\sin k_z \\
&\quad -2(-t^{111}_{Axz,yz} - t^{111}_{Bxz,yz} + t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\cos k_x\sin k_y\sin k_z
\end{aligned}$$

TABLE S2. Hopping integrals $t^{nlm}_{\alpha,\beta}$ along the direction [lmn] between the Ru atoms interlayer extracted from the tight-binding Hamiltonian with Wannier function as basis, where α and β are different orbitals. The connecting vector **T** is expressed in terms of the integer set [lmn] and the lattice constants a, b and c as **T** = l a **x** + m b **y** + n c **z**. The unit is meV. In our notation, $t^{000}_{\alpha,\beta} = \varepsilon^0_{\alpha,\beta}$.

$t^{nlm}_{xy,xy}$	$0\frac{1}{2}\frac{1}{2}$
$t_{1xy,3xy}$	3
$t_{1xy,3xz}$	$t_{13A}=13$ $t_{13B}=-2$
$t_{1xy,3yz}$	$t_{13A}=4$ $t_{13B}=8$
$t_{1xz,3xz}=t_{1yz,3yz}$	$t_{13A}=-27$ $t_{13B}=-1$
$t_{1xz,3yz}$	-12
$t_{1yz,3xz}$	-11

$$
\begin{aligned}
H_{4xy,4xz} = {} & \varepsilon^{0}_{xy,yz} + 2t^{100}_{xy,yz}\cos k_x + 2t^{010}_{xy,yz}\cos k_y + 2(t^{110}_{Axy,yz} + t^{110}_{Bxy,yz})\cos k_x \cos k_y + 2(t^{110}_{Axy,yz} - t^{110}_{Bxy,yz})\sin k_x \sin k_y \\
& +2(t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} + t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\cos k_x \cos k_y \cos k_z \\
& +2(t^{111}_{Axy,yz} - t^{111}_{Bxy,yz} - t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\sin k_x \sin k_y \cos k_z \\
& -2(-t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} - t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\sin k_x \cos k_y \sin k_z \\
& +2(-t^{111}_{Axy,yz} - t^{111}_{Bxy,yz} + t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\cos k_x \sin k_y \sin k_z \\
H_{4xy,4yz} = {} & \varepsilon^{0}_{xy,xz} + 2t^{100}_{xy,xz}\cos k_x + 2t^{010}_{xy,xz}\cos k_y + 2(t^{110}_{Axy,xz} + t^{110}_{Bxy,xz})\cos k_x \cos k_y + 2(t^{110}_{Axy,xz} - t^{110}_{Bxy,xz})\sin k_x \sin k_y \\
& +2(t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} + t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\cos k_x \cos k_y \cos k_z \\
& +2(t^{111}_{Axy,xz} - t^{111}_{Bxy,xz} - t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\sin k_x \sin k_y \cos k_z \\
& -2(-t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} - t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\sin k_x \cos k_y \sin k_z \\
& +2(-t^{111}_{Axy,xz} - t^{111}_{Bxy,xz} + t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\cos k_x \sin k_y \sin k_z \\
H_{4xz,4yz} = {} & \varepsilon^{0}_{xz,yz} + 2t^{100}_{xz,yz}\cos k_x + 2t^{010}_{xz,yz}\cos k_y + 2(t^{110}_{Axz,yz} + t^{110}_{Bxz,yz})\cos k_x \cos k_y + 2(t^{110}_{Axz,yz} - t^{110}_{Bxz,yz})\sin k_x \sin k_y \\
& +2(t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} + t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\cos k_x \cos k_y \cos k_z \\
& +2(t^{111}_{Axz,yz} - t^{111}_{Bxz,yz} - t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\sin k_x \sin k_y \cos k_z \\
& -2(-t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} - t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\sin k_x \cos k_y \sin k_z \\
& +2(-t^{111}_{Axz,yz} - t^{111}_{Bxz,yz} + t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\cos k_x \sin k_y \sin k_z
\end{aligned}
$$

while the non-diagonal terms of the spin-down, describing the hybridization between different atoms, are:

$$
\begin{aligned}
H_{2xy,4xy} &= 4t^{0\frac{1}{2}\frac{1}{2}}_{xy,xy}\cos\frac{k_y}{2}\cos\frac{k_z}{2} \\
H_{2xy,4xz} &= -2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,xz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,xz})\cos\frac{k_y}{2}\cos\frac{k_z}{2} - 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,xz} - t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,xz})\sin\frac{k_y}{2}\sin\frac{k_z}{2} \\
H_{2xy,4yz} &= -2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,yz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,yz})\cos\frac{k_y}{2}\cos\frac{k_z}{2} - 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,yz} - t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,yz})\sin\frac{k_y}{2}\sin\frac{k_z}{2} \\
H_{2xz,4xy} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,yz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,yz})\cos\frac{k_y}{2}\cos\frac{k_z}{2} - 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,yz} - t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,yz})\sin\frac{k_y}{2}\sin\frac{k_z}{2} \\
H_{2xz,4xz} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axz,xz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxz,xz})\cos\frac{k_y}{2}\cos\frac{k_z}{2} + 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axz,xz} - t^{0\frac{1}{2}\frac{1}{2}}_{Bxz,xz})\sin\frac{k_y}{2}\sin\frac{k_z}{2} \\
H_{2xz,4yz} &= 4t^{0\frac{1}{2}\frac{1}{2}}_{xz,yz}\cos\frac{k_y}{2}\cos\frac{k_z}{2} \\
H_{2yz,4xy} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,xz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,xz})\cos\frac{k_y}{2}\cos\frac{k_z}{2} - 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,xz} - t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,xz})\sin\frac{k_y}{2}\sin\frac{k_z}{2} \\
H_{2yz,4xz} &= 4t^{0\frac{1}{2}\frac{1}{2}}_{yz,xz}\cos\frac{k_y}{2}\cos\frac{k_z}{2} \\
H_{2yz,4yz} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axz,xz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxz,xz})\cos\frac{k_y}{2}\cos\frac{k_z}{2} - 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axz,xz} - t^{0\frac{1}{2}\frac{1}{2}}_{Bxz,xz})\sin\frac{k_y}{2}\sin\frac{k_z}{2}
\end{aligned}
$$

S4. MINIMAL MODEL FOR THE ALTERMAGNETISM

In the previous Section, we developed the complete formalism for the long-range neighbours hopping for the Ca_2RuO_4. In this Section, the minimal model for the altermagnetism is reported. We will create a minimal model where we will keep only the first neighbour hopping, along with the hopping necessary for the altermagnetism. We should note that the altermagnetic terms in red have the same sign inside the spin-up channel, which is the opposite of the spin-down channel.

For the spin-up Hamiltonian, the diagonal intralayer hybridization terms read:

$$
\begin{aligned}
H_{1xy,1xy} &= \varepsilon^0_{xy,xy} + 2t^{100}_{xy,xy}\cos k_x + 2t^{010}_{xy,xy}\cos k_y \\
H_{1xz,1xz} &= \varepsilon^0_{xz,xz} + 2t^{100}_{xz,xz}\cos k_x + 2t^{010}_{xz,xz}\cos k_y \\
H_{1yz,1yz} &= \varepsilon^0_{yz,yz} + 2t^{100}_{yz,yz}\cos k_x + 2t^{010}_{yz,yz}\cos k_y \\
H_{3xy,3xy} &= \varepsilon^0_{xy,xy} + 2t^{100}_{xy,xy}\cos k_x + 2t^{010}_{xy,xy}\cos k_y \\
H_{3xz,3xz} &= \varepsilon^0_{yz,yz} + 2t^{100}_{yz,yz}\cos k_x + 2t^{010}_{yz,yz}\cos k_y \\
H_{3yz,3yz} &= \varepsilon^0_{xz,xz} + 2t^{100}_{xz,xz}\cos k_x + 2t^{010}_{xz,xz}\cos k_y
\end{aligned}
$$

while the spin-up non-diagonal intralayer and interlayer hybridization terms

$$
\begin{aligned}
H_{1xy,1xz} &= -2(t^{110}_{Axy,xz} - t^{110}_{Bxy,xz})\sin k_x \sin k_y \\
&\quad +2(t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} + t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\cos k_x \cos k_y \cos k_z \\
&\quad +2(-t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} - t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\sin k_x \cos k_y \sin k_z \\
H_{1xy,1yz} &= -2(t^{110}_{Axy,yz} - t^{110}_{Bxy,yz})\sin k_x \sin k_y \\
&\quad +2(t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} + t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\cos k_x \cos k_y \cos k_z \\
&\quad +2(-t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} - t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\sin k_x \cos k_y \sin k_z \\
H_{1xz,1yz} &= +2(t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} + t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\cos k_x \cos k_y \cos k_z \\
&\quad +2(-t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} - t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\sin k_x \cos k_y \sin k_z \\
H_{3xy,3xz} &= -2(t^{110}_{Axy,yz} - t^{110}_{Bxy,yz})\sin k_x \sin k_y \\
&\quad -2(t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} + t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\cos k_x \cos k_y \cos k_z \\
&\quad -2(-t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} - t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz})\sin k_x \cos k_y \sin k_z \\
H_{3xy,3yz} &= -2(t^{110}_{Axy,xz} - t^{110}_{Bxy,xz})\sin k_x \sin k_y \\
&\quad -2(t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} + t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\cos k_x \cos k_y \cos k_z \\
&\quad -2(-t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} - t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz})\sin k_x \cos k_y \sin k_z \\
H_{3xz,3yz} &= +2(t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} + t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\cos k_x \cos k_y \cos k_z \\
&\quad +2(-t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} - t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz})\sin k_x \cos k_y \sin k_z
\end{aligned}
$$

For the spin-down Hamiltonian, the diagonal intralayer hybridization terms read:

$$
\begin{aligned}
H_{2xy,2xy} &= \varepsilon^0_{xy,xy} + 2t^{100}_{xy,xy}\cos k_x + 2t^{010}_{xy,xy}\cos k_y \\
H_{2xz,2xz} &= \varepsilon^0_{xz,xz} + 2t^{100}_{xz,xz}\cos k_x + 2t^{010}_{xz,xz}\cos k_y \\
H_{2yz,2yz} &= \varepsilon^0_{yz,yz} + 2t^{100}_{yz,yz}\cos k_x + 2t^{010}_{yz,yz}\cos k_y \\
H_{4xy,4xy} &= \varepsilon^0_{xy,xy} + 2t^{100}_{xy,xy}\cos k_x + 2t^{010}_{xy,xy}\cos k_y \\
H_{4xz,4xz} &= \varepsilon^0_{yz,yz} + 2t^{100}_{yz,yz}\cos k_x + 2t^{010}_{yz,yz}\cos k_y \\
H_{4yz,4yz} &= \varepsilon^0_{xz,xz} + 2t^{100}_{xz,xz}\cos k_x + 2t^{010}_{xz,xz}\cos k_y
\end{aligned}
$$

while the spin-down non-diagonal intralayer and interlayer hybridization terms

$$
\begin{aligned}
H_{2xy,2xz} &= +2(t^{110}_{Axy,xz} - t^{110}_{Bxy,xz}) \sin k_x \sin k_y \\
&+2(t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} + t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz}) \cos k_x \cos k_y \cos k_z \\
&+2(-t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} - t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz}) \sin k_x \cos k_y \sin k_z \\
H_{2xy,2yz} &= +2(t^{110}_{Axy,yz} - t^{110}_{Bxy,yz}) \sin k_x \sin k_y \\
&-2(t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} + t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz}) \cos k_x \cos k_y \cos k_z \\
&+2(-t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} - t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz}) \sin k_x \cos k_y \sin k_z \\
H_{2xz,2yz} &= +2(t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} + t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz}) \cos k_x \cos k_y \cos k_z \\
&-2(-t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} - t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz}) \sin k_x \cos k_y \sin k_z \\
H_{4xy,4xz} &= +2(t^{110}_{Axy,yz} - t^{110}_{Bxy,yz}) \sin k_x \sin k_y \\
&+2(t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} + t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz}) \cos k_x \cos k_y \cos k_z \\
&-2(-t^{111}_{Axy,yz} + t^{111}_{Bxy,yz} - t^{111}_{Cxy,yz} + t^{111}_{Dxy,yz}) \sin k_x \cos k_y \sin k_z \\
H_{4xy,4yz} &= +2(t^{110}_{Axy,xz} - t^{110}_{Bxy,xz}) \sin k_x \sin k_y \\
&+2(t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} + t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz}) \cos k_x \cos k_y \cos k_z \\
&-2(-t^{111}_{Axy,xz} + t^{111}_{Bxy,xz} - t^{111}_{Cxy,xz} + t^{111}_{Dxy,xz}) \sin k_x \cos k_y \sin k_z \\
H_{4xz,4yz} &= +2(t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} + t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz}) \cos k_x \cos k_y \cos k_z \\
&-2(-t^{111}_{Axz,yz} + t^{111}_{Bxz,yz} - t^{111}_{Cxz,yz} + t^{111}_{Dxz,yz}) \sin k_x \cos k_y \sin k_z
\end{aligned}
$$

For the spin-up Hamiltonian, the interlayer hybridization terms read:

$$
\begin{aligned}
H_{1xy,3xy} &= 0 \\
H_{1xy,3xz} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,xz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,xz}) \cos\frac{k_y}{2} \cos\frac{k_z}{2} \\
H_{1xy,3yz} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,yz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,yz}) \cos\frac{k_y}{2} \cos\frac{k_z}{2} \\
H_{1xz,3xy} &= -2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,yz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,yz}) \cos\frac{k_y}{2} \cos\frac{k_z}{2} \\
H_{1xz,3xz} &= -2(t^{0\frac{1}{2}\frac{1}{2}}_{Axz,xz} - t^{0\frac{1}{2}\frac{1}{2}}_{Bxz,xz}) \sin\frac{k_y}{2} \sin\frac{k_z}{2} \\
H_{1xz,3yz} &= 0 \\
H_{1yz,3xy} &= -2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,xz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,xz}) \cos\frac{k_y}{2} \cos\frac{k_z}{2} \\
H_{1yz,3xz} &= 0 \\
H_{1yz,3yz} &= +2(t^{0\frac{1}{2}\frac{1}{2}}_{Axz,xz} - t^{0\frac{1}{2}\frac{1}{2}}_{Bxz,xz}) \sin\frac{k_y}{2} \sin\frac{k_z}{2}
\end{aligned}
$$

For the spin-down Hamiltonian, the interlayer hybridization terms read:

$$\begin{aligned}
H_{2xy,4xy} &= 0 \\
H_{2xy,4xz} &= -2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,xz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,xz}) \cos\frac{k_y}{2}\cos\frac{k_z}{2} \\
H_{2xy,4yz} &= -2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,yz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,yz}) \cos\frac{k_y}{2}\cos\frac{k_z}{2} \\
H_{2xz,4xy} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,yz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,yz}) \cos\frac{k_y}{2}\cos\frac{k_z}{2} \\
H_{2xz,4xz} &= +2(t^{0\frac{1}{2}\frac{1}{2}}_{Axz,xz} - t^{0\frac{1}{2}\frac{1}{2}}_{Bxz,xz}) \sin\frac{k_y}{2}\sin\frac{k_z}{2} \\
H_{2xz,4yz} &= 0 \\
H_{2yz,4xy} &= 2(t^{0\frac{1}{2}\frac{1}{2}}_{Axy,xz} + t^{0\frac{1}{2}\frac{1}{2}}_{Bxy,xz}) \cos\frac{k_y}{2}\cos\frac{k_z}{2} \\
H_{2yz,4xz} &= 0 \\
H_{2yz,4yz} &= -2(t^{0\frac{1}{2}\frac{1}{2}}_{Axz,xz} - t^{0\frac{1}{2}\frac{1}{2}}_{Bxz,xz}) \sin\frac{k_y}{2}\sin\frac{k_z}{2}
\end{aligned}$$

We can see how the non-diagonal interlyer hybridizations $H_{xz,yz}$ host terms $t^{111}_{xz,yz}\sin k_x \cos k_y \sin k_z$, which change sign moving from spin-up to spin-down. These are altermagnetic terms which give rise to the bulk non-relativistic d_{xz} altermagnetism. Therefore the term $t^{111}_{xz,yz}$ is related to t^{3D}_{am} in our model. In this system, the non-diagonal intralyer hybridizations $H_{xy,\gamma z}$ contain contributions of the form $t^{110}_{xy,xz}\sin k_x \sin k_y$, which change sign when moving from spin-up to spin-down states. These are altermagnetic terms; however, this d_{xy} altermagnetism remains hidden in the bulk[11] and becomes apparent only at the surface[12] or in the stripe phase (see main text). Therefore the term $t^{110}_{xy,xz}$ is related to t^{2D}_{am} in our model. In addition, other terms also change sign between spin-up and spin-down states; these involve only cosine or only sine functions and appear in both interlayer and intralayer hybridization processes. Without these terms, the altermagnetism disappears even for the bulk, meaning that without these terms, both altermagnetic spin-splitting becomes hidden.

The tight-binding model captures all the symmetries of the system, but even though it contains only the minimal terms, it remains extremely complex; hence, in the main text, we focus on simplified versions of the model for Ca_2RuO_4.

S5. ANALYTICAL MODEL FOR RELATIVISTIC SPIN-MOMENTUM LOCKING

We calculated the band structure using the simplified 4×4 model Hamiltonian or an effective single-orbital Hamiltonian in terms of Pauli matrices for spin and site as described in the main text:

$$\mathcal{H}^0 = \varepsilon(\boldsymbol{k})\sigma_0^{spin}\sigma_0^{site} \tag{S1}$$

$$\begin{aligned}\mathcal{H}_{Sz}^{\mathrm{AM}} =& \Delta_z\sigma_z^{spin}\sigma_z^{site} \\ &+ 4t_{am}\sin k_x \sin k_y \sigma_0^{spin}\sigma_z^{site}\end{aligned} \tag{S2}$$

where t_{am} originates from the octahedral rotations, which are different on the opposite magnetic sites[13]. For the other two components, the hopping producing the spin-momentum locking is activated by the spin-orbit coupling λ via the antisymmetric exchange. We named Δ_x and Δ_y for the relative equations to be

$$\mathcal{H}_{Sx}^{\mathrm{AM}} = \Delta_x \sin(k_x)\sin(k_z)\sigma_x^{spin}\sigma_z^{site} \tag{S3}$$

and

$$\mathcal{H}_{Sy}^{\mathrm{AM}} = \Delta_y \sin(k_y)\sin(k_z)\sigma_y^{spin}\sigma_z^{site} \tag{S4}$$

For this model, it is also possible to find the analytic solution describing relativistic spin–momentum locking in Ca_2RuO_4. The four eigenvalues of the total Hamiltonian represented by the sum of the previous three equations are:

$$\begin{aligned}E^{1,2,3,4} =& \varepsilon(\boldsymbol{k}) \pm 4t_{am}\sin(k_x)\sin(k_z) \\ \pm&\sqrt{\Delta_x^2\sin^2(k_x)\sin^2(k_z) + \Delta_y^2\sin^2(k_y)\sin^2(k_z) + \Delta_z^2}\end{aligned} \tag{S5}$$

We can define:

$$Q = \sqrt{\Delta_x^2\sin^2(k_x)\sin^2(k_z) + \Delta_y^2\sin^2(k_y)\sin^2(k_z) + \Delta_z^2} > |\Delta_z|$$

$$P = \Delta_x^2\sin^2(k_x)\sin^2(k_z) - \Delta_y^2\sin^2(k_y)\sin^2(k_z) = \sin^2(k_z)(\Delta_x^2\sin^2(k_x) - \Delta_y^2\sin^2(k_y))$$

where the quantity P is invariant under the mirror operation with respect to the nodal planes; therefore, the spin–momentum locking is unaffected by the sign of P, and the eigenvalues can be written as:

$$E^1 = \varepsilon(\boldsymbol{k}) + 4t_{am}\sin(k_x)\sin(k_z) - Q$$

$$E^2 = \varepsilon(\boldsymbol{k}) + 4t_{am}\sin(k_x)\sin(k_z) + Q$$

$$E^3 = \varepsilon(\boldsymbol{k}) - 4t_{am}\sin(k_x)\sin(k_z) - Q$$

$$E^4 = \varepsilon(\boldsymbol{k}) - 4t_{am}\sin(k_x)\sin(k_z) + Q$$

The fourth analytic non-normalized eigenvectors are:

$$\vec{v}_1 = \begin{pmatrix} \frac{(Q-\Delta_z)\left(\Delta_x\sin(k_x)\sin(k_z) - i\Delta_y\sin(k_y)\sin(k_z)\right)}{P} & 1 & 0 & 0 \end{pmatrix}$$

$$\vec{v}_2 = \begin{pmatrix} \frac{(Q+\Delta_z)\left(\Delta_x\sin(k_x)\sin(k_z) - i\Delta_y\sin(k_y)\sin(k_z)\right)}{P} & 1 & 0 & 0 \end{pmatrix}$$

$$\vec{v}_3 = \begin{pmatrix} 0 & 0 & \frac{(Q+\Delta_z)\left(\Delta_x\sin(k_x)\sin(k_z) - i\Delta_y\sin(k_y)\sin(k_z)\right)}{P} & 1 \end{pmatrix}$$

$$\vec{v}_4 = \begin{pmatrix} 0 & 0 & \frac{(Q-\Delta_z)\big(\Delta_x \sin(k_x)\sin(k_z) - i\Delta_y \sin(k_y)\sin(k_z)\big)}{P} & 1 \end{pmatrix}$$

The expectation value of S$_x$ for the four eigenvalues is:

$$< S_x >_{E^1} \propto \frac{\Delta_x \sin(k_x)\sin(k_z)}{P}(Q-\Delta_z) \tag{S6}$$

$$< S_x >_{E^2} \propto \frac{\Delta_x \sin(k_x)\sin(k_z)}{P}(Q+\Delta_z) \tag{S7}$$

$$< S_x >_{E^3} \propto \frac{\Delta_x \sin(k_x)\sin(k_z)}{P}(Q+\Delta_z) \tag{S8}$$

$$< S_x >_{E^4} \propto \frac{\Delta_x \sin(k_x)\sin(k_z)}{P}(Q-\Delta_z) \tag{S9}$$

First of all, we note that if Δ_x=0, then the expectation value of $< S_x >$ is zero for all bands.

The expectation value of S$_y$ for the four eigenvalues is:

$$< S_y >_{E^1} \propto \frac{\Delta_y \sin(k_y)\sin(k_z)}{P}(Q-\Delta_z) \tag{S10}$$

$$< S_y >_{E^2} \propto \frac{\Delta_y \sin(k_y)\sin(k_z)}{P}(Q+\Delta_z) \tag{S11}$$

$$< S_y >_{E^3} \propto \frac{\Delta_y \sin(k_y)\sin(k_z)}{P}(Q+\Delta_z) \tag{S12}$$

$$< S_y >_{E^4} \propto \frac{\Delta_y \sin(k_y)\sin(k_z)}{P}(Q-\Delta_z) \tag{S13}$$

First of all, we note that if Δ_y=0, then the expectation value of $< S_y >$ is zero for all bands. We also add a 3D dispersion $\varepsilon_2(k_x, k_y, k_z)$ between the two magnetic sites with the very simple form of:

$$\mathcal{H}^0 = \varepsilon_2(k_x, k_y, k_z)\sigma_0^{spin}\sigma_x^{site} \tag{S14}$$

$$\varepsilon_2(k_x, k_y, k_z) = t_x \cos(k_x) + t_y \cos(k_y) + t_z \cos(k_z) \tag{S15}$$

and this last term is needed to prevent a site degeneration along the nodal line.

Using the analytical model described by equations (S1-S4), we calculate the band structure after fixing the model's parameters. For simplicity, the dispersion $\varepsilon(\boldsymbol{k})$ was set to zero; the values of other parameters are reported in the captions of the associated figures. The results are reported in Fig. S4 where we can see the effect of the three different momentum locking from the positive and negative values of the S$_x$, S$_y$ and S$_z$ components along the k-space directions. These results reproduce the spin–momentum locking presented in Fig. 4 of the main text, demonstrating that the 4×4 analytical model successfully captures this relativistic effect. The coordinates in the k-space are half of the R points reported in the Fig. 15 of the main text as R$_3$/2=$(-\pi/2, -\pi/2, \pi/2)$; R$_1$/2=$(\pi/2, -\pi/2, \pi/2)$; R$_2$/2=$(\pi/2, \pi/2, \pi/2)$ and R$_4$/2=$(-\pi/2, \pi/2, \pi/2)$. Using the analytical model described by Eqs. (S1)–(S5), we compute the band structure after fixing the model parameters. The corresponding results are shown in Fig. S4, where the effect of the three different spin-momentum lockings can be observed. These arise from the positive and negative values of the spin components S_x, S_y, and S_z along different directions. The coordinates in k-space correspond to the halfway of the R points reported in Fig. 15 of the main text, namely:

$$R_3/2 = \left(-\tfrac{\pi}{2}, -\tfrac{\pi}{2}, \tfrac{\pi}{2}\right), \quad R_1/2 = \left(\tfrac{\pi}{2}, -\tfrac{\pi}{2}, \tfrac{\pi}{2}\right), \quad R_2/2 = \left(\tfrac{\pi}{2}, \tfrac{\pi}{2}, \tfrac{\pi}{2}\right), \quad R_4/2 = \left(-\tfrac{\pi}{2}, \tfrac{\pi}{2}, \tfrac{\pi}{2}\right).$$

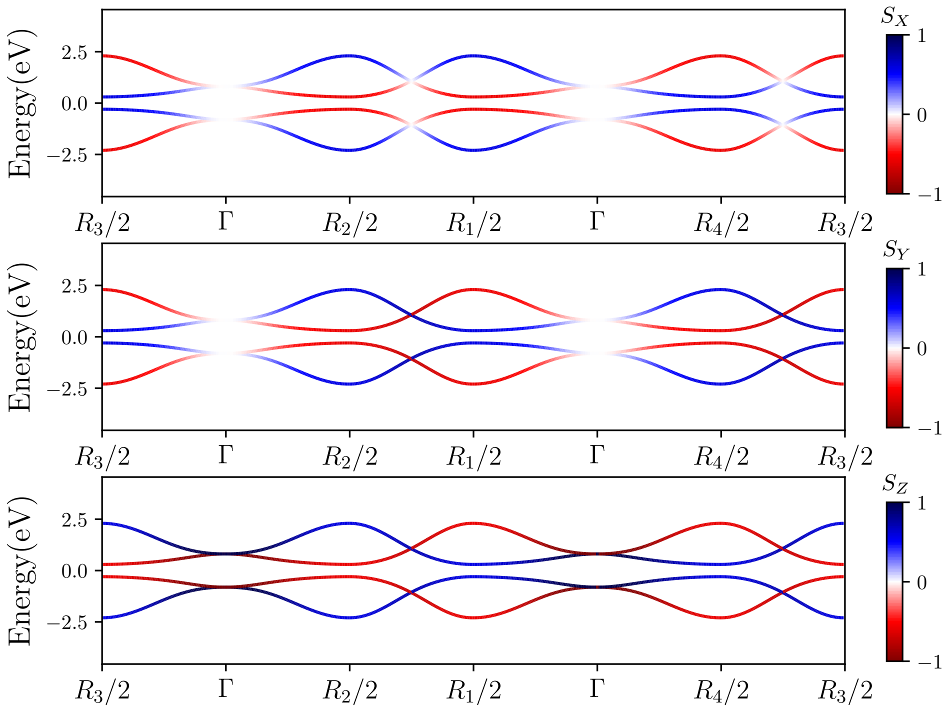

FIG. S4. Spin-resolved band structure for the S_x, S_y and S_z components. The numerical values used for the model are $t_{am} = 0.25$, $\Delta_z = \Delta_x$=Δ_y=0.75, $t_x = t_y = t_z = 0$ and $\alpha_x = \alpha_y = 0$.

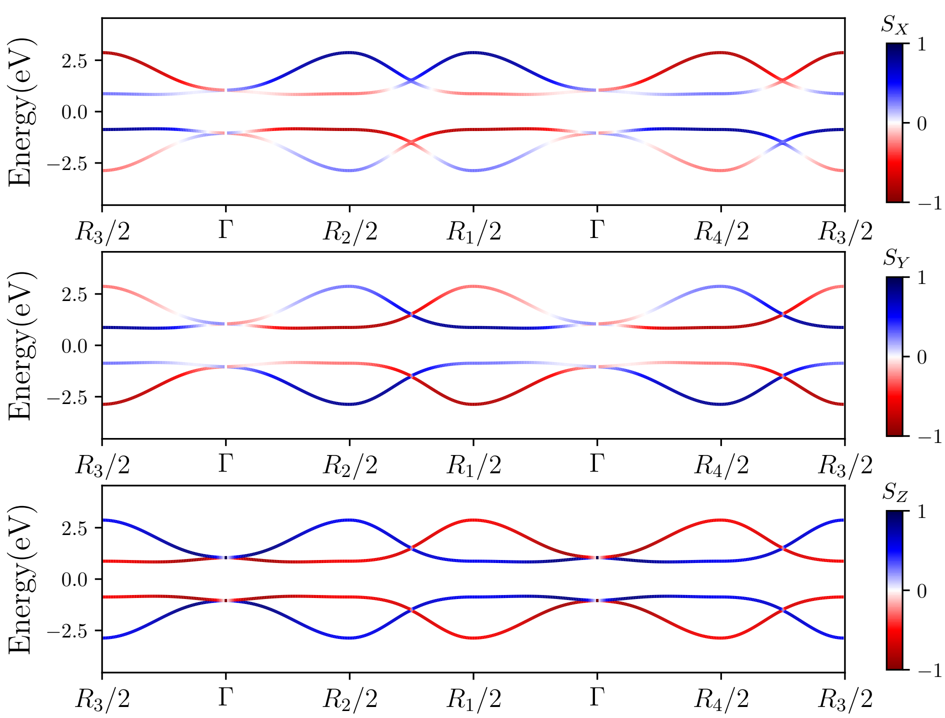

FIG. S5. Spin-resolved band structure for the S_x, S_y and S_z components. The numerical values used for the model are $t_{am} = 0.25$, $\Delta_z = \Delta_x$=Δ_y=1, $t_x = t_y = t_z = 0.2$ and $\alpha_x = \alpha_y = 0.5$.

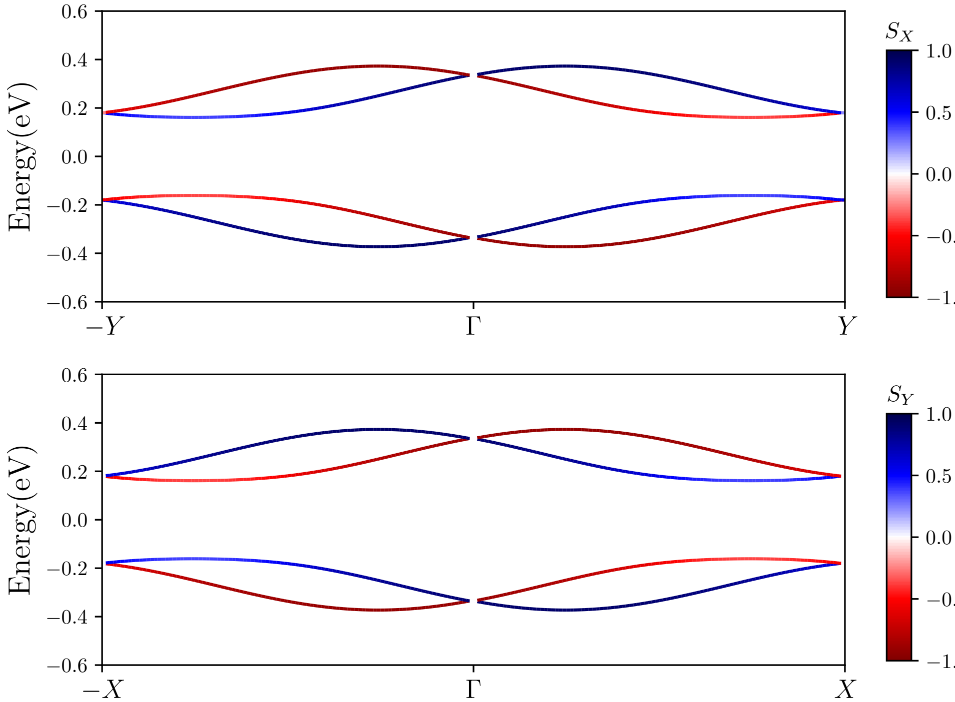

FIG. S6. Spin-momentum locking in the effective model. In the top panel is the spin-resolved band structure along the $-Y \to \Gamma \to Y$ path, and in the bottom panel is an analogous result for the $-X \to \Gamma \to X$ path. The weights correspond to the expectation value of the spin operator in the direction perpendicular to the path, as indicated by the colorbar title on the right. The numerical values used for the model are $t_{am} = 0.45$, $\Delta_z = \Delta_x$=Δ_y=0.15, $t_x = t_y = t_z = 0.2$ and $\alpha_x = \alpha_y = 0.1$.

We observe that midway between the points $R_1/2$ and $R_2/2$, the spin splitting vanishes for S_y and S_z due to the presence of the nodal plane. The same behavior occurs midway between $R_3/2$ and $R_4/2$. Other nodal planes are present in other directions, but they are not shown.

The next step is to add the Rashba-type spin-orbit coupling in the form of:

$$\mathcal{H}_R = (\alpha_y \sin(k_y)\sigma_x^{spin} - \alpha_x \sin(k_x)\sigma_y^{spin})\sigma_0^{site} \tag{S16}$$

The spin-resolved band structure is reported in Fig. S5. We observe that midway between the points $R_1/2$ and $R_2/2$, the spin splitting does not vanish for S_x, there is a finite polarization. The same behavior occurs midway between $R_3/2$ and $R_4/2$, however, the spin polarization has opposite sign, such that the vanishing magnetization is preserved. The same happens for S_y but along another direction.

Finally, in the Fig. S6, we report the Rashba effect along the nodal plane, plotting only the components that break the spin-momentum locking. The top panel Fig. S6 is the analog of 11(a) in the main text, where the S_x component is non-zero along the k-path from -Y to Γ and then to Y. The bottom panel of Fig. S6 is the analogue of Fig. 10(b) of the main text, where the S_y component is non-zero along the k-path from -X to Γ and then to X.

S6. MATRIX FORM OF THE TWO SPIN-MOMENTUM LOCKINGS HAMILTONIAN

We report the explicit matrix form of the simplified Hamiltonian for one orbital with two independent spin-momentum locking:

$$H^{\uparrow\uparrow} = \begin{pmatrix} \Delta_z + 4t_{am}^{3D} \sin k_x \sin k_z + 4t_{am}^{2D} \sin k_x \sin k_y & 0 \\ 0 & \Delta_z - 4t_{am}^{3D} \sin k_x \sin k_z - 4t_{am}^{2D} \sin k_x \sin k_y \end{pmatrix} \tag{S17}$$

and

$$H^{\downarrow\downarrow} = \begin{pmatrix} \Delta_z + 4t_{am}^{3D} \sin k_x \sin k_z + 4t_{am}^{2D} \sin k_x \sin k_y & 0 \\ 0 & \Delta_z - 4t_{am}^{3D} \sin k_x \sin k_z - 4t_{am}^{2D} \sin k_x \sin k_y \end{pmatrix} \tag{S18}$$

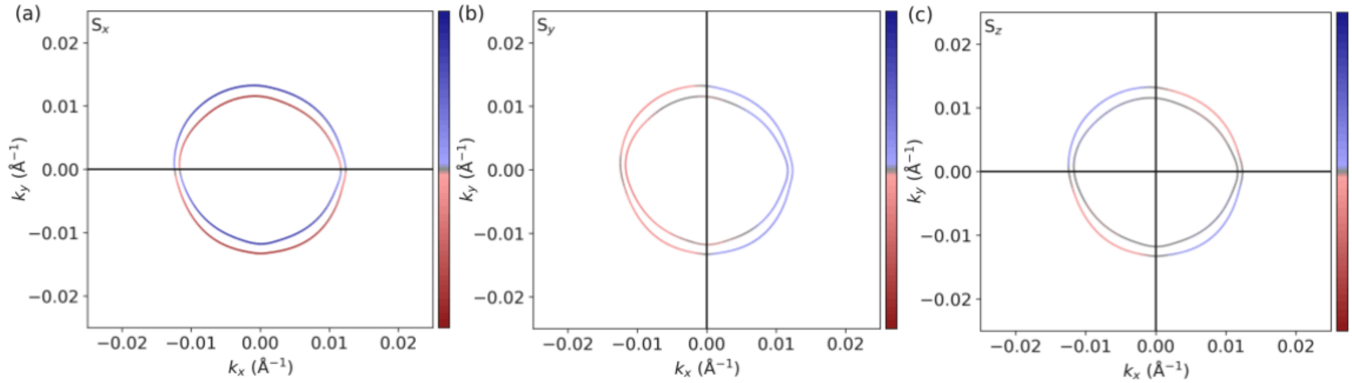

FIG. S7. Spin-resolved Fermi surface for the three spin components (a) S_x, (b) S_y, and (c) S_z of the A-centered Ca_2RuO_4 with ferroelectric distortions along the z-axis with Néel vector along the b-axis. The Fermi surface depicts a p_y-wave pattern for S_x and a p_x-wave pattern for S_y, while the S_z component still preserves its d_{xy} spin-momentum locking. The calculated Fermi surface accuracy was set to 0.011 Å^{-2}. In the 2D Fermi surfaces, the zero of the spin component is represented by the gray color. The Fermi surface was calculated at −0.3 eV relative to the Fermi level using an $11 \times 11 \times 4$ k-point grid.

S7. NODAL PLANES AND RASHBA EFFECT ON THE FERMI SURFACE

We consider the magnetic ground state of the A-centered structure, with the Néel vector oriented along the b axis and ferroelectric displacements along the z axis. The Rashba term has the form of:

$$\mathcal{H}_R = (\alpha_y \sin k_y \sigma_x^{spin} - \alpha_x \sin k_x \sigma_y^{spin})\sigma_0^{site} \tag{S19}$$

with S_x, which has a spin texture proportional to $\sin k_y$ and S_y, which has a spin texture proportional to $\sin k_x$. Upon including the effect of the altermagnetism, as discussed in the main text, the relativistic spin–momentum locking involves the p_y, p_x, and d_{xy} orbitals for the spin components S_x, S_y, and S_z, respectively. Fixing the energy at −0.3 eV relative to the valence-band maximum, we analyze the spin-resolved Fermi surface at k_z=0 in S7. The solid lines indicate symmetry-protected nodal planes across which the spin changes sign. We identify a single nodal plane for the S_x and S_y components, consistent with their p-wave character, and two nodal planes for the S_z component, reflecting its d-wave character. More in detail, the nodal plane for the S_x component is $k_y = 0$, which makes the spin–momentum locking p_y-wave, while the nodal plane for the S_y component is $k_x = 0$, which makes the spin–momentum locking p_x-wave. The S_z component has nodal planes at $k_x = 0$ and $k_y = 0$, which makes the spin–momentum locking d_{xy}-wave. These results are in agreement with the analysis presented in Table III of the main text.

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