Supplementary Information for

Efficient spin accumulation carried by slow relaxons in chiral tellurium

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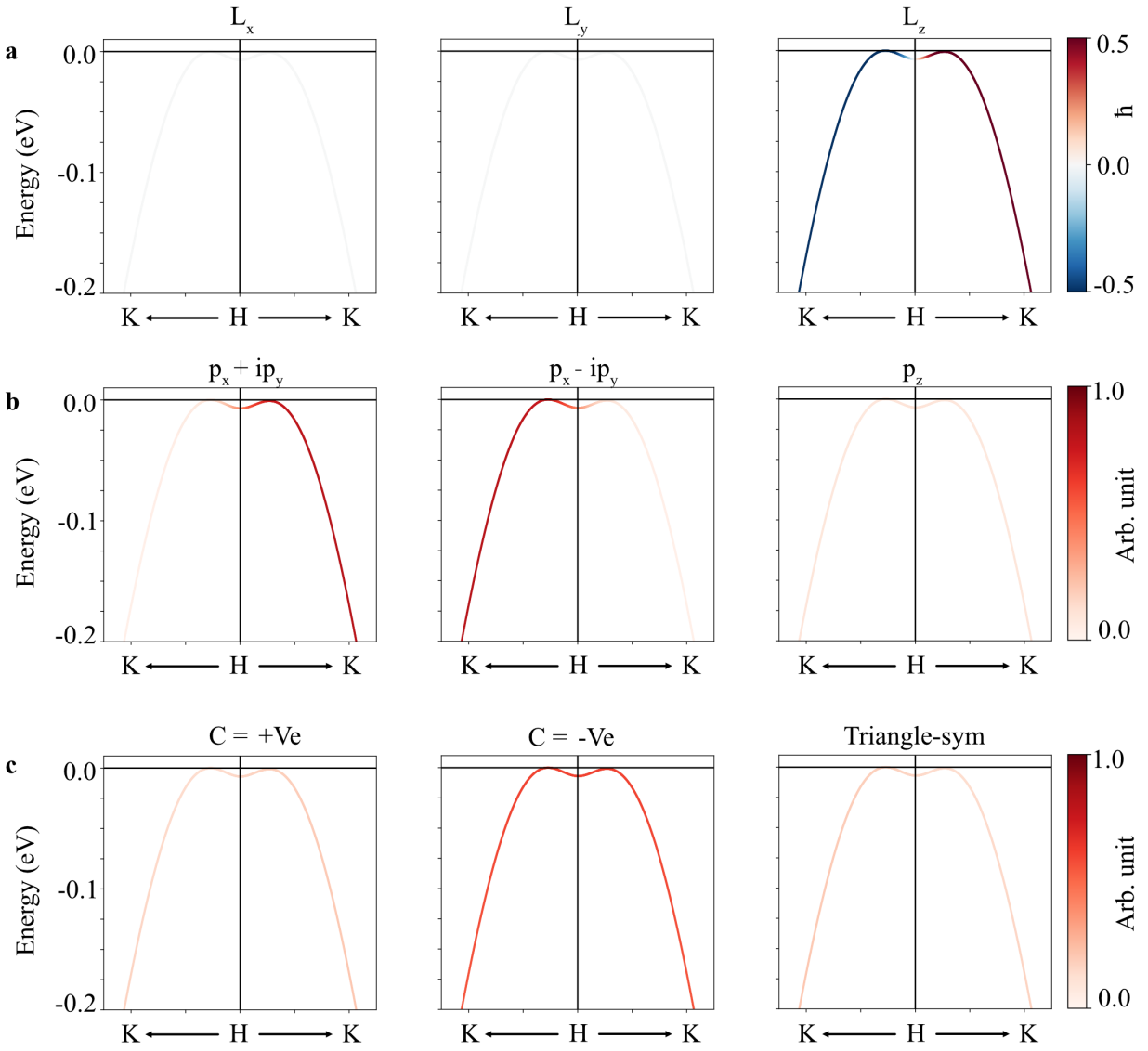

Supplementary figure 1: **Calculated valence band structure along the KHK line in the Brillouin zone in the left-handed Te.** Color shows **a** expectation value of the onsite orbital momentum, **b** wavefunction projection on atomic p-orbitals, and **c** projection on three sublattice states, where $\pm$ chirality corresponds to the state $(1, e^{\pm 2\pi i/3}, e^{\mp 2\pi i/3})/\sqrt{3}$ and the symmetric achiral state is $(1,1,1)/\sqrt{3}$.

Supplementary note 1

In Supplementary Fig. 1 we show calculated valence band structure along the KHK line in the Brillouin zone. Specifically, we separately identify chiral properties of the wavefunctions close to the low-doping hole pocket at the H point by projecting them onto different orbital and sublattice states. Panel **a** shows that the states are strongly polarized orbital momentum L_z. It is supported by panel **b**, which confirms that the right part of the band primarily are $p_x + ip_y$ states, whereas the left part are $p_x - ip_y$, since the p-orbital states are explicitly given in terms of $|L, L_z\rangle$ as:

$$\begin{aligned} |1,1\rangle &= -\frac{1}{\sqrt{2}}(p_x + ip_y), \\ |1,0\rangle &= p_z, \\ |1,-1\rangle &= \frac{1}{\sqrt{2}}(p_x - ip_y). \end{aligned} \tag{S1}$$

To obtain the orbital polarization separately from spin polarization, we calculate the expectation values of $\hat{L}_z = |1,1\rangle\langle 1,1| - |1,-1\rangle\langle 1,-1|$, which leads to the expression for orbital polarization in the main text. Since the strong SOC in Te ($\lambda \approx 0.45$ eV) splits the p-orbital states into a quadruplet and a doublet ($|J, J_z\rangle$ with $J = 3/2$ and $J = 1/2$) the partial spin and orbital polarization of states is likely a result of the mixing of oppositely-polarized states within the higher-energy quadruplet:

$$\begin{aligned} |\frac{3}{2},\frac{3}{2}\rangle &= -\frac{1}{\sqrt{2}}(p_x + ip_y)\uparrow, \\ |\frac{3}{2},\frac{1}{2}\rangle &= -\sqrt{\frac{1}{6}}(p_x + ip_y)\downarrow + \sqrt{\frac{2}{3}}p_z\uparrow, \\ |\frac{3}{2},-\frac{1}{2}\rangle &= \sqrt{\frac{2}{3}}p_z\downarrow + \sqrt{\frac{1}{6}}(p_x - ip_y)\uparrow, \\ |\frac{3}{2},-\frac{3}{2}\rangle &= \frac{1}{\sqrt{2}}(p_x - ip_y)\downarrow, \\ |\frac{1}{2},\frac{1}{2}\rangle &= -\frac{1}{\sqrt{3}}(p_x + ip_y)\downarrow - \frac{1}{\sqrt{3}}p_z\uparrow, \\ |\frac{1}{2},-\frac{1}{2}\rangle &= \frac{1}{\sqrt{3}}p_z\downarrow - \frac{1}{\sqrt{3}}(p_x - ip_y)\uparrow. \end{aligned} \tag{S2}$$

Note that these atomic wavefunctions naturally serve as the basis states in the PAOFLOW package, allowing us to calculate both spin and onsite orbital components by using $\hat{J}_z = \left(\hat{S}_z + \hat{L}_z\right) = \sum_{J,J_z} J_z\ |J, J_z\rangle\langle J, J_z|$.

Panel **c** shows that left-handed Te also imposes significant polarization in sublattice degrees of freedom. When the three sublattice states are written in the chiral basis, wherein $\pm$ chirality corresponds to the state $(1, e^{\pm 2\pi i/3}, e^{\mp 2\pi i/3})/\sqrt{3}$ and symmetric achiral state to $(1,1,1)/\sqrt{3}$, all the states close to the H point belong to the states with negative sublattice chirality.