

**Quantum anomalous Hall effect in nonmagnetic bismuth monolayer
with a high Chern number**

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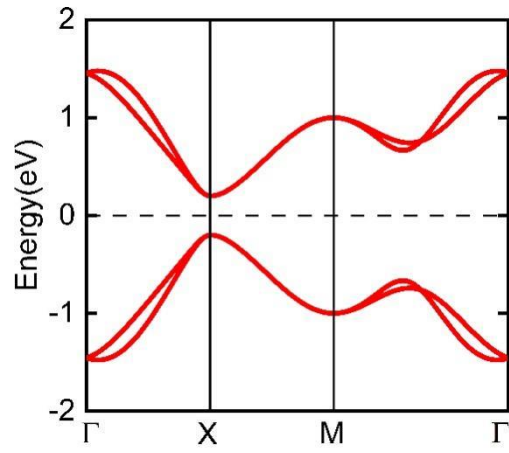


Figure S1. Band structure of the four-band tight-binding model for the parameters as $a = 0.1$ eV, $t = 0.3$ eV, $\lambda = 0.25$ eV and $\lambda_{\text{ROT}} = 0.2$ eV.

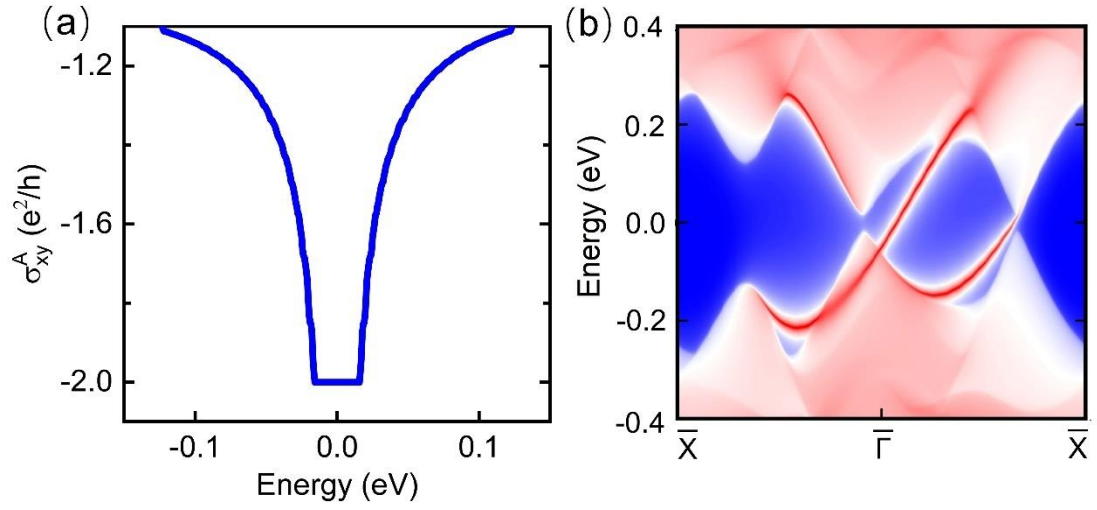


Figure S2. (a) Energy dependence of the anomalous Hall conductivity σ_{xy}^A and (b) the corresponding edge spectrum of TB model under the irradiation of right-handed CPL with light intensity $eA/\hbar = 0.15 \text{ \AA}^{-1}$, indicating that the Chern number is -2.

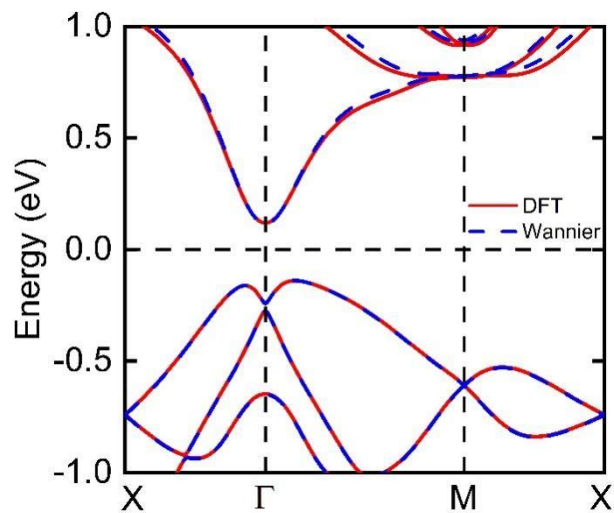


Figure S3. Comparison between the first-principles and Wannier fitting band structures.

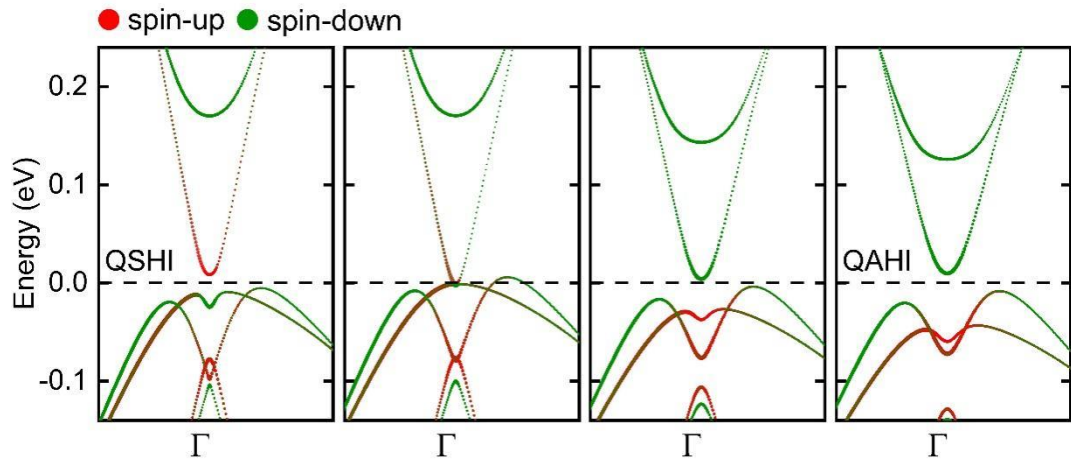


Figure S4. Band structure evolution for T-Bi under the irradiation of left-handed CPL with light intensity eA/\hbar of 0.35, 0.37, 0.4 and 0.42 \AA^{-1} , respectively. The red and green dots denote spin-up and spin-down component of bismuth, respectively.

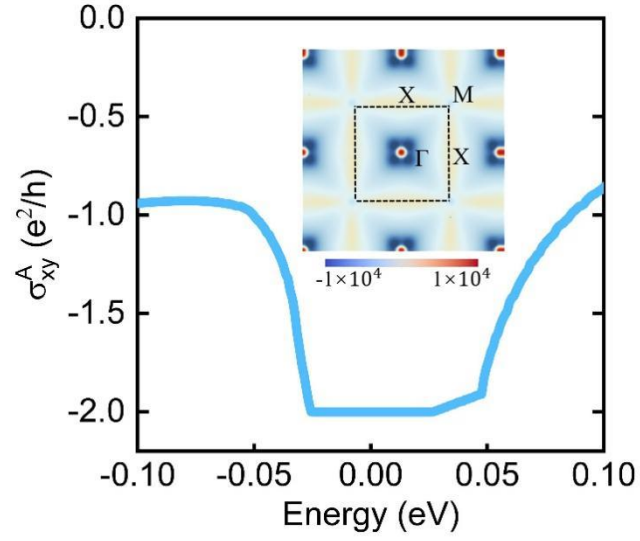


Figure S5. Energy dependence of the anomalous Hall conductivity σ_{xy}^A under the irradiation of right-handed CPL with light intensity $eA/\hbar = 0.5 \text{ \AA}^{-1}$, revealing a quantized value with Chern number $C = -2$ in the gapped regime. The inset shows the reciprocal-space distribution of Berry curvatures within the gapped regime.

Note S1. Floquet Theory

We first consider it under the irradiation of an external time-dependent circularly polarized light:

$$A(\tau) = A[\eta \sin(\omega\tau)e_1 + \cos(\omega\tau)e_2] \quad (\text{S1})$$

where ω is the frequency and A is the amplitude of the light, $\eta = \pm 1$ represent the chirality of the circularly polarized light. The Peierls substitution $k \rightarrow k + \frac{e}{\hbar} A(\tau)$ is used to take the effect of light into account in the Hamiltonian. The time-dependent tight-binding Hamiltonian will be written as

$$H(\vec{k}, \tau) = \sum_{m,n} \sum_j t_j^{mn}(\tau) e^{i\vec{k} \cdot \vec{R}_j} c_m^+(\vec{k}, \tau) c_n(\vec{k}, \tau) \quad (\text{S2})$$

where τ is the time, \vec{R}_j is the lattice vector and (m, n) is Wannier orbital index. The vector potential is coupled to the Hamiltonian through the minimal coupling of $t_j^{mn}(\tau) = t_j^{mn} e^{i\frac{e}{\hbar} \vec{A}(\tau) \cdot \vec{d}_j^{mn}}$, where \vec{d}_j^{mn} is the position vector between Wannier orbital m in the 0-cell and Wannier orbital n in the j -cell. Taking into account the lattice and time translation invariance, the time-dependent Hamiltonian can be effectively treated with Floquet theorem by performing dual the Fourier transformation [1-2]. Then an effective static Hamiltonian in the frequency and momentum space can be obtained

$$H_F(\vec{k}, \omega) = \sum_{m,n} \sum_{\alpha,\beta} [H_{\alpha-\beta}^{mn}(\vec{k}) + \alpha \hbar \omega \delta_{mn} \delta_{\alpha\beta}] c_{\alpha m}^+(\vec{k}) c_{\beta n}(\vec{k}) \quad (\text{S3})$$

where (α, β) is the Floquet index ranging from $-\infty$ to $+\infty$, $c_{\alpha m}^+$ ($c_{\beta n}$) is creation (annihilation) operator in the Floquet-Bloch picture, and the matrix element $H_{\alpha-\beta}^{mn}(\vec{k}, \omega)$ is

$$H_{\alpha-\beta}^{mn}(\vec{k}, \omega) = \sum_j e^{i\vec{k} \cdot \vec{R}_j} \left(\frac{1}{T} \int_0^T t_j^{mn} \times e^{i\frac{e}{\hbar} \vec{A}(\tau) \cdot \vec{d}_j^{mn}} e^{i(\alpha-\beta)\omega\tau} dt \right). \quad (\text{S4})$$

This Floquet-Bloch Hamiltonian can be written as the block matrix form

$$H_F(\bar{k}, \omega) = \begin{pmatrix} \ddots & \vdots & \vdots & \vdots & \ddots \\ \cdots & H_0 - \hbar\omega & H_{-1} & H_{-2} & \cdots \\ \cdots & H_1 & H_0 & H_{-1} & \cdots \\ \cdots & H_2 & H_1 & H_0 + \hbar\omega & \cdots \\ \ddots & \vdots & \vdots & \vdots & \ddots \end{pmatrix} \quad (\text{S5})$$

In this work, we use $q = 1$ for the Fourier transformation, model analysis and DFT calculations. The infinite Floquet sidebands can reduce to a finite dimension by including only a few lowest order photon processes of absorption or emission. Here, limited to a virtual photon process, one can distinctly observe the Floquet-Bloch band structures under CPL. Each matrix element in Eq. (S5) can be obtained by averaging the time-dependent Hamiltonian.

Reference

- [1] A. Gómez-León and G. Platero, Phys. Rev. Lett. 110, 200403 (2013).
- [2] K. F. Milfeld and R. E. Wyatt, Phys. Rev. A 27, 72 (1983).