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Floquet engineering of magnetism in topological insulator thin films

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Floquet engineering of magnetism in topological insulator thin films

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Abstract

Dynamic manipulation of magnetism in topological materials is demonstrated here via a Floquet engineering approach using circularly polarized light. Increasing the strength of the laser field, besides the expected topological phase transition (PT), the magnetically doped topological insulator thin film also undergoes a magnetic PT from ferromagnetism to paramagnetism, whose critical behavior strongly depends on the quantum quenching. In sharp contrast to the equilibrium case, the non-equilibrium Curie temperatures vary for different time scale and experimental setup, not all relying on change of topology. Our discoveries deepen the understanding of the relationship between topology and magnetism in the non-equilibrium regime and extend optoelectronic device applications to topological materials.

1. Introduction

The interplay between magnetism and topological state has attracted considerable attention in recent studies. Many exotic topological particles and macroscopic quantum phenomena have been realized, including Weyl fermions [1–7], antiferromagnetic Dirac fermions [8, 9], quantum anomalous Hall (QAH) effect [10–16], antiferromagnetic topological insulators (TI) [17, 18], and axion insulators [18–23]. In real materials, the emergence of magnetism breaks the time reversal symmetry and the exchange coupling can drive the system into a new topological phase. For example, the long-range ferromagnetic (FM) order can be formed by doping TI films with magnetic ions [10, 24–27], the Zeeman coupling splits degenerate bands and drives TI thin film to be a Chern insulator with the quantized edge conductance [11–15]. Meanwhile, in turn, these topological bands, both in bulk and on the surface, have the ability to form or enhance long-range ferromagnetism [10, 24, 25, 27]. Therefore, from a practical point of view, finding an efficient way to manipulate magnetic and topological orders will trigger intense research in the fields of spintronics and quantum information processing. The electric field-controlled ferromagnetism has been proposed theoretically [28] and achieved experimentally in a magnetically doped TI (MDTI) thin film [29], which is possible to be used in spintronic devices [28].

Different from conventional manipulations of topology and magnetism in equilibrium, Floquet engineering via ultrafast optical driving provides a new possibility to tune electronic properties of host

material dynamically in non-equilibrium [30–35]. The strong interaction of photons with charges, spins, and angular momenta can drive the system to a steady Floquet state, dramatically changing Floquet band structure and charge population [30, 32, 33]. This approach provides a new degree of freedom to manipulate the magnetic order [36, 37] and novel topological states [30, 31, 38–44]. By using Floquet engineering, semiconductor quantum wells [45–47] and two-dimensional (2D) materials [41, 48] can be driven to behave as Floquet TIs. Light-induced Hall conductance is predicted and observed in graphene, pointing towards a topological origin [49–56].

Herein, in the framework of the Floquet theorem, we study the electronic and magnetic properties of periodically driven TI and MDTI thin films. Under circularly polarized light (CPL), Floquet bands of steady states can be well-tuned, resulting in a photon-induced topological phase transition (PT). Furthermore, we develop a Floquet linear-response theory to calculate the magnetic susceptibility in MDTI thin films. We find that CPL can reduce its magnetic susceptibility, driving FM thin films to be paramagnetic. The decreasing behavior of the magnetic PT strongly depends on quantum quenching and dissipation. For example, in contrast to that on long time scale, the magnetic susceptibility decreases more quickly in a short time scale with the change of the laser field, resulting in a lower Curie temperature (T_C) correspondingly. Our work provides a new way to manipulate topological and magnetic properties in MDTI thin films via an optical approach and our predictions can be measured by magneto-optic and transport experiments.

2. Method

Under CPL, a non-equilibrium Floquet state is possible to be formed in quantum materials [31, 39, 40, 54]. In the Floquet theory [57–59], we can solve the time-dependent Schrödinger equation in the Hilbert space \mathcal{H} via mapping it to a time-independent infinite eigenvalue problem in an extended Hilbert space $\mathcal{H} \otimes \mathcal{L}_T$ [60, 61], where \mathcal{L}_T represents the space of ‘multi-photon-dressed’ states. Finally, the resulting states are the Floquet bands, whose occupations strongly depend on the quantum quenching and dissipation details.

The band structure and charge distribution in the initial equilibrium are shown in figure 1(a). If we switch on the laser suddenly and measure the physical observable in a short time (such as magneto-optical Kerr rotation angle [62], schematically shown in figure 1(d)), the time scale for this system to reach a steady state is short compared to the period of the laser [50, 51]. In this way (see figure 1(b)), electrons tend to stay in their original bands, and the population of the quenched Floquet system is mainly determined by the overlapping matrix between final and original states [61]. This approach is named the Sudden Approximation [50, 51]. On the other limit (see figure 1(c)), if the measurement is performed over a long time (such as anomalous Hall effect in ultrafast transport [55, 56], schematically shown in figure 1(e)), excited electrons are relaxed and the population of the Floquet state behaves like a Floquet Fermi–Dirac distribution with effective chemical potentials for electrons and holes [47, 63]. In this work, we calculate magnetic properties, especially the magnetic susceptibilities for MDTI thin films in these two extreme situations by the Floquet linear response theory [49, 51, 61, 64, 65].

3. Results

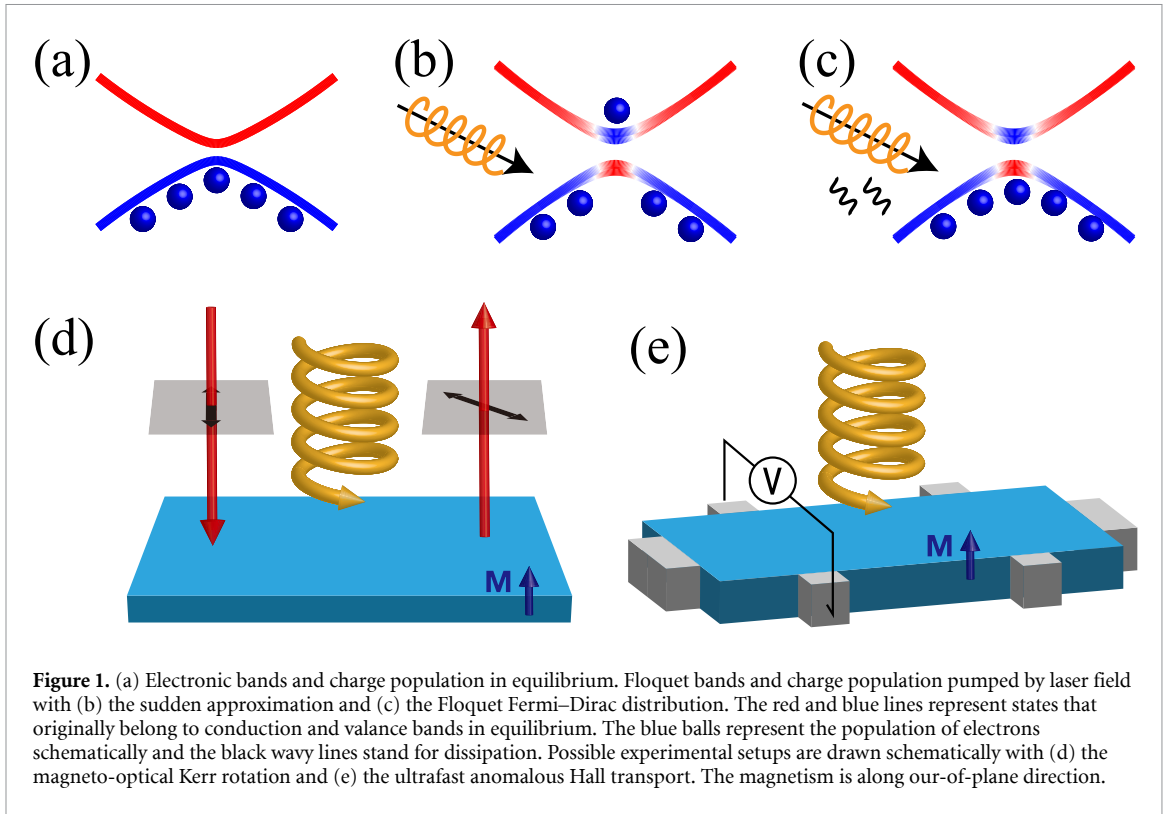
3.1. Effective model

To study MDTI thin films under CPL, we start from the 2D effective Hamiltonian for TI thin films [10, 28, 66, 67],

$$H_0(\mathbf{k}) = \epsilon(\mathbf{k}) + v_F k_y \sigma_1 \otimes \tau_3 - v_F k_x \sigma_2 \otimes \tau_3 + m(\mathbf{k}) \cdot I \otimes \tau_1 \quad (1)$$

written in the basis of $\{|u \uparrow\rangle, |u \downarrow\rangle, |d \uparrow\rangle, |d \downarrow\rangle\}$, \uparrow and \downarrow stand for the spin degree of freedom, $|u\rangle$ and $|d\rangle$ are surface states on the upper and down surfaces of the TI thin film. σ_i and τ_i ($i = 1, 2, 3$) are the Pauli matrices for spin and orbital, $k_{x,y}$ is the momentum in reciprocal space, v_F is the Fermi velocity, $\epsilon(\mathbf{k}) = \epsilon_0 + \epsilon_2(k_x^2 + k_y^2)$ is the energy shift, and $m(\mathbf{k}) = m_0 + m_2(k_x^2 + k_y^2)$ is the coupling between $|u\rangle$ and $|d\rangle$. Due to the inter-surface coupling, a finite gap opens around the Γ point and topological properties of thin films are determined by the sign of $m_0 m_2$ [10, 61, 68].

We include the effect of CPL by the Peierls substitution $\mathbf{k} \rightarrow \mathbf{k} - e\mathbf{A}(t)$, where $\mathbf{A}(t)$ is the laser amplitude and e is the electron charge. $\mathbf{A}(t) = A(\cos \omega t, \sin \omega t, 0)$ is for right-hand CPL, ω is the frequency with the photon energy of 3 eV. Time-independent infinite Floquet Hamiltonian $H_F(\mathbf{k})$ can be obtained in the framework of the Floquet theory [61]. By diagonalizing $H_F(\mathbf{k})$ with a cut-off on the frequency domain, we obtain the Floquet band structures [61]. In the high-frequency limit ($\hbar\omega \gg W$, \hbar is the reduced Planck constant and W is the bandwidth of the thin film), the low order expansion [30, 31, 60, 69] can be employed to reduce the infinite $H_F(\mathbf{k})$ to an effective Hamiltonian $H_{eff}(\mathbf{k})$ with the form of:



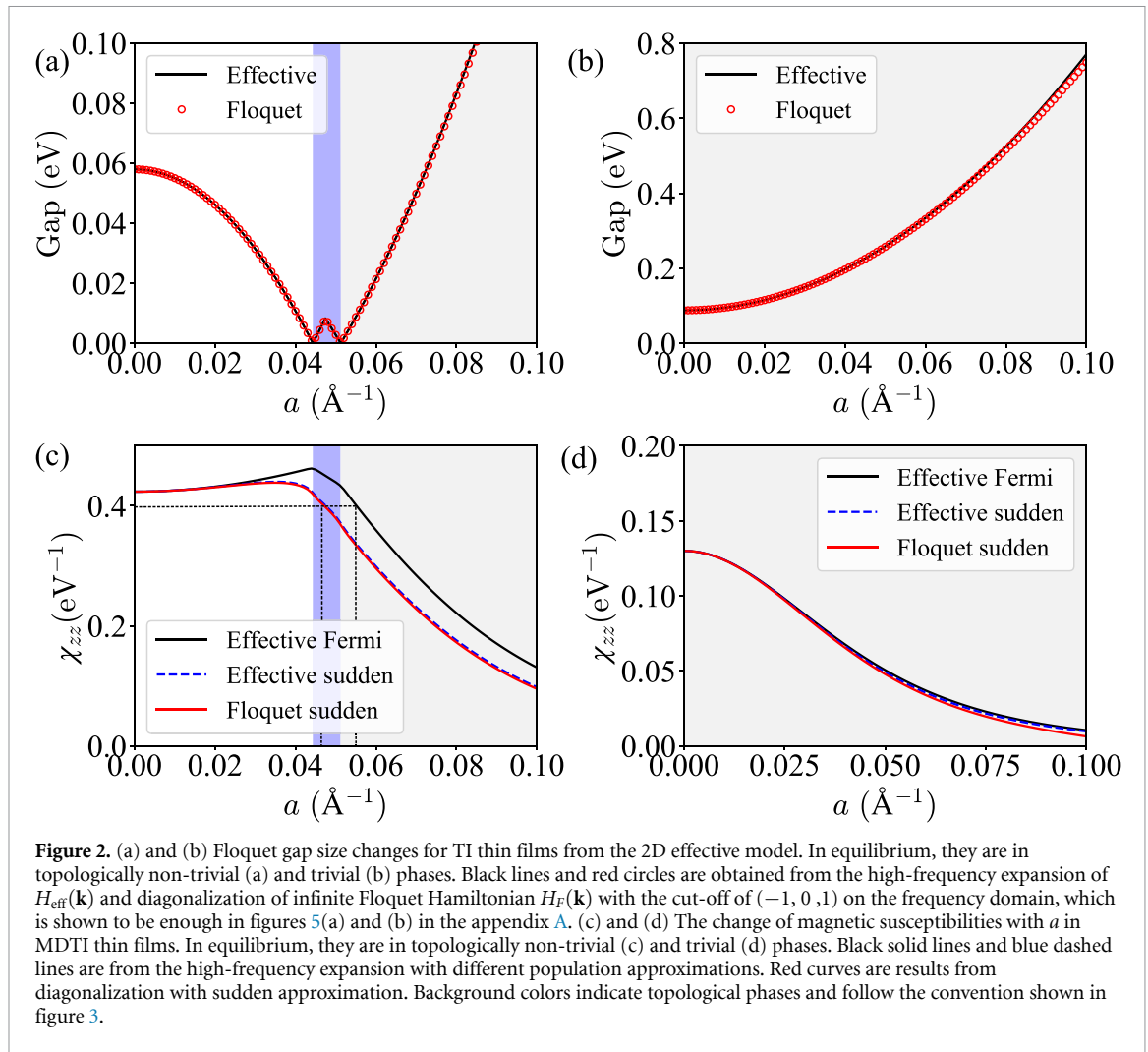
$$H_{\text{eff}}(\mathbf{k}) = H_0(\mathbf{k}) + \epsilon_2 a^2 + m_2 a^2 \cdot \mathbf{I} \otimes \tau_1 + \frac{a^2}{\hbar\omega} v_F^2 \cdot \sigma_3 \otimes \tau_0 - 2 \frac{a^2}{\hbar\omega} v_F m_2 (k_x \sigma_1 + k_y \sigma_2) \otimes \tau_2. \quad (2)$$

Here $a = eA$ is the effective laser amplitude with unit \AA^{-1} . This Hamiltonian has the same dimension as the original $H_0(\mathbf{k})$. Its second term corrects ϵ_0 (see equation (1)) and results in the total energy shift. The third term modifies $m_0 \rightarrow m_0 + m_2 a^2$ and induces a topological PT when the laser amplitude is large enough. The light intensity that triggers the topological PT in our system is around 10^9 – 10^{11} W cm^{-2} , which is experimentally feasible [43, 70, 71]. Interestingly, these terms do not depend on the laser frequency but rely on laser amplitude only. Moreover, CPL introduces two new terms at the end of equation (2), which are proportional to $a^2/\hbar\omega$ and smaller compared to correction terms under the high-frequency limit. The first one is momentum-independent and acts like the exchange coupling due to time reversal symmetry breaking. The second one is momentum-dependent and has a negligible effect for electronic bands around Γ point. To sum up, the change of band structures for TI thin films under CPL is dominated by correction terms and slightly modified by the frequency-dependent terms.

Numerical results demonstrate the analysis above is correct. In figures 2(a) and (b), we show the calculated direct Floquet gap size for TI thin films from the 2D effective model when fixing the frequency and changing the amplitude of the driving laser. In equilibrium, we use parameters shown in [28] for $H_0(\mathbf{k})$ where 4 quintuple layer (QL) TI thin film is in the quantum spin Hall (QSH) phase ($m_0 m_2 < 0$) and 3QL thin film is a normal insulator ($m_0 m_2 > 0$). When the laser is turned on, their electronic band structures are modified. Two curves with different colors illustrate the gap size in the Floquet band structures obtained through the high-frequency expansion (black line) and direct diagonalization respectively (red circles). The good agreement between the two methods suggests that current parameter settings reach the high-frequency limit.

When we increase the laser amplitude a for 4QL thin film, its Floquet gap at the Γ point decreases at first, undergoes two gap-closing processes, and finally increases again (see figure 2(a)). Such evolution indicates topological PTs from a NIB phase to a QAH phase, and finally to a NIA phase. Here, NIB is a state with a Floquet band structure similar to the QSH insulator without topological protection [28]. NIA stands for a normal insulator. The QAH phase (blue region) mainly originates from the light-induced exchange coupling in TI film. Since the exchange term is proportional to $a^2/\hbar\omega$, its phase region is very narrow in the high frequency. In contrast to the 4QL thin film, we observe no gap-closing process as the increase of light amplitude for 3QL film, meaning that it persists in NIA phase (see figure 2(b)).

When TI thin films are doped with magnetic ions, such as Cr [11, 26] and V [15] atoms, the long-range FM order can be formed in equilibrium. If we keep MDTI thin films insulating, the ferromagnetism is



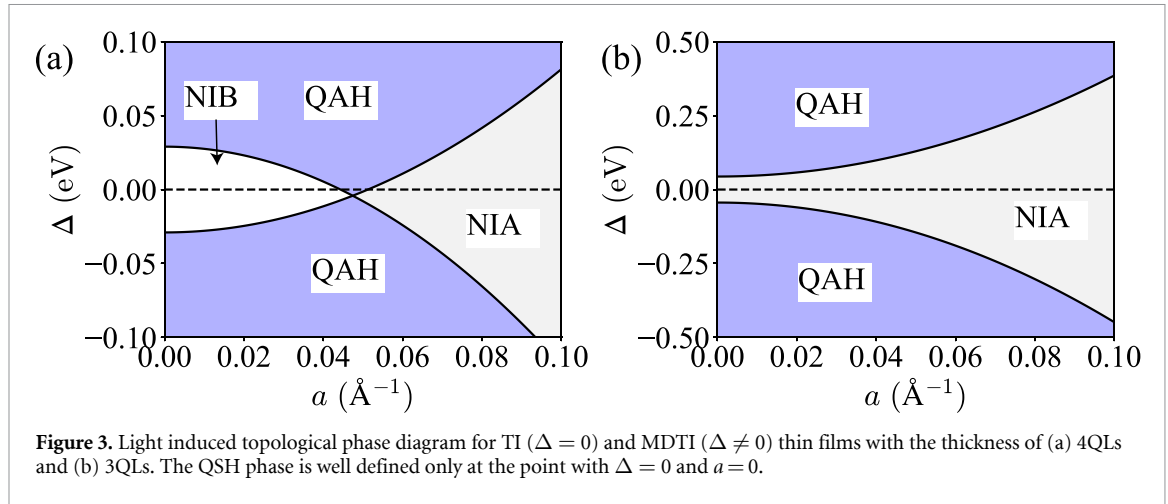
induced by the van Vleck mechanism [10, 26, 27, 61]. The low energy electronic structure around the Γ point can be described by the Hamiltonian: $H_0^{FM}(\mathbf{k}) = H_0(\mathbf{k}) + \Delta \cdot \sigma_3 \otimes I$, where Δ is the Zeeman term arising from FM order and is determined by the density of dopants and the magnetic susceptibility [10]. Interestingly, MDTI thin films undergo a FM to paramagnetic transition via changing dopant densities, accompanied by a topological PT [27]. Currently, the experimental preparation [11–13] and manipulation techniques [29] of MDTI thin films are relatively mature, and ultrafast laser pulse can be used to drive them far from equilibrium [39, 40].

We can fix the density of dopants in MDTI thin films to guarantee their equilibrium states to be FM for 4QLs and paramagnetic for 3QLs, and then apply CPL to drive the systems to the Floquet states. Thus the change of magnetic properties induced by the laser field is determined by the magnetic susceptibilities of electrons χ_{zz}^{Floq} , and it is calculated for Floquet states by the Kubo linear-response theory [61]:

$$\chi_{zz}^{\text{Floq}} = \sum_{\alpha \in \text{FBZ}, \beta, k} 2f_{\alpha} \frac{|\langle \langle \phi_{\alpha}(t) | s_z | \phi_{\beta}(t) \rangle \rangle|^2}{\epsilon_{\beta} - \epsilon_{\alpha}} \quad (3)$$

where α and β label the Floquet states, $\phi_{\alpha}(t)$ is the Floquet wavefunction with quasienergy ϵ_{α} , f_{α} is its population, s_z is the spin operator along z direction, and Floquet Brillouin zone (FBZ) [61]. In non-equilibrium, the occupation of Floquet states strongly depends on the frequency and amplitude of the driving laser [56, 72] as well as dissipation to a bath [47]. Here we only consider steady Floquet states formed in two limited situations with high frequency.

As shown in figures 2(c) and (d), the magnetic PT points change with different quenching schemes when the light intensity increases, although the trend of their variation χ_{zz}^{Floq} behaves similarly. For the 4QL MDTI thin film, the magnetic susceptibility increases slowly in the NIB phase and drops quickly in the QAH and NIA phases. When χ_{zz}^{Floq} reaches the critical value resulting in a magnetic PT, we observe different critical points a_c^{FM} for different quenching schemes (marked by a black dashed line in figure 2(c)). Compared with



T_C in equilibrium, CPL decreases its value by the Floquet engineering. Such decreasing tendency is mainly contributed by the change of gap size and matrix elements shown in equation (3).

In the regime of the sudden approximation, the critical value a_c^{FM} is smaller than that in the Floquet Fermi–Dirac distribution. Furthermore, a_c^{FM} in the two kinds of quenched schemes do not have the same value as the critical amplitude a_c^T for topological PT. Therefore, we conclude that, besides the modulation of energy levels, the light also changes the population of Floquet states, which plays an essential role in determining the magnetic behavior. This population dependence is unique for non-equilibrium states in contrast to previous studies [27, 28]. For 3QLs (see figure 2(d)), the decreasing behavior of χ_{zz}^{Floq} is observed but the entire system remains in the paramagnetic phase.

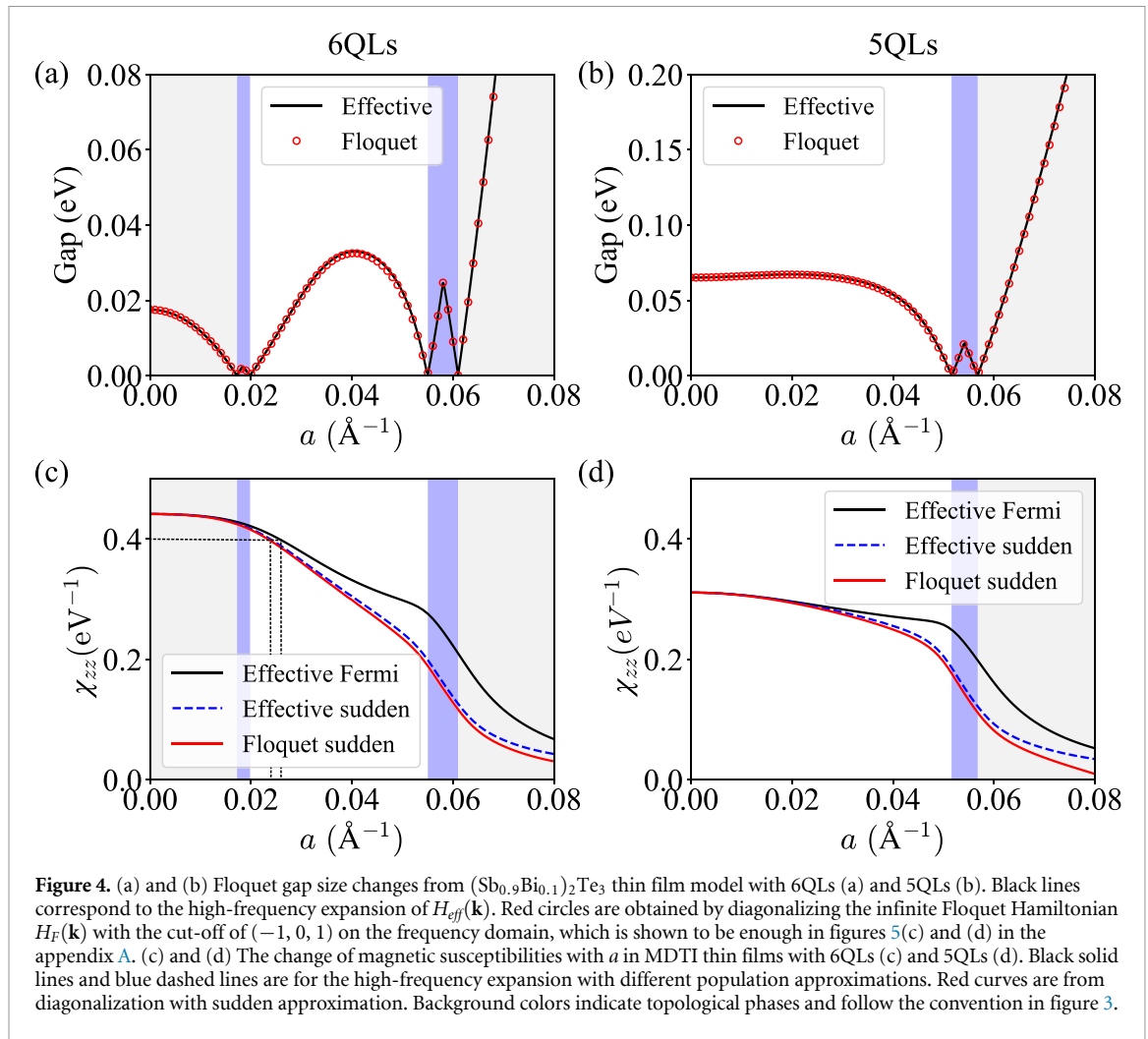
The topological phase diagram is shown in figure 3 for MDTI thin films, whose results are from the effective model considering the FM exchange coupling and the influence of CPL. If the intrinsic system is initially in QSH state (e.g. 4QLs), there are three phase regions as shown in figure 3(a), including a QAH phase and two trivial insulating phases NIA and NIB. The asymmetric feature with respect to Δ is due to the additional exchange terms induced by the driving laser (see equation (2)). For an initial state without topology (e.g. 3QLs), the phase diagram is much simpler with only transitions from the normal insulator to QAH state when Δ is large enough. We need to point out that under the aforementioned laser intensity, the system is possible to generate a static magnetic moment due to the nonlinear Edelstein effect [73]. However, it only shifts the critical points of the PT, rather than changing the qualitative conclusions.

3.2. Thin film model

The above discussions are based on the effective Hamiltonian. It is a relatively simple model to give us a clear physical picture of light-induced magnetic and topological PTs, but cannot predict all details precisely without including the contributions from quantum well states. To confirm the validity of the picture and describe a more realistic situation in experiments, we choose $(\text{Sb}_{0.9}\text{Bi}_{0.1})_2\text{Te}_3$ as an example, which has been fabricated in previous experiments [29, 66]. We consider its three-dimensional bulk Hamiltonian in a thin film construction with thickness d [61]. The confinement in the z direction quantizes the momentum on this axis. The contribution from QWSs can be fully considered in the thin film model [28, 68, 74, 75]. In our equilibrium calculations, $(\text{Sb}_{0.9}\text{Bi}_{0.1})_2\text{Te}_3$ thin film of 6QL is topologically trivial and its 5QL thin film is a QSH insulator.

To understand the effect induced by CPL in the thin film model, we do similar calculations as those by using the effective Hamiltonian. Figure 4 shows direct Floquet gap sizes and magnetic susceptibilities for $(\text{Sb}_{0.9}\text{Bi}_{0.1})_2\text{Te}_3$ thin films under CPL. Similar to results from the simple model, light-induced topological and magnetic PTs exist in intrinsic and doped thin films. A more complicated topological phase diagram is observed for 6QLs when a increases. They go through phases from NIA to QAH, to NIB, to QAH again, and finally enter a NIA phase. Such oscillatory crossover behaviour is dominated by quantum well states [61] and cannot be captured by the simple 2D effective model. For the 5QL thin film, the topological PT driven by the light is similar to that shown in figure 2(a), suggesting its topological characters can be well described by the 2D effective Hamiltonian.

When magnetic ions are doped at the density used in figure 2, the MDTI thin film of 6QL becomes FM while 5QLs do not, indicating that the 2D effective model is not well suited to describe the realistic magnetic properties for the 5QL thin film. The magnetic susceptibilities of the Floquet states decrease with the increase of light intensity and the slopes change differently in different topological phases. a_c^{FM} in magnetic PTs also



depends on the quantum quenching and does not correspond directly to the critical value of the topological PT. These results obtained from realistic thin film models are consistent with the conclusions from the 2D effective model, confirming the validity of our physical picture.

4. Conclusion

Using the Floquet theory, we study topological and magnetic properties for TI and MDTI thin films under CPL. In the high-frequency limit, topological PTs can be induced in TI thin films by increasing the intensity of the driving laser. Applying the same setup to MDTI thin films, we find magnetic PTs from FM to paramagnetic phases and their critical behaviors depend on the quantum quenches that influence the non-equilibrium charge population. Based on previous discussions using the Floquet-Green's function method [72], we argue that the high-frequency approximation is reasonable in a considerably large frequency regime, even when $\hbar\omega$ is comparable to W . On the other hand, when $\hbar\omega$ is smaller than W with the resonant photon absorption, χ_{zz}^{Floq} still behaves similarly to the non-resonant case except in photon absorption regions [61]. Thus, we are convinced that our conclusion could apply in a large parameter regime.

The phenomena proposed in this work are possible to be observed by different experimental setups. In pump-probe detection of the polar Kerr-rotation angle [62], the population effect could be described by the sudden approximation and the change of magnetism could be measured. Meanwhile, the Floquet Fermi-Dirac distribution is a good approximation for the transport measurement, such as the ultrafast anomalous Hall effect by laser-triggered photoconductive switches [55, 56]. In fact, due to the high photon energy and electric field strength required in our system, resonant interband absorption may occur and damage the sample during the experiment. However, some studies have shown that Floquet states can survive despite resonant absorption [43, 71]. We expect that light-driven topological and magnetic PTs can be realized in TI and MDTI thin films.

Data availability statement

All data that support the findings of this study are included within the article (and any supplementary files).

Acknowledgment

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Appendix A. Floquet theory

When a system is driven by the laser field, it is possible to enter a steady state with discrete time translation symmetry, named the Floquet state. Its Hamiltonian is a time-periodic $H(t) = H(t + T)$ with period $T = 2\pi/\omega$, where ω is the frequency of the light. For such kind of system, according to the Floquet theory, the solutions of Schrödinger equation have the form $\psi_\alpha(t) = e^{-i\epsilon_\alpha t}\phi_\alpha(t)$, in which $e^{-i\epsilon_\alpha t}$ is an aperiodic phase factor and $\phi_\alpha(t) = \phi_\alpha(t + T)$ is a periodic function. The Floquet states $\psi_\alpha(t)$ are eigenstates of the evolution operator over one period $U(t + T, t)$ [60], ϵ_α is the Floquet quasi-energy, and $\phi_\alpha(t)$ is the Floquet mode.

Periodic Floquet mode $\phi_\alpha(t)$ can be expanded by discrete Fourier series $\phi_\alpha(t) = \sum_m e^{-im\omega t} u_\alpha^m$, $m \in \mathbb{Z}$. When we substitute $\psi_\alpha(t)$ into the Schrödinger equation and expand the Floquet modes, we obtain [49]:

$$\sum_n H_{mn}(\mathbf{k}) u_\alpha^n(\mathbf{k}) = [\epsilon_\alpha(\mathbf{k}) + m\hbar\omega] u_\alpha^m(\mathbf{k}) \quad (\text{A1})$$

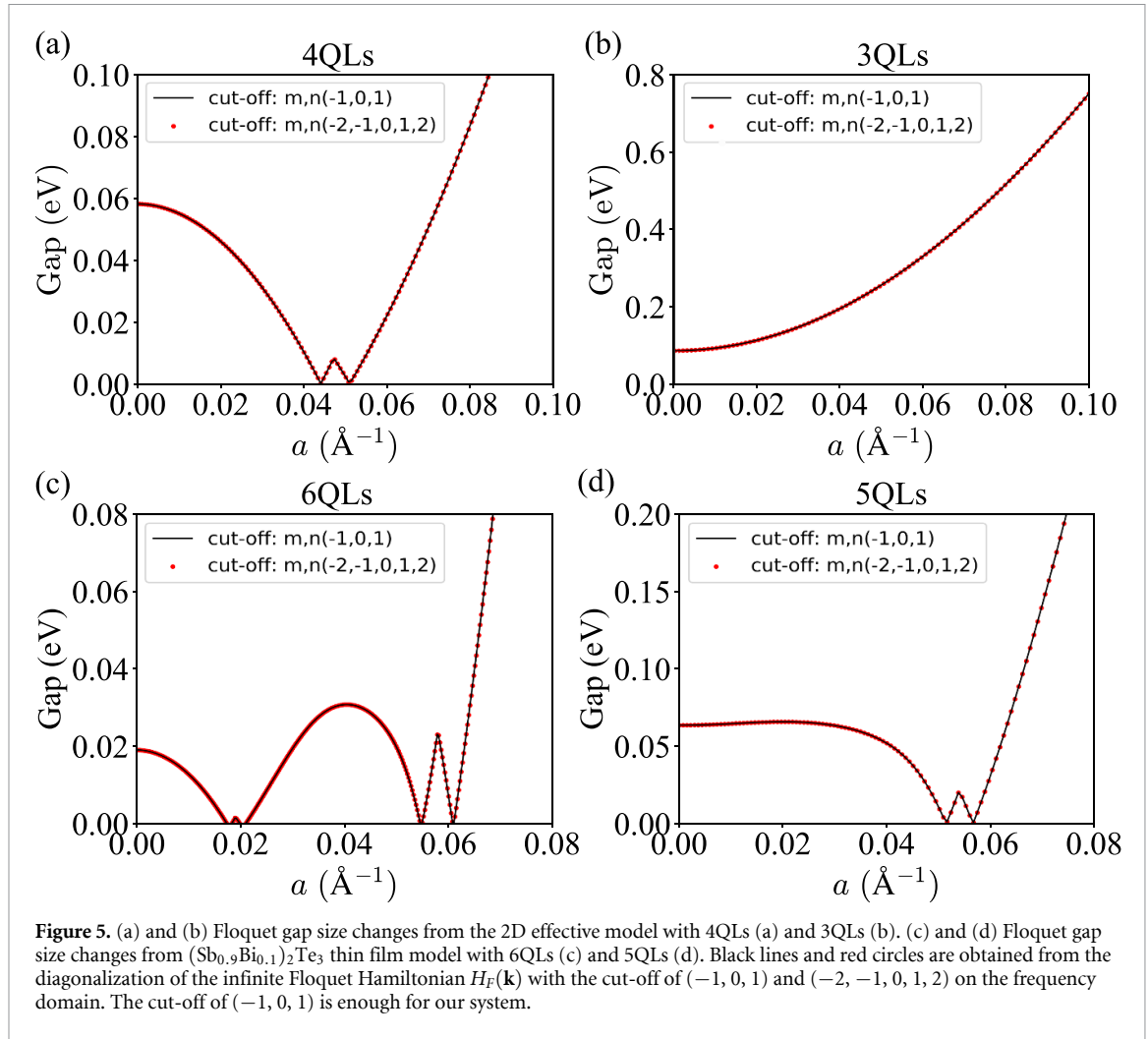
where $H_{mn}(\mathbf{k}) = \frac{1}{T} \int_0^T dt H(\mathbf{k}, t) e^{i(m-n)\omega t}$ and is time-independent. The infinite matrix $(H_{mn} - \delta_{mn}m\hbar\omega)$ is denoted as the infinite Floquet Hamiltonian $H_F(\mathbf{k})$ [49]. This time-independent equation is defined in the extended Hilbert space $\mathcal{H} \otimes \mathcal{L}_T$, in which \mathcal{H} is the original Hilbert space for the intrinsic system and \mathcal{L}_T is the space of T -periodic functions. In this work, the cut-off of the Floquet index m and n in the infinite Floquet Hamiltonian $H_F(\mathbf{k})$ is chosen to be $(-1, 0, 1)$, which is shown to be sufficient in figure 5.

The number of independent Floquet states should coincide with the dimension of $H(t)$. This means the infinite eigenstates of $H_F(\mathbf{k})$ are highly redundant. In fact, eigenstates whose eigenvalues differ by integer multiples of $\hbar\omega$ are related. For example, the Floquet modes $\phi_\alpha(t)$ and $e^{im\omega t}\phi_\alpha(t)$ with eigenenergy ϵ_α and $\epsilon_\alpha + m\hbar\omega$ represent the same Floquet state ψ_α . To remove such redundancy, we focus on states with quasienergies lying inside one $\hbar\omega$ region. Similar to the concept of the Brillouin zone (BZ), we name $[-\frac{\hbar\omega}{2}, \frac{\hbar\omega}{2})$ as the first FBZ. As an example, FBZ is shown by the yellow background in figure 10.

Under the high-frequency limit, the diagonal blocks are dominated by $m\hbar\omega$ term and weakly coupled with other blocks by $H_{mn}(\mathbf{k})$. By performing perturbation theory, the effective Hamiltonian [30, 31, 60, 69] can be derived as:

$$H_{\text{eff}}(\mathbf{k}) = H_{00} + \frac{[H_{10}, H_{01}]}{\hbar\omega} + O((\hbar\omega)^2). \quad (\text{A2})$$

Via applying this approach to the 2D effective thin film Hamiltonian (equation (1) in the main text), we get the time-independent Floquet effective Hamiltonian equation (2) in the main text.



Appendix B. 2D effective model

In the main text, we show in figure 2(a) that for 4QL film there are two gap-closing points with the increase of the effective light amplitude a , indicating topological PTs. For clarity, we plot here the corresponding band structures for typical amplitudes, see figure 6.

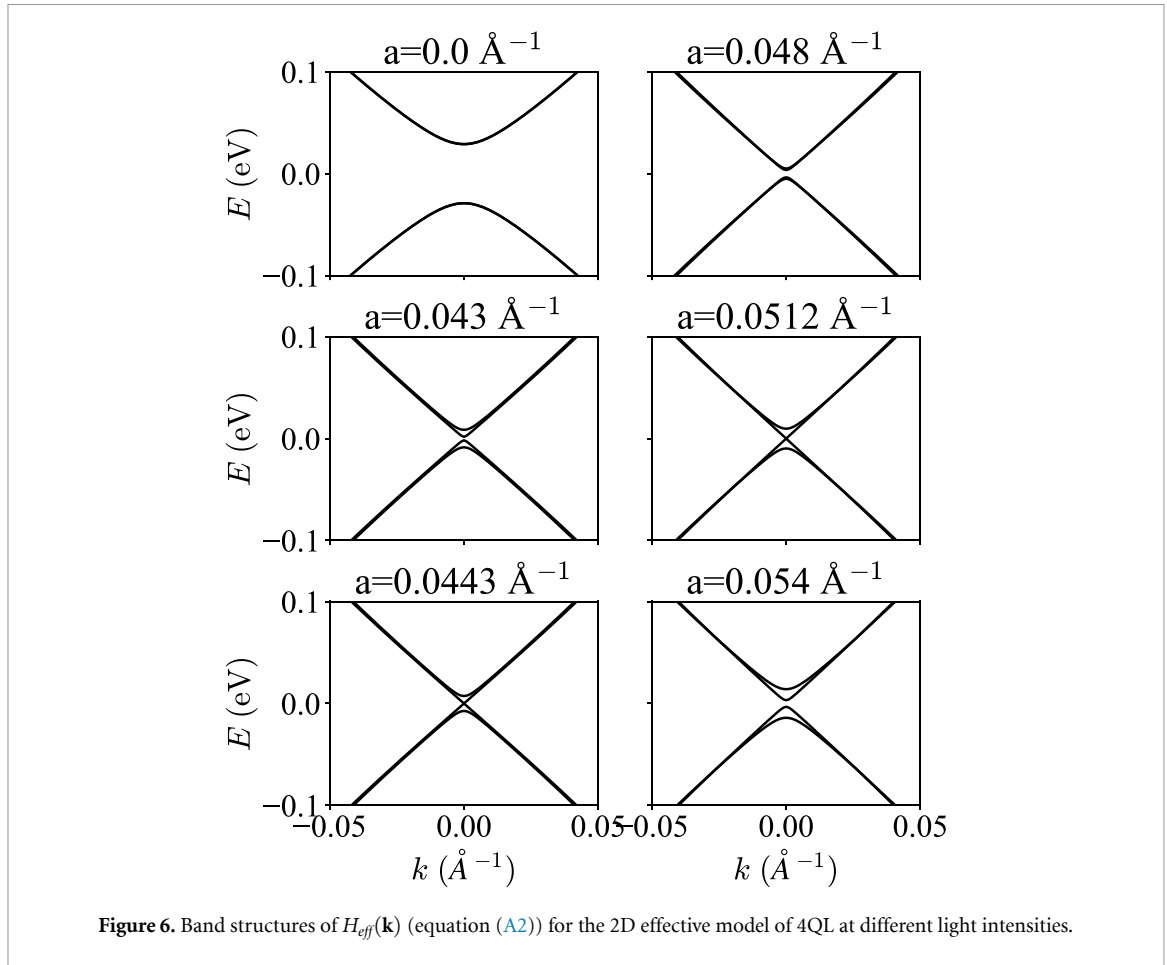
The gap-closing points can be analytically obtained from the low-order expanded Floquet effective Hamiltonian $H_{\text{eff}}(\mathbf{k})$ (see equation (2) in the main text). The eigenvalues of $H_{\text{eff}}(\mathbf{k})$ are:

$$\begin{aligned}
 E_1 &= \pm \sqrt{v_F^2 \left(\frac{2a^2}{\hbar\omega} m_2 + 1 \right)^2 k^2 + \left[a^2 \left(\frac{v_F^2}{\hbar\omega} - m_2 \right) - m(k) \right]^2} \\
 E_2 &= \pm \sqrt{v_F^2 \left(\frac{2a^2}{\hbar\omega} m_2 - 1 \right)^2 k^2 + \left[a^2 \left(\frac{v_F^2}{\hbar\omega} + m_2 \right) + m(k) \right]^2}.
 \end{aligned} \tag{B1}$$

Here $m(k) = m_0 + m_2(k_x^2 + k_y^2)$. Because the model system has particle-hole symmetry, the gap-closing points must occur at zero energy ($E_1 = 0$ or $E_2 = 0$). In total, there are two circumstances to host such condition with zero energy solutions.

Firstly, they can lie at $k = 0$ with $m_0 = -a^2(m_2 \pm \frac{v_F^2}{\hbar\omega})$. Usually, $\frac{v_F^2}{\hbar\omega}$ has small value compared to that of m_2 [28] and can be ignored. Thus, the sign of $m_0 m_2$ must be negative, meaning the system is in the QSH phase initially. The two critical amplitudes a for two topological transitions are separated by $\Delta a \approx \frac{|m_0|^{1/2}}{|m_2|^{3/2}} \frac{v_F^2}{\hbar\omega}$ in the high-frequency limit. Such value is very small, corresponding to a narrow region for the QAH phase shown in figures 2(a) and (b) in the main text.

The other circumstance is at $k \neq 0$ together with $\frac{2a^2}{\hbar\omega} m_2 = 1$ and $a^2 \left(\frac{v_F^2}{\hbar\omega} + m_2 \right) + m_0 = 0$. Substituting the former condition into the latter one, we have $\frac{\hbar\omega}{2} + \frac{v_F^2}{2m_2} + m_0 = 0$. In the high-frequency limit, $\hbar\omega \gg W \sim m_0$, so we can not find a suitable solution for this condition.



Appendix C. Kubo formula

We use the time-dependent Kubo formula derived from linear response theory to calculate the magnetic susceptibilities χ_{zz} of the Floquet system. Following the notion in [64], we have:

$$\chi_{zz}(t, t') = -i\theta(t - t') \langle \Psi_0 | [s_z^I(t), s_z^I(t')] | \Psi_0 \rangle. \quad (\text{C1})$$

Here Ψ_0 represents the initial state, the subscript I represents interaction picture, s_z is the spin operator along the z direction, and $\theta(t, t')$ is the Heaviside function. Written in the Schrödinger picture, equation (C1) becomes,

$$\chi_{zz}(t, t') = -i\theta(t - t') \langle \Psi_0 | U(t_0, t) s_z U(t, t') s_z U(t', t_0) - U(t_0, t') s_z U(t', t) s_z U(t, t_0) | \Psi_0 \rangle \quad (\text{C2})$$

with the evolution operator [60]:

$$\begin{aligned} U(t, t') &= \sum_{\alpha \in \text{FBZ}} |\psi_\alpha(t)\rangle \langle \psi_\alpha(t')| \\ &= \sum_{\alpha \in \text{FBZ}} e^{-i\epsilon_\alpha(t-t')} |\phi_\alpha(t)\rangle \langle \phi_\alpha(t')| \end{aligned} \quad (\text{C3})$$

given by the Floquet state. If the system without laser driving is in thermal equilibrium, the initial state Ψ_0 is a thermal ensemble. The response formula becomes,

$$\chi_{zz}(t, t') = -i\theta(t - t') \sum_i f_i \langle \psi_i | [s_z^I(t), s_z^I(t')] | \psi_i \rangle \quad (\text{C4})$$

where ψ_i represents the eigenstate of the initial equilibrium Hamiltonian and follows Fermi–Dirac distribution f_i . $\chi_{zz}(t, t')$ depends on both t and t' in the Floquet systems due to the lack of time translation invariance. However, in reality, we only care about the mean response over several periods. This can be done by averaging over the mean time $\frac{t+t'}{2}$ [51]. The static spin susceptibility is obtained by a further integration over $t - t'$.

Appendix D. Sudden approximation

In this section, we show how we apply the sudden approximation to both $H_{\text{eff}}(\mathbf{k})$ and $H_F(\mathbf{k})$ in our calculations. We assume that the system is initially in thermal equilibrium, and for simplicity, we omit k summation in the following equations.

For Floquet effective Hamiltonian $H_{\text{eff}}(\mathbf{k})$ in the Hilbert space of \mathcal{H} , after the light is turned on, the evolution of the system can be described by the eigenstates ψ_p of $H_{\text{eff}}(\mathbf{k})$. By substituting $|\psi_i\rangle = \sum_p \langle \psi_p | \psi_i \rangle |\psi_p\rangle$ and $U(t, t') = \sum_a e^{-i\epsilon_p(t-t')} |\psi_p\rangle \langle \psi_p|$ into equation (C4) and integrating over $\frac{t+t'}{2}$ and $t-t'$, we have:

$$\chi_{zz} = \sum_{pq} 2f_p \frac{\langle \psi_p | s_z | \psi_q \rangle \langle \psi_q | s_z | \psi_p \rangle}{\epsilon_q - \epsilon_p} \quad (\text{D1})$$

with $f_p = \sum_i f_i |\langle \psi_i | \psi_p \rangle|^2$.

For the infinite Floquet Hamiltonian $H_F(\mathbf{k})$ in the Hilbert space of $\mathcal{H} \otimes \mathcal{L}_T$, the initial eigenstate ψ_i is expanded by independent Floquet basis $\psi_\alpha(t)$ (Floquet states inside the first FBZ):

$$\begin{aligned} \psi_i &= \sum_{\alpha \in \text{FBZ}} \langle \psi_\alpha(t_0) | \psi_i \rangle \psi_\alpha(t_0) \\ &= \sum_{\alpha \in \text{FBZ}} b_{i\alpha} \psi_\alpha(t_0) \\ &= \sum_{\alpha \in \text{FBZ}} b_{i\alpha} e^{-i\epsilon_\alpha t_0} \phi_\alpha(t_0). \end{aligned} \quad (\text{D2})$$

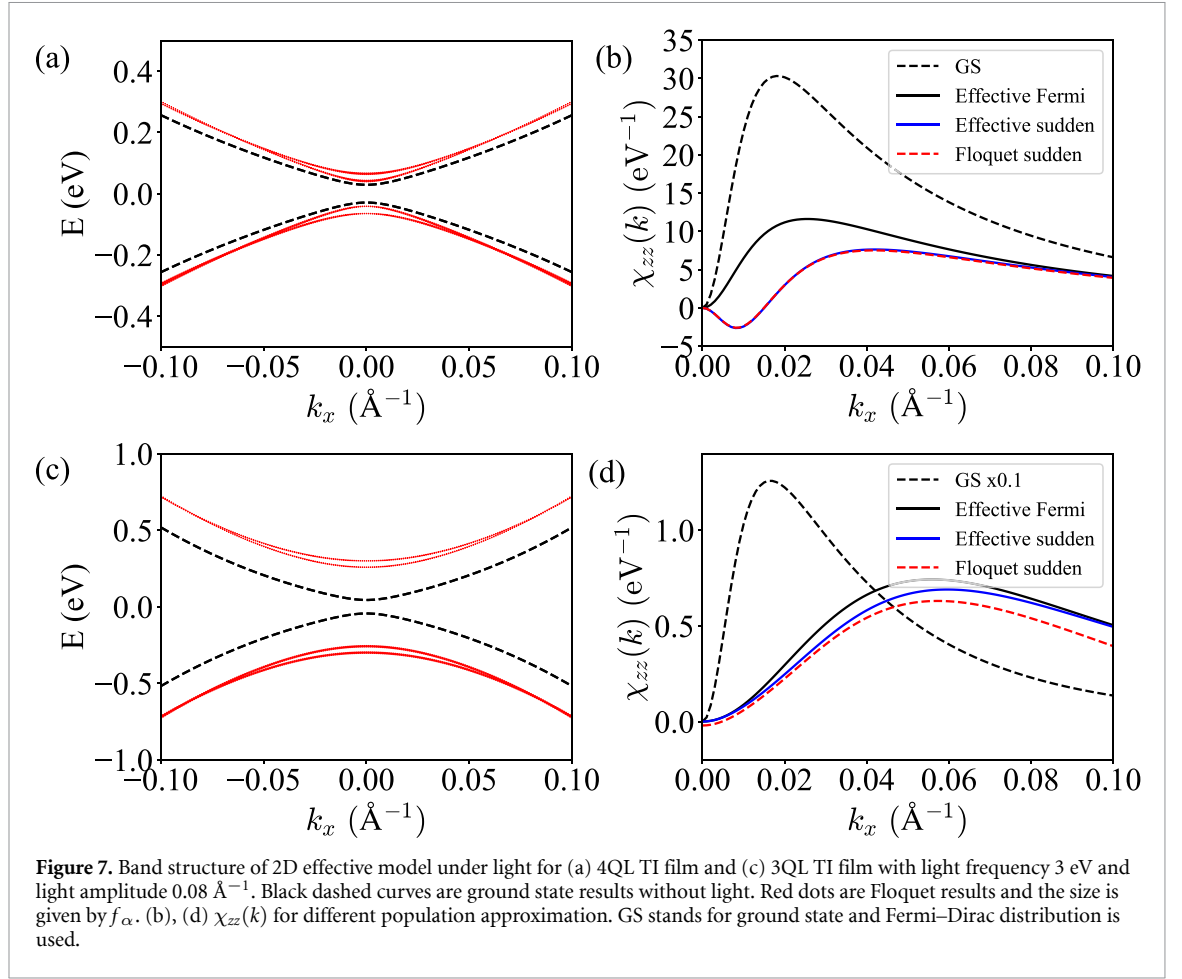
Substitute equations (C3) and (D2) in equation (C4), we get the static spin susceptibility for the Floquet system:

$$\chi_{zz} = \sum_{\alpha \in \text{FBZ}, \beta} 2f_\alpha \frac{|\langle \langle \phi_\alpha(t) | s_z | \phi_\beta(t) \rangle \rangle|^2}{\epsilon_\beta - \epsilon_\alpha}. \quad (\text{D3})$$

Here $f_\alpha = \sum_i f_i |b_{i\alpha}|^2 = \sum_i f_i |\langle \psi_\alpha(t_0) | \psi_i \rangle|^2$, acting like the population of Floquet states. Notice that β goes over all possible quasienergies while α only goes over independent Floquet states. The double bracket is introduced as $\langle \langle \cdot | \cdot \rangle \rangle = \frac{1}{T} \int_0^T \langle \cdot | \cdot \rangle$ [49].

To show the difference between equations (D1) and (D3), band structures and the k dependent magnetic susceptibility $\chi_{zz}(k)$ obtained from $H_{\text{eff}}(\mathbf{k})$ and $H_F(\mathbf{k})$ respectively are compared in figure 7. As the amplitude we use here (0.08 \AA^{-1}) is rather large, the Floquet bands (red solid lines in figures 7(a) and (c)) is quite different from equilibrium (black dashed lines in figures 7(a) and (c)). The susceptibility (figures 7(b) and (d)) is also different in both shape and size compared with equilibrium (black dashed lines in figures 7(b) and (d)). The difference between results obtained from equations (D1) and (D3) (blue and red curves in figure 7(d)) is not very sharp, implying that the system is still in the high-frequency regime.

For clarity, we also calculate the magnetic susceptibility under the Floquet Fermi–Dirac distribution approximation, which is equation (D1) with f_a replaced by the Fermi–Dirac distribution. The difference between the results from sudden approximation (blue and red curves in figures 7(b) and (d)) and the Floquet Fermi–Dirac approximation (black curve in figures 7(b) and (d)) is significant when band inversion occurs. We observe an obvious population inversion near $k_x = 0$ for 4QLs (figure 7(b)) rather than 3QLs (figure 7(d)).



Appendix E. The long-range FM from mean-field theory

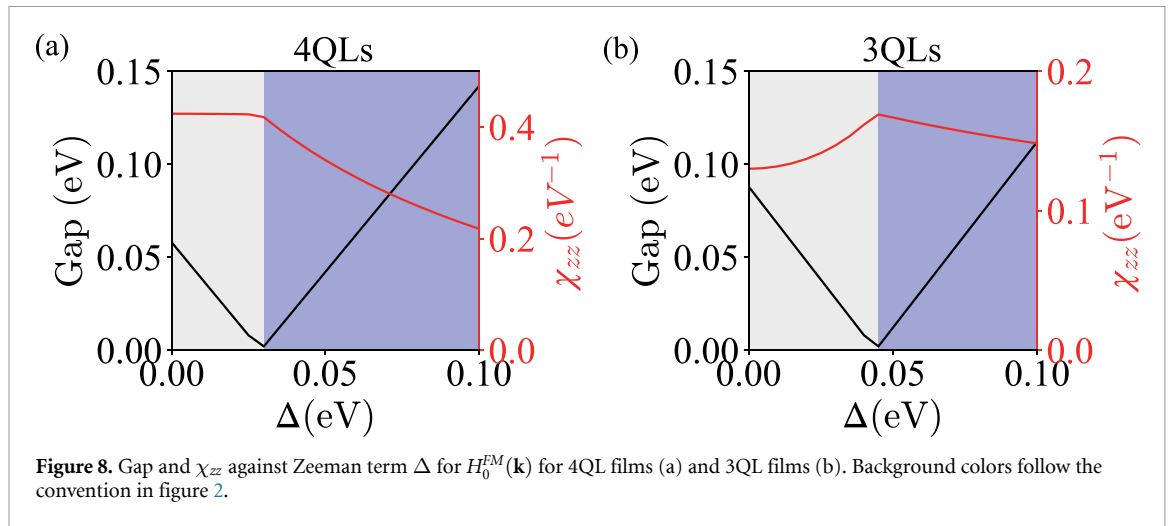
Magnetic properties of this system from the coupling of local moments can be described by $H_{LM} = -\sum_{ij} \mathcal{J} S_i \cdot S_j$. Here S_i represents local moment at site i and \mathcal{J} represents effective exchange interactions. To realize robust FM order, we need \mathcal{J} to be positive.

In our system, two mechanisms [10, 28] mainly contribute to \mathcal{J} . The first mechanism is the RKKY interaction, namely, a local moment magnetizes nearby band electrons and these band electrons further affect local moments at other sites. This interaction $\mathcal{J}_F = J^2 \chi$ is FM and proportional to the density of doped magnetic ions and the magnetic susceptibility of band electrons, where J is the exchange integral between local moment S_i and band electrons. The second mechanism is the antiferromagnetic superexchange interaction between local moments, which contributes to negative \mathcal{J} and is proportional to the density of dopant and the inverse Hubbard U of the local moments $\mathcal{J} \propto -J^2/U$. When we fix the dopant density, we need the magnetic susceptibility to satisfy $\chi > 1/U$ to realize FM order. For typical transition metal ions, it is reasonable to choose $U = 2.5 \text{ eV}$ [28]. Thus, in our system, the critical spin susceptibility for a possible magnetic PT is around 0.4 eV^{-1} . The corresponding critical light amplitudes are indicated in figures 2 and 4 in the main text.

When FM order is developed, the 2D effective Hamiltonian becomes,

$$H_0^{FM}(\mathbf{k}) = H_0(\mathbf{k}) + \Delta \cdot \sigma_3 \otimes I \quad (\text{E1})$$

with an additional Zeeman term Δ . As a result, the magnetic susceptibility χ_{zz} for H_0^{FM} now also depends on Δ . We plot the gap and χ_{zz} against Δ in figure 8. The system undergoes a transformation from the trivial phase to the QAH phase at $\Delta = 0.03 \text{ eV}$ for 4QL films ($\Delta = 0.045 \text{ eV}$ for 3QLs). During these topological PTs, the spin susceptibility curves also exhibit kinks and then decrease abruptly. We mention that the increase in figure 8(b) is likely due to the band inversion induced by the Zeeman term for 3QL films if the long range FM order is developed. Additionally, Δ depends on χ_{zz} , necessitating a self-consistent process to solve for both Δ and χ_{zz} as well as to estimate the Curie temperature.



Appendix F. Thin film models

Now we fully consider the quantum confinement effects in TI thin films by constructing a film Hamiltonian from a bulk effective $k \cdot p$ model, which includes contributions from the quantum well states. Following the notion in [68, 74], we start from the effective $k \cdot p$ model of a Bi_2Se_3 -type 3D TI at the Γ point in the basis of $|P1_z^+ \uparrow\rangle, |P2_z^- \uparrow\rangle, |P1_z^+ \downarrow\rangle, |P2_z^- \downarrow\rangle$:

$$H = \epsilon(k) + \begin{pmatrix} \mathcal{M}(k) & A_1 k_z & 0 & A_2 k_- \\ A_1 k_z & -\mathcal{M}(k) & A_2 k_- & 0 \\ 0 & A_2 k_+ & \mathcal{M}(k) & -A_1 k_z \\ A_2 k_+ & 0 & -A_1 k_z & -\mathcal{M}(k) \end{pmatrix} + O(k^2) \quad (\text{F1})$$

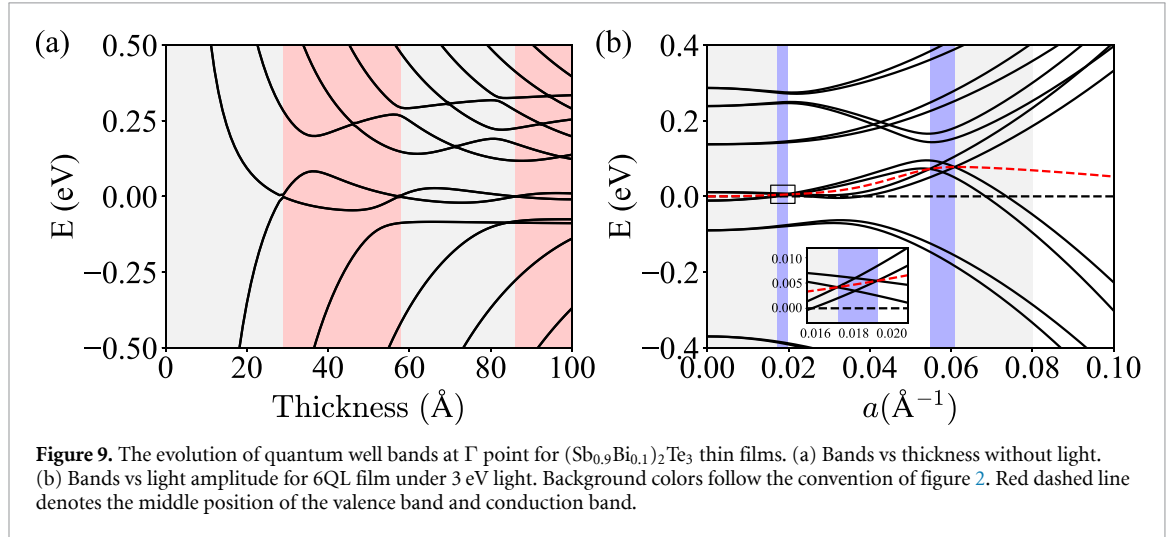
where $k_{\pm} = k_x \pm ik_y$, $\epsilon(k) = \epsilon_0 + \epsilon_1 k_z^2 + \epsilon_2 (k_x^2 + k_y^2)$ and $\mathcal{M}(k) = m_0 + m_1 k_z^2 + m_2 (k_x^2 + k_y^2)$. To construct the Hamiltonian for thin films with fixed boundary condition, we replace k_z by $-i\partial_z$ and solve equation (F1) on the quantum well basis [75] $\psi_n(z) = \sqrt{\frac{2}{d}} \sin(\frac{n\pi z}{d})$, where $n = 1, 2, 3, \dots$ and d is the thickness of the thin film. The following calculations on 6QL ($d = 60 \text{ \AA}$) and 5QL ($d = 50 \text{ \AA}$) ($\text{Sb}_{0.9}\text{Bi}_{0.1}$) $_2\text{Te}_3$ films are with parameters obtained from [68] by linear interpolation. The parameters are listed in table 1.

According to [75], the topological properties of TI thin films oscillate between a trivial phase and a QSH phase when the thickness increases. This is because more quantum well bands invert as the films become thicker. We plot the band evolution of ($\text{Sb}_{0.9}\text{Bi}_{0.1}$) $_2\text{Te}_3$ thin films with the change of thickness in figure 9(a).

In this work, we find that the applied laser field also alters the band inversion behavior of the quantum well states. We choose 6QL film as an example. When we increase the light amplitude (see figure 9(b)), these degenerate bands split and invert, inducing four critical laser amplitudes for four gap-closing processes. The splitting of the double-degenerate bands is due to the break of time reversal symmetry by right-handed CPL. The middle position of the gap is denoted by the dashed red line in figure 9(b).

Table 1. $k \cdot p$ parameters for $(\text{Sb}_{0.9}\text{Bi}_{0.1})_2\text{Te}_3$ films. Unit for ϵ_0 and m_0 is eV. Unit for ϵ_1 , ϵ_2 , m_1 and m_2 is eV \AA^2 . Unit for A_1 and A_2 is eV \AA .

ϵ_0	ϵ_1	ϵ_2	m_0	m_1	m_2	A_1	A_2
-0.0171	-10.496	-4.734	-0.228	17.955	49.397	0.786	3.347



Appendix G. Problems for sudden approximation

As stated in the main text, the population of Floquet bands under sudden approximation is determined by the overlap between initial and final states when the light is applied. It results in a non-thermal population and could give rise to oscillations of $\chi_{zz}(k)$ near Floquet band crossings (or avoided-crossings).

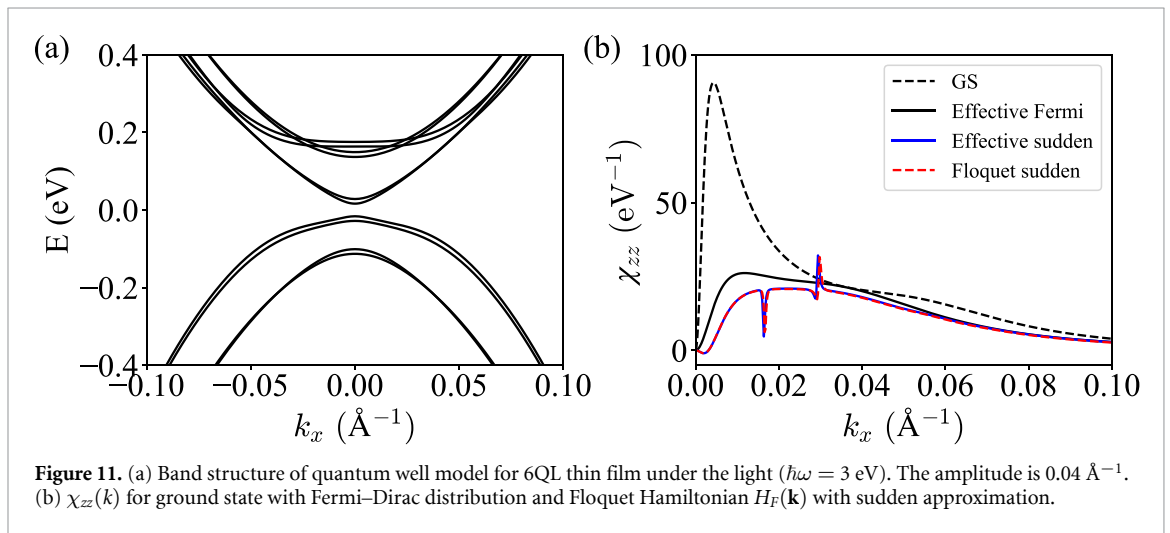
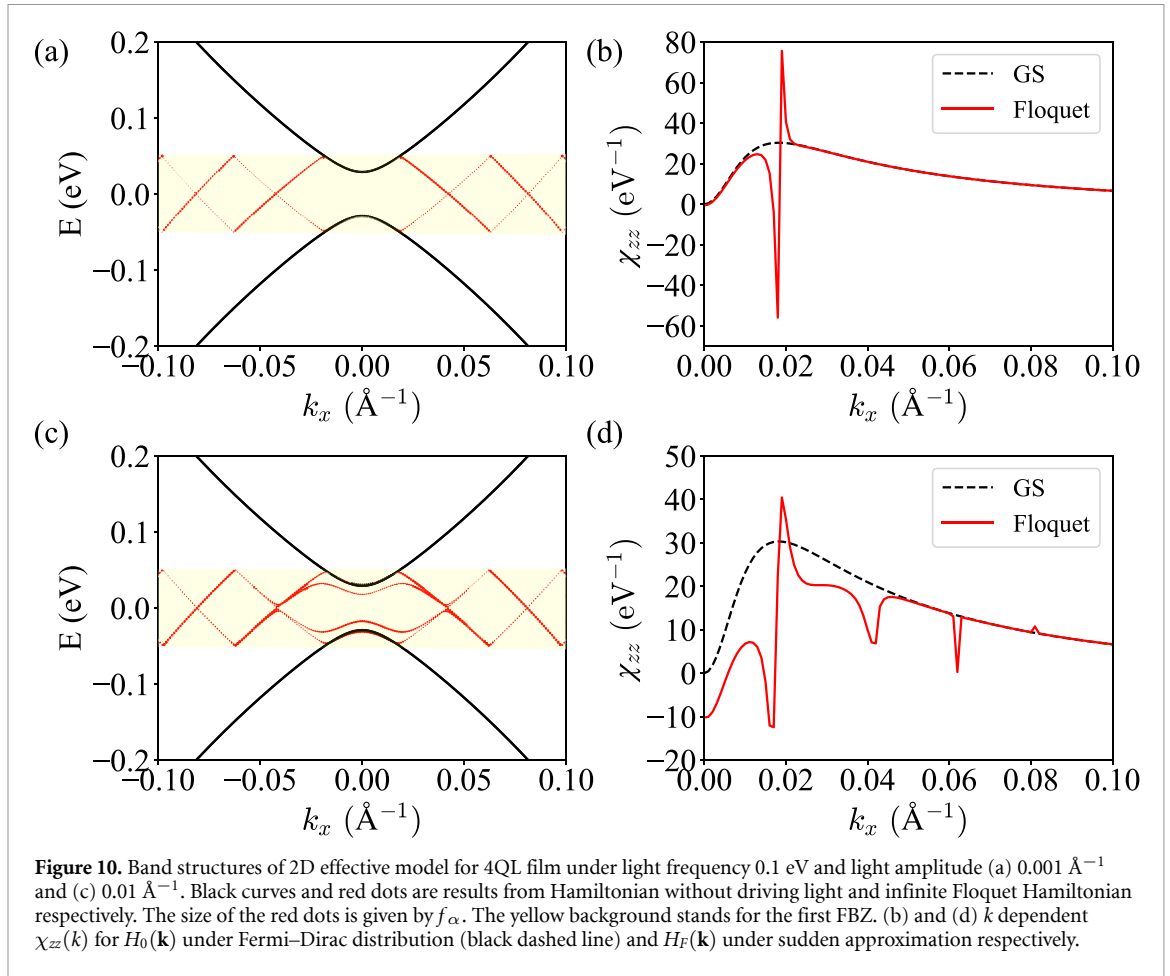
There are two types of band crossings in the Floquet systems, one happens between different photon sectors (e.g. the Floquet band with $m = 0$ crosses with $m = 1$) when the laser frequency is small ($\hbar\omega < W$). And the other happens within one photon sector (e.g. Floquet bands cross each other with same m).

We first discuss the type of crossings between photon sectors with low frequency. Figure 10 presents an example for the 4QL 2D effective model with light driving, where the light frequency ($\hbar\omega = 0.1$ eV) is chosen to be smaller than the bandwidth to induce band crossings between photon sectors. We consider two laser amplitudes: figures 10(a) and (c) are for amplitude 0.001 \AA^{-1} and 0.01 \AA^{-1} , respectively. The black curves and red dots represent the band structures from Hamiltonian $H_0(\mathbf{k})$ without driving light and Floquet Hamiltonian $H_F(\mathbf{k})$, respectively. For $H_F(\mathbf{k})$, only bands inside FBZ are plotted. Figures 10(b) and (d) show the k dependent spin susceptibility.

Near band crossing points, as shown in figure 10(b), $\chi_{zz}(k)$ diverges due to a small energy difference on the denominator of equation (D3) and a nonzero population difference on the numerator. Comparing figures 10(c) with (a), under larger amplitude, we observe contributions from higher Floquet bands and broadening of the divergent peaks. The broadening occurs because the sizes of the avoided-crossing gaps increase with the increase of the light amplitude.

In our framework with sudden approximation, the effect of dissipation is not considered. Based on the Ono and Ishihara's results from the Floquet-Green's function [72], the magnetic susceptibility is continuous with increasing light amplitude. We argue that such kind of divergence is possible to be smeared out by the dissipation.

Figure 11 illustrates the types of crossings that occur inside one photon sector. Here we choose the quantum well model for a 6QL thin film as an example, with a light frequency of 3 eV that is consistent with the main text and larger than the bandwidth to avoid inter-sector crossing. The band structure for $a = 0.04 \text{ \AA}^{-1}$ is shown in figure 11(a), and $\chi_{zz}(k)$ is displayed in figure 11(b). We observe divergent peaks in susceptibility when bands cross with each other.



Appendix H. Light intensity

In our work, we adopt effective light amplitude a for simplicity as it is in unit of Å⁻¹. In this section, we discuss the relation between light intensity and the effective light amplitude a . The relation between light intensity and light electric field is $I = \frac{1}{2}cn\epsilon_0E^2$, in which ϵ_0 is the vacuum permittivity, c is the speed of light, and n is the refractive index of the TI thin film [76]. In the experiment, the light intensity can be achieved as high as 10¹² W cm⁻², which corresponds to 0.2/ $\hbar\omega$ for effective light amplitude a . Thus our proposed critical amplitudes are possible to be achieved in experiments.

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