February 26, 2024

Dear Editor,

Thank you for securing expert reviews for our article and for allowing us to revise our manuscript as per the reviewer's comments.
We are very grateful to the reviewer for the time and effort in reviewing our manuscript. We feel that the received feedback from the referee has helped us improve both content and presentation of the paper. We have addressed all the reviewer' comments, as presented in our point-by-point responses below.

Sincerely,
Authors

THE REPORT OF THE REFEREE

The referee states:
This manuscript presents a theoretical study on the manipulation of topological phases in thin films of Bi2Se3 using circularly polarized light via Floquet engineering. It explores the potential for creating quantum anomalous Hall, high-Chern number, and axion insulator phases, each characterized by distinct Chern numbers. These phases are of great interest for their unique properties and potential applications in spintronics and optoelectronics. I support its publication, provided the authors address the following issues.

Our reply:
We thank the referee for recognizing the merit of our work. In what follows, we carefully address all the received questions and remarks.

The referee states:
The key results of the paper are primarily derived from calculating Chern numbers for different thin film thicknesses, light wavelengths, and intensities. Importantly, the authors consider the fact that the driving field decays inside the sample. I believe that presenting the explicit expression (presumably, a formula that sums over the layer index, while in-plane wave vectors remain good quantum numbers) for calculating the Chern number with the layer-dependent vector potential, which decays exponentially inside the bulk, is crucial.

Our reply:
In the calculations of the Chern number, the effect of illumination is effectively added to the onsite energy of the Hamiltonian (according to the site position and layer number), which is possible for the real space Hamiltonian. Also, to check the results of the Chern number, we have performed calculations related to Hall conductance, confirming our results.

In response to this comment, we have added the following clarification in the manuscript:
In calculating the Chern number, the influence of illumination is added to the onsite energy of the Hamiltonian, considering the layer index in the sample. The Chern number can be defined using $\widehat{\mathbf{R}}=\mathbf{R}/R$ as:

$$C = 2\int_{Bz} \hat{\boldsymbol{R}}.\left(\frac{\partial \hat{\boldsymbol{R}}}{\partial k_x} \times \frac{\partial \hat{\boldsymbol{R}}}{\partial k_y}\right)\frac{dk_x dk_y}{4\pi},$$

where **R** = ($-v_F k_y$, $v_F k_x$, M). The calculated Chern number was validated by assessing Hall conductance.

The referee states:
Similarly, it is crucial to present the explicit expression for calculating the orbital magnetization Mz(Ez), taking into account the decay of the driving field inside the sample. It is important to note that in Floquet systems, the concept of a "ground state" loses its traditional meaning - Equation (10) directly references Ref. [60], which is a study of an equilibrium state.

Our reply:
We agree and thank the reviewer for the comment.

To accommodate the remark of the reviewer, we have added these sentences in the manuscript:
It is worth noting that in Floquet systems, the idea of a ground state loses its conventional meaning, which is typically used to describe an equilibrium state. Therefore, just like the computations related to the Chern invariant, we incorporate the impact of applied light to the onsite energies of the Hamiltonian. Then we use Eq. (10) to calculate the orbital magnetization.

The referee suggests:
*Suggested: After the initial stage of identifying Floquet engineering as an interesting method for manipulating material properties, researchers gradually realized that the filling of the Floquet bands does not simply follow Fermi-Dirac statistics, and the so-called Floquet 'topological' states "may not give rise to quantized transport" [see Nature Reviews Physics 2, 229–244 (2020)]. To date, experimental transport observations have also failed to produce a quantized plateau [see Nature Physics 16, 38–41 (2020)]. Hence, it is recommended to highlight this aspect to raise awareness within the community. The topological classifications of Floquet systems are not adequately captured by the effective Hamiltonian.

Our reply:
We thank the referee for this suggestion, which we added to the manuscript to raise respective awareness within the community.

To accommodate this remark of the referee, we added the following clarification in the discussion:
One should note the recent predictions that the compound CPL can lead to rich topological phase transitions in antiferromagnetic topological insulator MnBi2Te4 films [31]. However, further research has shown that the filling of the Floquet bands in some cases does not conform to Fermi-Dirac statistics. As a result, the so-called Floquet topological states may not lead to quantized transport [32].

The referee suggests:

*Suggested: The symbols Ax, Ay, Az in the tight-binding Hamiltonian and the periodic field A use the same notation. A change of notation is advised.

Our reply:
We appreciate the suggestion from the referee and have changed the hopping parameters from Ax, Ay, Az to Dx, Dy, Dz .

In response to this comment, we changed the hopping parameters from Ax, Ay, Az to Dx, Dy, Dz.

The referee suggests:
*Suggested: Calculating the maximum permitted power density of the laser illumination (e.g., W/cm2) instead of just using Ã, would benefit the reader. And up to now, only transient Floquet states have been observed when driven by laser, as the laser rapidly heats the sample. It would be beneficial for the reader if this aspect were mentioned.

Our reply:
In this study, we have focused solely on the impact of light on the breaking of TRS and the resulting gap in surface states, which is influenced by the intensity A. Other factors, such as heating, have not been taken into account. Therefore, we have presented our calculations based on A alone. We do agree with the referee that closer connection to the laser power density and other realistic factors would be helpful to the experimentalists and could yield exciting results, but the required details of such an analysis exceed the scope of the present manuscript (focused on fundamental aspects of CPL-TI interaction and the resulting phases).