

Attosecond pulse generation from Weyl semimetals

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The generation of attosecond pulses from higher-order harmonics is an essential milestone toward enabling time-resolved spectroscopy of electronic processes in matter on the attosecond timescale. The avenue for high-harmonic generation from solids has proliferated since its discovery a decade ago. However, isolated attosecond pulse generation from solids remains scarce. We report a theoretical proposal that leverages Weyl semimetals for highly efficient attosecond pulse generation in the far-ultraviolet region. The generation of isolated attosecond pulses from Weyl semimetals is achieved using two-color laser pulses, which generate a broad higher-harmonic plateau ranging from 3 to 11 eV. Attosecond pulses with comparable durations – spanning approximately 500 to 900 as – are generated in both collinear and orthogonal configurations of the two-color fields. The strength and duration of the attosecond pulses can be optimized by tailoring the polarization direction and relative strength of the two-color field.

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I. INTRODUCTION

Early experimental demonstrations of high-order harmonic generation (HHG) in gases [1,2] opened avenues for exploring strong-field-driven nonlinear optical phenomena [3–6]. Rooted in the mechanisms of electron tunneling and recombination [7,8], HHG has become a powerful probe for interrogating ultrafast electronic motion on its natural timescale [9–18]. Beyond enabling time-resolved studies of electron dynamics, HHG has also broken the femtosecond barrier, making it possible to generate attosecond (as) pulses [19–22]. Remarkably, an isolated attosecond pulse with a duration of just 43 as – currently the shortest ever generated, to the best of our knowledge – was reported in 2017 [23]. The profound impact of HHG in advancing our understanding of ultrafast electron dynamics was recognized with the award of the 2023 Nobel Prize in Physics, highlighting the transformative role of attosecond physics in contemporary science and technology [24–26].

Significant advancements in laser technology over the past decade have enabled the generation of intense midinfrared laser pulses. These developments led to the experimental observation of nonperturbative HHG from zinc oxide in 2011 [27]. Since then, HHG from solids has emerged as a versatile method for probing a wide range of nonequilibrium phenomena across diverse classes of solid-state systems [28–55].

Compared to HHG in gases, solid HHG offers several advantages: The higher electron density in solids results in a relatively stronger harmonic yield. Furthermore, the presence of intense and multiple plateaus in the harmonic spectra makes solid HHG an attractive route for generating attosecond pulses [56]. These characteristics position HHG from solids as a lucrative and practical platform for realizing compact, tabletop sources of bright, coherent attosecond pulses. Despite its potential, attosecond pulse generation from solid HHG remains one of the least explored frontiers in strong-field physics.

This work presents a theoretical scheme for synthesizing isolated attosecond pulses by exploiting solid HHG. Our approach employs a two-color driving field composed of an intense, nonresonant fundamental pulse (ω) and a relatively weaker second-harmonic pulse (2ω) to drive harmonic emission from topological semimetals. To generate higher-order harmonics, we use Weyl semimetals (WSMs), a class of topological quantum materials characterized by broken inversion and/or time-reversal symmetry. WSMs host Weyl nodes—the source and sink of Berry curvature—and exhibit topological robustness against external perturbations [57]. A recent experiment demonstrated that the inversion-broken WSM β -WP₂ yields efficient higher-order harmonics under relatively low driving intensities [58], and its generation efficiency surpasses that of other materials under similar laser parameters. Thus, owing to their topological robustness against external perturbations and their efficient HHG response, WSMs are well suited for synthesizing isolated attosecond pulses. Moreover, the presence of the 2ω field alongside the fundamental ω pulse enables tailoring of the emitted harmonic spectrum, as demonstrated across various solid systems [59–65].

In the following, we demonstrate how the addition of a weaker 2ω linearly polarized pulse, in the presence of an

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intense linear ω pulse, significantly modifies HHG of time-reversal-broken (TRB) WSMs and facilitates the synthesis of attosecond pulses with a duration of ~ 630 as for the collinear configuration of the ω and 2ω pulses. In contrast, the orthogonal configuration, where the ω and 2ω pulses are polarized perpendicular to each other, yields longer pulses of ~ 900 as. We observe that the temporal structure of the synthesized attosecond pulses can be controlled by adjusting the relative delay between the ω and 2ω fields. We have also investigated the sensitivity of the pulse duration to the intensity ratio between the 2ω and ω pulses, as well as to variations in the ω pulse intensity. We have extended our study from the case of type-I WSMs with two degenerate Weyl nodes located at the Fermi energy, to type-II WSMs with nondegenerate nodes, i.e., shifted away from the Fermi level [66]. In the case of type-II WSMs, Weyl nodes are tilted rather than upright, which commonly occurs in material candidates for WSMs such as transition-metal monpnictides [67]. An investigation is carried out to understand how the energy splitting between Weyl nodes and their tilt influence attosecond pulse generation. The orthogonal configuration is found to be more efficient in generating attosecond bursts from a type-II WSM, producing pulses as short as 640 as. It is worth mentioning that recent years have witnessed several theoretical proposals for generating attosecond pulses from solid-state HHG [68–72], along with notable advancements on the experimental front [73,74].

II. NUMERICAL METHOD

We simulate laser-driven electron dynamics in WSMs by solving the equations of motion within the density matrix formalism. Specifically, we solve the coupled semiconductor Bloch equations numerically in the Houston basis, following the approach described in Refs. [75,76]. Numerical integration of the semiconductor Bloch equations is performed using a fourth-order Runge-Kutta algorithm with a time step of 0.014 fs, together with a $80 \times 80 \times 80$ $\mathbf{k}$ -point grid for Brillouin-zone sampling. We employ a pair of linearly polarized pulses with fundamental corresponding to the wavelength of 3200 nm and its second harmonic. In the collinear setup, both pulses are polarized along the x axis, while the ω pulse is polarized along the x axis and the 2ω pulse along the y axis in the orthogonal configuration. The relative strength of the vector potential for the 2ω pulse with respect to the ω pulse is characterized by the ratio $\mathcal{R}$, which is fixed at 0.25 unless stated otherwise. The electric field strength of the 2ω pulse can be increased by increasing $\mathcal{R}$. The pulse duration of both the pulses is fixed at ~ 80 fs and the peak intensity of the ω pulse is fixed at 5×10^{11} W/cm², unless stated otherwise. The high-harmonic spectrum is obtained by computing the Fourier transform of the time derivative of the electronic current as $|\mathcal{FT}(\partial \mathbf{J}(t)/\partial t)|^2$, where $\mathbf{J}(t)$ denotes the total current induced by the incident laser pulse. A dephasing time of 1.5 fs is used to account for the decoherence between electrons and holes [77,78].

A tight-binding Hamiltonian of the form $\mathcal{H} = d_0(\mathbf{k})\sigma_0 + \sum_{i=1}^3 \mathbf{d}_i(\mathbf{k}) \cdot \sigma_i$ is adopted for TRB type-I and type-II WSMs with σ_0 and σ_i as the identity and Pauli matrices, respectively

[78]. The components of $\mathbf{d}(\mathbf{k})$ for a TRB WSM are given by

$$\begin{aligned} \mathbf{d}(\mathbf{k}) = & t[t_c\{\cos(k_z a) + \cos(k_x a) - 1\} + t_g \sin(k_z a), \\ & \sin(k_x a), \sin(k_y a), \{\cos(k_z a) - \cos(k_0 a) \\ & + 2 - \cos(k_x a) - \cos(k_y a)\}]. \end{aligned} \quad (1)$$

Here, k_0 specifies the location of the Weyl nodes, while the parameters t_c and t_g control the tilt and energy splitting of the nodes, respectively. For a WSM with $t_c = t_g = 0$, the Weyl nodes are located at $k = [0, 0, \pm\pi/(2a)]$, i.e., at $k_0 = \pm\pi/(2a)$ with $a = 6.28$ Å as the lattice constant of a cubic lattice. We have considered an isotropic hopping parameter of 1.8 eV in this work. When $t_c = t_g = 0$, the material behaves as a type-I WSM with upright Weyl cones. Increasing t_c beyond unity introduces a tilt in the cones, marking the transition to a type-II WSM. At $t_c = 2$, the cones are strongly tilted in opposite directions, resulting in the coexistence of electron and hole pockets near the Weyl points [78].

III. RESULTS AND DISCUSSION

High-harmonic spectra corresponding to a type-I WSM with degenerate Weyl nodes located at the Fermi level are presented in Fig. 1. When driven by a single ω pulse, only odd-order harmonics are generated, consistent with the inversion symmetry of the WSM [see Fig. 1(a)]. In addition, anomalous odd-order harmonics are generated in the direction perpendicular to the laser polarization due to the absence of time-reversal symmetry, in agreement with previous findings [77,79–83]. Although the inclusion of a weaker 2ω field does not significantly enhance the overall harmonic intensity, it plays a crucial role in reshaping the spectral characteristics, as demonstrated in Fig. 1(b). The combined $\omega - 2\omega$ driving field produces even-order harmonics in addition to the odd ones, even when the 2ω component is relatively weak. Notably, the intensity of the fourth harmonic is comparable to that of the fifth harmonic in the collinear configuration [Fig. 1(b)], and even marginally stronger in the orthogonal configuration [Fig. 1(c)]. These observations highlight the strong sensitivity of the high-harmonic response to the interference between the ω and 2ω pulses. Both collinear and orthogonal $\omega - 2\omega$ configurations generate a harmonic plateau extending from approximately 3 to 11 eV. In the orthogonal configuration, however, harmonic peaks are not well-resolved in the plateau region as compared to the collinear case. Overall, the spectral features show only modest sensitivity to the relative time delay between the ω and 2ω pulses when varied from perfect overlap to a full cycle delay of the ω field.

As evident from the spectra, harmonic orders corresponding to the energy range 3–11 eV form a plateau, which can be used to synthesize attosecond pulses. The temporal profiles of the synthesized pulses derived from this plateau are shown in Fig. 2 for the collinear and orthogonal configurations. In the collinear cases [Figs. 2(a)–2(c)], the full width at half maximum (FWHM) of the resulting single pulses lies in the attosecond regime, with durations of 630, 630, and 680 as for time delays of 0, 0.5T, and T, respectively. While the sub-cycle delay does not significantly change the pulse duration, it enables control over the temporal structure of the attosecond pulses, as evident in Figs. 2(a)–2(c) for the collinear

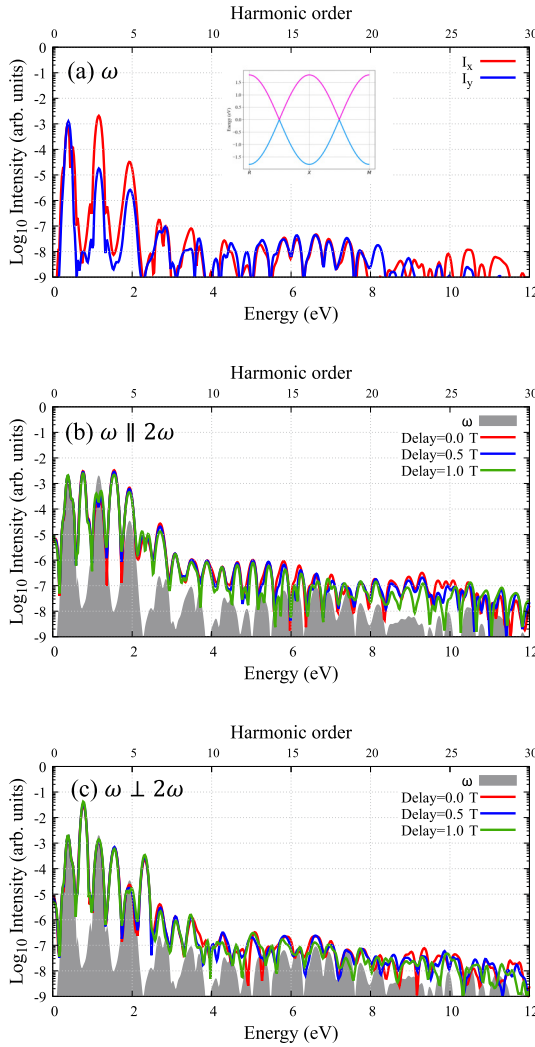


FIG. 1. High-harmonic spectra of a Weyl semimetal driven by combined ω and 2ω laser pulses with relative time delays of 0, 0.5 T, and T with T as the single cycle time period of the ω pulse. (a) High-harmonic spectra corresponds to a single ω pulse. Panels (b) and (c) correspond to collinear and orthogonal polarization configurations, respectively. Harmonic spectra generated by a single ω pulse are shown in grey for reference. The inset displays the energy band structure of the type-I Weyl semimetal, featuring two degenerate Weyl nodes positioned at the Fermi level, i.e., at $k = [0, 0, \pm\pi/(2a)]$.

configuration. In the case of the orthogonal configuration, the duration of the synthesized attosecond pulses varies from ~ 670 to 880 as the subcycle delay is tuned, as shown in Figs. 2(d)–2(f). The synthesized pulses are noticeably longer and exhibit pronounced temporal structures in the orthogonal configuration compared to the collinear case. Thus, the collinear configuration yields shorter, nearly structure-free attosecond pulses. These results highlight the feasibility of generating isolated attosecond pulses using a combined $\omega - 2\omega$ configuration and demonstrate that polarization geometry and subcycle delay provide effective control knobs for tailoring their temporal characteristics. At this juncture, one may question the feasibility of attosecond pulse generation from other potential solid-state systems, such as monolayer

MoS₂. As illustrated in the Appendix, inversion-symmetric WSMs generate shorter single attosecond pulses compared to MoS₂ under comparable conditions, thereby justifying and reinforcing our choice of WSMs as the primary system of interest (see Appendix).

The sensitivity of the temporal profile of the synthesized attosecond pulse to the polarization configuration and time delay between the ω and 2ω pulses can be attributed to the critical role of the total vector potential. The resultant vector potential differs significantly between collinear and orthogonal configurations, as well as for varying delays, leading to distinct interband transitions and intraband electron dynamics. These variations, in turn, influence the harmonic plateau structure and ultimately affect the efficiency and structure of attosecond pulse generation. As shown in Fig. 1, although the harmonic spectrum generated by a single ω pulse is nearly as intense as that produced by the combined $\omega - 2\omega$ fields, the plateaus differ significantly—most notably by the absence of even-order harmonics in the single-pulse case. Moreover, for the collinear configuration [Fig. 1(b)], the spectrum displays distinct and well-resolved harmonics above 4 eV, whereas the orthogonal configuration [Fig. 1(c)] results in a comparatively less resolved harmonic structure in the same energy range. This suggests that the interference between the ω and 2ω pulses in orthogonal configuration suppresses contributions from long electron trajectories [84]. Consequently, the attosecond pulses generated in the orthogonal configuration exhibit considerable temporal substructures and are not fully isolated, in contrast to the nearly isolated and cleaner attosecond pulses obtained in the collinear configuration.

So far, our discussion has focused on attosecond pulse generation using $\omega - 2\omega$ configuration with a fixed ratio $\mathcal{R} = 0.25$ and the intensity of the ω pulse at 5×10^{11} W/cm². To establish a more comprehensive understanding of the $\omega - 2\omega$ setup in controlling attosecond pulse characteristics, it is essential to investigate how the pulse duration varies with respect to $\mathcal{R}$ and the intensity of the ω pulse. This analysis will strengthen the importance of the $\omega - 2\omega$ field in tailoring attosecond pulse durations. We focus our discussion on two representative subcycle delays, 0.0 T and 0.5 T, in the collinear configuration, as they correspond to the most promising conditions for generating isolated attosecond pulses.

Figure 3(a) illustrates the variation of the synthesized pulse duration as a function of the ratio $\mathcal{R}$ for two different delays while keeping the peak intensity of the fundamental ω pulse fixed at 5×10^{11} W/cm². For both delays, the pulse duration decreases steadily as $\mathcal{R}$ increases up to approximately 0.5. Beyond this point, the 2ω component becomes dominant over the ω pulse, which leads to a nonmonotonic dependence of the attosecond pulse duration on $\mathcal{R}$. This behavior clearly indicates that the 2ω pulse must remain weaker than the ω pulse to support optimal gating conditions for generating isolated attosecond pulses. Figure 3(b) presents the sensitivity of the pulse duration to the peak intensity of the ω pulse for a fixed $\mathcal{R} = 0.25$. An inverse relationship is observed: the pulse duration decreases as the ω pulse intensity increases. However, the degree of temporal structuring of the synthesized pulse varies across different intensities, suggesting that shortening the pulse alone does not guarantee an optimal temporal profile. These results collectively demonstrate that the

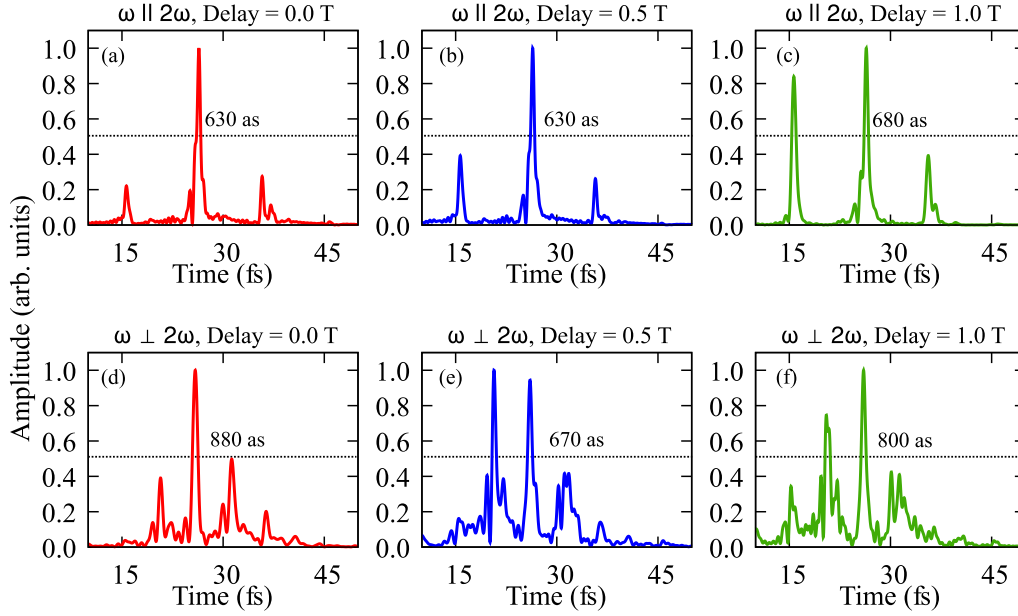


FIG. 2. Reconstructed attosecond pulses from the harmonic spectra in Fig. 1, normalized in amplitude, are shown in panels (a)–(c) at different subcycle delays in collinear polarization configuration. Panels (d)–(f) display the corresponding pulses for the orthogonal configuration. The FWHM duration of the dominant attosecond peak is indicated in each panel.

intensity of the ω pulse, the ratio $\mathcal{R}$, and the relative polarization between the two pulses must be tuned in a coordinated manner to realize an isolated attosecond pulse.

To delve deeper into the underlying mechanisms of attosecond pulse generation, we analyze the time-frequency

profiles of high-harmonics along the polarization direction for different delays. This analysis is based on the Gabor transform of the harmonic spectra presented in Fig. 1(b). While the overall time-frequency profiles exhibit similar trends in energy emission across the three delay configurations, notable differences arise that significantly influence the pulse duration and amplitude. A negative atto-chirp is observed within subcycles in Figs. 4(a) and 4(b), which favors isolated attosecond pulses. In contrast, Fig. 4(c) indicates a reduction in the negative atto-chirp, which broadens the pulse duration. Moreover, higher-energy harmonic emissions are considerably weaker for the T delay compared to the zero and 0.5 T delays. In the case of zero delay, the harmonic intensity remains consistently strong up to ~ 11 eV, contributing to the enhanced amplitude of the synthesized pulse.

Thus far, our discussion has been limited to WSMs with degenerate Weyl nodes located at the Fermi level. There exist scenarios in which the Weyl nodes are not degenerate and may lie away from the Fermi level. Moreover, WSMs can exhibit tilted, rather than upright, linear dispersions, leading to the type-II WSM phase. It is therefore important to examine whether the conclusions drawn so far regarding attosecond pulse generation remain valid under these modified conditions. Our additional simulations for a type-II WSM indicate that isolated attosecond pulses are still attainable; however, in contrast to the type-I case, the orthogonal polarization configuration proves to be more favorable than the collinear one. The collinear configuration in type-II systems results in pronounced temporal substructures in the synthesized pulses.

Figure 5(a) shows the harmonic spectra of the type-II WSM in the orthogonal configuration with $\mathcal{R} = 0.25$, along with the spectrum generated by a single ω pulse for comparison. The combined effects of a finite tilting and energy splitting between the Weyl nodes break inversion symmetry in the type-II WSM, as reflected in the band

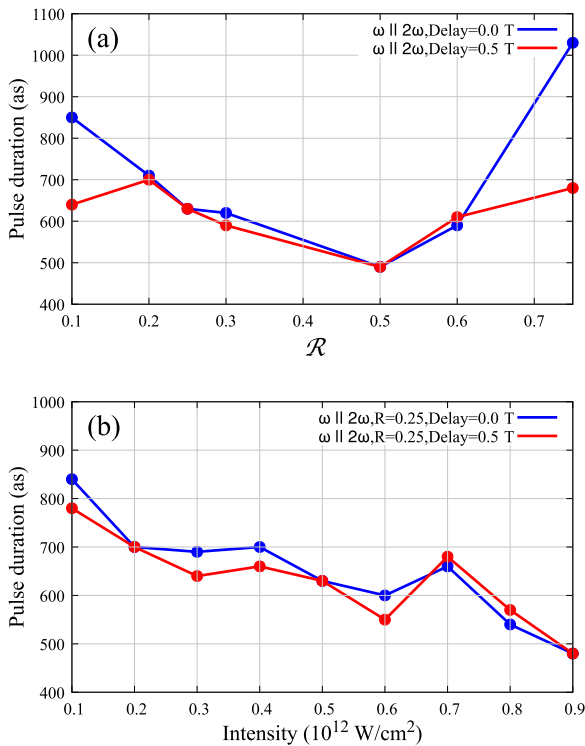


FIG. 3. Sensitivity of the synthesized attosecond pulse duration to (a) the ratio $\mathcal{R}$ in the collinear configuration at a fixed ω -pulse peak intensity of 5×10^{11} W/cm² and (b) the peak intensity of the ω pulse at a fixed ratio $\mathcal{R} = 0.25$.

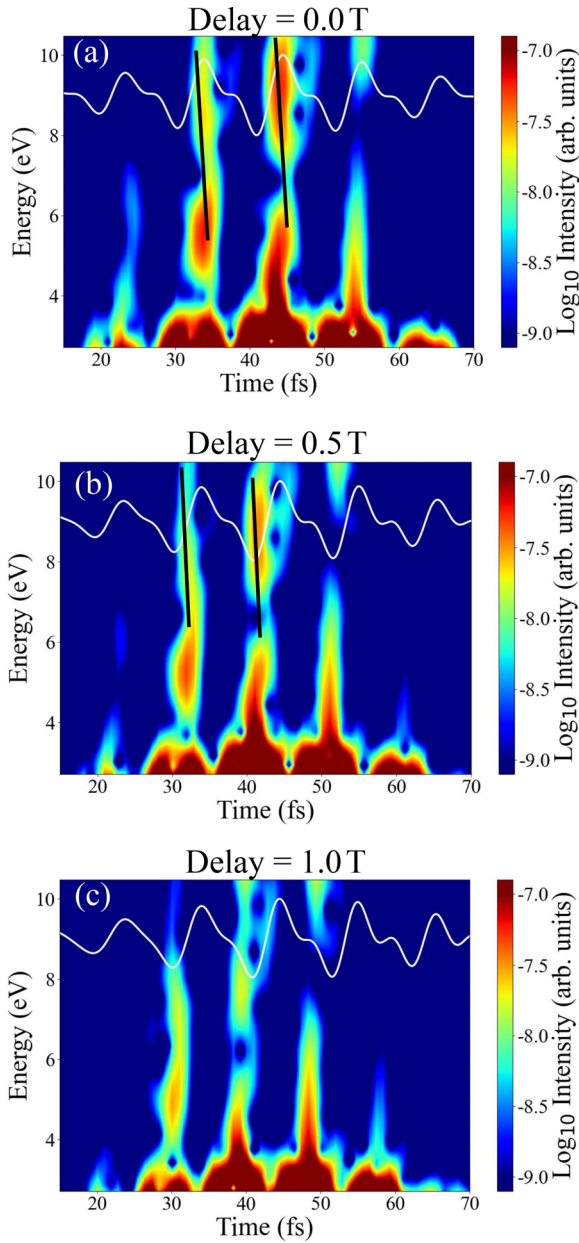


FIG. 4. Time-frequency profiles of the high-harmonic spectra in Fig. 1 for the collinear configuration at subcycle delays of (a) 0.0 T, (b) 0.5 T, and (c) 1.0 T. The corresponding $\omega - 2\omega$ electric field waveform along the polarization direction is overlaid as a white curve.

structure shown in the inset, which leads to the emergence of even-order harmonics under a single ω pulse [78]. Introducing a relatively weak 2ω field further enhances the intensity of these even harmonics. Moreover, the yield of odd-order harmonics is substantially higher than that in the type-I WSM (see Fig. 1), despite the lower driving intensity used for the type-II case (10^{11} W/cm²). However, the plateau region exhibits noticeably reduced intensity in the orthogonal configuration for the type-II WSM. Figure 5(b) displays the attosecond pulse of duration 640 as as obtained from the harmonic spectrum in Fig. 5(a). Although the pulse shows a dominant isolated peak, the reconstructed waveform still exhibits considerable temporal substructures. This indicates

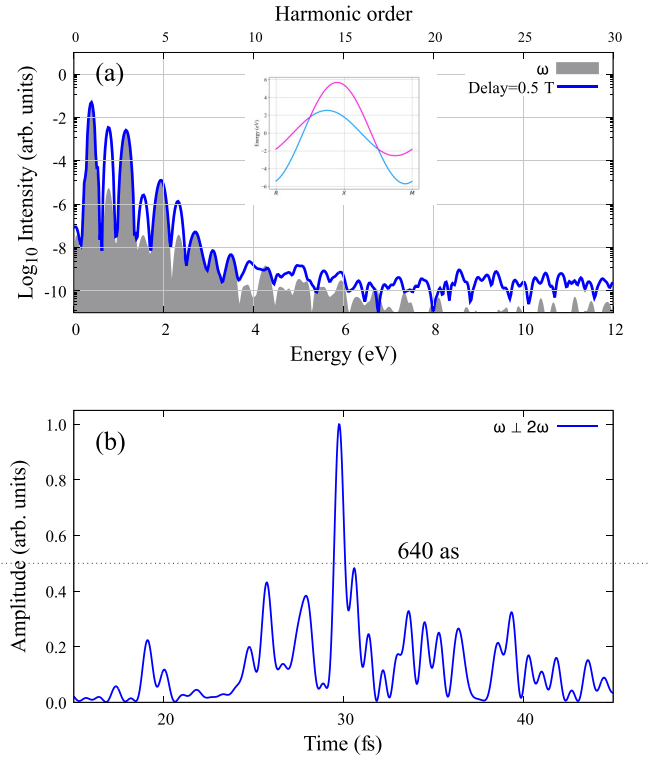


FIG. 5. (a) High-harmonic spectrum of a type-II WSM driven by an orthogonal $\omega - 2\omega$ field with a delay of 0.5 T, $\mathcal{R} = 0.25$, and an ω -pulse peak intensity of 10^{11} W/cm². For comparison, the spectrum generated by a single ω pulse is shown as the grey background. The inset illustrates the band structure of the type-II WSM for $t_c = 2$ and $t_g = 1$. (b) Synthesized attosecond pulse reconstructed from the spectrum in panel (a), with the FWHM duration of the dominant peak indicated.

that, while type-II WSMs are capable of generating isolated attosecond pulses, further optimization of both the interaction conditions and driving-field parameters is required to achieve a well-resolved isolated pulse. However, Weyl semimetal materials with smaller tilt and energy split can be viable for attosecond pulse generation [85].

IV. CONCLUSION

In summary, we studied the generation of isolated attosecond pulses in Weyl semimetals using collinear and orthogonal $\omega - 2\omega$ pulses. Weyl semimetals' harmonic spectra features broad plateaus, which have been utilized to produce isolated attosecond pulses. Furthermore, we identified that the optimal combination of polarization and relative intensity of the two-color field is required to achieve the shortest and intense attosecond pulses. We also investigated attosecond pulse generation under type-II configuration of Weyl nodes in WSMs. Our findings on attosecond pulse generation are expected to be applicable for inversion-symmetry-broken WSMs. Owing to the relatively high electron density and intrinsic periodicity of solids compared to gases, attosecond pulses from solid HHG hold immense potential for enabling attosecond pump-attosecond probe experiments on compact, table-top platforms [86–88]. Our findings therefore open another

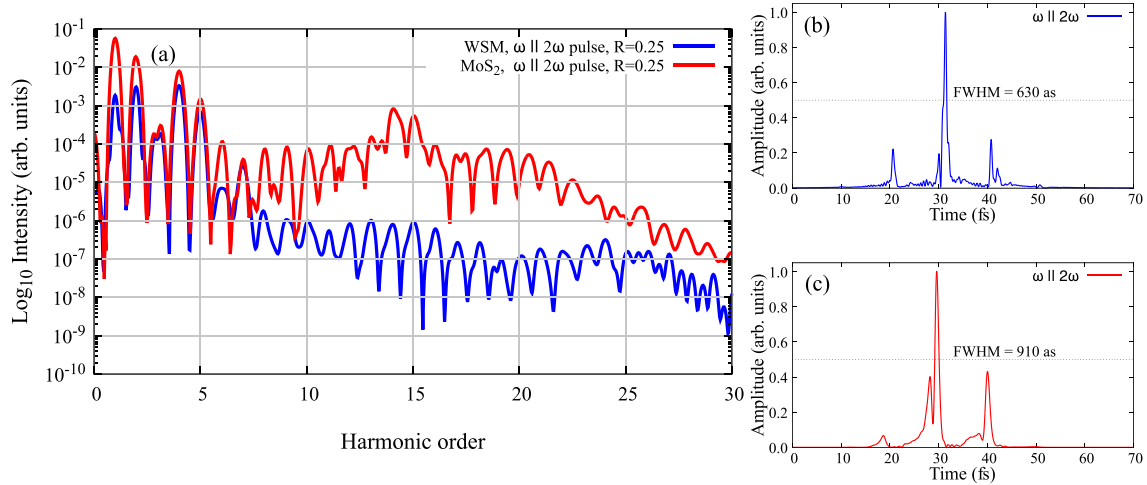


FIG. 6. (a) High-harmonic spectra from an inversion-symmetric WSM and monolayer MoS_2 driven by a $\omega - 2\omega$ field in a parallel configuration, along with the reconstructed attosecond pulses for (b) the WSM and (c) monolayer MoS_2 . The electronic structure of MoS_2 is described using a tight-binding Hamiltonian as given in Ref. [65]. A 300×300 $\mathbf{k}$ -point grid is employed for Brillouin-zone sampling, with a dephasing time of 4 fs. All other parameters are the same as those used in Fig. 1(b).

avenue for advancing attosecond spectroscopy and exploring ultrafast electron dynamics in matter.

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DATA AVAILABILITY

The data that support the findings of this article are not publicly available. The data are available from the authors upon reasonable request.

APPENDIX

In this Appendix, we compare attosecond pulse generation from inversion-symmetric WSMs with that from inversion-broken monolayer MoS_2 . It is well-known that inversion-symmetric WSMs generate exclusively odd-order harmonics when driven by a linearly polarized pulse, as shown in Fig. 1(a). However, even-order harmonics emerge when an inversion-symmetric WSM is driven by a two-color $\omega - 2\omega$ field. In contrast, monolayer MoS_2 , which lacks inversion symmetry, naturally produces even-order harmonics. As evident from Fig. 6, the attosecond burst generated from the WSM is narrower (630 as) and isolated in nature, whereas the pulse obtained from MoS_2 is broader (~ 900 as). Although the synthesized attosecond pulse from MoS_2 exhibits a higher peak strength compared to that from the WSM, the latter offers shorter isolated attosecond pulse.

It is worth emphasizing that the higher-order harmonics generated by the single- ω pulse in Fig. 1(a) seem broadened and less resolved, an effect arising from the finite pulse duration as discussed in Refs. [89–93]. Increasing the pulse length from 8 cycles to 16 cycles, where one cycle corresponds to the time period of the ω pulse (~ 10.7 fs), results in sharper and

well-resolved higher-order harmonics, as evident in Fig. 7(a). However, the harmonics generated by $\omega - 2\omega$ pulse exhibit well-resolved odd and even harmonics for eight-cycle pulse duration as well as for longer pulses, as shown in Fig. 7(b). This demonstrates that the two-color scheme is intrinsically immune against finite-pulse effects. Consequently, we have presented our results using the shorter pulse duration, which preserves the physical fidelity of the spectra while remaining computationally efficient.

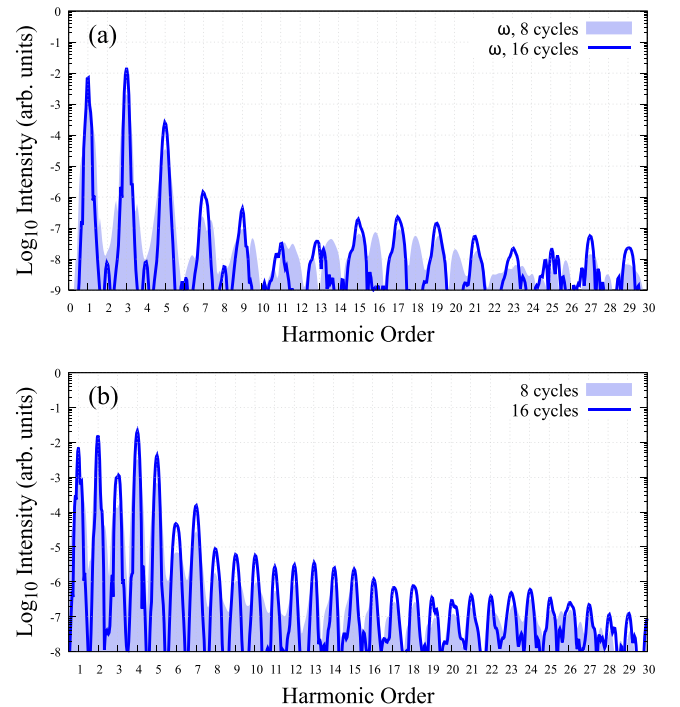


FIG. 7. High harmonics generated using (a) a single ω pulse and (b) a two-color $\omega - 2\omega$ pulse with both pulses of duration 8 cycles and 16 cycles.

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