Strong Nonlinear Optical Response and Transient Symmetry Switch in Type-II Weyl Semimetal $\beta$-WP$_2$

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1. Introduction

As topological quantum materials, Weyl semimetals (WSMs) exhibit a unique linear band dispersion in the close vicinity of the Fermi energy described by the Weyl equation. WSMs with peculiar band structures exhibit novel nonlinear optical enhancement phenomena, even for light at optical wavelengths. While many intriguing nonlinear optical effects are constantly uncovered in type-I WSMs, few experimental works focus on basic nonlinear optical properties in type-II WSMs. Here a static and time-resolved second harmonic generation (SHG) is performed on the 3D type-II WSM candidate $\beta$-WP$_2$. Although $\beta$-WP$_2$ exhibits extremely high conductivity and a high carrier density ($\approx 10^{21}$ cm$^{-3}$), the SHG is unscreened by conduction electrons, and a rather strong SHG response is observed, comparing with non-topological polar metals. Additionally, the time-resolved SHG experiment traces ultrafast symmetry switch and reveals that polar metal $\beta$-WP$_2$ tends to form an inversion symmetric metastable state after photo-excitation. Intense femtosecond laser pulse can optically drive symmetry switch and tune nonlinear optical response on ultrafast timescales although the interlayer coupling of $\beta$-WP$_2$ is very strong. These findings are illuminating for the polar metal nonlinear optics and provide a new perspective for future ultrafast topological optoelectronic devices.

Weyl nodes with left and right chirality occur in the crossing of conduction and valence bands. These nodes act as monopoles of Berry curvature in momentum space. The emergent low-energy quasiparticle excitations in WSMs are regarded as massless fermions. Unlike type-I WSMs, the Lorentz invariance is violated and the Weyl cone is tilted in the momentum space in type-II WSMs. The novel topological band structure can be manifested in intriguing electronic and optical properties in condensed matter physics.

Recently, WSMs have aroused tremendous interest for their spectacular nonlinear optical properties and potential technological applications. Some nonlinear optical phenomena, such as the giant second harmonic generation, colossal mid-infrared bulk photovoltaic effect, and chiral Terahertz wave emission have been observed in type-I WSM TaAs. While it was proposed that type-II Weyl fermions could emerge from nonmagnetic layered compound T$_3$P$_2$ and 3D compound $\beta$-WP$_2$ [8,20] and 3D compound $\beta$-WP$_2$/MoP$_2$ [21] Minority works mainly focus on their primary nonlinear optical properties. [22,23] SHG is a fundamental nonlinear optical process and a sensitive probe for tracing crystal symmetry switch [24,26] Electric-dipole (ED) allowed SHG response is expected to be dominant in the bulk crystal without inversion symmetry and the SHG intensity could reflect the magnitude of second-order susceptibility $\chi^{(2)}$, defined by the relation $P^{(2)} = 2\varepsilon_0\chi^{(2)}E_jE_k$. The magnitude of $\chi^{(2)}$ is significant in nonlinear optics and closely connected to some other nonlinear optical processes such as optical rectification and photocurrent generation.

Moreover, the possibility to manipulate matter exotic quantum phases using ultrashort light pulses has been extensively explored theoretically and was recently revealed by different experiment means in a variety of typical 2D/3D material systems including manganites, charge density wave materials, topological materials, and transitional metal oxides via impulsively driven lattice symmetry changes. However, few works have focused on the ultrafast manipulation of 3D WSMs nonlinear optical properties.

Here we report an experimental study of static and time-resolved SHG in the 3D type-II WSM candidate bulk $\beta$-WP$_2$...
single crystal. In static SHG measurement, we verify the C2v point group of β-WP2 and estimate the size of the second-order susceptibility χ(2). Although β-WP2 exhibits extremely good metallic properties with a high carrier density, we observe a rather strong SHG response at 800 nm excitation, and the maximum nonlinear susceptibility |χ(2)zxx| of WP2 is approximately the same level of the electro-optics crystal GaAs(39) and ZnTe(38). Additionally, we find that light can non-thermally drive novel transient symmetry switch on hundreds of picoseconds timescales. The maximum SHG response decreases at about 70% with 4 mJ/cm² pump fluence. It indicates that β-WP2 has tendency to switch to the inversion symmetric (topological trivial) metastable state and the topological nonlinear optical properties could be tuned by light. The ultrafast switch is accompanied by a distinct softening of the A1 phonon that is generally related to the structural instability. In previous work, light-driven interlayer shear displacements plays an essential role in 2D layered type-II WSMs Td-WTe2/XTe2 (X = W,Mo), driving a structural phase transition from Td to IT' phase.[34,35] For 3D compound β-WP2, the strength of interlayer coupling is very strong and robust, light-induced structural switch mechanism or other mechanisms should be considered. The results would have implications for optical control of the symmetry and topological properties of the transition metal phosphides and related materials.

2. Results and Discussions

β-WP2 is a 3D compound and typically needle-shaped with shiny luster (Optical micrograph of β-WP2 is shown in Figure S1, Supporting Information). Previous Raman experimental results indirectly indicate that β-WP2 crystallizes into a non-centrosymmetric orthorhombic Cmc21 (No.36) space group.[39–41] A simple illustration of the crystal structure is shown in Figure 1a. It contains a twofold screw axis along the crystalline z-axis, a glide plane perpendicular to the y-axis, and a mirror plane perpendicular to the x-axis. Comparing with the first predicted type-II WSMs candidate layered Td-WTe2/MoTe2, the interlayer coupling of β-WP2 are much stronger while crystal point group is the same as C2v. No structural or other phase transition has been observed in β-WP2 at 4–300 K range yet.[39,40,42]

Since β-WP2 is a nonmagnetic compound, inversion symmetry breaking is prerequisite for topological nontrivial band structure. It is expected to produce fundamental SHG response from ED contribution. Our model reflecting static/time-resolved SHG optical measurement setup is depicted in Figure 1b. In our setup, the WP2 sample is kept static while the polarization of incidence or outgoing beam is rotated at least. The long x-axis of WP2 xz plane is attached on the sample holder along the parallel direction. The electric field polarizations of the obliquely incident fundamental beam (in) and outgoing SHG beam (out) can be independently selected to lie either parallel or perpendicular to the crystal high-symmetry axis. The SHG response data are acquired by measuring the parallel (P-out) and perpendicular (S-out) component of SHG intensity reflected from the (010) surface of WP2 as a function of the polarization at 45° incidence beam. In crystals with C2v point group symmetry, five independent non-vanishing elements of χ(2), are χzzz, χzzx, χyyz, χyzy, χyyy. Hence rotating oblique 45° xz plane fundamental incidence polarization could simultaneously detect ED-SHG responses from all the symmetry-allowed tensor elements. The fit lobes for P-out and S-out configurations are

\[ I_{P-out}(\theta) = \frac{1}{4} |\chi_{zzz} + \chi_{yzy}|^2 \sin^2(2\theta), \]

\[ I_{S-out}(\theta) = \frac{1}{2} |\chi_{zzx} + \chi_{yyz}| \cos^2(\theta) + |\chi_{yzy}|^2, \]

respectively, where

**Figure 1.** Sample structure and characterizations for β-WP2. a) Crystal structure of β-WP2. b) Schematic of the reflection static/time-resolved SHG spectroscopy geometry, focus lens is omitted. Polarization SHG scans c) for the parallel-polarized (P-out) and d) for the perpendicular-polarized SH photons (S-out) relative to the scattering plane while the incidence wired-polarizer is rotated. The solid curves are theory fit results to C2v point group symmetry. The polarization of incidence is kept 45° and the polarization of outgoing is kept parallel as depicted in (c) with 45° incidence for fluence dependent TR-SHG measurement.
the polarization of incidence rotates at an angle \( \theta \). The results of the two scan patterns are shown in Figure 1c,d. All the solid fit curves for SHG responses derived from the \( C_{2v} \) point group. Detailed formula derivation procedure for SHG patterns can be found in Supporting Information. Our static rotation SHG results offer a very direct and explicit evidence that the point group symmetry of \( \beta\text{-WP}_2 \) is \( C_{2v} \).

The intensity of ED-SHG response reflects the magnitude of second-order nonlinear susceptibility. And the order of magnitude of \( \chi^{(2)} \) is significative and generally related to some other nonlinear optical processes such as optical rectification, difference frequency generation, and generating photocurrents. Our optical setup and measuring scheme for the estimation of \( \chi^{(2)} \) refers to previous work in TaAs.\(^{[16]} \) Since it is liable to distinguish each tensor component \( \chi^{(2)} \) contribution, the angle of incidence is set as near-normalized (11\(^{\circ}\)). We used insulating (111) GaAs wafer (\( \rho>1 \times 10^9 \ \Omega \ cm \)) and 1mm-thick (110) ZnTe archetypal electro-optic crystals as benchmarks. Both benchmark crystals have large and well-characterized SHG responses, only one non-vanishing tensor \( \chi^{(2)}_{xzy} \) at fundamental 800 nm wavelength (\( \hbar \omega=1.55 \text{ eV} \)) for their high symmetry point group (\( T_d \)). The SHG probe fluence is kept with 0.30 mJ cm\(^{-2} \) for the measurements. Surprisingly, the SHG response of \( \beta\text{-WP}_2 \) is rather strong that unscreened by conduction electrons. As seen in the Figure 2a–c, the signal intensity is normalized to the peak SHG intensity in ZnTe. The peak intensity formulas are derived in Supporting Information. In literature, the experimental refractive index of bulk GaAs, ZnTe, and \( \beta\text{-WP}_2 \) can be found. Taking \( |\chi^{(2)}_{xzy}| \) value of ZnTe as reference, by Blumberg and Pershan expression,\(^{[43]} \) we could calculate the \( |\chi^{(2)}_{xzy}| \) of \( \beta\text{-WP}_2 \) is about 150 pm V\(^{-1} \). The order of the estimated \( |\chi^{(2)}_{xzy}| \) is approximate same to the electro-optics crystal GaAs, ZnTe, and significantly larger than topological trivial polar metal LiOsO\(_3\) at 800 nm fundamental wavelength.\(^{[44]} \)

The magnitude of ED-SHG response depends on several possible factors. Generally, it is recognized that the intensity of SHG is closely-linked to the degree of the crystal polarization. So insulating ferroelectric is more ideal for strong ED-SHG response in a sense. Appearance of conduction electrons in a crystal can screen and weaken the polarization and SHG responses.\(^{[45]} \) Since \( \beta\text{-WP}_2 \) exhibits extremely high conductivity and a high free carrier density, it is unexpected to observe strong SHG signal. Meanwhile, strong SHG response was hardly observed in topological trivial polar metal such as LiOsO\(_3\).\(^{[44]} \) Obviously, the conventional displacive ferroelectric polarization perspective inadequately illuminates the strong SHG response from topological polar metal. The strong SHG response may arise from the peculiar band topology contributions in theoretical model proposed by Morimoto and Nagaosa.\(^{[46]} \) This theory deduced concise expression with explicit geometrical meaning for \( \chi^{(2)} \) and its possible relation to the existence of Weyl nodes. However, the energy of fundamental excitation at optical wavelength is away from the low energies Weyl physics. Recently, resonance-enhanced effect from other bands was proposed and consistent with their first-principle calculation.\(^{[47,48]} \) Previous infrared spectroscopy studies indicate much higher carrier densities in \( \beta\text{-WP}_2 \),\(^{[39,49]} \) thus it is reasonable to expect that the SHG response is weakened in view of carriers screening effect in ferroelectric polarization perspective when compared with type-I WSM TaAs. The origin of strong SHG response in polar WSMs is still controversial, topological band contributions, band resonance-enhanced and carriers screening effect are all possible influences for the SHG response.\(^{[16,47,48,50]} \) In \( \beta\text{-WP}_2 \), the Weyl nodes are located hundreds of meV below Fermi energy.\(^{[3,21]} \) The relative large energy scale provides an ideal platform for studying the strong topological nonlinear optical response. Since the value of \( \chi^{(2)} \) is dependent on the frequency, more theoretical and band structure based calculation are needed to explain the attractive strong topological nonlinear optical responses in type-I and type-II polar WSMs.

Symmetry plays a central role in conventional and topological phases of matter, intense femtosecond laser pulse can optically induce transient symmetry change and make novel metastable or hidden states in some quantum materials.\(^{[32,51–53]} \) Electric dipole allowed SHG arises from a non-zero second-order susceptibility \( \chi^{(2)} \), as shown in non-centrosymmetric topological systems.\(^{[16,22,46]} \) Thus, time-resolved SHG measurement is a very sensitive technique for monitoring ultrafast transient symmetry switch processes in these topological materials. Our optical setup for tr-SHG measurements is shown in Figure 1b. The pump pulse is normal to the \( x-z \) plane, the polarization of incidence and reflecting probe pulse is set to make the static SHG response at maximum value, with 45° incidence P-out configuration. (The polarization direction of incidence is depicted in Figure 1c.)

After the femtosecond pump pulse arrives (\( \Delta t = 1.5 \text{ ps} \)), the SHG intensity decreases saliently. Figure 3a shows the measured SHG time traces from WP\(_2\) at various pump fluences. At

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**Figure 2.** Benchmark static SHG measurements. Benchmark 11° incident 800 nm SHG experiments on a) (110) ZnTe, b) (111) intrinsic insulating GaAs wafer (\( \rho>1 \times 10^9 \ \Omega \ cm \)), and c) (010) \( \beta\text{-WP}_2 \), the signal is normalized to the maximum SHG intensity of ZnTe.
a rather low fluence excitation (about 0.5 mJ cm\(^{-2}\), red curve), the time-dependent SHG intensity begins to show a remarkable reduction. When increasing pump fluence to 4.0 mJ cm\(^{-2}\), the SHG intensity plummets drastically, about 70\% reduction as shown in Figure 3b. Continue to increase pump fluence until the damage threshold, the value of \(\Delta I_{\text{max}}/I\) keeps almost unchanged. In order to cross-check whether there are irreversible modifications, the fluence-dependent measurements are also performed from high to low fluence (see Figure S3, Supporting Information). No distinct difference can be identified, indicating the transient effect is reversible. And the much longer timescales intensity variation is shown in inset of Figure 3c. The whole decay process persists over one hundred picoseconds and slowly relaxes to the initial state before the next pump pulse arrives, consisting of metastable state behavior. Figure 3c shows the temperature-dependent relative change of the SHG intensity at fixed pump fluence of about 2.5 mJ cm\(^{-2}\) as a function of the delay time. Rather slight maximum SHG intensity reduction difference of temperature variations in Figure 3c and no high temperature structural transition was observed, manifesting non-thermal symmetry switch process. Varying the linear polarization of the excitation pulse in the \(xz\) plane relative to the x-axis as shown in Figure 3d, no distinct amplitude change could be found in \(\Delta I_{\text{max}}/I\). The results suggest the tr-SHG responses are almost isotropic in the pump polarization. Comparing with the previous work in layered T\(_{\beta}\)-WTe\(_2\), light with sufficient electric field strength can drive it to a completely topological trivial state, indicating almost no SHG response.\(^{[34]}\) Noting that the SHG intensity saturates to a nonzero value ultimately. Such residual signals are commonly observed across photoinduced phase transitions.\(^{[33,35,54,55]}\) They may be attributed to extrinsic/intrinsic reasons such as spatial nonuniformity of the pump intensity, the saturated pump light absorption or strong interatomic bonding prevents atoms moving to the entire inversion symmetry positions with SHG response decreasing to almost undetectable. Based on our results, we conclude that the crystal has strong tendency to the inversion-symmetric state.

Following near-infrared (1.55 eV) light excitation, Figure 4a,b shows pronounced SHG signal changes in the two polarization scan lobes before/after the pump pulse arriving. The overall SHG intensity of all lobes at 10 ps shows a mutual tendency to decrease with the maximum SHG intensity variations occurring in the maximum static SHG positions (such as 45\° for P-out and 0\° for S-out ). Under 2 and 4 mJ cm\(^{-2}\) pump excitation, the maximum SHG intensity decreases nearly 38\% and nearly 64\% for the S-out setup, respectively. As the P/S-out SHG scan lobes in Figure 4a,b show, the SHG polar patterns are still consistent with the assumed equilibrium C\(_{2v}\) point group symmetry fits at 10 ps under both 2 and 4 mJ cm\(^{-2}\) pump fluence but with much less SHG response intensity. Based on the above results, the dominant SHG tensor element \(|\chi^{(2)}_{xxz}|\) in WP\(_2\) can be deduced to a large reduction of nearly 80\%. Considering the pump electric field polarization and perturbations are in the \(xz\)-plane, the results suggest a possible connection between the polar axis and the strong optical nonlinearity of the \(\beta\)-WP\(_2\). The snapshots of SHG polarization patterns also indicate the specific reversible ultrafast symmetry switch process. In a word, an intense laser pulse could manipulate the nonlinear optical properties drastically in WP\(_2\) in an ultrafast fashion.
In general, the ED-SHG response depends on the lattice symmetry so that it is often taken as a sensitive tool to detect the structural symmetry change of a compound such as GaAs. It is natural to take the structural change into account for the transient symmetry switch. Interlayer shear displacements could be driven by intense light in layered $\text{T}_\text{d}$-**$\text{XTe}_2(\text{W,Mo})$**, resulting in SHG decreasing. And the intensity of low-frequency shear mode phonon would recede in a time-resolved reflectivity experiment with the structural transition. However, previous Raman results of $\beta$-**$\text{WP}_2$** point that no such low-frequency shear motion phonon exists. In order to get further insight into the latent switch mechanism, we also carry out the fluence-dependent ultrafast optical pump-probe reflectivity experiment to investigate coherent phonon oscillations. The data under different pump fluences are shown in Figure 5a, the positive signal background before time-zero comes from stray pump light and has little impact on the phonon frequency. All the decay processes can be fit by a double-exponential function:

$$\Delta \frac{R}{R} = A_1 \exp\left(-t/\tau_1\right) + A_2 \exp\left(-t/\tau_2\right) + C,$$

where $A_1, A_2$ in the formula represents the amplitude of the photo-induced reflectivity change, $\tau_1, \tau_2$ stands for the two fast relaxation time, and $C$ is constant for long lived thermal diffusion process. After subtracting the background decay processes, only one $A_1$ phonon mode could be extracted from time domain coherent oscillation by Fourier transform over the entire fluence range. This $A_1$ mode phonon frequency is about 5.4 THz ($\approx 180 \text{ cm}^{-1}$) in Figure 5b under 500 µJ cm$^{-2}$ pump excitation, which is in accord with the previous static Raman scattering measurements. The dependence of the phonon frequency on the pump fluence is depicted in Figure 5c. With the pump fluence further increasing, the $A_1$ phonon frequency in Figure 5c shows continuous red-shifted from 5.4 to 5.1 THz ($\approx 5\%$ frequency red-shifted than equilibrium state), indicating a distinct phonon mode softening behavior. Similar softening behavior has been observed in fluence-dependent transient reflectivity experiments of tellurium (Te), bismuth (Bi), and tin selenium (SnSe) crystals before. This behavior could be generally regarded as evidence for the structural instability.

The $A_1$ phonon frequency is relevant to the atom positions in previous theoretical and experiment results. In the W atoms displacements along the $A_1$ phonon coordinate (see Figure S3, Supporting Information), the lattice tends to distort, resulting to form the hypothesis inversion symmetry (IS) monoclinic phase from IS breaking orthorhombic structure. As the result, the polar degree of the lattice would be weakened accompanied with SHG intensity decreasing. SHG is a sensitive tool to reflect the polar degree of the crystal, so the softening of $A_1$ phonon could be considered as an indirect evidence for supporting the structural instability. Some other mechanisms have also been proposed for SHG decreasing. In semiconductor metasurfaces, the transient SHG decrease is caused by electronic unbalance and band filling effects. Free-carriers injection by light causes permittivity dramatic change. Some qualitative discussions

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**Figure 4.** Snapshots of transient SHG patterns. With a linearly-polarized pump excitation, the SHG patterns in P/S-out configurations with 45° probe incidence for various delay time and pump fluence. As shown in (a,b), the maximum intensity of polar patterns reveals pronounced decreasing at 10 ps while the pattern shapes are almost unchanged. All the pattern fits assuming the crystal point symmetries of $C_\text{2v}$ are shown as solid curves.

**Figure 5.** The fluence-dependent coherent vibrational dynamics. a) The transient pump-induced reflectivity at 800 nm with 0.5–3.5 mJ cm$^{-2}$ fluence. b) Fourier transformation of the oscillation after subtracting the background. c) The pump fluence dependence of $A_1$ phonon mode frequency.
that rule out these electronic mechanisms can be found in Section S6, Supporting Information. In this work, we tentatively propose that a structural symmetry switch makes transient SHG decreasing. However, time-dependent optical reflectivity just monitors the $A_2$ vibration mode and can not provide direct messages about atomic positions. Detailed time-resolved X-ray diffraction is necessary to elucidate the dynamics of the atomic positions. Relevant ab initio calculation to simulate atom positions/lattice structural evolution following impulsive optical excitation is worth pursuing in the future.

3. Conclusion

In summary, we performed combined static and time-resolved SHG measurements in Type-II Weyl semimetal $\beta$-WP$_2$. As a topological polar metal with high carrier density, WP$_2$ exhibits rather strong SHG responses with maximum $|\chi^{(2)}|$ about 150 pm V$^{-1}$, much larger than topological trivial polar metal LiOsO$_4$ with 1.55 eV photon excitation. The static SHG results enrich the visions of intriguing polar metal nonlinear optics and enable potential topological optoelectronic devices. After photo-excitation, intense laser pulse could optically cause novel symmetry switch and tune nonlinear optical responses on ultrafast timescales. The results indicate that $\beta$-WP$_2$ has strong tendency to the topological trivial state. We tentatively propose that driving mechanism is attributed to the structural switch. Our study opens up experimental possibilities for ultrafast optical manipulation of the topological nonlinear optics properties of solids. Further time-resolved X-ray diffraction and ab initio calculation should be pursued for the transient lattice and related topological electronic structure switch in future.

4. Experimental Section

Crystal Growth and Structure Characterization: Single crystals of $\beta$-WP$_2$ were grown via chemical vapor transport with iodine as the transport agent. All capillary processes were operated in a sealed glove box. Detailed growth method can be found in ref. [39]. The as-grown 3D crystals with needle-shape exhibited multiple shiny surface facets. $\beta$-WP$_2$ is air stable. Crystal structure and crystal orientation were confirmed using single crystal X-ray diffractometer (SXRD) Bruker D8 Venture with Mo-K$_\alpha$ radiation ($\lambda = 0.71 \text{Å}$) at room temperature. The determined lattice parameters were consistent with the previous report in the literature.

Optics Set-Up for Reflecting Static/Time-Resolved SHG and Pump-Probe Measurement: The optics set-up for measuring reflecting static/ time-resolved SHG is depicted in Figure 1b. The measurements were performed in a Ti:sapphire amplified laser (Spitfire Ace) system of 800-nm center-wavelength pulses for a duration of 35 fs (1 kHz repetition rate). Generator pulses passed through a mechanical chopper that provided amplitude modulation at 377 Hz. And the pulses were focused at 11°-45° incidence onto the sample. The spot sizes of pump and probe were measured at about 150 and 120 μm in 45° incidence configuration by a 100-μm diameter pinhole, respectively. The pump was in normal incidence relative to $xz$ plane with a half waveplate (808 nm, zero-order) to vary linear polarization. The probe fluence of the time-resolved SHG was set as 0.30 mJ cm$^{-2}$. A quarter waveplate (808 nm, zero-order) and a rotatable wire-grid polarizer in the incidence beam path were used to generate circular polarization and to vary the direction of linear polarization. Both the outgoing fundamental and the second-harmonic beams were directed to a short-pass, band-pass filter group that ensured only the second-harmonic light reached the photomultiplier tube photodetector (PMM01, Thorlabs, Bias voltage set as 0.65 V). Another rotatable wire-grid polarizer placed before the PMT allowed for analysis of the polarization of the second-harmonic beam. Temperature-dependence measurements were performed by mounting the WP$_2$ sample on a holder of helium-free closed-cycle cryostat (Montana Instruments). Benchmark measurements on $\beta$-WP$_2$, ZnTe, and GaAs were performed in ambient conditions and the samples were mounted flat on an xyz-microrometer stage to maximize the signal at room temperature.

For the fundamental time-dependent reflectivity measurement, a standard pump-probe scheme can be found elsewhere.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

second harmonic generation, ultrafast symmetry switch, Weyl semimetals

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