Helicity-dependent THz emission induced by ultrafast spin photocurrent in nodal-line semimetal candidate Mg$_3$Bi$_2$

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Helicity-dependent ultrafast spin current generated by circularly polarized photons in topological materials holds the crux to many technological improvements, such as quantum communications, on-chip communication processing and storage. Here, we present the manipulation of helicity-dependent terahertz emission generated in a nodal line semimetal candidate Mg$_3$Bi$_2$ by using photon polarization states. The terahertz emission is mainly ascribed to the helicity-dependent photocurrent that is originated from circular photogalvanic effects, and the helicity-independent photocurrent that is attributed to linear photogalvanic effect. Our work will inspire more explorations into novel nodal line semimetals and open up new opportunities for developing ultrafast optoelectronics in the topological system.

Keywords: terahertz; spin photocurrent; nodal-line semimetal; topological material

Introduction

Generation and control of an ultrafast spin-photocurrent in topological materials are of great significance$^1$. On the one hand, the spin-photocurrent generated in a picosecond timescale permits ultrafast optical manipulation$^2$. Proverbially, efficient devices could be realized for data storage and processing if the spin degree of freedom is utilized$^3$. However, the processing of information calls for the generation and control of spin current in transient time to achieve ultrafast operations, especially at frequencies coming up to the undeveloped terahertz band$^4$. Fortunately, the unique chiral behavior of spin photocurrent in topological materials permits the ultrafast operation under circularly polarized light without an external bias field$^5$. On the other hand, the spin-photocurrent induced electromagnetic wave in the terahertz frequency domain has been proven to be an efficient helical terahertz (THz) emitter$^6,7$. It is worth mentioning that the generation and manipulation of helical THz pulses in the past mainly depend on the complicated pulse shaping or two color manipulation of the incident laser while emitters possess no intrinsic optical chirality$^8$. Therefore, the ability to explore the topological materials with peculiar spin-momentum locking or chiral properties is of crucial importance.

Nodal-line semimetals (NLSs), which host one-dimensional closed-loop or line degeneracies formed by the crossing of two bands, have been proposed recently as a family of topological phase materials$^{9-11}$. The
distinguishing characteristic of NLSs is their two
dimensional (2D) drumhead-like surface states, which
are embedded into the 'direct gap' between conduction
and valence band in the 2D projection of the nodal
ring9,12–14. Up to now, NLSs have been intensively studied
in many compounds such as PtSn415, Cu3PdN14, PbTaSe211,
TTaSe215, MgBi216. Among them, MgBi2 is a significant
thermoelectric material with excellent performance in the
process of energy conversion17. The good thermoelectric
performance of MgBi2-based materials may be closely
related to its topological properties20. Mg3Bi2 is predicted
to be NLSs in the absence of spin-orbit coupling (SOC)
and become a strong topological insulator when SOC is
considered16. Taking SOC into consideration in the
calculation, the drumhead surface band would split and
transform into topological surface states, with a small
bulk energy gap of about 35 meV opening at the line
nodes16. However, the topological properties can hardly
be enclosed by experiments due to the heavily p-doped
Mg vacancies16,18. Thus, Mg3Bi2 is usually called as a
candidate for NLSs. Fortunately, a recent work has
revealed that Mg3Bi2 is a semimetal with topological
surface states by in-situ angle resolved photoemission
spectroscopy (ARPES)18, and further investigation for the
optical properties of Mg3Bi2 is still in need.

In this paper, we exploit the chiral-controlled spin
photocurrent in nodal line semimetal candidate Mg3Bi2
under the photon injection, we found the polarity and
chirality of the photocurrents can be manipulated by
polarized near-infrared optical pump on a picosecond
timescale. In addition, we demonstrate that the chirality-dependent terahertz emission is mainly
originated from the circular photogalvanic effect (CPGE),
which is associated with the angular momentum
selection rules of topological surface states.

Results and discussion

Materials growth and characterization

Mg3Bi2 is a layered Kagome lattice structure with the
space group of P3m1, as shown in Figs. 1(a) and 1(b).
Mg and Bi atoms assemble in layers naturally with five
atomic layers stacking along the (001) crystal
orientation18. Mg3Bi2 thin films were grown on the
transparent sapphire (Al2O3) substrates by using
molecular beam epitaxy. A Se capping layer is essential to
protect the Mg3Bi2 film from oxidation in the atmosphere
and prevent the thin film from the evaporation of
pulsed-laser illumination19. The details of sample
preparation are discussed in the supplementary
information. Figures 1(d) and 1(e) display the in-situ
RHEED (reflection high-energy electron diffraction)
patterns along $\bar{\Gamma}-\bar{\bar{K}}$ and $\bar{\Gamma}-\bar{\bar{M}}$ directions, and the sharp
streaks indicating the high quality of the film. XRD
spectrum in Fig. 1(f) shows the (001), (002), (003), (004),
and (005) diffraction peaks of Mg3Bi2. In-plane lattice
constant deduced from the RHEED streak spacing is

![Fig. 1](image-url) | Characterization of the Mg3Bi2 film. Crystal structure of Mg3Bi2 from (a) side view and (b) top view. (c) The bulk and the projected (001)
Brillouin zones of Mg3Bi2. RHEED patterns of the Mg3Bi2 film grown on the sapphire substrate, with the incident electron beam along the (d) $\bar{\Gamma}-\bar{\bar{K}}$
and (e) $\bar{\Gamma}-\bar{\bar{M}}$ directions, respectively. (f) XRD patterns of the Mg3Bi2 film, with all the (001), (002), (003), (004), and (005) diffraction peaks marked.

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about 0. 4763 nm, which is in good agreement with the results from references18,20.

Terahertz emission analysis

Terahertz time-domain spectroscopy (TDS) is a crucial method to confirm the properties of topological states and the dynamics of charge carriers on a sub-picosecond timescale7,19,21–25. In the process of exploring topological properties, it is significant to separate the surface signals from the bulk backgrounds26–28. Fortunately, it has been verified that circularly polarized light could give rise to the asymmetric depopulation of spin-polarized surface states by optical selection rule and then generate a spin-polarized photocurrent6,19,26,29,30. This transient process is known as the circular photogalvanic effect and can be detected by a femtosecond laser pulse. Based on Maxwell equations, the transient variation in the current density on picosecond timescale will generate electromagnetic radiation with the frequencies located in the THz region (1 THz =1 ps⁻¹), which can be described by

\[ E_{\text{THz}} \propto \frac{\partial}{\partial t} J. \]  

Here, \( E_{\text{THz}} \) represents the radiated electric field, and \( J \) expresses the transient current31,32. The schematic view of this process can be described in Fig. 2.

Fig. 2 | Schematic view of helicity-dependent terahertz radiation process.

In addition, THz spectroscopy is a viable contactless alternative that can complement conventional transport measurement without the necessity for electrodes or background signal that may disturb the system29,33. This is especially important for the study of chiral-photocurrent in a topological system, and this technique has recently provided valuable information in topological insulators such as Bi2Se324,30, Sb2Te319, as well as in Dirac and Weyl semimetals, like graphene12,34, TaAs35 and so on. Besides, the chirality-dependent photocurrents resulting from TSSs can be controlled by the polarization of the incident light, which demonstrates the potential for optoelectronics19,30,36. To date, no dynamical behavior of ultrafast photocurrent and its associated electromagnetic wave has been explored in Mg3Bi2, even in nodal line semimetals.

The schematic of the experimental setup is shown in Fig. 3(a) and the diagram of our terahertz emission system is depicted in Fig. S1. The Mg3Bi2 thin film was set up in the transmission direction of the pump light. The optical pulses with a central wavelength of 800 nm (1.55 eV) were focused on the crystal surface at an incident angle of \( \theta \) to generate ultrafast photocurrent and then the terahertz wave was radiated. The vertical and horizontal components (along the \( E_x \) and \( E_y \) direction) of the emitted THz pulses were detected by electro-optical sampling, respectively. The polarization and chirality of optical pulses are controlled by rotating a quarter-wave plate (QWP) with an angle of \( \alpha \). The impact from bulk transient currents of Mg3Bi2 may contribute to the terahertz emissions in out-of-plane (perpendicular to the surface of the samples) direction, and these can be canceled by polarization settings of terahertz detection19.

The helicity-dependent terahertz emissions along the vertical direction (\( E_x \)) were measured by rotating a quarter-wave plate with an angle of \( \alpha \) at incident angle \( \theta = -45^\circ \) and \( +45^\circ \), respectively, which were shown in Fig. 3. The polar plots of terahertz waveforms (0~4.5 ps) in Figs. 3(b) and 3(c) illustrate a clear polarity reversal at different helicity of optical excitations. According to the polarization trajectory, the polarization and chirality of the THz wave can be manipulated by the switching of circular or linear polarization of the incident light. Such controllability stems from the chiral ultrafast photocurrents derived from the polarized femtosecond optical pulses. Twofold symmetry was observed in the \( \alpha \)-scan patterns, which gives more profound insights into the origins of terahertz emissions.

Figures 3(d) and 3(e) show the time domain terahertz waveforms of the Mg3Bi2 thin film for incident angle \( \theta = -45^\circ \) and \( +45^\circ \) respectively, with linearly polarized (LP, \( \alpha = 0^\circ \), red line), right-hand circularly polarized (RHCP, \( \alpha = 45^\circ \), black line), and left-hand circularly polarized (LHCP, \( \alpha = 135^\circ \), blue line) optical excitations. The key observation is that signals obtained from the right and left-handed circularly polarized light are completely in opposite phase for both \( \theta = -45^\circ \) and \( +45^\circ \), and for helicity-fixed optical excitation (both RHCP and LHCP),
a similar polarity reversal was also found when the polarized light incident angle $\theta$ alternates from -45° in Fig. 3(d) to 45° in Fig. 3(e). This phenomenon usually results from the reversed incident photon helicity. THz signals from the linearly polarized light were also in the opposite phase for $\theta = -45°$ and $+45°$. Obviously, both the magnitude and temporal shape of the THz waveform depend strongly on the light polarization and the incident angle. These results are consistent with the scenario for the helicity-dependent terahertz emission: the spin-polarized current generated by incident photon polarization is the major contributor to this process\textsuperscript{19,30–32,35}.

To further comprehend the origin of THz emission in MgB\textsubscript{2}, it is essential to figure out the mechanisms behind the generation of time-resolved photocurrent. We

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**Fig. 3** | (a) Schematic of the THz emission configuration. Polar plots of terahertz waveforms (0~4.65 ps) as a function of $\alpha$ at (b) $\theta = -45°$ and (c) $+45°$, respectively, for $\varphi = 0°$. The colors represent the amplitude of the terahertz emissions. Terahertz waveforms for excitation with left-hand circularly polarized, linearly polarized, and right-hand circularly polarized optical pulses at (d) $\theta = -45°$ and (e) $+45°$, respectively.
extracted the $\alpha$-dependent THz amplitudes at the specific moment ($t = 2.46$ ps), which shows the largest variation of light response and can be controlled by rotating the QWP by an angle of $\alpha$, as shown in Figs. 4(a) and 4(b). Based on the description of helicity-dependent photocurrents in topological insulators, the terahertz electric field can be inferred as $E_{THz}(t) \propto \partial f(\alpha,t) / \partial t$. Thus, the $\alpha$-dependent peak values can be well fitted using the equation as follows:

$$E_{\text{THz}(\alpha)} = C \sin(2\alpha) + L_1 \sin(4\alpha) + L_2 \cos(4\alpha) + D. \quad (2)$$

In this equation, the coefficient $C$ represents the contribution from helicity-independent circular photogalvanic effect. $L_1$ describes the helicity-independent linear photogalvanic effect. It has been reported that both $C$ and $L_1$ are related to the topological surface states, and they are predicted to be associated with the Berry phase in spin-orbital coupled quantum well structures theoretically. It is worth noting that the LPGE depends on the crystal symmetry or the linear polarization of light and is generally only excited on the surface of the sample. $L_2$ represents the photon drag effect (PDE), which is linked by the linear momentum transfer between incident photons and electrons. Optical rectification (OR), as a second-order nonlinear optical process, is the major process to coefficient $D$, which is following the well-known second-harmonic generation in noncentrosymmetric materials.

Based on our results, the amplitude and phase of $E_{\text{THz}}$ are dominated by $C$ (CPGE, 34.87%) and $L_1$ (LPGE, 51.72%), which were revealed in Figs. 4(c), 4(d) and Fig. S2. The extracted CPGE component of the emitted THz electric field follows a sinusoidal dependence of $2\alpha$ whose periodicity matches a change for the spin direction of the incident light. However, the LPGE part shows a $4\alpha$ periodicity because it has no response to the helicity of the incident ray. Both of them are coincident with the rotational symmetry of the Dirac cone of TIs. Another $\sim 11.46\%$ component is from the polarization independent optical rectification effect, while PDE can be omitted (only 1.95%). The above results indicate that the THz emission is generated by the topological surface

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**Fig. 4** (a) and (b) represent the $\alpha$-dependent terahertz amplitude at $t = 2.46$ ps. The red solid lines are the best fit with Eq. (2). The symbol $\nearrow$, the counterclockwise arrow $\nearrow$, and the clockwise arrow $\nearrow$ denote linearly polarized (black: $\alpha = 0^\circ$), left-hand circularly polarized (pink: $\alpha = 45^\circ$), and right-hand circularly polarized (green: $\alpha = 135^\circ$) incident photons, respectively. (c) and (d) display the $\alpha$-dependent coefficients $C$ (red line), $L_1$ (blue line), $L_2$ (orange line), and $D$ (blue line) extracted [using Eq. (2)] individually from (a) and (b).
states of the Mg₃Bi₂ thin film.

Terahertz signals in the \( E_y \) direction appear in a polarization-independent manner, which are substantially different from \( E_x \), as shown in Fig. S3. On the one hand, signals in \( E_y \) direction are relatively weak compared with that in \( E_x \) direction. On the other hand, according to the fitting result, \( E_y \) is dominated by a polarization-independent \( D \). \( L_2 \) plays a minor role, while \( C \) and \( L_1 \) can be neglected. These results suggest that the helicity-controlled terahertz emission mainly results from the polarization-dependent photocurrent in \( E_x \) direction, rather than the polarization-independent photocurrent in \( E_y \) direction. Such distinct features in \( E_x \) and \( E_y \) directions may lead to an elliptically polarized transient THz field.

As another controllable measurement, we explore the relationship between the THz emission response and the power of incident optical pulses centered at 800 nm, with the polarization state fixed to right-hand circular polarization. It is apparent that the increase of the laser power augments the amplitude of the emitted THz waves but does not change the shape of the waveform, as shown in Fig. 5(a). Through fast Fourier transformations (FFT) of the THz waveforms, Figure 5(b) displays the frequency domain that covers the frequency range up to 2 THz. On the other hand, the peak value for all of the spectra is centered around 0.55 THz, which is in very good agreement with the results for the time-domain analysis. Additionally, as illustrated in Fig. 5(c), a linear behavior is indicated in the dependence of the THz peak amplitude on the incident pump power up to 1800 \( \mu \)J/cm². Figure 5(d) displays the azimuthal-scan (\( \varphi \)-scan) results for the peak-to-peak amplitudes of the terahertz radiations with linearly polarized optical pulses at nearly normal incidence (\( \vartheta=0º \)) from Mg₃Bi₂ thin films. The azimuthal scan strongly depends on the crystal structure of samples. Obviously, Mg₃Bi₂ owns a centrosymmetric lattice structure for bulk, but the inversion symmetry of the top and bottom surface is broken. We deduce that the anisotropy of the azimuthal-scan results is related to the broken inversion symmetry of the surface.

To shed more light on the emission mechanism, terahertz radiation arising from the linearly polarized light can be further investigated. A half-wave plate is used to clarify the relationship between \( E_{\text{THz}} \) with the linear polarization angle \( \alpha \). The incident angle \( \vartheta \) is fixed at 45º. The relationship between pump polarization angle and

![Fig. 5](https://example.com/fig5.png)

Fig. 5 | (a) THz waveforms at different photoexcitation power with the wavelength center in 800 nm. (b) Fast Fourier transformations (FFT) spectra for the directly measured terahertz waveforms that are shown in (a). (c) THz amplitude versus the power of the incident laser beam. (d) The azimuthal dependent (\( \varphi \)-dependent) absolute amplitude of THz emission waveforms with linear polarized optical pulses.
terahertz amplitude near the peak values is shown in Fig. 6(a). We found that $E_{\text{THz}}$ induced by the linearly polarized light can be well described by a sinusoidal function with a period of 180°. The amplitude of the wave excited by linearly polarized light is approximately three times smaller than that of the wave excited by circularly polarized light, compared with Figs. 4(a) and 4(b). When the polarization angle of the pump beam $\alpha$ is changed from 0° to 90°, the polarity of the emitted THz pulse is reversed, as shown in Fig. 6(b). No THz emission is detected at a polarization angle of $\alpha = 45°$. This phenomenon can be described by a second-order nonlinear optical process. The different intensity and period of terahertz signals excited by linearly or circularly polarized light further verified the dominant role of topological surface states in spintronic terahertz emission.

Summary and discussions

In conclusion, for the first time, we demonstrated that spin photocurrent induced helicity-dependent terahertz emission could be realized in nodal-line semimetal candidate Mg$_3$Bi$_2$. The polarization directions and magnitudes of helical THz wave can be easily controlled without any THz waveplate. The polarization control is mainly originated from polarity-dependent photocurrent, and then can be effectively manipulated by circular photogalvanic effect. Such an intrinsic spintronic emitter shows promising characteristics and is easily accessible. Finally, this work will open up intriguing opportunities for fundamental studies of novel THz emission, THz spintronics, and THz modulation devices by using topological materials.

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Author contributions
M. Y. Tong and Y. Z. Hu contributed equally to this work. J. Tian and M. Y. Tong conceived the idea and designed the research; M. Y. Tong and X. N. Xie fabricated all the samples; Y. Z. Hu performed the optical measurement. T. Jiang, Z. Y. Wang, X. A. Cheng and X. G. Zhu analysed the data; M. Y. Tong and Y. Z. Hu co-wrote the manuscript. All authors discussed and commented on the manuscript.

Competing interests
The authors declare no competing financial interests.