

Topological materials as promising candidates for tuneable helicity-dependent terahertz emitters

Jessica L. Boland^{*a}, Djamshid A. Damry^a, Chelsea Q. Xia^b, Yahya Saboon^a, Abdul Mannan^a, Piet Schönherr^b, Dharmalingam Prabhakaran^b, Laura M. Herz^b, Thorsten Hesjedal^b, Michael B Johnston^b.

^a Photon Science Institute, Department of Electrical and Electronic Engineering, University of Manchester, Manchester M13 9PL, U.K;

^b Department of Physics, University of Oxford, Clarendon Laboratory, Parks Road, Oxford OX1 3PU, U.K.

ABSTRACT

Topological materials have rapidly gained interest as contenders for development of coherent, controllable terahertz emitters. Possessing Weyl nodes either at the surface or within the bulk, they host spin-polarised, helicity-dependent currents that offer possibility to control the emitted THz pulse by changing the polarization of the optical pulses generating the radiation. Here, we show that upon near-infrared excitation at oblique incidence, multi-cycle pulses are generated with a narrow bandwidth of ~ 0.4 THz for cadmium arsenide bulk crystals and nanowire ensembles. Both the bandwidth and peak emission frequency of the generated THz radiation can be tuned by respectively varying the photon helicity and angle of incidence of the photoexcitation light.

Keywords: terahertz, emission, Dirac semi-metals, nanowires, helicity-dependent, topological materials

1. INTRODUCTION

Terahertz (THz) emitters with tuneable peak emission frequency, bandwidth and polarization control are essential for several applications, including imaging, security, 6G communication and sensing. Topological materials, in particular, have emerged as promising candidates for a new range of THz sources [1]. They have already been shown excellent device performance as broadband photodetectors and THz modulators [2]. These topological materials all possess Weyl nodes, which host topological-protected massless Dirac fermions. In topological insulators (TIs), these nodes occur at the surface, so that the material is insulating/semiconducting in the bulk but highly conducting at the surface. In Weyl semi-metals (WSMs) and Dirac semi-metals (DSMs), the Weyl nodes are within the bulk of the material, so a linear, gapless energy-momentum dispersion is observed in 3 dimensions. For WSMs, the nodes have opposite chirality, whereas they are degenerate for DSMs. However, in all cases, they can host spin-polarized, helicity-dependent photocurrents due to spin-momentum locking, which can lead to subsequent helicity-dependent THz emission [3].

2. RESULTS

In this work, we present helicity-dependent THz emission from two prime topological candidates: a centrosymmetric DSM cadmium arsenide (Cd_3As_2) single crystal and a non-centrosymmetric WSM Cd_3As_2 nanowire ensemble [4]. We perform terahertz emission spectroscopy using a near-infrared ($\sim 1.55\text{eV}$) pulse to photoexcite the samples above the bandgap energy and then measure the generated THz pulses in transmission via electro-optic sampling with a ZnTe crystal. For both the bulk crystal and nanowire ensemble, the THz emission increased linearly with increasing optical pump fluence but demonstrated a strong dependence on pump polarization. Under photoexcitation at normal incidence, a maximum (minimum) was observed in the THz emission for linearly-polarized light when the polarization was parallel (perpendicular) to the nanowire axis. In contrast, when photoexcited at oblique incidence, a maximum was observed in the emitted terahertz amplitude for circularly-polarized light. By fitting these polarization dependences of the emitted THz pulses, we determine that the photocurrent response is dominated by circular photogalvanic (CPGE) currents when photoexcited at oblique incidence, whereas photon drag and bulk thermal currents are responsible for THz emission at normal incidence.

Figure 1 shows a terahertz waveform and its corresponding FFT spectrum generated by the nanowire ensemble when photoexcited at oblique incidence with a NIR optical pulse with linear ($\Phi = 0^\circ$), and circular ($\Phi = \pm 45^\circ$) polarization. A clear change in polarity is observed when the nanowires are photoexcited with opposite photon helicity ($\Phi = \pm 45^\circ$), as expected for emission due to CPGE. For both the single crystal and nanowire ensemble, the emission frequency can also be tuned between 0.23 and 1.40 THz by tuning the angle of incidence of photoexcitation, highlighting DSMs as potential building blocks for an all-optical THz source for coherent quantum optoelectronic devices.

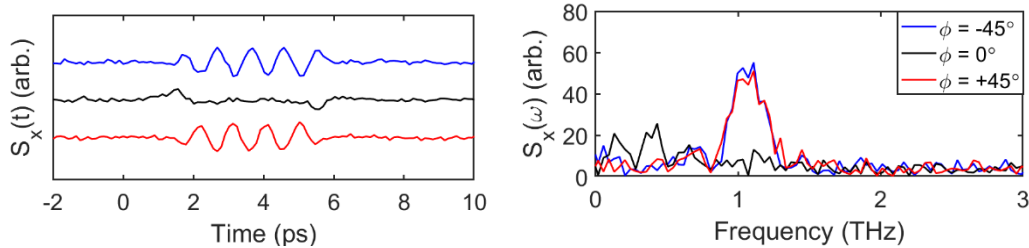


Figure 1. The electro-optic signal of the emitted THz waveform (left) and its corresponding FFT spectrum (right) measured for the nanowire ensemble under NIR photoexcitation at oblique incidence of -45 degrees for right-handed circular (blue) polarization, linear polarization (black) and left-handed circular polarization (red) [4].

However, the non-centrosymmetric nanowire ensemble displays a higher peak emission frequency (~ 1.5 THz) compared to the centrosymmetric single crystal (~ 0.8 THz), indicating a shorter relaxation time. Previous magneto-transport and terahertz spectroscopic studies on Cd_3As_2 nanowire ensembles have also demonstrated a transition from the metallic behavior observed in bulk crystals and thin films to semiconducting behavior [5, 6]. This semiconducting behavior could also account for the higher peak emission frequency. These nanowire electronic properties, along with their inherent polarization anisotropy, could provide additional control of their THz emission. Previous studies have also shown that the efficiency of a THz emitter can be substantially improved by exploiting the high surface-to-volume ratio and plasmonic enhancement due to nanowire geometry, making them exciting candidates for further study.

3. CONCLUSIONS

In conclusion, we present helicity-dependent THz emission from topological DSM Cd_3As_2 nanowires and demonstrate that it can be optically-switched from broadband to narrowband emission by switching the polarization of the optical pump pulse from linear to circular polarization respectively. Upon photoexcitation with circular polarization, the peak emission frequency could also be tuned from 0.23 and 1.40 THz by tuning the angle of incidence from 0 to 45 degrees. Our work highlights the promise of these Dirac semi-metal nanowires (and other topological nanostructures) for development of controllable all-optical THz sources.

REFERENCES

- [1] Weber, C. P.; "Ultrafast investigation and control of Dirac and Weyl semimetals," *Journal of Applied Physics*, 129, 070901 (2021)
- [2] Dai, Z.; Manjappa, M.; Yang, Y.; Tan, T. C. W.; Qiang, B.; Han, S.; Wong, L. J.; Xiu, F.; Liu, W.; Singh, R.; "High Mobility 3D Dirac Semimetal (Cd_3As_2) for Ultrafast Photoactive Terahertz Photonics," *Advanced Functional Materials*, 31, 2011011 (2021)
- [3] Gao, Y.; Kaushik, S.; Philip, E. J.; Li, Z.; Qin, Y.; Liu, Y. P.; Zhang, W. L.; Su, Y. L.; Chen, X.; Weng, H.; et al. "Chiral terahertz wave emission from the Weyl semimetal TaAs," *Nature Communications*, 11, 720 (2020)
- [4] Boland, J.L.; Damry, D.A.; Xia, C.Q.; Schönherr, P.; Prabhakaran, D.; Herz, L.N.; Hesjedal, T. and Johnston, M.B. "Narrowband, Angle-Tunable, Helicity-Dependent Terahertz Emission from Nanowires of the Topological Dirac Semimetal Cd_3As_2 ," *ACS Photonics*, 10 (5), 1473-1484 (2023)
- [5] Singh, A.; Schönherr, P.; Barnes, C.; and Hesjedal T. "Ultra-high Carrier Mobility in Cd_3As_2 nanowires" *Phys. Status Solidi RRL*, 17, 2200365 (2023)
- [6] Boland, J.L.; Damry, D.A.; Xia, C.Q.; Filip, M.; Schönherr, P.; Prabhakaran, D.; Herz, L.N.; Hesjedal, T. and Johnston, M.B. "Tracking Electron & Hole Dynamics in 3D Dirac Semimetals," 46th International Conference on Infrared, Millimeter and Terahertz Waves (IRMMW-THz), Chengdu, China, pp. 1-2, doi: 10.1109/IRMMW-THz50926.2021.9567041. (2021)