

Helicity-dependent ultrafast photocurrents in Weyl magnet Mn₃Sn



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This manuscript has been previously reviewed at another Nature Portfolio journal. This document only contains reviewer comments and rebuttal letters for versions considered at Communications Physics.

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors basically answered my questions and suggestions. Before publication I want to ask the author to clarify (1) in configuration 1, why CPGE and LPGE do not exist, which are discussed in Sb₂Te₃ (for example Appl. Phys. Lett. 118, 011102 (2021)); (2) I noticed another literature (Applied Physics Letters 115(18):182402 2019), please discuss the difference between your work and the literature.

Reviewer #2 (Remarks to the Author):

The revised manuscript has provided a complete and detailed analysis of the THz emission in Mn₃Sn. I agree with the authors' conclusion that photon drag is the main mechanism for the THz emission signals observed. The authors have also provided detailed responses to all my questions and comments. I believe that the manuscript presents sufficient novel and important findings to be published in Communications Physics.

Reviewer #3 (Remarks to the Author):

I read carefully the revised manuscript. The authors answered well my questions and comments. I thus recommend its publication in Communications Physics.

Reviewer #1 (Remarks to the Author):

The authors basically answered my questions and suggestions. Before publication I want to ask the author to clarify (1) in configuration 1, why CPGE and LPGE do not exist, which are discussed in Sb₂Te₃ (for example Appl. Phys. Lett. 118, 011102 (2021)); (2) I noticed another literature (Applied Physics Letters 115(18):182402 2019), please discuss the difference between your work and the literature.

We are grateful the Reviewer found our previous reply satisfactory and we are happy to clarify these latest points:

- (1) The bulk CPGE/LPGE require inversion symmetry breaking. Our results are insensitive to the intrinsic spin ordering of the material, suggesting that the generated photocurrents do not arise as a result of the magnetic ordering in the material and we focus on the non-magnetic symmetry analysis where the bulk of the sample respects inversion symmetry.

In the article Appl. Phys. Lett. 118, 011102 (2021) the PGE is associated to the surface break of symmetry and surface contributions of the PGE are indeed allowed in principle. However, in our analysis we are able to conclude that the linear contribution of the surface PGE (surface-LPGE) might be present, but is a minor contribution to the THz emission. This is possible by repeating the experiment with different tilting angles (that is not done in the cited article). The surface-LPGE effect contains lowest-order terms that are even under tilting, we instead observe that the odd terms are the highest in amplitude (Fig.2d of the main text). We want to underline, however, that this might be different in different materials. In the mentioned article the surface-LPGE is explained with the different electron density along the Sb–Te bond after optical pumping, which gives rise to the transient photocurrent that radiates the THz wave.

As for the circular contribution (surface-CPGE), we would not expect any contribution from this effect at normal incidence. But, we see it. Again from the tilting angle analysis we are able to observe that this component grows with increasing tilting angle and the small component at normal incidence is explained with a small unintentional rotation of the sample with respect to the pump direction. This is also described in more details in the reply to point 3 of Reviewer 2 in our previous correspondence. So, repeating the experiment with different tilting angles was key in our case to identify the main contribution to the THz emission, the bulk photon-drag effect.

- (2) In the Sb₂Te₃ work discussed, there is a clear an substantial dependence on magnetic field, suggesting that centro-symmetry is non-negligibly broken by the magnetic component. In contrast, we find no dependence on magnetic field or spin configuration (section E of the main text – Magnetic field dependence). Moreover, the authors claim that “the polarization direction of the laser pulse does not affect the polarization direction of coherent THz radiation”, excluding the PDE and PGE. In this case the origin of the THz emission is entirely associated with the magneto-dipole radiation, which is connected to the net magnetisation of the film. Clearly, the

authors see a flip in sign of the THz emission upon reversing the magnetic field, consistently with this picture. But to really compare our sample with their sample, magnetometry measurements would be necessary to check on the residual ferromagnetism, which unfortunately are not provided. We note that this ferromagnetism will depend on the quality of the growth and on the thickness of the sample (30 nm in the cited paper and 80 nm in our work). We conclude that in our sample the magnetic dipole emission is negligible with regard to the other contributions.