Peer Review File

Manuscript Title: Observation of a linked loop quantum state in a topological magnet

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Referee #1 (Remarks to the Author):

The theoretical band structure of solids is fascinating. Such, that it can easily inspire an artist. Bands can form saddles and valleys, Fermi surfaces often appear as "cups", "barrels" and "cigars", not to mention that they can have "necks", "hands" and look like a "monster". Because of the four dimensions caused by components of momentum and energy, it is easy to find even more exotic combinations of points, curves and surfaces. Obviously, many of these k-space creatures (or their absence, i.e., gaps) do define physical properties of the material, but should any analogy to real space objects be published in Nature, attracting enormous resources from the scientific community?

In any case, the nodal lines and loops, merely locus of degenerate or non-degenerate band crossings, have been noticed earlier. The case when nodal line crosses the BZ boundary and can be seen closed in repeated zone scheme has been published too. Even the case when such artificially constructed loops appear linked is also already documented. Moreover, it was also claimed earlier, that ARPES data on Co_2MnGa support the presence of nodal loops. The present manuscript, where all mentioned above results are referenced, finds that the nodal loops in Co_2MnGa are linked.

Let us assume, that such a band structure is indeed important, even though the nodes are far below the Fermi level and thus have elusive practical meaning. In order to support theory with the ARPES data, the authors present:

- One intensity map in Fig. 2d, which is symmetrized in unknown way, recorded from unknown portion of k-space, integrated within unknown energy interval and smoothed and separated from the background using an unknown procedure. It would be very instructive to see the corresponding raw dataset. This is a 3D-material. Which k_z-values are probed? It cannot be a "plane", by definition, because single photon energy probes a sphere in the k-space. If it runs through X1 point, what is the evidence for that? Why this photon energy is chosen? Did authors record a periodic structures as a function of photon energy?

- Two intensity maps in Fig. 2e,f, again, without important experimental parameters (see above). More significantly, it is not clear how these datasets were measured. Did the authors perform Fermi surface mapping for each energy (which step?) from the 500 to 800 eV? A couple of representative raw datasets would certainly help.

- Two momentum-energy cuts in Fig. 3a. Two dispersing features are seen merging near E_f. According to the authors they should make a Weyl crossing at significant binding energy, as schematically shown in Fig. 3e. This is not seen in the data.

Presented derivatives from the ARPES data are certainly not sufficient to support the claims made in the paper. Neither a single Weyl crossing is shown, nor the momentum location of the nodal loops (if any).

The other presented in the manuscript ARPES data are symmetrized (!) momentum-energy cuts in Fig. 5a which is unacceptable, since dispersion can be stopped at any arbitrary point and then reflected completely distorting the underlying picture. Only providing raw datasets can authors begin to sound convincing.

How were the data points in Fig. 3c obtained? Are there any supporting ARPES data? How were the error

bars determined?

It is a ferromagnet below Curie temperature. Are there any domains? How do they influence photoemission? How was the magnetic field measured? What is the influence of this field on photoelectrons?

Referee #2 (Remarks to the Author):

By using soft X-ray ARPES experimental techniques and first-principle calculations, the authors confirmed the linked nodal lines in ferromagnet Co2MnGa which was theoretically proposed in Ref [21]. Experimental confirmation of nodal link quantum states would be interesting. However, these states were already reported in Ref [20] although they only observe one surface which confirmed the existence of Weyl loops. However, the nodal link structure is easy to be interpreted according to the symmetry analysis. So I don't recommend it to be published in Nature.

The experimental data are with high-quality and the analysis and presentation are well established. It's a good experimental work and it deserves published in other journals.

Referee #3 (Remarks to the Author):

The manuscript entitled « Observation of a linked loop quantum state » reports on a soft x-ray ARPES study of the electronic properties of Co2MnGa. Combining ferromagnetism with the presence of mirror symmetries, this material is expected to host lines of Weyl points, named Weyl loops. The authors claim that due to the presence of multiple mirror planes (orthogonal to the x, y, and z axes), multiple loops should be present. The authors speculate about the possibility that those loops interconnect and link each other, thus realizing a complex knot structure. The authors do an effort to elaborate this analogy with knot theory, by classifying the system in terms of the linking number of the states forming the Fermi surface.
I have serious doubts about the interpretation of the data, which in my opinion do not support the claim of the manuscript.

1) As first thing, I want to make a strong point about the data presentation. Unfortunately the ARPES community is now heavily relying on images, rather than energy and momentum dispersion curves. As a matter of fact, images are very often manipulated in the way they are displayed. I think we can long debate about the use or abuse of color saturation, what I think we should not accept, as a community, is data symmetrization.
The authors do not provide clear details about the process of symmetrization (C4 symmetry, or two mirror planes). In figure 2, have the authors symmetrized the measured band dispersion, or directly the constant energy cuts?
The band dispersions in Figure 5 (a) are also symmetrized. For those images, the definition of the mirror plane with respect to the band dispersion is critical because it can severely affect the band crossing. How the authors define the plane of symmetry?
Considering all those issues, I think that the raw data should be shown as well.

2) The major concern I have is about the presentation of the data in an extended zone, that is not what has been measured experimentally. I think that the correct description of the data is provided in Figure 2 (and partially in Figure 5): the authors have measured the dispersion of a bulk state in the 3 mutually orthogonal planes as indicated with violet, red and orange colors. The interpretation that is given in Figure 4 and partially in Figure 5 is a wrong geometrical construction.
We can easily understand that the working assumption is wrong, by the fact that in Figure 2 the Fermi surface contour extends beyond the square projection of the bulk Brillouin zone on the plane passing through the X point. This is telling us that the correct description should be given in terms of the 3D bulk

Brillouin zone, as naturally expected for a 3D bulk state. This is a well-known problem also for the Fermi Surface of noble metals, which exhibit similar bulk Brillouin zone.

2.1) I think the authors should show clearly the whole tomography of the 3D Fermi surface, of which the contours in Figure 2 show only 3 cuts on the 3 planes. This can be easily done by calculations, but can be done also experimentally. In general, I think that showing this closed lines in a 3D Brillouin zone is misleading. The Fermi surface must be a surface, it can be a close surface (3D) or it can be open in 1 or 2 direction in the case of systems with reduced dimensionality. Such a tomography for different planes would bear the additional advantage to show how the Weyl loop evolve far from the planes passing through X.

2.2) At 540 eV an angular acceptance of ± 10° is already sufficient to cover ± 2Å-1, and the momentum window gets as large as ± 2.5Å-1 at 800 eV. This should be sufficient to directly probe the high-order Brillouin zones. This would allow to observe the linking structure which is now only sketched.

2.3) For completeness, the authors should show also the Brillouin zone projection on different planes, for example passing through Γ.

3) Related to the issue of the data representation in the reduced/extended zone, I think a key point is the number of states observed. In the data in the whole manuscript, I see one single contour. Figure 2 simply shows that this single contour obeys the symmetry of the crystal. In order to observe a link structure, the authors should observe the doubling of the states.

4) From a theoretical point of view, it is quite unclear the physical interpretation of the linking number. It is computed from Figure 5 on the basis of the extend zone replica, but I see no topological implication or connection to the Barry phase. By doping the material, in principle the dimension of the Fermi contour could be reduced and the linking could be changed, or is it enforced by something? If there is nothing imposing and protecting this linking number, then it is just a number, and I do not see why it should be important.

5) A fallout of the previous question is which are the implication of this specific linking structure in the transport, or magnetic, properties of the material? The authors speculate about a bridge between physics and knot theory, axion Lagrangian and non –Abelian node loop charges, but these concepts are very vague. I think that if the authors want to meet the requirements of Nature, they should strongly motivate how their findings can open new field of research, by explaining in which direction and how the scientific community could move.

Minor question: 6) Why in the data of Figure 2 e, the intensity around the Γ point is not observed, in contrast to panel d?

Author Rebuttals to Initial Comments:

Re: *Nature* 2020-10-18080, Observation of a linked loop quantum state

We sincerely thank the referees for taking the time to study our manuscript and offer criticism. In the following we respond to the comments of the referees, point by point.

REPORT OF REFEREE #1

Referee #1, Q1: The theoretical band structure of solids is fascinating. Such, that it can easily inspire an artist. Bands can form saddles and valleys, Fermi surfaces often appear as

"cups", "barrels" and "cigars", not to mention that they can have "necks", "hands" and look like a "monster". Because of the four dimensions caused by components of momentumand energy, it is easy to find even more exotic combinations of points, curves and surfaces. Obviously, many of these *k*-space creatures (or their absence, i.e. gaps) do define physical properties of the material, but should any analogy to real space objects be published in *Nature*, attracting enormous resources from the scientific community?

In any case, the nodal lines and loops, merely loci of degenerate or non-degenerate band crossings, have been noticed earlier. The case when nodal line crosses the BZ boundary and can be seen closed in repeated zone scheme has been published too. Even the case when such artificially constructed loops appear linked is also already documented. Moreover, it was also claimed earlier, that ARPES data on Co_2MnGa support the presence of nodal loops. The present manuscript, where all mentioned above results are referenced, finds that the nodal loops in Co_2MnGa are linked.

Authors: We sincerely thank the referee for taking the time to review our manuscript. We appreciate that the referee accurately summarized our essential observation that "the nodal loops in Co_2MnGa are linked".

We summarize our key contributions,

1. We directly observe 3 intricately intertwined Weyl loops in Co_2MnGa using state-of-the-art soft X-ray angle-resolved photoemission spectroscopy (SX-ARPES).

2. Based on our experimental data, we explicitly draw the link diagram for the linked Weyl loops; we determine the linking number to be $(2, 2, 2)$.

3. Unexpectedly, we find that the link is geometrically essential, establishing a new bridge between physics and knot theory on the three-torus, $\mathscr{T}^3$.

In our revised manuscript,

1. We have now, for the first time, predicted & observed signatures of topological Seifert boundary states associated with the bulk link, indicating a novel Seifert bulk-boundary correspondence (new Fig. 5).

2. We have included all additional raw data as requested in the revised Extended Data.

Considerable theoretical interest: Recent theoretical effort has uncovered a close relationship between node loop linking and the gravitational θ angle [*Phys. Rev. B* **95**, 094512 (2017)]; a topological contribution of the node loop linking number to the thermal magneto-electric coefficient [*Phys. Rev. Lett.* **119**, 147001 (2017)]; and an axion Lagrangian θ angle fixed by the the node loop linking number [*Phys. Rev. B* **96**, 081114(R) (2017)]. Linked node loops have further been associated with non-Abelian topological charges [*Science* **365**, 1273 (2019)], that are further predicted to exhibit exotic phenomenain superconductors [*Phys. Rev. B* **103**, 224523 (2021)] and dielectric photonic crystals [arXiv:2102.12546], as well as novel topological phase transitions [*Phys. Rev. B* **103**, L121101 (2021)]. It is clear that **experimentally demonstrating a linked node loop state is interesting, relevant and timely**. However, to date no linked node loops have been identified in any quantum material, photonic crystal or mechanical metamaterial. In the present manuscript, we provide a **direct spectroscopic observation of linked node loops for the first time**, opening this exciting frontier for experimental exploration as well as further theoretical investigation.

Significance & implications: To further address the referee's remark, we have explored two physical phenomena associated with linked Weyl loops.

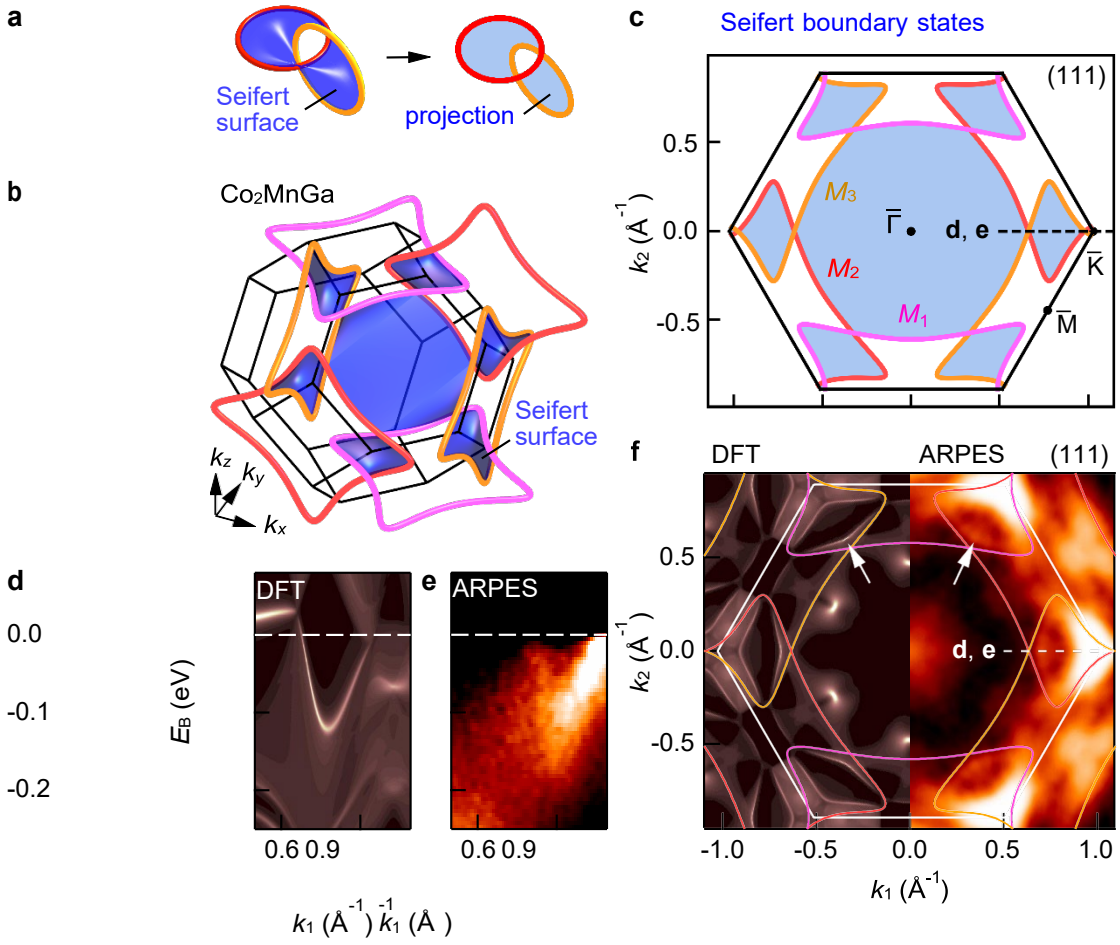


FIG. R1: **Seifert bulk-boundary correspondence. a**, A Seifert surface is defined as a three-dimensional surface bounded by a link, shown for the example of a Hopf link. Its two-dimensional projection produces alternating filled and empty regions pinned together at characteristic touching points. **b**, In a condensed matter system, the Seifert surface is taken as a surface bounded bythe linked loop nodes in three-dimensional momentum space (k_x, k_y, k_z), shown for the case of the linked loop nodes observed in Co_2MnGa. **c**, The projection of the Seifert surface into the surface Brillouin zone is associated with topological boundary modes (blue regions) which touch at points in momentum space. Energy axis collapsed for clarity. **d**, *Ab initio* calculation of the surfacestates through the touching point, exhibiting pairs of boundary modes pinned together at the Weyl loops. **e**, Surface-sensitive vacuum ultraviolet (VUV) ARPES energy-momentum cut through the touching point, exhibiting signatures of the pinned Seifert boundary modes, consistent with *ab initio* calculations. Photon energy $h\nu$ = 63 eV. **f**, Fermi surface in *ab initio* calculation (left) and VUV-ARPES (right) exhibiting Seifert boundary modes that stretch across the topological regions, connecting different Weyl loops, consistent with the Seifert projection.

1. **Topological Seifert boundary modes.** In our revised manuscript, we present signatures of topological boundary states associated with the Seifert surface of the bulk Weyl loop link (main text Fig. 5, shown in this document as Fig. R1). Mathematically, a Seifert surface is a three-dimensional surface bounded by a knot or link [*Math. Ann.* **110**, 571 (1935)]. For example, in the minimal case of a Hopf link, the Seifert surface exhibits a branched structure which 'wraps' around the link (Fig. R1a, left). Upon projection intotwo dimensions, the Seifert surface produces a set of filled regions which touch at discrete points (Fig. R1a, right). For a linked loop quantum state in a condensed matter system,the Seifert surface is taken to live in bulk momentum space (k_x, k_y, k_z) with the energyaxis collapsed. The topological boundary modes on all cleaving planes are given by the projection of the Seifert surface [*Rev. Mod. Phys.* **93**, 015005 (2021), *Comm. Phys.* **4**, 47 (2021), *Phys. Rev. B* **99**, 161115(R) (2019), *Commun. Phys.* **2**, 135 (2019)]. These Seifert boundary modes then exhibit a unique configuration of filled regions pinned together at touching points in momentum space, associated with the bulk links. This relationship between the link, its Seifert surface and the topological boundary modes suggests a **Seifertbulk-boundary correspondence**, where the linked loop nodes in the bulk protect Seifert boundary states and, conversely, the characteristic configuration of Seifert boundary states encodes the linking number. By examining the (111) sample surface, **we observe experimental signatures of these Seifert boundary states**, consistent between *ab initio* calculation and surface-sensitive vacuum ultraviolet (VUV) ARPES (Fig. R1d-f).

2. **Linked Weyl loop axion electrodynamics.** The axion θ term may give rise to a topological magneto-electric effect in electronic structures hosting linked node loops [*Phys. Rev. B* **96**, 081114(R) (2017), *Phys. Rev. B* **95**, 094512 (2017), *Phys. Rev. Lett.* **119**, 147001 (2017)]. To achieve this effect in Co_2MnGa, we expect that it is first necessary to gap the Weyl loops by breaking mirror symmetry. This could potentially be achieved in thin film samples of Co_2MnGa via an asymmetric substrate-induced lattice strain. Fermi level tuning or perturbation of the electronic structure may then be needed to place the Fermi level in the Weyl loop gap, possibly achieved by engineering the material's magnetic properties via the film substrate, temperature, carrier doping or chemical substitution. Transport or optics experiments may then detect quantized axion electrodynamics, as previously investigated in topological insulators [*Science* **354**, 1124 (2016)]. In the present case the axion response is

expected to be quantized not to the topological insulator $\mathbb{Z}_2$ invariant, but rather to the Weyl loop linking number. In our revised manuscript, we have pointed out that: "Moreover, the linked loop state in Co_2MnGa, as well as in other materials, may give rise to exotic response quantized to the linking number, such as a link-quantized topological magneto-electric effect [25, 32, 33, 48].", citing these four references.

Referee #1, Q2: Let us assume, that such a band structure is indeed important, even though the nodes are far below the Fermi level and thus have elusive practical meaning.

Authors: We thank the referee for the remark. We kindly note that the nodal loops in Co_2MnGa are understood to cross the Fermi level, playing an essential role in its transport and other practical properties up to room temperature, as reported previously by several groups [*Nat. Phys.* **14**, 1119 (2018), *Science* **365**, 1278 (2019), *NPG Asia Materials* **11** 16(2019), *Phys. Rev. B* **101**, 060406(R) (2020)]. In our photoemission spectra, we also find that the linked node loops disperse to the Fermi level within experimental resolution (see, for example, Figs. R4, R5).

In the revised main text, we have noted that the Weyl loops in Co_2MnGa are understood to dominate essential transport properties of the material, "The extracted dispersion reaches the Fermi level within experimental resolution, suggesting that the observed Weyl loops are relevant for transport and other low-energy response. This result is consistent with previous reports that Weyl loops play a dominant role in the giant anomalous Hall effect and other exotic transport properties of Co_2MnGa [20, 40–42].", citing the four references above.

Referee #1, Q3: In order to support theory with the ARPES data, the authors present:

- One intensity map in Fig. 2d, which is symmetrized in an unknown way, recorded from an unknown portion of k-space, integrated within an unknown energy interval and smoothed and separated from the background using an unknown procedure. It would be very in- structive to see the corresponding raw dataset. This is a 3D-material. Which k_z-values are probed? It cannot be a "plane", by definition, because a single photon energy probes a

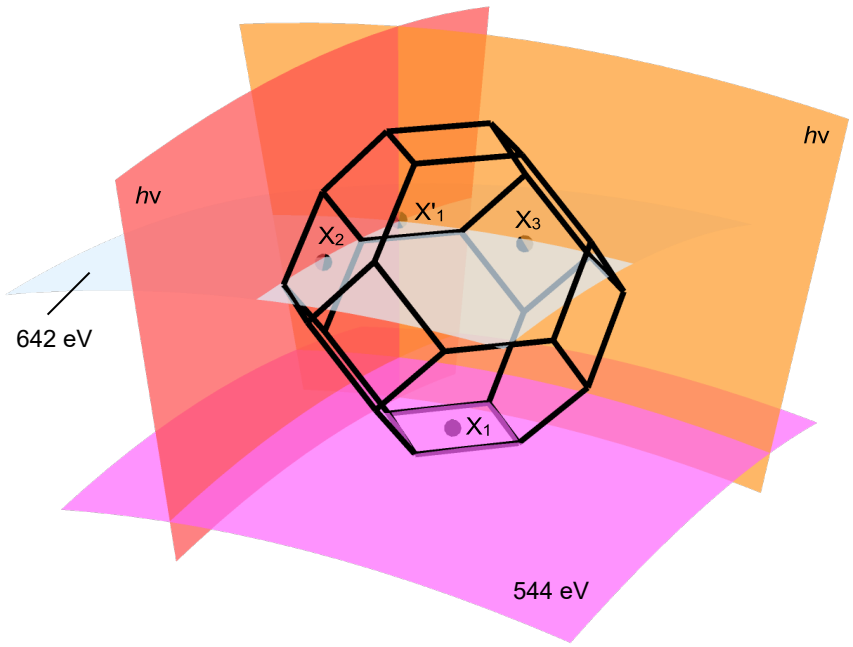


FIG. R2: **Measured Fermi surfaces in an extended zone scheme.** The Brillouin zone corresponds to $\Gamma_{(066)}$ in the primitive reciprocal basis.

sphere in the k-space. If it runs through the X_1 point, what is the evidence for that? Why was this photon energy chosen? Did the authors record periodic structures as a function of photon energy?

Authors: We thank the referee for the careful examination of our data and we apologize that we did not clearly list these technical parameters.

The Fermi surface in Fig. 2d was symmetrized about M_x and M_{xy}, consistent with the symmetries of the crystal. The symmetrization simplifies presentation of the data for the general reader, but does not play any role in drawing conclusions from the data. The inci- dent photon energy $h\nu$ = 544 eV corresponds to the top face of the fifth Brillouin zone up in k_z, with $k_x = k_y = 0$. Data were integrated in a window of $\pm$38 meV around the Fermi level,

consistent with the nominal energy resolution of the measurement δE = 75 meV. The background was removed by a fixed intensity cutoff. The momentum-space region shown is closeto normal emission and can consequently be well-approximated by a plane at fixed $k_z = X_1$.The photon energy was calibrated to k_z using the photon energy dependence acquired from 500 eV to 800 eV, which clearly exhibited periodic structures across multiple Brillouin zones.

In our revised manuscript, we have included the raw Fermi surface used to generated Fig. 2d (Extended Data Fig. 6a, Fig. R3a). The raw data exhibits a clear diamond-shapedloop contour (Weyl loop) encircling the X point. We further include the photon energy dependence indicating that $h\nu$ = 544 eV corresponds approximately to $k_z = X_1$ (Extended Data Fig. 6b, Fig. R3b). Lastly, we plot the nominal momentum-space surface probed by the mapping at $h\nu$ = 544 eV, illustrating that it is well-approximated by a plane throughX_1 in the momentum range of interest (Extended Data Fig. 10, Fig. R2). In the revised Methods, we have noted that, "The constant-energy cuts were symmetrized about M_x and M_{xy} (Fig. 1d), M_x and M_{xz} (Fig. 1e) and M_y and M_{yz} (Fig. 1f)." and "A background was removed from the photoemission spectra by a fixed intensity cutoff (raw, unsymmetrized data in Extended Data Figs. R3, R4, R5)."

Referee #1, Q4: - Two intensity maps in Fig. 2e, f, again, without important experimental parameters (see above). More significantly, it is not clear how these datasets were measured. Did the authors perform Fermi surface mapping for each energy (which step?) from the 500 to 800 eV? A couple of representative raw datasets would certainly help.

Authors: Again we thank the referee for the remark. The out-of-plane Fermi surface in Fig. 2e was acquired through a standard photon energy dependence. The measurementwas calibrated to slice through the X_2 point closest to normal emission, corresponding toa sample rotation angle of $\sim 5^\circ$ which was systematically varied at each photon energy to remain in the M_2 mirror plane. The step size was 2 eV and at each photon energy a single high-quality energy-momentum cut was acquired. Fig. 2f was acquired in the same way, but the sample was rotated by 90° so that the measurement sliced through X_3. For both of the Fermi surfaces, data were integrated in a window of $\pm$25 meV around the Fermi level.

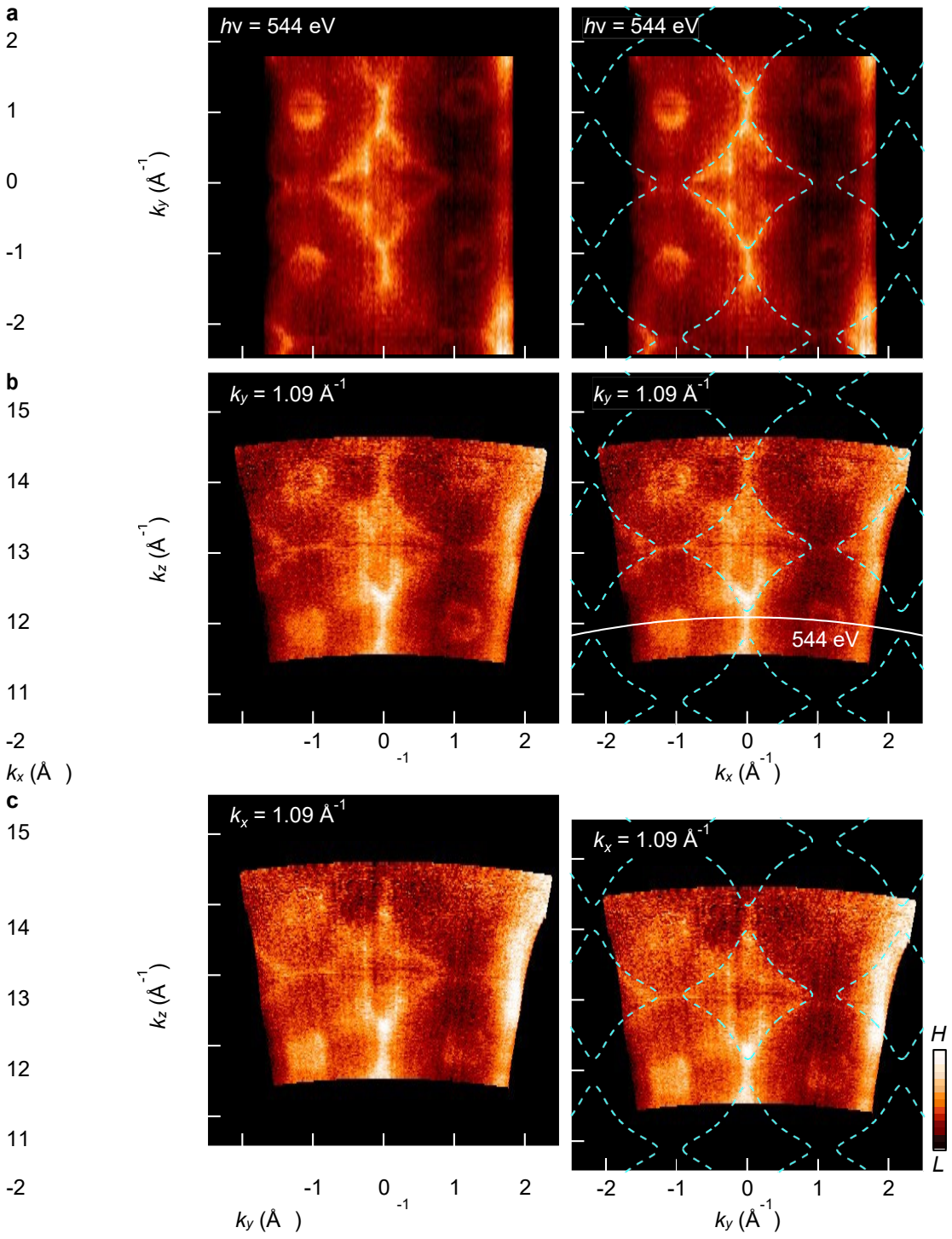


FIG. R3: **Unsymmetrized Fermi surfaces. a-c**, Left: photoemission spectra displayed in Fig. 2d-f, without symmetrization. Right: the same spectra, with the experimentally-determined Weyl loop trajectory overlaid across multiple Brillouin zones. The irrelevant Γ pocket is consistently observed in all unsymmetrized spectra. Signatures of Weyl loops are observed around all *X* points.

The resulting images were symmetrized in the same way as Fig. 2d, corresponding to M_x and M_{xz} (Fig. 2e) and M_y and M_{yz} (Fig. 2f), all consistent with the symmetries of the crystal. As in the case of Fig. 2d, the background was removed by a fixed intensity cutoff. The symmetrization and background subtraction simplifies presentation of the data for the general reader, but does not play any role in drawing conclusions from the data.

In our revised Extended Data, we have included the two raw Fermi surfaces used to generate Figs. 2e, f (Extended Data Fig. 6b, c, Fig. R3b, c). Again, all datasets exhibit multiple diamond-shaped loop contours (Weyl loops) encircling the X points. We also plot the nominal momentum-space surfaces probed by the two photon-energy dependences (Extended Data Fig. 10, Fig. R2). In the revised Fig. 2 caption we have included the photon energy step size, "Out-of-plane Fermi surface acquired on the same Co_2MnGa sample by an ARPES photon energy dependence from 500 eV to 800 eV in steps of 2 eV". In the revised Methods we indicated, "The high-symmetry energy-momentum cuts were similarly symmetrized about M_x, M_y or M_z, as appropriate and consistent with the nominal symmetries of the crystal (Fig. 4a)." Lastly, we noted that, "The Fermi surfaces were binned in an energy window of ±38 meV (Fig. 1d) and ±25 meV (Fig. 1e,f) around E_F.".

Referee #1, Q5: - Two momentum-energy cuts in Fig. 3a. Two dispersing features are seen merging near E_f. According to the authors they should make a Weyl crossing at significant binding energy, as schematically shown in Fig. 3e. This is not seen in the data.

Presented derivatives from the ARPES data are certainly not sufficient to support the claims made in the paper. Neither a single Weyl crossing is shown, nor the momentum location of the nodal loops (if any).

Authors: We thank the referee for carefully studying our work. Indeed, a cut near $k_a = 0$ Å^{-1} exhibits signatures of a Weyl crossing at ~ 0.1 eV below the Fermi level. This can be clearly seen in our photoemission data and we apologize that we did not include this systematic in our initial submission.

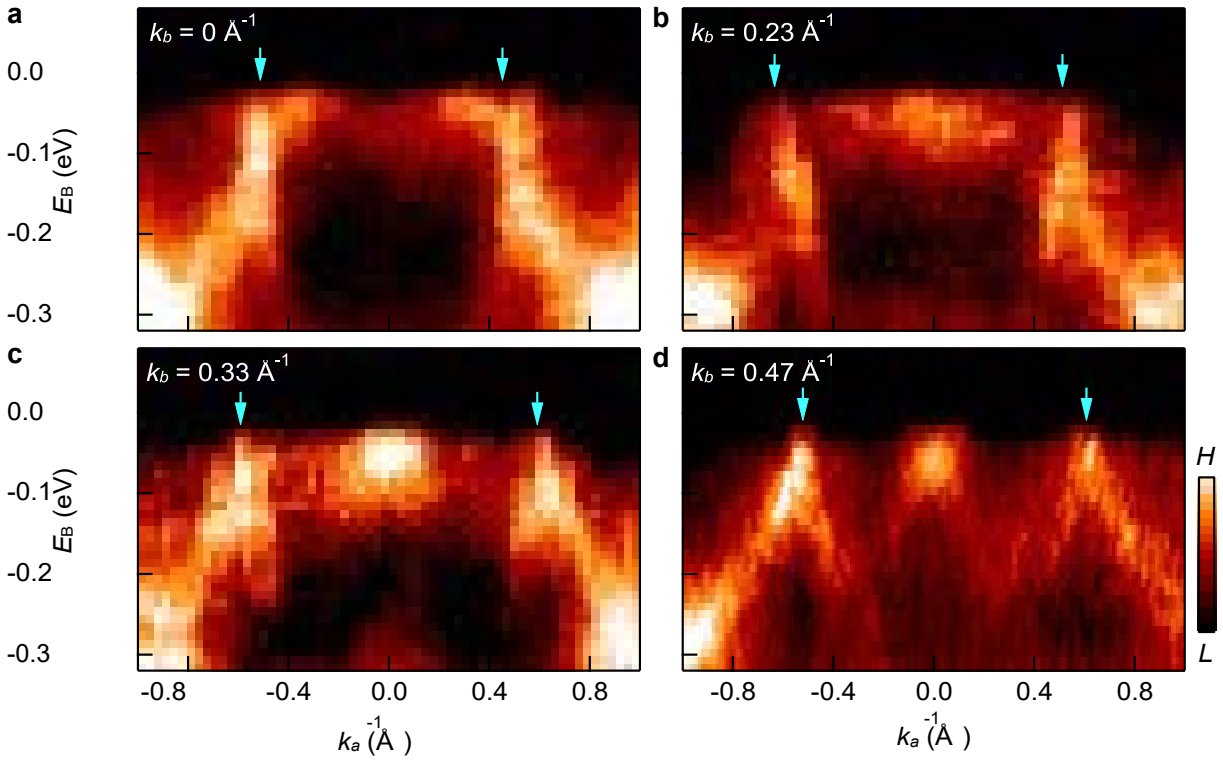


FIG. R4: **Energy-momentum cuts through the Weyl loop.** Photoemission spectra used to extract Fig. 2c.

In the revised Extended Data, we have included this raw energy-momentum cut (Extended Data Fig. 7a, Fig. R4a). We have also included the raw data for Fig. 3a. We have further included additional raw energy-momentum cuts used to track the momentum location of the Weyl loop (Extended Data Fig. 7b-c, Fig. R4b-c).

Referee #1, Q6: The other presented in the manuscript ARPES data are symmetrized (!) momentum-energy cuts in Fig. 5a which is unacceptable, since dispersion can be stopped at any arbitrary point and then reflected completely distorting the underlying picture. Only providing raw datasets can authors begin to sound convincing.

Authors: We again thank the referee for the remark and we apologize that our initial submission was not clear on this point. The momentum-energy cuts in Fig. 5a aresymmetrized according to the nominal mirror symmetries of the crystal.

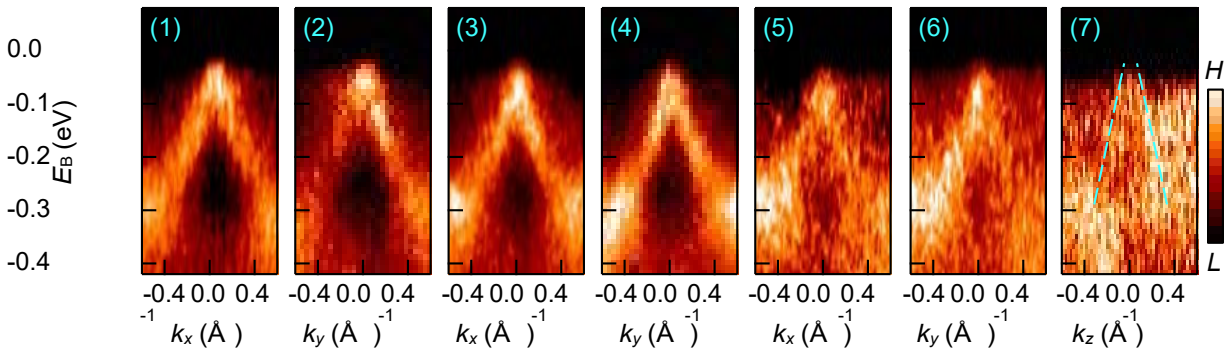


FIG. R5: **Unsymmetrized energy-momentum cuts.** Photoemission spectra displayed in main text Fig. 5a, without symmetrization.

In the revised Extended Data, we have provided the full complement of raw, unsymmetrized datasets for Fig. 5a (Extended Data Fig. 8, Fig. R5). The raw data are consistent with the symmetrized Fig. 5a and all datasets clearly suggest the presence of linked Weyl loops.

Referee #1, Q7: How were the data points in Fig. 3c obtained? Are there any supporting ARPES data? How were the error bars determined?

Authors: We thank the referee for the remark and we apologize that we did not present sufficient source data for Fig. 3c. Each point in Fig. 3c is based on a distinct energy-momentum dataset. The error bars of ± 0.1 Å^{-1} represent the typical linewidth of these source spectra (see, for example, Fig. 3a) and are further consistent with the nominal momentum resolution of the measurement, ~ 0.04 Å^{-1}.

In the revised Extended Data, we have presented additional raw energy-momentum datasets to support the data points and error bars in Fig. 3c, (Extended Data Fig. 7, Fig. R4).

Referee #1, Q8: It is a ferromagnet below Curie temperature. Are there any domains? How do they influence photoemission? How was the magnetic field measured? What is the influence of this field on photoelectrons?

Authors: We thank the referee for the interesting question. As suggested by the referee, in- deed we perform our measurements at T = 16 K, below the Curie temperature of T_C = 690 K and deep in the ferromagnetic phase. We find that our photoemission spectra are typicallyvery sharp with narrow linewidth, suggesting that neither magnetic domains nor surface stray fields significantly broaden or distort the data. Sharp photoemission spectra have also recently been observed in other magnetic materials of interest to the community [*Nature* **555**, 638 (2018), *Nature* **576**, 423 (2019), *Science* **365**, 1282 (2019)], including specifically in Co_2MnGa [*Science* **365**, 1278 (2019)]. The absence of a stray field distortion in our data may be due to our high-energy soft X-ray probe, producing high-energy outgoing photo- electrons which are negligibly deflected by the stray field. It may also be that the varying magnetization direction of different domains perturbs the electronic structure on an energyscale smaller than the energy resolution of our measurement or the typical energy scale ofthe features of interest. This conclusion is further supported by the excellent agreement between our photoemission spectra and *ab initio* calculations (see, for example, Fig. 3a, b). These considerations suggest that broadening of the lineshape due to magnetic domains or surface stray fields can be neglected in the present work. In the revised manuscript, we have added a brief comment on this point.

Summary: We briefly summarize our main points in this reply letter,

1. **Theoretical & experimental evidence for Seifert boundary states:** In our revised manuscript, we present signatures of Seifert boundary states in ARPES for the first time, providing an example of the Seifert bulk-boundary correspondence in quantum matter (Fig. R1).

2. **Link-quantized topological magneto-electric effect:** Recent theoretical work indicates that linked node loops may be a rich source of phenomena relating to the ax- ion θ angle, the gravitational θ angle and non-Abelian topological charges, producing exotic phenomena in quantum matter, photonic crystals and mechanical metamateri- als. However, to date linked node loops have not been reported in any such system. The availability of an experimental platform to explore the theoretical proposals is clearly urgently needed. In the present manuscript, we provide for the first time a direct observation of linked node loops in a quantum material. We further propose

that the linked Weyl loops specifically in Co_2MnGa may give rise to topological axion electrodynamics quantized to the Weyl loop linking number.

3. **Source data:** We have provided all raw data as additional Extended Data, as well as clearly indicated the symmetrization and background subtraction procedures that we used. Crucially, all conclusions are drawn directly from raw datasets produced by the spectrometer. The background subtraction only serves to improve the presentation for the general reader.

We have attempted to clarify these points in our revised submission. We again thank the referee for her/his remarks and for helping us improve our manuscript.

REPORT OF REFEREE #2

Referee #2, Q1: By using soft X-ray ARPES experimental techniques and first-principle calculations, the authors confirmed the linked nodal lines in the ferromagnet Co_2MnGa which were theoretically proposed in Ref. [21]. Experimental confirmation of nodal link quantum states would be interesting. However, these states were already reported in Ref. [20] although they only observe one surface which confirmed the existence of Weyl loops. However, the nodal link structure is easy to be interpreted according to the symmetry analysis. So I don't recommend it to be published in *Nature*.

The experimental data are of high quality and the analysis and presentation are well-established. It's a good experimental work and it deserves publication in other journals.

Authors: We sincerely thank the referee for taking the time to review our work and offer comments. We are thrilled that the referee finds that the "[e]xperimental confirmationof nodal link quantum states would be interesting" and that "the authors confirmed the linked nodal lines in the ferromagnet Co_2MnGa".

We summarize our key contributions,

1. We directly observe 3 intricately intertwined Weyl loops in Co_2MnGa using state-of-the-art soft X-ray angle-resolved photoemission spectroscopy (SX-ARPES).
2. Based on our experimental data, we explicitly draw the link diagram for the linked Weyl loops; we determine the linking number to be $(2, 2, 2)$.
3. Unexpectedly, we find that the link is geometrically essential, establishing a new bridge between physics and knot theory on the three-torus, $\mathcal{T}^3$.

In our revised manuscript,

1. We have now, for the first time, predicted & observed signatures of topological Seifert boundary states associated with the bulk link, indicating a novel Seifert bulk-boundary correspondence (new Fig. 5).

2. We have included all additional raw data as requested in the revised Extended Data.

Previous experiments are insufficient: Ref. [20], [*Science* **365**, 1278 (2019)] did not consider or discuss in any way the linking number or composite linked loop structure in Co_2MnGa. That work was only able to observe a restricted momentum-space region of a small non-linked Weyl loop living around the K point of the bulk Brillouin zone. This smallloop was examined because Ref. [20] relies only on surface-sensitive ultraviolet ARPES, which provides limited out-of-plane k_z resolution and does not allow characterization of thelink-forming Weyl loops. By contrast, in the present work we use bulk-sensitive soft X-ray ARPES, which offers high k_z resolution and access to multiple full bulk Brillouin zones. This powerful experimental probe is essential to measuring the Weyl loop linking number, achieved for the first time in our work.

Symmetry analysis is insufficient: Trivial link diagrams exist which respect the symmetries of the system. Because Ref. [20] only characterized a restricted region ofan irrelevant Weyl loop, our results cannot be obtained by applying symmetry analysisto that previous work. In fact, Ref. [21] [*Phys. Rev. Lett.* **119**, 156401 (2017)] points out that a line node Hopf link can undergo a transition into a trivial link as the size ofthe loops is varied (see SI Section C. of that work). In general, linking number phase transitions exist which can change the linking number while preserving the symmetriesof the system, in analogy to traditional topological phases of matter such as topological insulators. Clearly, an experimental measurement of the complete momentum-space trajectory of the Weyl loop is essential to determine the linking number. In our work,we measure the complete Weyl loop trajectory for the first time, allowing us to demon- strate a linked loop quantum state, draw its link diagram and determine the linking number.

The observed link disagrees with the theoretical prediction: The previous work,Ref. [21], only suggested that a Hopf-like link many arise in Co_2MnGa, but did not theoretically predict the link diagram, calculate the associated linking numbers or predict that the Co_2MnGa link is geometrically essential. Crucially, in contrast to the prediction of Ref. [21], our systematic experimental measurements have shown that the link is not a Hopflink. While a Hopf link has two components, the linked node loops which we observe form

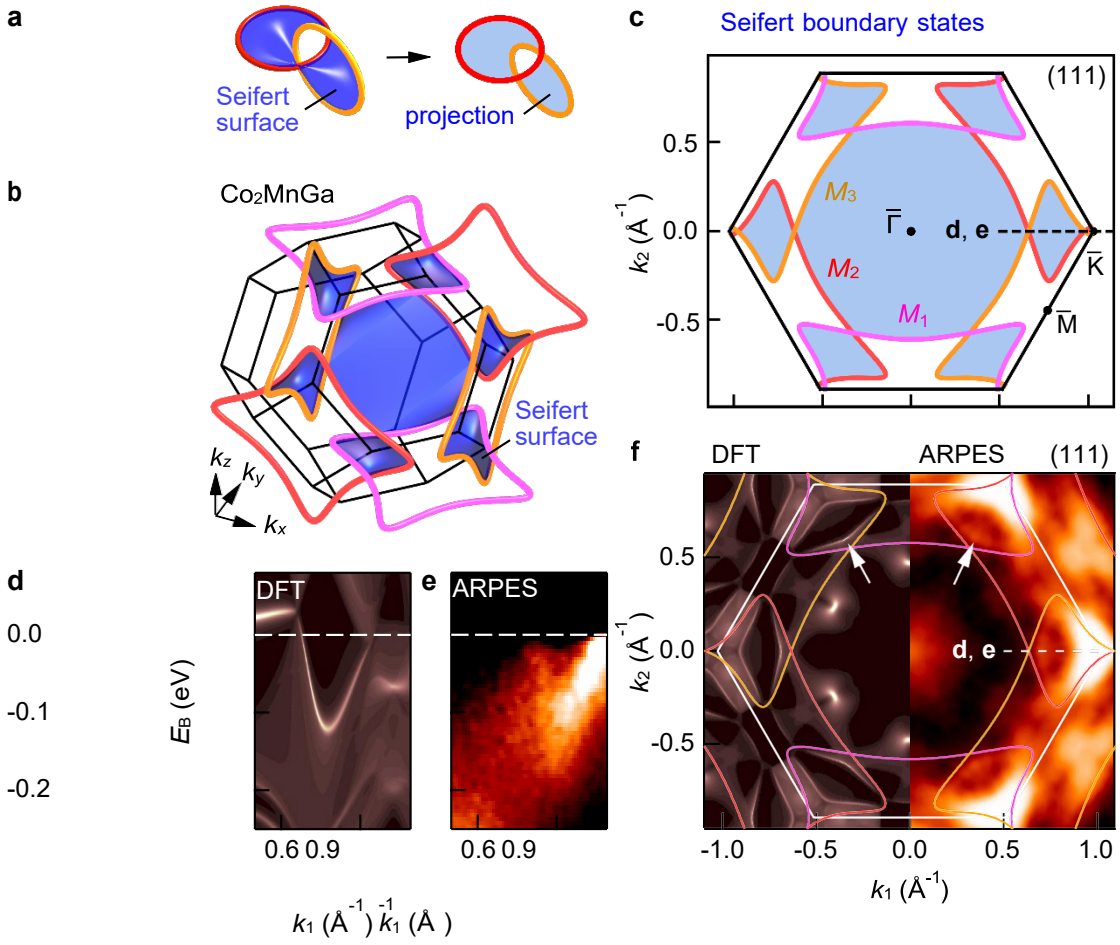


FIG. R6: **Seifert bulk-boundary correspondence. a**, A Seifert surface is defined as a three-dimensional surface bounded by a link, shown for the example of a Hopf link. Its two-dimensional projection produces alternating filled and empty regions pinned together at characteristic touching points. **b**, In a condensed matter system, the Seifert surface is taken as a surface bounded bythe linked loop nodes in three-dimensional momentum space (k_x, k_y, k_z), shown for the case of the linked loop nodes observed in Co_2MnGa. **c**, The projection of the Seifert surface into the surface Brillouin zone is associated with topological boundary modes (blue regions) which touch at points in momentum space. Energy axis collapsed for clarity. **d**, *Ab initio* calculation of the surfacestates through the touching point, exhibiting pairs of boundary modes pinned together at the Weyl loops. **e**, Surface-sensitive vacuum ultraviolet (VUV) ARPES energy-momentum cut through the touching point, exhibiting signatures of the pinned Seifert boundary modes, consistent with *ab initio* calculations. Photon energy $h\nu$ = 63 eV. **f**, Fermi surface in *ab initio* calculation (left) and VUV-ARPES (right) exhibiting Seifert boundary modes that stretch across the topological regions, connecting different Weyl loops, consistent with the Seifert projection.

a three-component link. Additionally, a Hopf link by definition has linking number $\nu = 1$, but the linking number between each pair of components in the experimentally-observed link is $\nu = 2$.

Considerable theoretical interest, but no experimental platforms: Recent theoretical effort has uncovered a close relationship between node loop linking and the gravitational θ angle [*Phys. Rev. B* **95**, 094512 (2017)]; a topological contribution of the node loop linking number to the thermal magneto-electric coefficient [*Phys. Rev. Lett.* **119**, 147001 (2017)]; and an axion Lagrangian θ angle fixed by the the node loop linking number [*Phys. Rev. B* **96**, 081114(R) (2017)]. Linked node loops have further been associated with non-Abelian topological charges [*Science* **365**, 1273 (2019)],that are further predicted to exhibit exotic phenomena in superconductors [*Phys. Rev. B* **103**, 224523 (2021)] and dielectric photonic crystals [arXiv:2102.12546], as well as novel topological phase transitions [*Phys. Rev. B* **103**, L121101 (2021)]. It is clear that experimentally demonstrating a linked node loop state is interesting, relevant and timely. However, to date no linked node loops have been identified in any quantum material, photonic crystal or mechanical metamaterial. In the present manuscript, we provide a direct spectroscopic observation of linked node loops for the first time, openingthis exciting frontier for experimental exploration as well as further theoretical investigation.

Observation of a geometrically-essential link on the three-torus: Typical examples of topology in condensed matter physics consider winding numbers from real or momentumspace into a target order parameter space. For example, in a magnetic vortex the local magnetization winds along a closed path in real space (Fig. 1a of our manuscript). By contrast, in the present case, our experimental results indicate linked Weyl loops,representing a form of topology which is qualitatively different from the order parameter winding considered over decades of research in condensed matter physics. An exotic featureof this new form of topology is that it has allowed the realization of a geometrically-essentiallink on $\mathcal{T}^3$. In particular, through our state-of-the-art experiments we are able to directly observe that the linked loop quantum state wraps around the bulk Brillouin zone. Toour knowledge this is the first observation of any linked or knotted structure which wraps around $\mathcal{T}^3$. We believe that such an advance merits consideration for publication in *Nature*.

Significance & implications: We propose two phenomena associated with linked Weyl loops in Co_2MnGa, to underline the significance and impact of our work.

1. **Topological Seifert boundary modes.** In our revised manuscript, we present signatures of topological boundary states associated with the Seifert surface of the bulk Weyl loop link (main text Fig. 5, shown in this document as Fig. R6). Mathematically, a Seifert surface is a three-dimensional surface bounded by a knot or link [*Math. Ann.* **110**, 571 (1935)]. For example, in the minimal case of a Hopf link, the Seifert surface exhibits a branched structure which 'wraps' around the link (Fig. R6a, left). Upon projection intotwo dimensions, the Seifert surface produces a set of filled regions which touch at discrete points (Fig. R6a, right). For a linked loop quantum state in a condensed matter system,the Seifert surface is taken to live in bulk momentum space (k_x, k_y, k_z) with the energyaxis collapsed. The topological boundary modes on all cleaving planes are given by the projection of the Seifert surface [*Rev. Mod. Phys.* **93**, 015005 (2021), *Comm. Phys.* **4**, 47 (2021), *Phys. Rev. B* **99**, 161115(R) (2019), *Commun. Phys.* **2**, 135 (2019)]. These Seifert boundary modes then exhibit a unique configuration of filled regions pinned together at touching points in momentum space, associated with the bulk links. This relationship between the link, its Seifert surface and the topological boundary modes suggests a **Seifertbulk-boundary correspondence**, where the linked loop nodes in the bulk protect Seifert boundary states and, conversely, the characteristic configuration of Seifert boundary states encodes the linking number. By examining the (111) sample surface, **we observe experimental signatures of these Seifert boundary states**, consistent between *ab initio* calculation and surface-sensitive vacuum ultraviolet (VUV) ARPES (Fig. R6d-f).

2. **Linked Weyl loop axion electrodynamics.** The axion θ term may give rise to a topological magneto-electric effect in electronic structures hosting linked node loops [*Phys. Rev. B* **96**, 081114(R) (2017), *Phys. Rev. B* **95**, 094512 (2017), *Phys. Rev. Lett.* **119**, 147001 (2017)]. To achieve this effect in Co_2MnGa, we expect that it is first necessary to gap the Weyl loops by breaking mirror symmetry. This could potentially be achieved inthin film samples of Co_2MnGa via an asymmetric substrate-induced lattice strain. Fermi level tuning or perturbation of the electronic structure may then be needed to place the

Fermi level in the Weyl loop gap, possibly achieved by engineering the material's magnetic properties via the film substrate, temperature, carrier doping or chemical substitution. Transport or optics experiments may then detect quantized axion electrodynamics, as previously investigated in topological insulators [*Science* **354**, 1124 (2016)]. In the present case the axion response is expected to be quantized not to the topological insulator $\mathbb{Z}_2$ invariant, but rather to the Weyl loop linking number. In our revised manuscript, wehave pointed out that: "Moreover, the linked loop state in Co_2MnGa, as well as in other materials, may give rise to exotic response quantized to the linking number, such as a link-quantized topological magneto-electric effect [25, 32, 33, 48].", citing these four references.

We again thank the referee for her/his remarks and for helping us improve our manuscript.

REPORT OF REFEREE #3

Referee #3, Q1: The manuscript entitled "Observation of a linked loop quantum state" reports on a soft X-ray ARPES study of the electronic properties of Co_2MnGa. Combining ferromagnetism with the presence of mirror symmetries, this material is expected to host lines of Weyl points, named Weyl loops. The authors claim that due to the presence of multiple mirror planes (orthogonal to the x, y, and z axes), multiple loops should be present.The authors speculate about the possibility that those loops interconnect and link each other,thus realizing a complex knot structure. The authors do an effort to elaborate this analogy with knot theory, by classifying the system in terms of the linking number of the states forming the Fermi surface.

Authors: We sincerely thank the referee for taking the time to examine our manuscript. The referee has carefully and accurately summarized our essential result. We particularly appreciate that the referee noticed that we made "an effort to elaborate th[e] analogy with knot theory", representing a qualitatively new manifestation of topology in physics.

We summarize our key contributions,

1. We directly observe 3 intricately intertwined Weyl loops in Co_2MnGa using state-of-the-art soft X-ray angle-resolved photoemission spectroscopy (SX-ARPES).
2. Based on our experimental data, we explicitly draw the link diagram for the linked Weyl loops; we determine the linking number to be $(2, 2, 2)$.
3. Unexpectedly, we find that the link is geometrically essential, establishing a new bridge between physics and knot theory on the three-torus, $\mathcal{T}^3$.

In our revised manuscript,

1. We have now, for the first time, predicted & observed signatures of topological Seifert boundary states associated with the bulk link, indicating a novel Seifert bulk-boundary correspondence (new Fig. 5).
2. We have included all additional raw data as requested in the revised Extended Data.

Referee #3, Q2: I have serious doubts about the interpretation of the data, which in my opinion do not support the claim of the manuscript.

1) As first thing, I want to make a strong point about the data presentation. Unfortu- nately the ARPES community is now heavily relying on images, rather than energy and momentum dispersion curves. As a matter of fact, images are very often manipulatedin the way they are displayed. I think we can long debate about the use or abuse ofcolor saturation, what I think we should not accept, as a community, is data symmetrization.

The authors do not provide clear details about the process of symmetrization (C_4 symme- try, or two mirror planes). In figure 2, have the authors symmetrized the measured band dispersion, or directly the constant energy cuts?

Authors: We thank the referee for the careful examination of our data and we apologize that we did not fully discuss these technical details.

The Fermi surface in Fig. 2d was symmetrized about M_x and M_{xy}, consistent with the symmetries of the crystal. Similarly, Fig. 2e was symmetrized about M_x and M_{xz} and Fig. 2f about M_y and M_{yz}, all consistent with the symmetries of the crystal. In all cases, we directly symmetrized the constant-energy cut. The symmetrization simplifies presentationof the data for the general reader, but does not play any role in drawing conclusions fromthe data.

In our revised Extended Data, we have included these three raw Fermi surfaces as Extended Data Fig. 6 (Fig. R7). The raw datasets clearly exhibit a diamond-shaped loop contour (Weyl loop) encircling all X points. In the revised Methods, we have noted that, "The constant-energy cuts were symmetrized about M_x and M_{xy} (Fig. 1d), M_x and M_{xz} (Fig. 1e) and M_y and M_{yz} (Fig. 1f)."

Referee #3, Q3: The band dispersions in Figure 5 (a) are also symmetrized. For those images, the definition of the mirror plane with respect to the band dispersion is critical be-

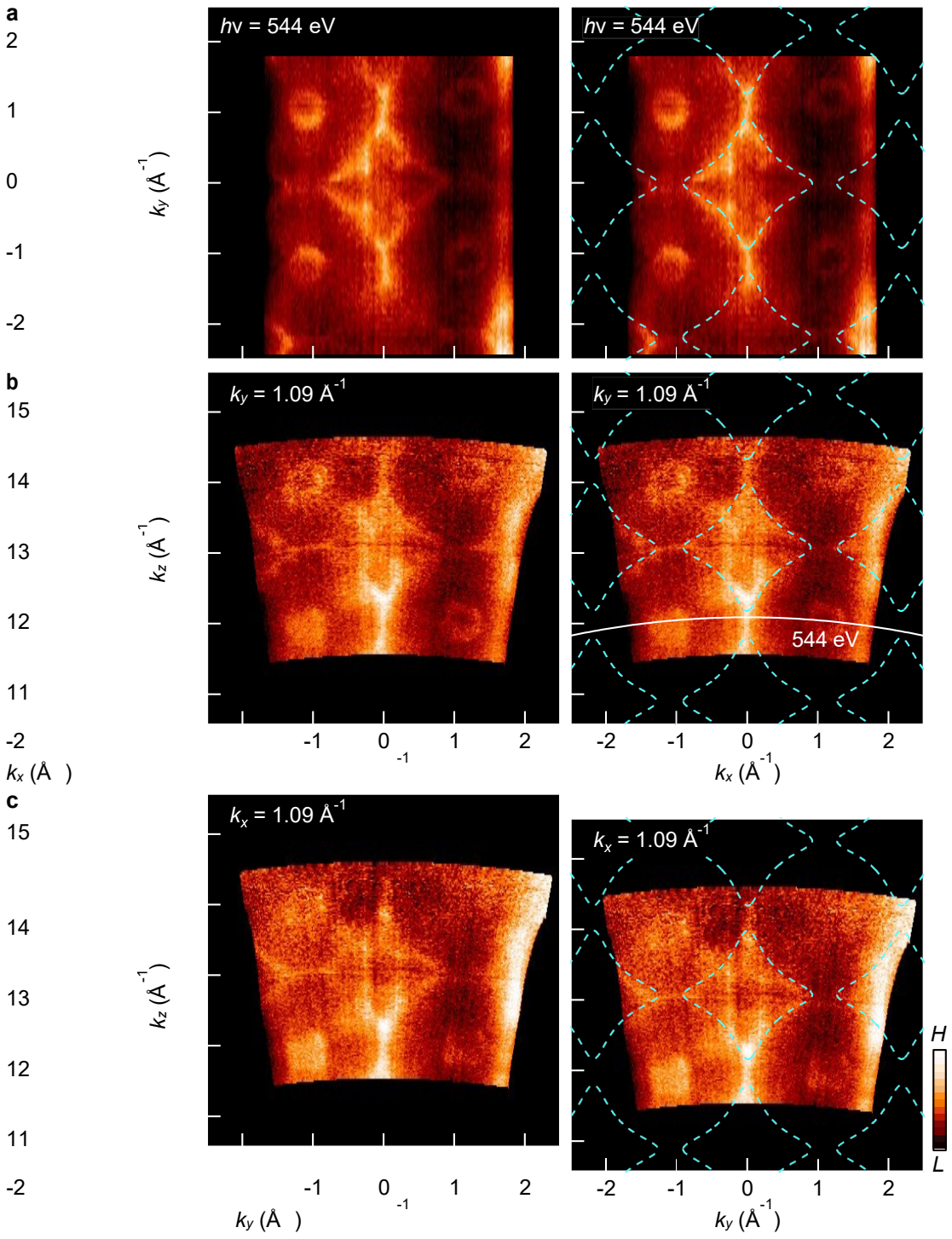


FIG. R7: **Unsymmetrized Fermi surfaces.** Left: photoemission spectra displayed in main text Fig. 2a, without symmetrization or filtering. Right: the same spectra, with the experimentally- determined Weyl loop trajectory overlaid across multiple Brillouin zones.

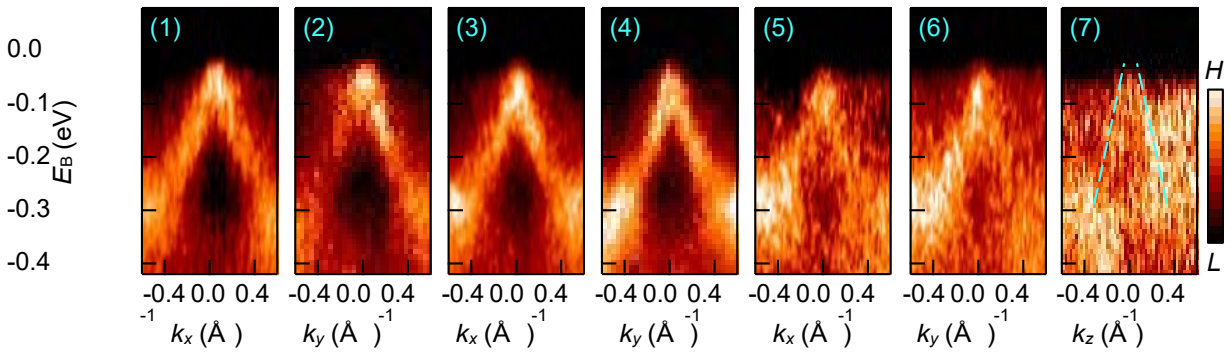


FIG. R8: **Unsymmetrized energy-momentum cuts.** Photoemission spectra displayed in main text Fig. 5a, without symmetrization or filtering.

cause it can severely affect the band crossing. How the authors define the plane of symmetry?

Considering all those issues, I think that the raw data should be shown as well.

Authors: We again thank the referee for the remark and we apologize that our initial submission was not clear on this point. The momentum-energy cuts in Fig. 5a are symmetrized according to the nominal mirror symmetries of the crystal. The symmetrization simplifies presentation of the data for the general reader, but plays no role in drawing conclusions from the data.

In our revised manuscript, we have provided the full complement of raw datasets as Ex- tended Data Fig. 8 (Fig. R8). We have also noted in the Methods, "The high-symmetry energy-momentum cuts were similarly symmetrized about M_x, M_y or M_z, as appropriate and consistent with the nominal symmetries of the crystal (Fig. 4a)." The raw data are consistent with the symmetrized Fig. 5a. All datasets clearly indicate a Weyl cone feature and provide further evidence of linked Weyl loops in Co_2MnGa.

Referee #3, Q4: 2) The major concern I have is about the presentation of the data in an extended zone, that is not what has been measured experimentally. I think that the correct description of the data is provided in Figure 2 (and partially in Figure 5): the authors have measured the dispersion of a bulk state in the 3 mutually orthogonal planes as indicated

with violet, red and orange colors. The interpretation that is given in Figure 4 and partially in Figure 5 is a wrong geometrical construction.

We can easily understand that the working assumption is wrong, by the fact that in Figure2 the Fermi surface contour extends beyond the square projection of the bulk Brillouin zoneon the plane passing through the *X* point. This is telling us that the correct description should be given in terms of the 3D bulk Brillouin zone, as naturally expected for a 3D bulk state. This is a well-known problem also for the Fermi Surface of noble metals, which exhibitsimilar bulk Brillouin zone.

Authors: We thank the referee for the comment and we apologize that we did not sufficiently help the reader navigate momentum space. First, as the referee no doubt un- derstands, soft X-ray ARPES naturally probes the bulk electronic structure in an extended zone scheme. Consequently, all of our ARPES data should be understood from the point of view of the extended zone scheme and we have used the extended zone three-dimensional bulk Brillouin zone throughout our work. Please note that the square drawn in Fig. 2d is*not* the two-dimensional projection of the bulk Brillouin zone but simply the top face ofthe bulk Brillouin zone. The fact that the Fermi surface contours of Fig. 2 extend beyondthe square face simply indicates that those states live in the neighboring bulk Brillouin zones.

In the revised manuscript, we plot the nominal measured momentum-space slices in Ex- tended Data Fig. 10 (Fig. R9). We believe this should help the reader more easily parseour ARPES data.

Referee #3, Q5: 2.1) I think the authors should show clearly the whole tomography of the 3D Fermi surface, of which the contours in Figure 2 show only 3 cuts on the 3 planes. This can be easily done by calculations, but can be done also experimentally. In general, I think that showing this closed lines in a 3D Brillouin zone is misleading. The Fermi surface must be a surface, it can be a close surface (3D) or it can be open in 1 or 2 direction in the case of systems with reduced dimensionality. Such a tomography for different planes wouldbear the additional advantage to show how the Weyl loop evolve far from the planes passing

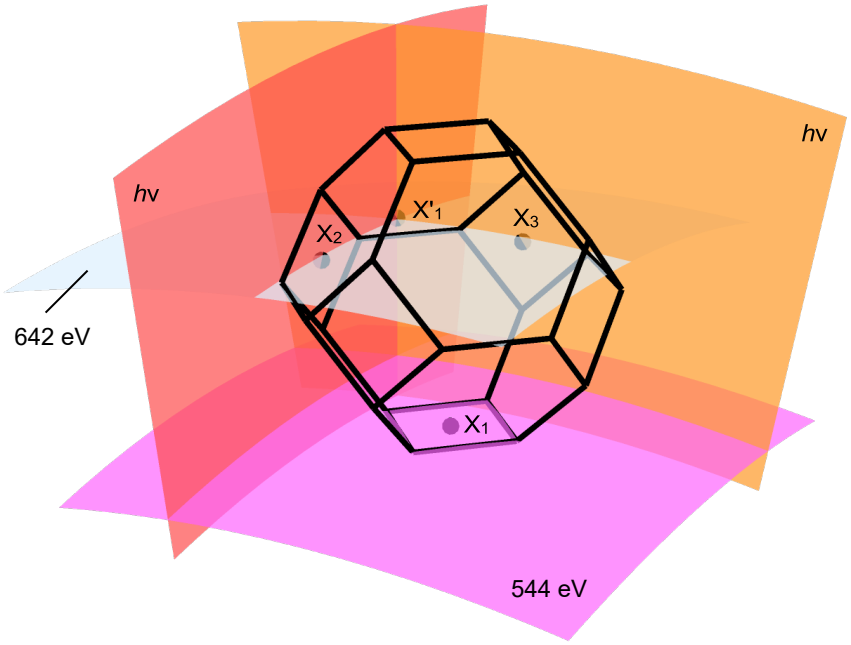


FIG. R9: **Measured Fermi surfaces in an extended zone scheme.** The Brillouin zone corresponds to $\Gamma_{(066)}$ in the primitive reciprocal basis.

through *X*.

Authors: We thank the referee for the insightful suggestion. In our revised Extended Data,we have included a three-dimensional Fermi surface obtained by *ab initio* calculations nearthe experimental Fermi level (Extended Data Fig. 9, Fig. R10). This Fermi surface clearly illustrates Weyl loop links in the electronic structure, providing additional evidence for a linked loop quantum state.

Referee #3, Q6: 2.2) At 540 eV an angular acceptance of $\pm 10^{\circ}$ is already sufficient to cover $\pm$ 2 Å^{-1}, and the momentum window gets as large ± 2.5 Å^{-1} at 800 eV. This should as

be sufficient to directly probe the high-order Brillouin zones. This would allow to observe the linking structure which is now only sketched.

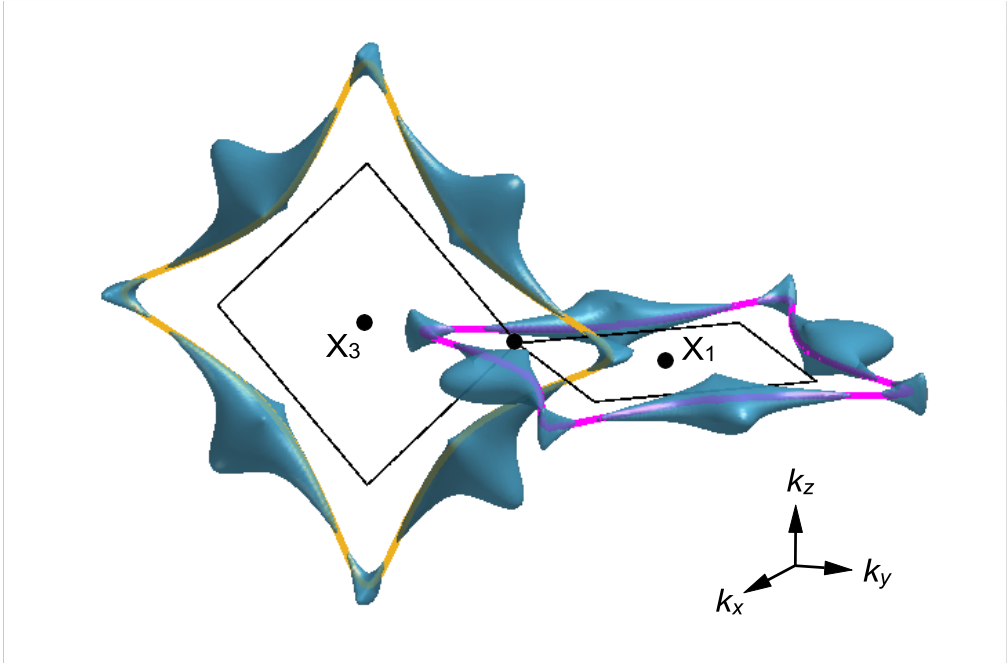


FIG. R10: **Linked Weyl loop Fermi surface.** Constant-energy slice of the pockets (navy) making up two linked Weyl loops obtained by *ab initio* calculation, at binding energy $E_B = -10$ meV, below the experimental Fermi level. The Fermi surface pockets touch at a set of discrete points, where the Weyl loop disperses through this particular E_B. For reference, the full Weyl loop trajectories are indicated, collapsed in energy (orange around X_3, magenta around X_1). We observe that the Weyl loop Fermi surface pockets form a linked structure.

Authors: We thank the referee for the remark and for the careful check of the accessible momentum range. Examining a wider momentum range may be interesting, but doesnot necessarily allow an improved observation of the linking structure. For any given high-symmetry plane, for example the one indicated in Fig. 2d, expanding the range will cover additional distant X_1 points, but the Weyl loops associated with these well-separated X_1 points do not link. Rather, we observe that links are only formed between neighboring pairs of mutually-orthogonal Weyl loops, such as X_1 and a neighboring X_2. Due to the intrinsically three-dimensional nature of the link, we systematically examined Weyl loops living in multiple mutually-orthogonal slices of momentum space in order to determine the link structure.

In our new Extended Data Fig. 6, we have plotted the Fermi surfaces for Figs 2.d-f in the full measurement range captured by the spectrometer (Fig. R7).

Referee #3, Q7: 2.3) For completeness, the authors should show also the Brillouin zone projection on different planes, for example passing through Γ.

Authors: We again thank the referee for the suggestion and we apologize that we did not sufficiently illustrate the Brillouin zone structure in our initial submission. In our revised Extended Data, we have plotted a high-symmetry Γ slice through the bulk Brillouin zonein an extended zone scheme (Extended Data Fig. 2d, Fig. R11d).

Referee #3, Q8: 3) Related to the issue of the data representation in the reduced/extended zone, I think a key point is the number of states observed. In the data in the whole manuscript, I see one single contour. Figure 2 simply shows that this single contour obeys the symmetry of the crystal. In order to observe a link structure, the authors should observe the doubling of the states.

Authors: We thank the referee for the remark. First, we clarify that our analysis suggests that the contours in Fig. 2d-f are three distinct and separate contours, rather than three slices of a single large three-dimensional contour. Based on our systematic experimental data, comparison with *ab initio* calculation and symmetry considerations, we conclude in our work that these three contours (Fig. 2d-f) trace out three distinct, symmetry-relatedand linked Weyl loops.

As the referee notes, it is possible to observe signatures of multiple linked Weyl loops in a single dataset. For example, we kindly point the referee to our Extended Data Fig. 5b,c, where we present energy-momentum cuts exhibiting pairs of linked Weyl loops. In our revised manuscript, we have further included an additional Fermi surface acquired at $h\nu$ = 642 eV which exhibits signatures of all three mutually-orthogonal Weyl loops (Extended Data Fig. 5d, Fig. R12d.

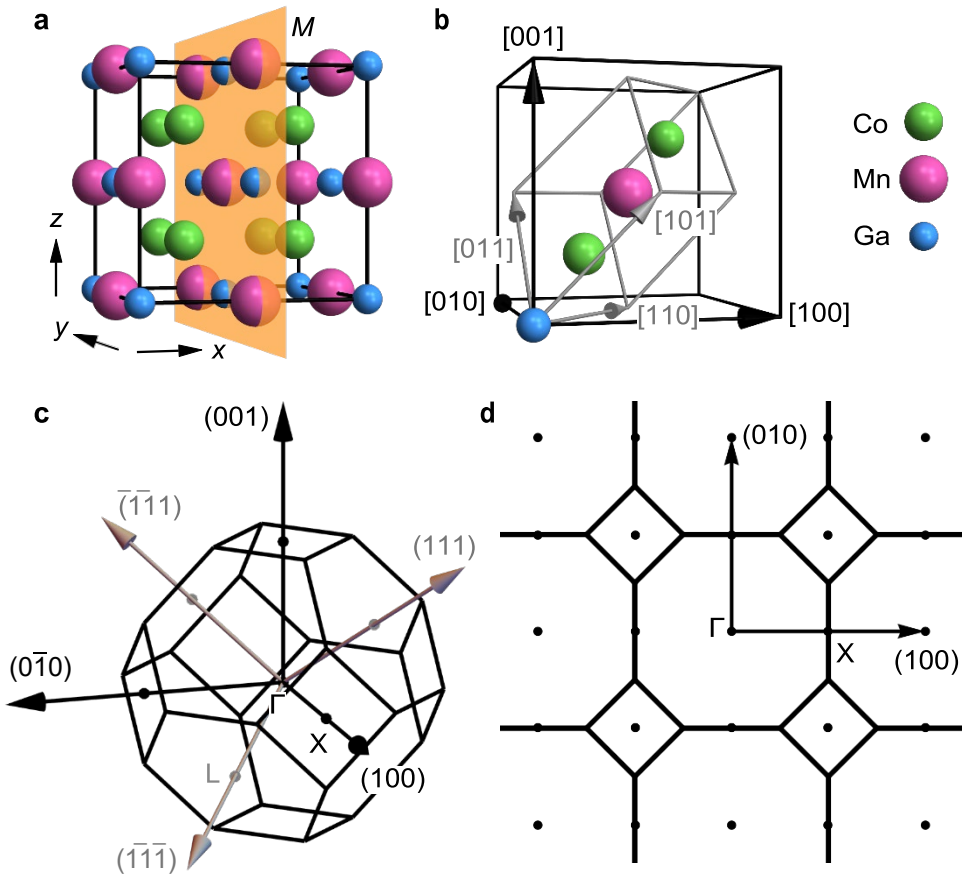


FIG. R11: **Crystal structure and Brillouin zone of Co_2MnGa. a**, Conventional unit cell with representative crystallographic mirror plane *M* (orange). **b**, The primitive unit cell (grey) includes one formula unit. **c**, Brillouin zone, with reciprocal lattice basis vectors (grey). In the reciprocal lattice basis, the M_1 plane corresponds to (001), M_2 corresponds to (010) and M_3 corresponds to (100). **d**, Slice through Γ in an extended zone scheme.

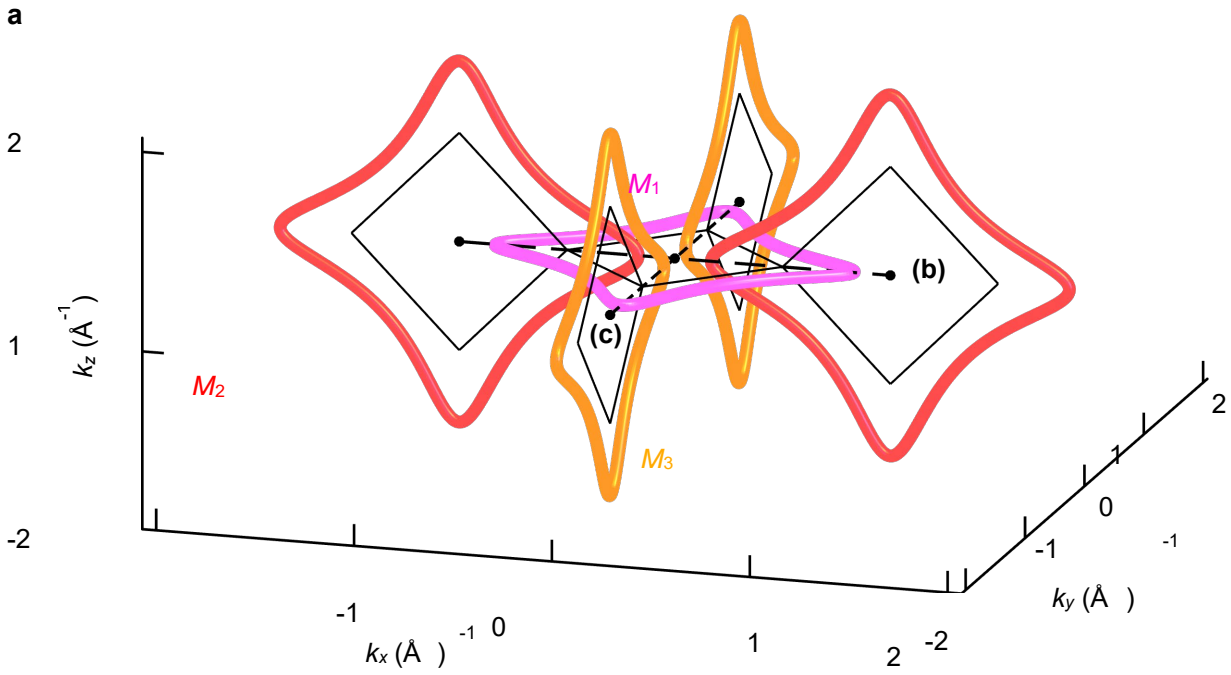
Referee #3, Q9: 4) From a theoretical point of view, it is quite unclear the physical interpretation of the linking number. It is computed from Figure 5 on the basis of the extended zone replica, but I see no topological implication or connection to the Berry phase. By doping the material, in principle the dimension of the Fermi contour could be reduced and the linking could be changed, or is it enforced by something? If there is nothing imposing and protecting this linking number, then it is just a number, and I do not see why it should be important.

Authors: We thank the referee for the remark. The linking number is protected in a way similar to traditional topological invariants characterizing topological insulators and Weyl semimetals.

For topological insulators, we say that the surface state Dirac cone is protected in the sense that it is robust to small, arbitrary, symmetry-preserving perturbations. Such perturbations may change the detailed surface state dispersion, but cannot change the fact that the Dirac cone connects the bulk valence and conductions bands. For the case of $\mathbb{Z}_2$ topological insu- lators, the relevant symmetry is time-reversal symmetry—if ferromagnetism is introduced, the Dirac cone surface state can gap out and be removed. Large symmetry-preserving perturbations of the electronic structure can invert the bulk band gap, removing the Dirac cone surface state via a topological phase transition. Regarding doping the material,topological insulators do not enjoy protection against carrier doping, which can freely change the position of the Fermi level and render the Dirac cone surface state relevant or irrelevant to transport response.

For Weyl semimetals, we similarly say that the Weyl points are robust to small, arbitrary, symmetry-preserving perturbations. Such perturbations may move the Weyl points around in momentum space, but cannot gap individual Weyl points. The relevant symmetriesfor Weyl semimetals are typically translation symmetry and $U(1)$ gauge symmetry—if a charge density wave or superconductivity is introduced, a gap can open in individual Weyl points. Large symmetry-preserving perturbations can bring pairs of Weyl points together in momentum space and annihilate them, opening a gap via a topological phase transition. Again, carrier doping the material may significantly change the Fermi surface by shifting the Weyl points far above or below the Fermi level.

Regarding our experimental observations in the present case, we again expect that the linked Weyl loops are robust to small, arbitrary, symmetry-preserving perturbations. Such perturbations may change the detailed trajectory of the Weyl loop in momentum space, but cannot change the linking number. We expect that the essential symmetry is mirror symmetry—if a mirror-breaking lattice distortion were introduced, the Weyl loops could gap out, removing the link. Large mirror-respecting perturbations are expected to unlink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M1
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(b)
(c)
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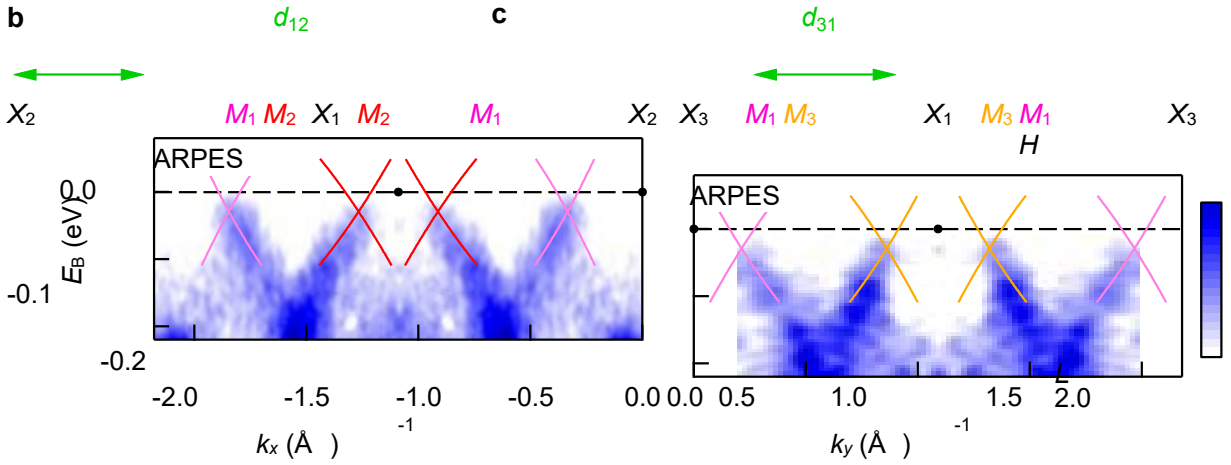
ky (Å-1)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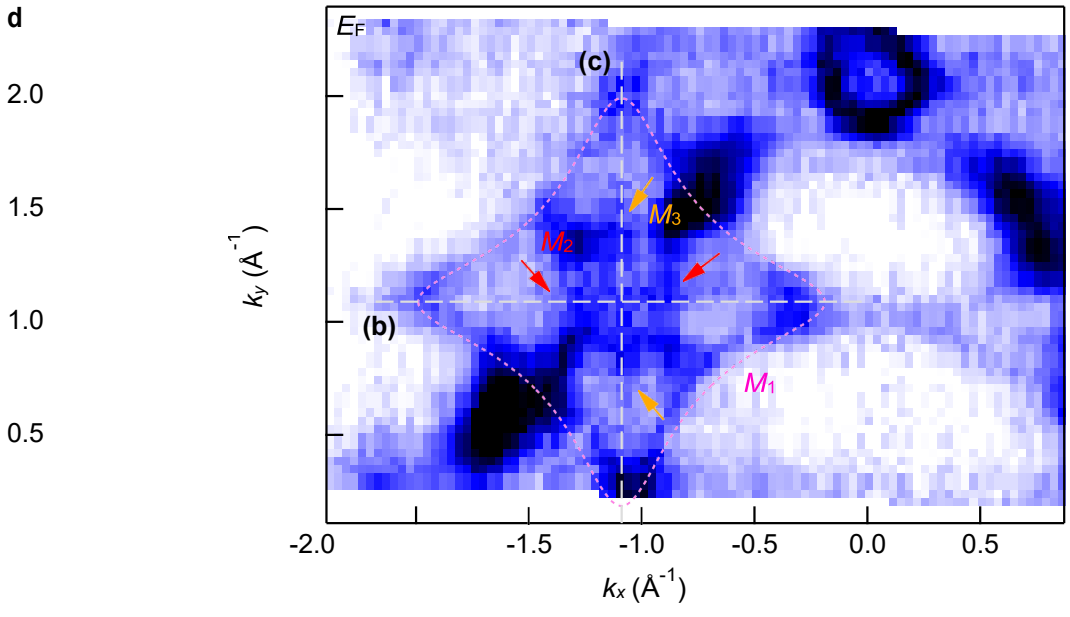
d
EF
(c)
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M2
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ky (Å-1)
kx (Å-1)

FIG. R12: **Independent measurement of the link depth. a**, M_1, M_2 and M_3 Weyl loops, with trajectories obtained from the analytical model (see main text), showing that M_1 links M_2 twice and M_3 twice. Energy-momentum photoemission slices along the high-symmetry paths **b**,$X_1 - X_2$ and **c**, $X_3 - X_1$ obtained at photon energy $h\nu$ = 642 eV. We observe d_{12} = 0.56 ± 0.1 Å^{-1} and d_{31} = 0.61 ± 0.1 Å^{-1}, consistent with Extended Data Fig. 4. **d**, Fermi surface acquired at $h\nu$ = 642 eV, exhibiting an in-plane Weyl loop contour, M_1. We further observe spectral weight emanating along k_x and k_y from the center of M_1, corresponding to the linearly dispersive branches in (**b**, **c**), again suggesting that M_1 is linked by M_2 and M_3.

the Weyl loops, changing the linking number via a topological phase transition. In this sense, the Weyl loop linking number is symmetry-protected, robust and meaningful.

In our revised main text, we have attempted to briefly clarify this point, writing "By analogy with topological insulators and Weyl point semimetals, this Weyl loop link is expectedto be stable under arbitrary, small, symmetry-preserving perturbations of the electronic structure.".

Referee #3, Q10: 5) A fallout of the previous question is which are the implicationof this specific linking structure in the transport, or magnetic, properties of the material? The authors speculate about a bridge between physics and knot theory, axion Lagrangianand non-Abelian node loop charges, but these concepts are very vague. I think that ifthe authors want to meet the requirements of *Nature*, they should strongly motivate how their findings can open new field of research, by explaining in which direction and how the scientific community could move.

Authors: We appreciate the interesting question regarding future implications of our work. We propose two phenomena associated with linked Weyl loops in Co_2MnGa which the community could pursue.

1. **Topological Seifert boundary modes.** In our revised manuscript, we present signatures of topological boundary states associated with the Seifert surface of the bulk

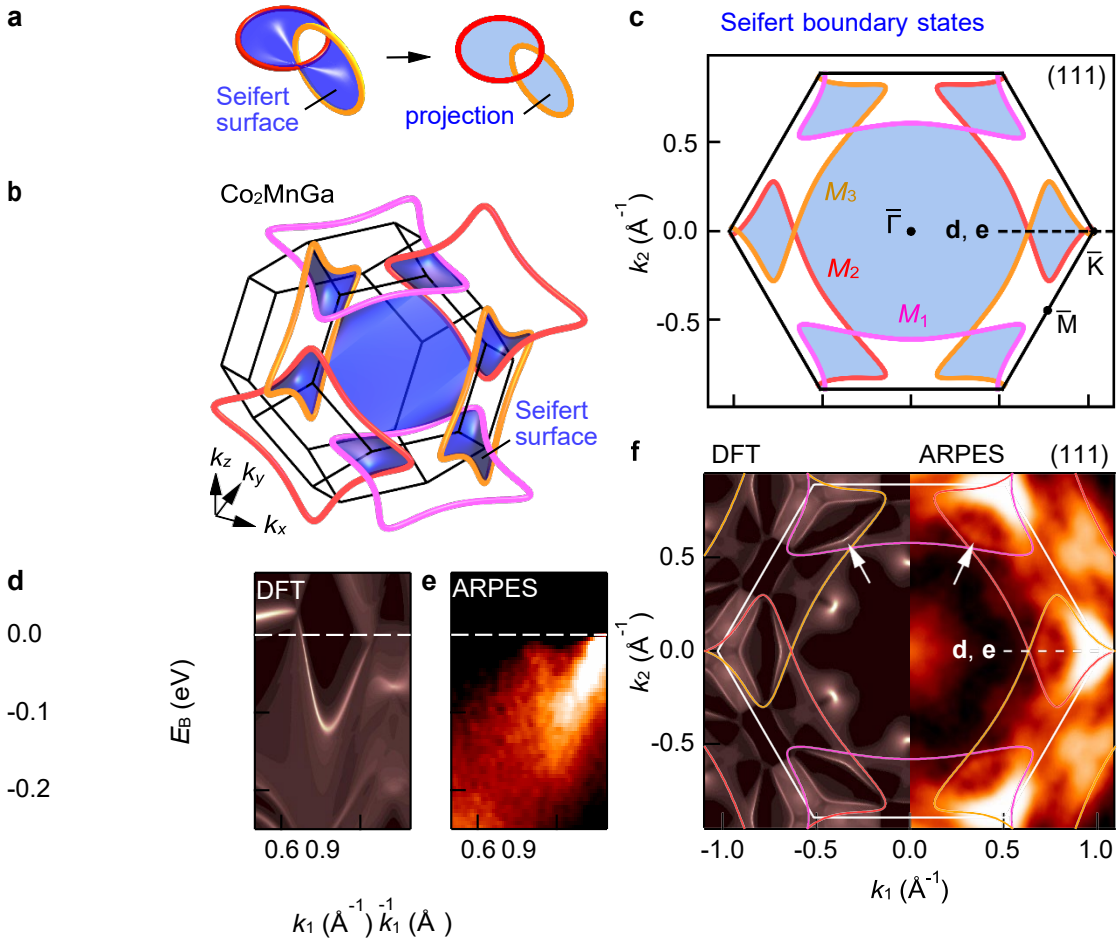

FIG. R13: **Seifert bulk-boundary correspondence. a**, A Seifert surface is defined as a three-dimensional surface bounded by a link, shown for the example of a Hopf link. Its two-dimensional projection produces alternating filled and empty regions pinned together at characteristic touching points. **b**, In a condensed matter system, the Seifert surface is taken as a surface bounded bythe linked loop nodes in three-dimensional momentum space (k_x, k_y, k_z), shown for the case of the linked loop nodes observed in Co_2MnGa. **c**, The projection of the Seifert surface into the surface Brillouin zone is associated with topological boundary modes (blue regions) which touch at points in momentum space. Energy axis collapsed for clarity. **d**, *Ab initio* calculation of the surfacestates through the touching point, exhibiting pairs of boundary modes pinned together at the Weyl loops. **e**, Surface-sensitive vacuum ultraviolet (VUV) ARPES energy-momentum cut through the touching point, exhibiting signatures of the pinned Seifert boundary modes, consistent with *ab initio* calculations. Photon energy $h\nu$ = 63 eV. **f**, Fermi surface in *ab initio* calculation (left) and VUV-ARPES (right) exhibiting Seifert boundary modes that stretch across the topological regions, connecting different Weyl loops, consistent with the Seifert projection.

Weyl loop link (main text Fig. 5, shown in this document as Fig. R13). Mathematically, a Seifert surface is a three-dimensional surface bounded by a knot or link [*Math. Ann.* **110**, 571 (1935)]. For example, in the minimal case of a Hopf link, the Seifert surface exhibits a branched structure which 'wraps' around the link (Fig. R13a, left). Upon projection into two dimensions, the Seifert surface produces a set of filled regions which touch at discrete points (Fig. R13a, right). For a linked loop quantum state in a condensed matter system,the Seifert surface is taken to live in bulk momentum space (k_x, k_y, k_z) with the energyaxis collapsed. The topological boundary modes on all cleaving planes are given by the projection of the Seifert surface [*Rev. Mod. Phys.* **93**, 015005 (2021), *Comm. Phys.* **4**, 47 (2021), *Phys. Rev. B* **99**, 161115(R) (2019), *Commun. Phys.* **2**, 135 (2019)]. These Seifert boundary modes then exhibit a unique configuration of filled regions pinned together at touching points in momentum space, associated with the bulk links. This relationship between the link, its Seifert surface and the topological boundary modes suggests a **Seifertbulk-boundary correspondence**, where the linked loop nodes in the bulk protect Seifert boundary states and, conversely, the characteristic configuration of Seifert boundary states encodes the linking number. By examining the (111) sample surface, **we observe experimental signatures of these Seifert boundary states**, consistent between *ab initio* calculation and surface-sensitive vacuum ultraviolet (VUV) ARPES (Fig. R13d-f).

2. **Linked Weyl loop axion electrodynamics.** The axion θ term may give rise to a topological magneto-electric effect in electronic structures hosting linked node loops [*Phys. Rev. B* **96**, 081114(R) (2017), *Phys. Rev. B* **95**, 094512 (2017), *Phys. Rev. Lett.* **119**, 147001 (2017)]. To achieve this effect in Co_2MnGa, we expect that it is first necessary to gap the Weyl loops by breaking mirror symmetry. This could potentially be achieved in thin film samples of Co_2MnGa via an asymmetric substrate-induced lattice strain. Fermi level tuningor perturbation of the electronic structure may then be needed to place the Fermi level inthe Weyl loop gap, possibly achieved by engineering the material's magnetic properties viathe film substrate, temperature, carrier doping or chemical substitution. Transport or optics experiments may then detect quantized axion electrodynamics, as previously investigated in topological insulators [*Science* **354**, 1124 (2016)]. In the present case the axion response is expected to be quantized not to the topological insulator $\mathbb{Z}_2$ invariant, but rather to the Weyl loop linking number. In our revised manuscript, we have pointed out that: "Moreover, the

linked loop state in Co_2MnGa, as well as in other materials, may give rise to exotic response quantized to the linking number, such as a link-quantized topological magneto-electric effect [25, 32, 33, 48].", citing these four references.

Referee #3, Q11: Minor question: 6) Why in the data of Figure 2e, the intensity around the Γ point is not observed, in contrast to panel d?

Authors: We again thank the referee for the comment and for taking the time to closely examine our data. We do find that the intensity of the irrelevant Γ states is suppressed in the out-of-plane photon energy dependence (Fig. 2e) compared to the in-plane Fermi surface (Fig. 2d). This may be due to intrinsic limits on k_z resolution associated with thephoton energy dependence, which may reduce the signal-to-noise ratio of certain states.

In our revised manuscript, we have included the raw datasets for both of these measurementsas Extended Data Fig. 6 (Fig. R7). Notably, in the raw photon energy dependence, we can clearly observe intensity around the Γ points, similar to the in-plane Fermi surface. Inthe associated figure caption we have noted that, "The irrelevant Γ pocket is consistently observed in all unsymmetrized spectra."

Summary: We summarize our main points,

1. **Source data:** We have provided all raw data as additional Extended Data, as well as clearly indicated the symmetrization procedures that we used. Crucially, all conclusions are drawn directly from raw datasets produced by the spectrometer. The symmetrization only serves to improve the presentation for the general reader.
2. **Theoretical & experimental evidence for Seifert boundary states:** In our revised manuscript, we present signatures of Seifert boundary states in ARPES for the first time, providing an example of the Seifert bulk-boundary correspondence in quantum matter (Fig. R13).
3. **Link-quantized topological magneto-electric effect:** Recent theoretical work indicates that linked node loops may be a rich source of phenomena relating to the ax-

ion θ angle, the gravitational θ angle and non-Abelian topological charges, producingexotic phenomena in quantum matter, photonic crystals and mechanical metamateri- als. However, to date linked node loops have not been reported in any such system. The availability of an experimental platform to explore the theoretical proposals is clearly urgently needed. In the present manuscript, we provide for the first time a direct observation of linked node loops in a quantum material. We further propose that the linked Weyl loops specifically in Co_2MnGa may give rise to topological axionelectrodynamics quantized to the Weyl loop linking number.

We have attempted to clarify these points in our revised submission. We again thank the referee for her/his remarks and for helping us improve our manuscript.

Reviewer Reports on the First Revision:

Referees' comments:

Referee #1 (Remarks to the Author):

The authors have done a good job answering my questions and comments. It is indeed an interesting result. I have nothing against the publication.

Referee #2 (Remarks to the Author):

In the revised manuscript, the authors added two contents to show the significance of the Weyl nodal link. One is the Seifert boundary modes., the other one is the proposed linked Weyl loop axion electrodynamics. Although the surface states of the Weyl nodal link could be explained by the existence of the Berry phase, Seifert boundary modes still could be of interest. However, I don't think these two points would be of broad interest to the physicists. I would not change my opinion that this paper is not suitable for Nature. Besides, the following comments should be addressed for future submission elsewhere.

1. In the manuscript, there are three kinds of nodal line marked as orange (M3), pink (M1) and red (M2) colors. Each of the three nodal loops is related to the others by the rotational symmetries. The type of these three nodal loops is one of the three types of nodal loop in Ref [20] [21], i.e. the yellow lines in Fig 3 d,e,g,h and i of Ref. [21]. The link of the yellow loops is called as Hopf link in Ref. [21] because they only consider the link of two loops instead of link of all nodal loops. That's the point that the authors claimed to be very important of this manuscript. However, the authors should consider the links extended to the whole momenta space. In such case, the link number will not be (2,2,2) and would be complicated. They think the way that only two loops are considered as a Hopf link in Ref.[20] [21] is wrong. The same logical is that only three loops are taking into account is also wrong. We should consider the links of all loops extended to the whole momenta space.

2. From the reply: "The observed link disagrees with the theoretical prediction: The previous work, Ref. [21], only suggested that a Hopf-like link many arise in Co2MnGa, but did not theoretically predict the link diagram, calculate the associated linking numbers or predict that the Co2MnGa link is geometrically essential."
Actually, Ref. [21] gives the link structure of the whole nodal lines. See Fig.3 in Ref.[21]. The yellow lines in Fig 3d are the whole structure of the link of the nodal loops mentioned in this manuscript.

Referee #3 (Remarks to the Author):

I thank the Authors for their careful rebuttal. I think that the inclusion of the raw data is extremely useful to better visualize the spectroscopic features under discussion.
I still have doubts about the data interpretation, and moreover I feel like few of the questions that I have raised in my previous report are still waiting for clarification. I cannot recommend the publication of the article in the present form.

1) The central point that requires further explanation is the actual form of the Fermi surface. The authors claimed that "our analysis suggests that the contours in Fig. 2d-f are three distinct and separate contours, rather than three slices of a single large three-dimensional contour". The central point of the manuscript is the unique way those lines are linked, something that is pictorially illustrated in several Figures but in my opinion still needs to be experimentally confirmed. Now that raw data are available to the readers, by putting together the information from Extended data Fig. 10 and 9, I have produced a simple sketch that shows what the authors have indeed measured, or at least what I have understood. (See Figure in attachment).

In panel a, I have added a white line that corresponds to the plane investigated as a function of kz. The dashed line forming the closed Weyl loop is highlighted by a green square. It lies completely on one side of the white line (for ky<1.09A-1), it does not cross the line/plane. Similarly if we consider panel b, the Weyl loop lies at kz larger than the plane corresponding to 544 eV photon energy. If I have understood well, by combining this information in the same 3D plot (panel c), the green rectangles they do not cross each other. I do not see any linking between them. The same argument is valid for the orange plane.
This is, in my opinion, what is contained in the experimental data: the description given in Figure 3 of the main manuscript does not correspond to what has been experimentally measured.

2) In general, the possibility to form complex link-loops is granted (if I understood carefully the DFT calculations shown in the Extended Data Fig. 9) by the fact that the electronic properties are formed by highly elongated Fermi sheets, in contrast to the common wisdom of a bulk 3D Fermi surface. This is indeed extremely interesting, and it would be highly appreciated to explain it from a microscopic point of view (in terms of orbitals or Wannier functions).
While the origin of such a feature might be matter for a future theoretical investigation, I think it is mandatory to confirm this from an experimental point of view. This is, in fact, the solid foundation for confirming the nature of the link-Weyl loops.
This can easily be done by exploring the evolution of the Fermi surface in the kx-ky planes in the region across the bulk X1 point, for example for few selected photon energies between 520 and 570 eV. According to Extended Data Fig. 9, the Weyl loops around X1 is restricted to a small range of kz, and it would be interesting to understand its evolution.
This might help the authors to further support the presence of a link structure.

3) Moreover, although the authors draw lines and cones in the dispersion of the data, for example in Extended Data Fig. 5, I cannot see those features, I always see one of the states traced by the colored lines

4) In general, I think that it is difficult, if not impossible, for a general audience to understand the physics contained in the data. Although the Authors share now more information about the data, and the data analysis, I have always the feeling that the data presentation is excessively complicated.
I invite the Authors to add important information to their data, to guide the reader in the correct interpretation. Whenever it is possible, it would be useful to indicate the projections of the different Brillouin zone (distinguishing their order with different colors).

5) I find the calculated Fermi surface shown in Extended Data Fig. 9 very simplified, it would be nice to have something like what is commonly found in other publications on the same subject, see for example the

Fermi surface IrF4 in Fig.3 d of the seminal work of Bzdušek et al. (Nature 538, 75 (2016)).
In think that such a representation of the 3D Fermi surface within the reduced Brillouin zone is fundamental to describe the link-structure. Additionally, a careful description of the calculated band structure and Fermi surface is also important to draw a comparison with calculations already present in the literature, as for example the data shown in Figure 2e in the pioneering work of Belopolski et al. on the same material (Science 365 1278 (2019)).

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6) Concerning the new part, I have read with interest the attempt of the Authors to introduce new theoretical elements from knot theory in the electronic properties of this interesting material. However, as for the link-structure, I have the impression that the data are insufficient to fully support the claim.

6.1) First of all, the Authors compare two completely different sets of data: SXARPES (more bulk sensitive) on the (001) termination, and VUV (more surface sensitive) on the (111) termination. This makes impossible for the readers to compare the results in order to isolate the surface from the bulk states. While the calculated band structure of Figure 5 d contains many bands, the ARPES data of Fig. 5 e shows one parabolic band. This might correspond to the surface state, or not, especially because they occupy different position in the momentum space (the measured band is shifted of more than 0.1A-1 towards higher k value).
In the comparison between surface and bulk states, it is important to notice that in the literature the Weyl-lines have been observed with VUV ARPES at 50 eV (Science 365 1278 (2019)). We can conclude that the use of 63 eV is not sufficient to suppress completely the bulk spectral features in favor of the surface state. I invite the Authors to make a more careful comparison between theory and experiments at different photon energies in the VUV and soft X-ray in order to fully elucidate the surface and bulk dispersion in the (111) surface termination

6.2) The topological nature of the surface state has not been discussed. This surface state might be trivial. It nicely follows the dispersion of the bulk bands, and it might be some kind of surface resonance. The authors should discuss, at least on a theoretical basis, how breaking the mirror symmetries and the Weyl-lines is affecting the surface state dispersion.

Author Rebuttals to First Revision:

Re: *Nature* 2020-10-18080, Observation of a linked loop quantum state

We sincerely thank the referees for taking the time to review our manuscript in a second round of review. In the following we respond to the remaining comments of the referees, point by point.

REPORT OF REFEREE #1

Referee #1, Q1: The authors have done a good job answering my questions and comments. It is indeed an interesting result. I have nothing against the publication.

Authors: We thank the referee for supporting publication of our work.

REPORT OF REFEREE #2

Referee #2, Q1: In the revised manuscript, the authors added two contents to show the significance of the Weyl nodal link. One is the Seifert boundary modes, the other one is the proposed linked Weyl loop axion electrodynamics. Although the surface states of the Weyl nodal link could be explained by the existence of the Berry phase, Seifert boundary modes still could be of interest. However, I don't think these two points would be of broad interest to the physicists. I would not change my opinion that this paper is not suitable for *Nature*.

Authors: We sincerely thank the referee for studying our revised manuscript. The observation of an intricate link formed by electrons in a quantum material will no doubt be of broad interest to physicists, chemists and engineers. Even non-scientist readers of *Nature* may be fascinated by the notion of a quantum link and may find the link structure aesthet- ically appealing. Knots and links are one of the oldest human technologies; they appearas powerful artistic and religious symbols across human cultures; and in the modern era they have been objects of profound scientific and mathematical interest [J. C. Turner, P.van de Griend, *History and Science of Knots* (1996); C. C. Adams, *The Knot Book* (1994)]. Our discovery of a highly non-trivial quantum linked structure in Co_2MnGa represents an important contribution to this tradition. We elaborate on the significance of our work,

1. **Topology can drive new subfields of science:** Discoveries of new manifestations of topology often open rich new research directions.

    - *Organic chemistry:* Breakthroughs in template-directed synthesis of **linked polymer macrocycles** in the early 1980s opened the study of linked catenanes and rotaxanes [J. P. Sauvage, C. Dietrich-Buchecker, *Molecular Catenanes, Rotaxanes and Knots* (1999)], leading to the development of link-based molecular switches, molecular rotary motors and high-specificity electrochemical sen- sors. Linked organic polymers have come to represent a **triumph of synthetic supramolecular chemistry** [*Ang. Chemie*, **54**, 6110 (2015)].
    - *Quantum mechanics:* The discovery that the **Jones polynomial** of knot theory could be associated with vacuum expectation values of Wilson loops in Chern- Simons theory catalyzed the development of **topological quantum field theory**

[E. Witten, *Comm. Math. Phys.* **121**, 351 (1989); L. H. Kauffman, R. A. Baad- hio, *Quantum Topology* (1993)]. This then contributed to particle physics via the Khovanov homology; deepened our understanding of the electric-magnetic dual- ity, magnetic monopoles and fractional electronic charges; and contributed to thesupersymmetric theory of stochastic dynamics [T. Ohtsuki, *Quantum Invariants* (2001)].

- *Condensed matter physics, order parameters:* Windings of order parameters in real space are a unifying theme of condensed matter, encompassing disclinations in liquid crystals; vortices in superconductors and superfluid ^{4}He; and magnetic skyrmions, whose invariants are proposed as the basis for next-generation com- puting memory and logic.
- *Condensed matter physics, band topology:* Momentum-space wavefunction wind- ing is associated with emergent Dirac fermions in two- and three-dimensional topological insulators; Weyl fermions in topological semimetals; and the quantum Hall effect, which sets the prevailing von Klitzing standard of electrical resistance.

Our results provide a new and qualitatively different example of topology in nature: the linking of Weyl loops in electronic structure. The historical importance of earlier topological phenomena suggests that linked node loops are also likely to become a fruitful topic of research.

2. **Seifert boundary modes:** We appreciate that the referee feels that "Seifert bound- ary modes still could be of interest". Indeed, there has been intense interest in ob- serving linked loop states with Seifert boundary modes throughout physics, not only in electronic structures, but also in optics experiments, electric circuits, mechanical systems and cold-atom platforms [*Rev. Mod. Phys.* **93**, 015005]. For the first time, we experimentally demonstrate this unconventional linked topological structure.

3. **Quantized magneto-electric effect:** The axion θ term may give rise to a topological magneto-electric effect in electronic structures hosting linked node loops [*Phys. Rev. B* **96**, 081114(R) (2017), *Phys. Rev. B* **95**, 094512 (2017), *Phys. Rev. Lett.* **119**, 147001 (2017)]. To achieve this effect in Co_2MnGa, we expect that it is first necessary togap the Weyl loops by breaking mirror symmetry. This could potentially be achieved

in thin film samples of Co_2MnGa via an asymmetric substrate-induced lattice strain. Fermi level tuning or perturbation of the electronic structure may then be neededto place the Fermi level in the Weyl loop gap, possibly achieved by engineering the material's magnetic properties via the film substrate, temperature, carrier doping or chemical substitution. Transport or optics experiments may then detect quantized axion electrodynamics, as investigated in topological insulators [*Science* **354**, 1124 (2016)]. In the present case the axion response is expected to be quantized not to the topological insulator Z_2 invariant, but instead to the Weyl loop linking number.

4. **Direct photoemission observation:** Complex magnetic loop node structures have recently attracted considerable interest but **even state-of-the-art analyses have relied on indirect probes** such as de Haas-van Alphen oscillations of topological protectorates in MnSi [*Nature* **594**, 374 (2021)] and the giant anomalous Nernst effect arising from a Weyl loop network in Fe_3Ga [*Nature* **581**, 53 (2020)]. We provide the first photoemission tomography of a linked loop quantum state, directly measure its linking number, establish a new bridge between physics and knot theory, and observe signatures of a Seifert bulk-boundary correspondence. Our work represents a **breakthrough comparable to the recent transport works**, published in *Nature*.

5. **Large class of candidate materials:** Dirac loops, Weyl loops and other classes of loop nodes are ubiquitous in crystals. In our present case, the linked loop phase is driven by a combination of mirror symmetry and ferromagnetism. We note that 160of the 230 space groups contain a mirror or glide symmetry and there are several thou-sand known ferromagnets, suggesting that more **linked Weyl loop phases await discovery**. Other classes of loop nodes, such as Dirac loops protected by a combi-nation of inversion and time-reversal symmetry, further broaden the list of candidate materials. Future materials discoveries of linked node loop states will likely providea **rich materials playground for exploring link-quantized magneto-electric effects**, other exotic response and device applications.

6. **Non-Abelian braiding:** Nodal loops can be characterized by non-Abelian topolog- ical charges [*Science* **365**, 1273 (2019)], which may allow braiding operations [*Nat. Phys.* **16**, 1137 (2020)]. These results suggest that **magnetic control of the link- ing number** in a loop node quantum phase, such as the one we discover in Co_2MnGa,

may allow a novel form of topological computation.

The discovery of a new example of topology in science is valuable and likely to be of interestto a broad audience of physicists, chemists, engineers and the general public.

Referee #2, Q2: Besides, the following comments should be addressed for future submission elsewhere.

1. In the manuscript, there are three kinds of nodal line marked as orange (M_3), pink (M_1) and red (M_2) colors. Each of the three nodal loops is related to the others by the rotational symmetries. The type of these three nodal loops is one of the three types of nodal loop in Ref. [20], [21], i.e. the yellow lines in Fig. 3d, e, g, h and i of Ref. [21]. The link of the yellow loops is called as Hopf link in Ref. [21] because they only consider the link of two loops instead of link of all nodal loops. That's the point that the authors claimed to be very important of this manuscript. However, the authors should consider the links extended to the whole momenta space. In such case, the link number will not be (2,2,2) and would be complicated. They think the way that only two loops are considered as a Hopf link in Ref. [20], [21] is wrong. The same logical is that only three loops are taking into account is also wrong. We should consider the links of all loops extended to the whole momenta space.

Authors: We thank the referee for carefully examining our work and the earlier literature. We appreciate that the referee highlighted our message that the system exhibits threelinked loops, giving linking number (2, 2, 2). It is important to consider **all loops in a single Brillouin zone** (a reduced zone scheme). Considering fewer loops by ignoring physically-distinct link components, as is done in Ref. [20], does not correctly characterize the linking number. On the other hand, considering the whole of momentum space (an extended zone scheme) introduces redundant copies of the electronic states and is also incorrect. While it may be useful to plot several extra copies of the loops in order to visualize the link structure, this redundancy must be taken into account when interpretingthe linking number. For example, although 6 loops are plotted in Fig. 4b, loops of the same color refer to redundant copies of the same physical states. In total, there are three distinct loops, one of each color. Linking between exactly these three distinct loops determines the

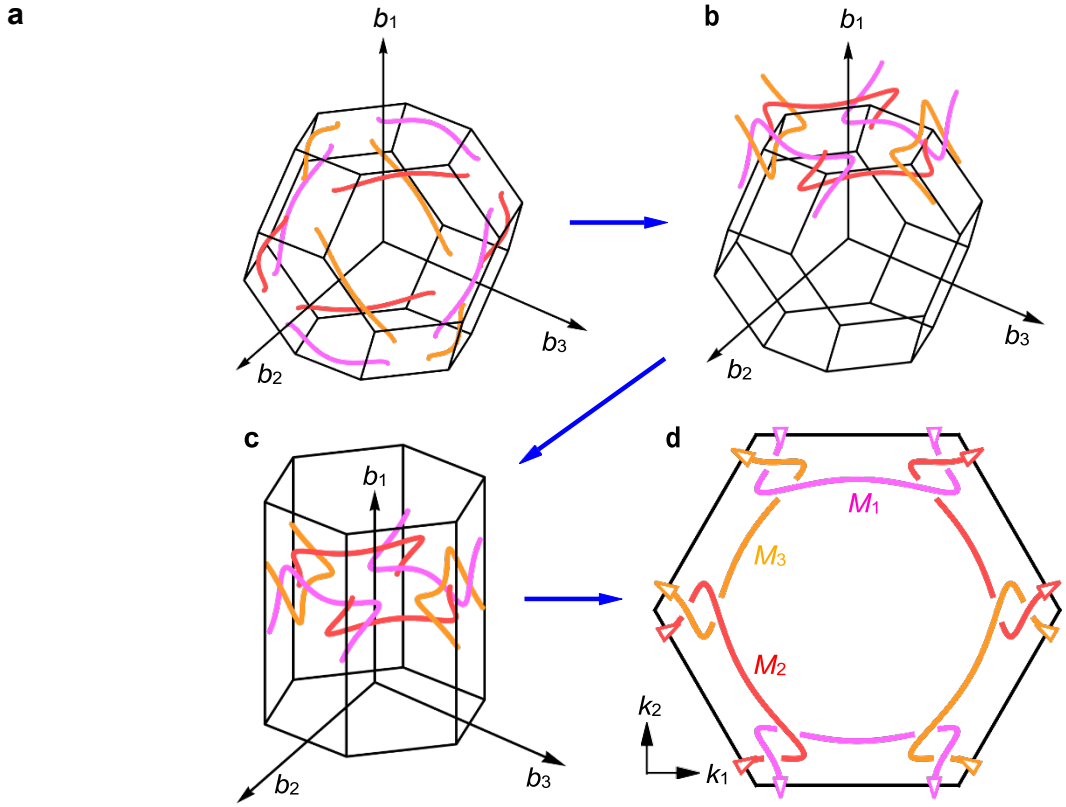

FIG. R1: **Analysis of the linking number. a**, Single copy of the linked node loops, plotted in the first bulk Brillouin zone. **b**, Again a single copy of the linked node loops, but plottedaround a hexagonal face of the Brillouin zone to make the linking more evident. **c**, The samesingle copy as in (b), plotted in a non-standard reciprocal primitive unit cell. The chosen primitive cell is a cylindrical prism with the same volume as the bulk Brillouin zone, but which naturally displays the linking for a single copy of the loops and which can be directly projected in the (111),or b_1 direction. **d**, Link diagram in the (111) surface Brillouin zone following projection, clearly exhibiting linking number $\nu = 2$ between each pair of loops (main text Fig. 4e).

linking number.

We kindly note that considering a single copy of physically-distinct states is typical in condensed matter physics. For example, the density of states of a band structure requiresan **integral over a single Brillouin zone**, so that each one-electron level is counted

exactly once [Eq. 8.57, Ashcroft & Mermin (1967)],

$$g(\varepsilon) = \int_{BZ^3} \delta\left(\varepsilon - \bar{\varepsilon}(k)\right) \frac{d^3k}{4\pi} \quad \text{[integral over one Brillouin zone]} \tag{1}$$

Similarly, a calculation of a Chern number also requires an **integral over a single two-dimensional Brillouin zone** [Eq. 4.4.6, Xiao-Gang Wen (2004)],

$$\sigma_{xy} = \frac{e^2}{h}\frac{1}{2\pi} \int_{BZ} d^2k \Omega_z(k) \quad \text{[integral over one Brillouin zone]} \tag{2}$$

In the present case, **it is necessary and sufficient to consider all Weyl loops associated with physically-distinct electronic states.** To further clarify this point, in our revised Supplementary Information we plot a single copy of the Weyl loops in a way which highlights the linking (Fig. R1a,b). Equivalently, we can partition extended momentum space with a non-standard reciprocal primitive unit cell (Fig. R1c). Although it is not the Brillouin zone, this alternative primitive cell has the same volume as the Brillouin zone and contains one copy of the electronic states. This choice of primitive cell naturally pulls out a single set of three loops in a way that manifests the linking structure. This primitive cell fur-ther lends itself to projection along (111) or b_1. We can then project the links into the (111) surface Brillouin zone, while preserving the out-of-plane information using an over/under notation. This procedure yields our link diagram and clearly demonstrates linking number$(2, 2, 2)$ for the Co_2MnGa node loops (Fig. R1d, main text Fig. 4e).

Referee #2, Q3: 2. From the reply: "The observed link disagrees with the theoretical prediction: The previous work, Ref. [21], only suggested that a Hopf-like link many arise in Co_2MnGa, but did not theoretically predict the link diagram, calculate the associated linking numbers or predict that the Co_2MnGa link is geometrically essential." Actually, Ref. [21] gives the link structure of the whole nodal lines. See Fig. 3 in Ref. [21]. The yellow lines in Fig. 3d are the whole structure of the link of the nodal loops mentioned in this manuscript.

Authors: We thank the referee for the remark and for carefully reading our first round reply. Ref. [21] clearly claims that Co_2MnGa hosts a Hopf link. For example, the title of Fig.3 in Ref. [21] reads "Hopf links and nodal chains in Co_2MnGa". A Hopf link by definition is

associated with linking number $\upsilon = 1$. However, through our careful experimental study of Co_2MnGa, we have observed that in fact the linked Weyl loops all exhibit $\upsilon = 2$. Moreover, our experimental results have allowed us to draw the link diagram for the Co_2MnGa link (Fig. 4e), which was not considered in Ref. [21]. Lastly, our experimental results clearly indicate that the link is geometrically essential, wrapping around the bulk Brillouin zone three-torus, which was not previously discussed in Ref. [21] or any other work. Most importantly, we have not only considered the theory, but demonstrated this exotic link experimentally forthe first time.

REPORT OF REFEREE #3

Referee #3, Q1: I thank the authors for their careful rebuttal. I think that the inclusion of the raw data is extremely useful to better visualize the spectroscopic features under discussion.

Authors: We sincerely thank the referee for carefully considering our revised manuscript and we are happy to hear that the referee finds the raw data useful.

Referee #3, Q2: I still have doubts about the data interpretation, and moreover I feel like few of the questions that I have raised in my previous report are still waiting for clarification. I cannot recommend the publication of the article in the present form.

1) The central point that requires further explanation is the actual form of the Fermi surface. The authors claimed that "our analysis suggests that the contours in Fig. 2d-f are three distinct and separate contours, rather than three slices of a single large three-dimensional contour". The central point of the manuscript is the unique way those lines are linked, something that is pictorially illustrated in several Figures but in my opinion still needs tobe experimentally confirmed. Now that raw data are available to the readers, by putting together the information from Extended data Fig. 10 and 9, I have produced a simple sketch that shows what the authors have indeed measured, or at least what I have understood.

In panel a, I have added a white line that corresponds to the plane investigated as a function of k_z. The dashed line forming the closed Weyl loop is highlighted by a green square. It lies completely on one side of the white line (for k_y < 1.09Å), it does not cross the line/plane. Similarly if we consider panel b, the Weyl loop lies at k_z larger than the plane correspondingto 544 eV photon energy. If I have understood well, by combining this information in the same 3D plot (panel c), the green rectangles they do not cross each other. I do not see any linking between them. The same argument is valid for the orange plane. This is, in my opinion, what is contained in the experimental data: the description given in Figure 3 ofthe main manuscript does not correspond to what has been experimentally measured.

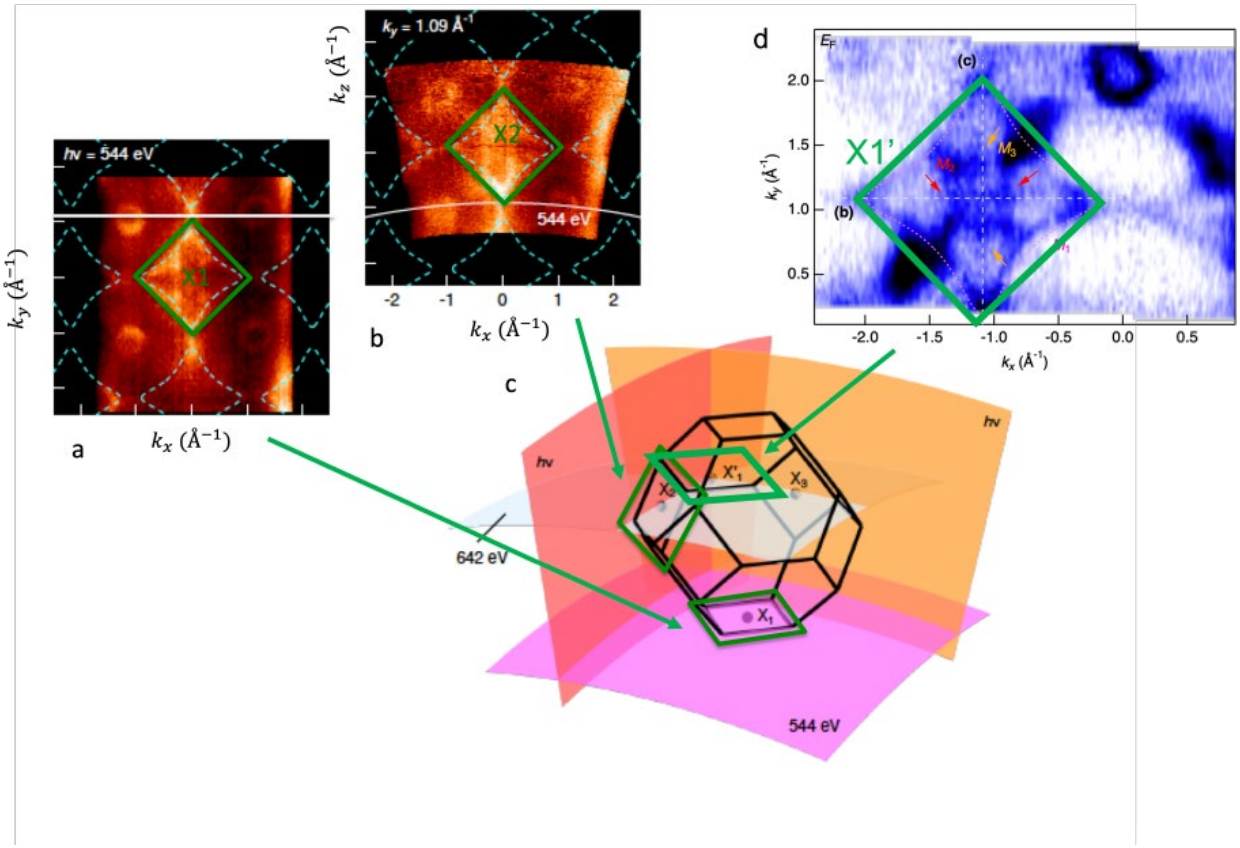

FIG. R2: **Measured Weyl loops. a**, Measurement of the X_1 Weyl loop and **d**, an equivalent X_1' Weyl loop in another Brillouin zone. **b**, Measurement of the X_2 Weyl loop. **c**, These three measured loops in an extended zone scheme. The X_1' and X_2 loops are directly observed to link, see also Extended Data Figs. 5b,c.

Authors: We thank the referee for the technical comment and associated sketch of our presented Fermi surfaces. In our analysis, we have shifted the momentum axes by reciprocal lattice vectors in order to present the datasets in the vicinity of the first Brillouin zone, as indicated by the axis scaling in main text Fig. 3. Of course, it is also possible to observe the linking in the full extended zone scheme, for example using our additional Fermi surfaceat 642 eV presented in Extended Data Fig. 5. Including this contour in the referee's figure, we can conclude the linked node loops directly in the extended scheme (Fig. R2). The analysis in Fig. 3 and Extended Data Fig. 5 unambiguously demonstrates linked node loops in Co_2MnGa. To address this point, in our revised manuscript we have noted in the Fig. 3 caption, "Momentum axes shifted by reciprocal lattice vectors to present the datasets in

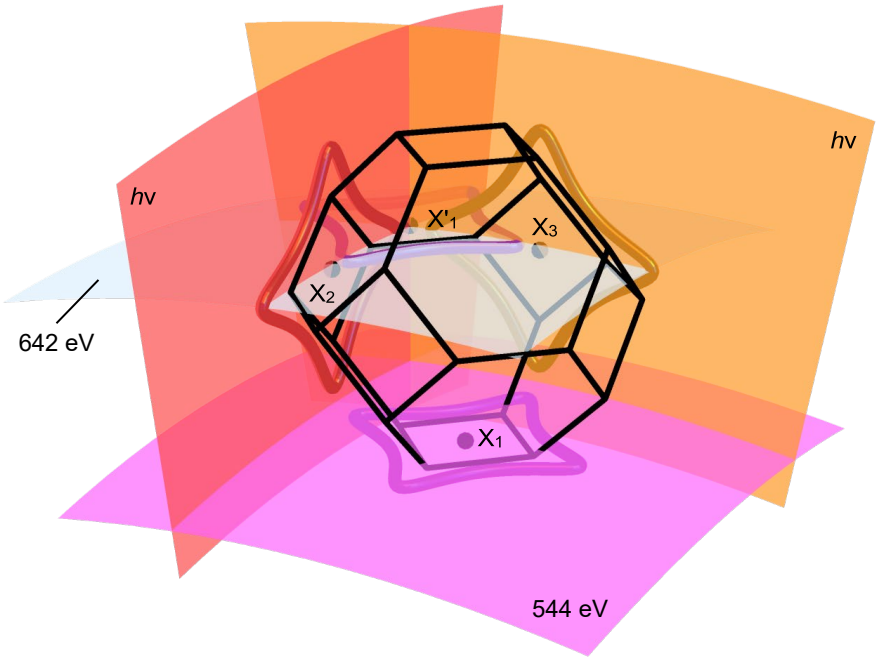

FIG. R3: **Measured Fermi surfaces in an extended zone scheme.** The Brillouin zone corresponds to $\Gamma_{(066)}$ in the primitive reciprocal basis.

the vicinity of the first bulk Brillouin zone." We have further plotted the Weyl loops in the extended zone scheme in a revised Extended Data Fig. 10 (Fig. R3).

Referee #3, Q3: 2) In general, the possibility to form complex link-loops is granted (ifI understood carefully the DFT calculations shown in the Extended Data Fig. 9) by thefact that the electronic properties are formed by highly elongated Fermi sheets, in contrastto the common wisdom of a bulk 3D Fermi surface. This is indeed extremely interesting, and it would be highly appreciated to explain it from a microscopic point of view (in terms of orbitals or Wannier functions). While the origin of such a feature might be matter for a future theoretical investigation, I think it is mandatory to confirm this from an experimental point of view. This is, in fact, the solid foundation for confirming the nature of the link-Weyl

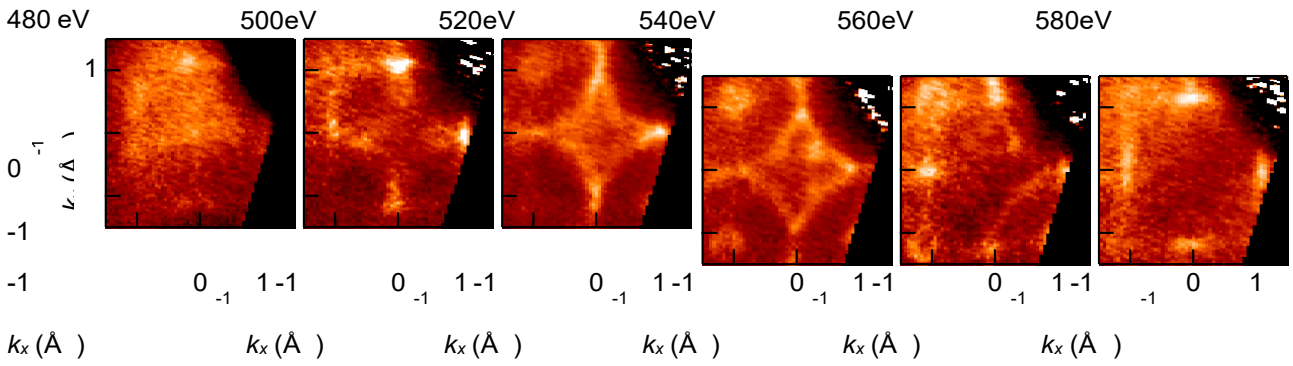

FIG. R4: **Evolution of the X_1 Weyl loop in k_z.** Fermi surfaces acquired by SX-ARPES at several photon energies. The Weyl loop contour vanishes as k_z shifts away from X_1, at ~ 540 eV.

loops. This can easily be done by exploring the evolution of the Fermi surface in the k_x-k_y planes in the region across the bulk X_1 point, for example for few selected photon energies between 520 and 570 eV. According to Extended Data Fig. 9, the Weyl loops around $X_1$is restricted to a small range of k_z, and it would be interesting to understand its evolution. This might help the authors to further support the presence of a link structure.

Authors: We thank the referee for the comment and we are thrilled that the referee finds that our observed exotic electronic structure "is indeed extremely interesting" and may deserve "future theoretical investigation". Following the referee's suggestion we have acquired additional SX-ARPES data and indeed found that the Weyl loop around X_1 is restricted to a small range of k_z (Fig. R4). The clearest loop-shaped contour is observedat ~ 540 eV, close to X_1 as pinpointed in our photon energy dependences. Moving away from X_1, we observe some perturbation of the Weyl loop contour, for example at 520 eV. However, the loop rapidly vanishes as we move further away in k_z. Beyond ±40 eV we find that the loop contributes little spectral weight. This sets a bound of $\delta k_z \sim \pm 0.25$Å^{-1} on the k_z extent of the X_1 loop. Given the convolution with the intrinsic k_z broadening of the SX-ARPES measurement, we can estimate the anisotropy of the Weyl loop Fermi sheets to be $a \sim \delta k_{||}/\delta k_z \sim 0.8$Å$^{-1}/0.2$Å$^{-1} \sim 4$. The highly anisotropic character of the Fermi sheet further supports the presence of Weyl loops confined to the momentum-space mirror planes, which together form an exotic linked Weyl loop structure.

Referee #3, Q4: 3) Moreover, although the authors draw lines and cones in the dispersion of the data, for example in Extended Data Fig. 5, I cannot see those features, I always see one of the states traced by the colored lines.

Authors: We thank the referee for the remark and we apologize that we did not discuss this technical point in our earlier revision. In Extended Data Fig. 5, the photoemission scattering plane coincides with the crystallographic mirror plane of the sample, so that we observe appreciable photoemission cross-section for states of only one mirror eigenvalue. Since the Weyl loop is a protected crossing of bands with opposite mirror eigenvalues, itis expected that **one branch from each cone is suppressed in a mirror-symmetric measurement geometry**. In fact, this effect provides additional, independent evidence that the Weyl loop is a mirror-symmetry-protected band crossing. We note that spectra ac- quired away from a mirror-symmetric measurement geometry readily exhibit both branches,as we have shown in Figs. 2a, 4a as well as Extended Data Figs. 7. We have revisedthe Extended Data Fig. 5 caption accordingly, writing "For both cuts, exactly one branchof each Weyl cone exhibits appreciable photoemission cross-section, as expected from the mirror-symmetric measurement geometry [53]." Here Ref. [53] refers to Fig. 5 (pg. 34-35) of [*Sync. Rad. News* **27**, 31-40 (2014)], which discusses in the context of SX-ARPES this well-known photoemission geometry effect.

Referee #3, Q5: 4) In general, I think that it is difficult, if not impossible, for a general audience to understand the physics contained in the data. Although the authors share now more information about the data, and the data analysis, I have always the feeling thatthe data presentation is excessively complicated. I invite the authors to add important information to their data, to guide the reader in the correct interpretation. Whenever itis possible, it would be useful to indicate the projections of the different Brillouin zones (distinguishing their order with different colors).

Authors: We thank the referee for the remark and we apologize that we have not sufficiently guided the reader in parsing our data. The linked Weyl loop structure is indeed counter-intuitive and we have made every effort to introduce this exotic object in a simple step-by-

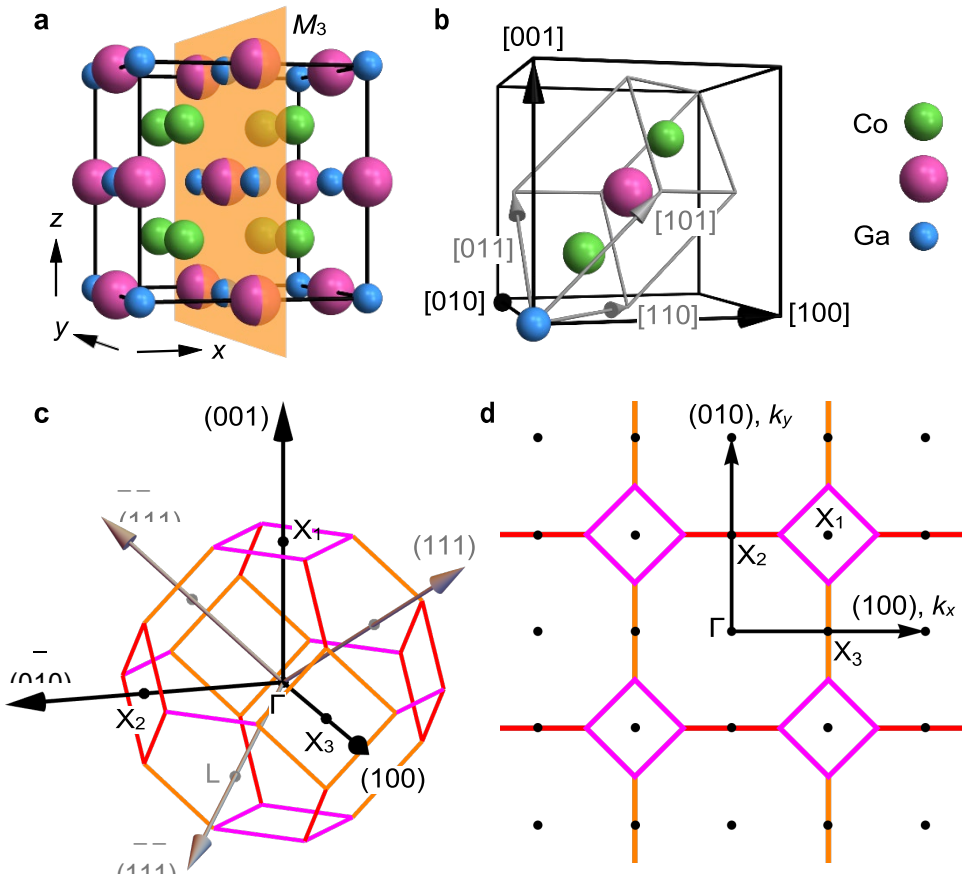

FIG. R5: **Crystal structure and Brillouin zone of Co_2MnGa. a**, Conventional unit cell with representative crystallographic mirror plane *M* (orange). **b**, The primitive unit cell (grey) includes one formula unit. **c**, Brillouin zone, with conventional reciprocal lattice basis vectors (black). Brillouin zone edges color-coded to correspond to the mirror planes: magenta M_1 plane, (001); red M_2 plane, (010), orange M_3 plane, (100). **d**, Slice through Γ in an extended zone scheme.

step way. In our revised manuscript, we note "To simplify the presentation of our results, we first discuss the Weyl loops one at a time (Figs. 1, 2); then consider their linking two ata time (Fig. 3); and finally consider the full three-component link (Fig. 4)." We emphasize that in discussing the link, we work in bulk momentum space with the three-dimensional Brillouin zone, which is not projected to two dimensions. In our revised manuscript, we have upgraded our Extended Data Fig. 2 (Fig. R5) to a color-coded bulk Brillouin zone matched with the three mirror plane colors. This color-coding should further help the reader

see where the three Weyl loops live in the bulk Brillouin zone.

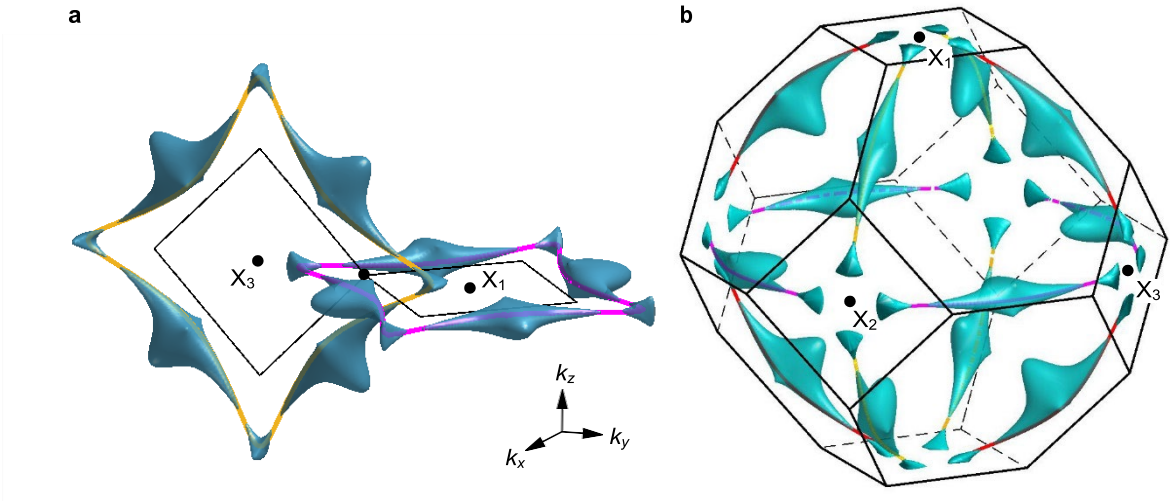

FIG. R6: **Linked Weyl loop Fermi surface.** Constant-energy slice of the pockets (navy) making up the linked Weyl loops obtained by *ab initio* calculation, at binding energy $E_B = -10$ meV below the experimental Fermi level. Plotted **a**, in an extended zone scheme (only two loops shown for simplicity) and **b**, the reduced Brillouin zone (all three loops shown). The Fermi surface pockets touch at a set of discrete points, where the Weyl loop disperses through this particular E_B. For reference, the full Weyl loop trajectories are indicated, collapsed in energy (magenta around X_1, red around X_2, orange around X_3). The Weyl loop Fermi surface pockets form a linked structure.

Referee #3, Q6: 5) I find the calculated Fermi surface shown in Extended Data Fig. 9 very simplified, it would be nice to have something like what is commonly found in other publications on the same subject, see for example the Fermi surface IrF_4 in Fig. 3d of the seminal work of Bzdušek *et al.* (*Nature* **538**, 75 (2016)). In think that such a representation of the 3D Fermi surface within the reduced Brillouin zone is fundamental to describe the link structure. Additionally, a careful description of the calculated band structure and Fermi surface is also important to draw a comparison with calculations already present inthe literature, as for example the data shown in Fig. 2e in the pioneering work of Belopolski*et al.* on the same material (*Science* **365**, 1278 (2019)).

Authors: We thank the referee for the useful suggestion. In our revised Extended DataFig. 9 (Fig. R6) we have included the linked *ab initio* Fermi surface plotted in the reduced Brillouin zone. Each loop is clearly observed to wrap around the Brillouin zone boundary, passing out and in through the square faces. This Fermi surface further illustrates that theWeyl loops form a geometrically essential link on the three-torus. Our calculations are fully consistent with the earlier works. For example, the large 'boomerang-like' Fermi surface pockets making up the loops clearly match in their overall shape with the feature (c) visiblein the *ab initio* calculation of Fig. 2E in [*Science* **365**, 1278 (2019)].

Referee #3, Q7: 6) Concerning the new part, I have read with interest the attempt of the authors to introduce new theoretical elements from knot theory in the electronic propertiesof this interesting material.

Authors: We are thrilled that the referee finds our new knot theory and Seifert surface results interesting.

Referee #3, Q8: However, as for the link-structure, I have the impression that the data are insufficient to fully support the claim.

6.1) First of all, the authors compare two completely different sets of data: SX-ARPES(more bulk sensitive) on the (001) termination, and VUV (more surface sensitive) on the (111) termination. This makes it impossible for the readers to compare the results in order to isolate the surface from the bulk states.

While the calculated band structure of Fig. 5d contains many bands, the ARPES dataof Fig. 5e shows one parabolic band. This might correspond to the surface state, or not, especially because they occupy different position in the momentum space (the measured band is shifted of more than 0.1 $\mathring{A}^{-1}$ towards a higher k value).

In the comparison between surface and bulk states, it is important to notice that in the literature the Weyl lines have been observed with VUV-ARPES at 50 eV (*Science* **365**,

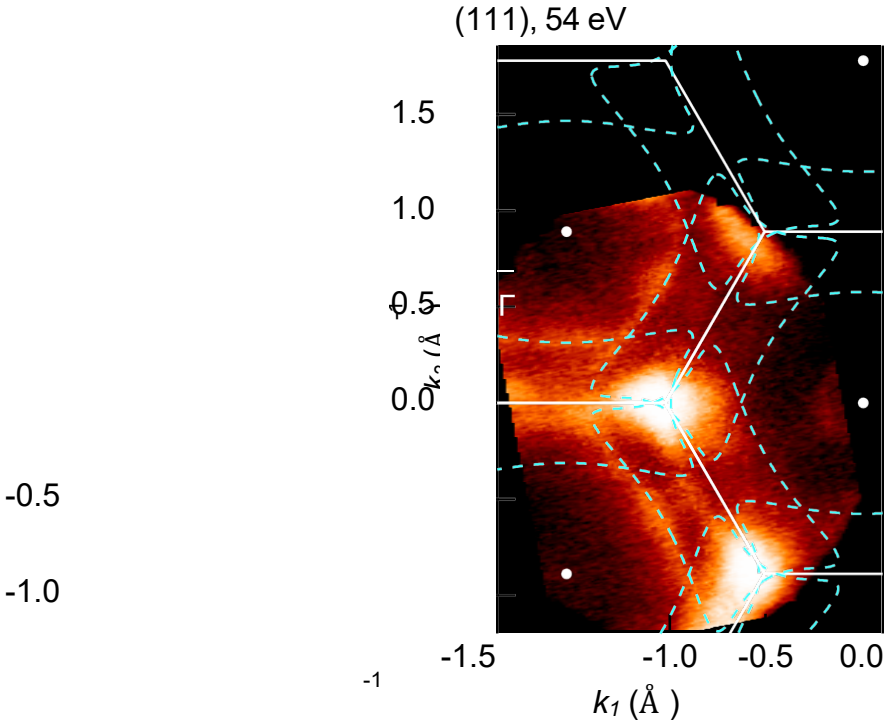

FIG. R7: **Linked Weyl loops in VUV-ARPES.** Fermi surfaces acquired at $h\nu = 54$ eV, with the Weyl loop trajectory extracted from SX-ARPES superimposed (cyan dashed lines). TheWeyl loop observed by SX-ARPES on (001) matches well that observed by VUV-ARPES on (111), allowing us to pinpoint the bulk states in our VUV spectra. The large overlap of the Weyl loops again demonstrates a bulk linked loop structure.

1278 (2019)). We can conclude that the use of 63 eV is not sufficient to suppress completely the bulk spectral features in favor of the surface state. I invite the authors to make a more careful comparison between theory and experiments at different photon energies in the VUV and soft X-ray in order to fully elucidate the surface and bulk dispersion in the (111) surface termination.

Authors: We thank the referee for the remark. To more carefully compare our SX- and VUV-ARPES results, we consider our Weyl loop trajectory obtained from our systematic SX-ARPES measurements (main text, Fig. 2c) and project it on (111). We then super- impose the projected Weyl loop on a (111) Fermi surface acquired by VUV-ARPES, at $h\nu = 54$ eV (Fig. R7). We observe clear signatures of large Weyl loops, which exhibit

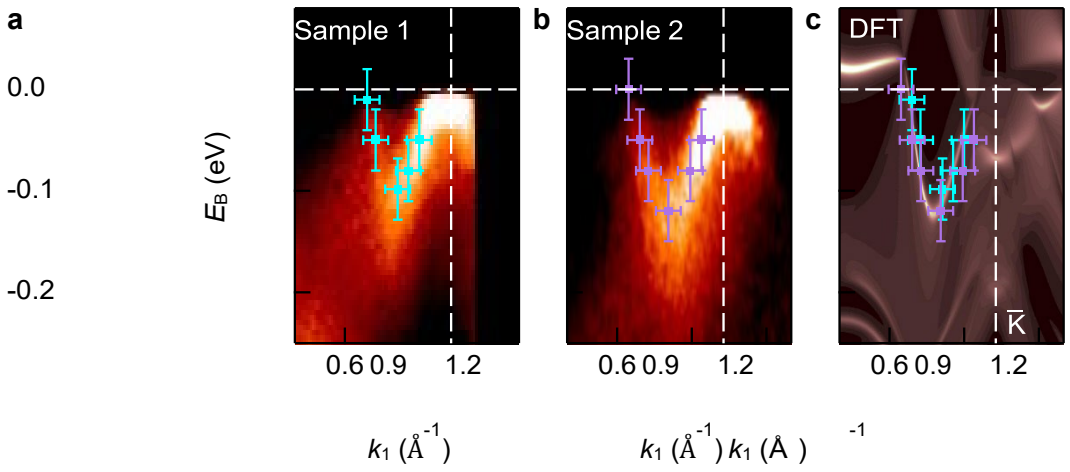

FIG. R8: **Seifert boundary mode. a**, **b**, ARPES energy-momentum cut through the Seifert boundary modes for two sample cleaves, acquired at $h\nu$ = 63 eV, with momentum axis fixed by alignment to the Weyl loops (see Fig. R7). **c**, Analogous *ab initio* calculation, with ARPES dispersion superimposed (cyan and purple markers). The ARPES and *ab initio* results show good agreement, providing evidence for Seifert boundary modes on the (111) surface of Co_2MnGa.

excellent quantitative agreement with the projected Weyl loop trajectory. Moreover, we clearly observe that the Weyl loop contours exhibit large overlap on our (111) Fermi surface. Given the oblique orientation of (111) relative to the (100), (010) and (001) mirror planes hosting the Weyl loops, the observation of **clear overlapping Weyl loops on** (111) **provides an independent demonstration of a linked loop quantum state**.

The clear identification of the bulk Weyl loops in our VUV-ARPES spectra further allows us to disentangle bulk and surface states in the low-photon-energy regime. In particular, based on our bulk *ab initio* calculations, we expect no irrelevant bulk states near the Weyl loop linking point. At the same time, our surface state calculations exhibit clear arc-like Seifert boundary modes stretching across linked loops in this region of the surface Brillouin zone. We observe a corresponding arc-like state in VUV-ARPES, connecting the Weyl loops near the linking point (main text Fig. 5f). In this way, our SX-ARPES Weyl loop trajectory, VUV-ARPES Fermi surface and *ab initio* results to- gether suggest the observation of a Seifert boundary mode on the (111) surface of Co_2MnGa.

The observed shift of the Seifert boundary mode between VUV-ARPES and *ab initio* calculation may be due to a small uncertainty in the determination of $\bar{\Gamma}$ in our photoemission spectra. As a point of comparison, this error falls within our estimated error barof $\delta k = \pm 0.1 \text{Å}^{-1}$ noted in main text Fig. 2c. To further address the referee's remark, we have recalibrated $\bar{\Gamma}$ in our VUV-ARPES Fermi surfaces using the SX-ARPES Weyl loop trajectory (Fig. R7). The recalibrated $\bar{\Gamma}$ produces close **quantitative agreement in the Seifert boundary mode dispersion between experiment and theory** (Fig. R8a, c).

Lastly, we **reproduced the Seifert boundary mode** with additional VUV-ARPES measurements on a second Co_2MnGa sample. We **again observed quantitative agreement** in the dispersions between the VUV-ARPES and surface *ab initio* calculations (Fig. R8). Our new VUV-ARPES measurements on a second sample; quantitative comparison of the Weyl loop between SX-ARPES and VUV-ARPES; and the quantitative agreement with surface *ab initio* calculations all suggest the observation of Seifert boundary modes arisingfrom the linked loop quantum state.

Referee #3, Q9: 6.2) The topological nature of the surface state has not been discussed. This surface state might be trivial. It nicely follows the dispersion of the bulk bands, andit might be some kind of surface resonance. The authors should discuss, at least on a theoretical basis, how breaking the mirror symmetries and the Weyl lines is affecting the surface state dispersion.

Authors: We thank the referee for the remark and we apologize that we did not explore this question more thoroughly in our previous revision. The observed surface state isindeed topological and is protected by a quantized winding number. We can theoretically demonstrate that it is topological by examining its behavior under modified surface potential in *ab initio* calculation (Fig. R9). Under such a perturbation, we find that the surface state remains robustly pinned to the linked Weyl loops, showing that it is a Seifert boundary mode and not a topologically-trivial surface resonance.

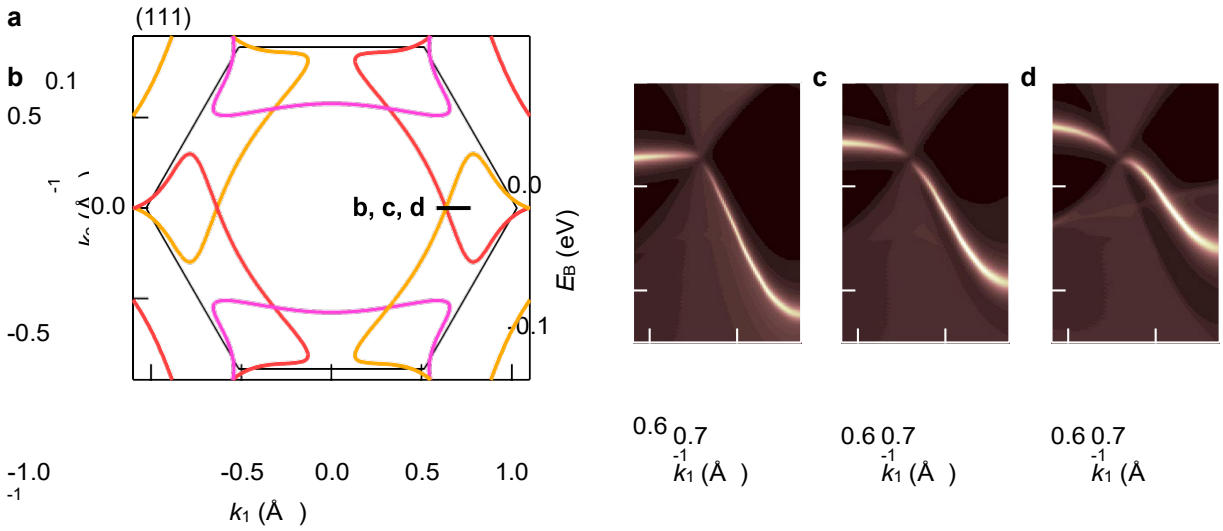

FIG. R9: **Robust pinning of Seifert boundary modes. b**, Zoomed-in *ab initio* calculation of the surface states through the Weyl loop 'linking point' on (111), path indicated in **a**. **b**, **c**, Same as (a), but with modified surface chemical potential. The Seifert boundary modes continue to be pinned to the touching point, again demonstrating their topological nature.

If mirror symmetry were to be broken in the system, the Weyl loops would gap out. If a gap opened everywhere along the Weyl loop trajectory, the system would enter a triv-ial phase. Alternatively, the loop may leave behind some discrete number of gapless Weyl points, producing a Weyl point semimetal. The Seifert boundary modes would then evolve either into trivial surface states or topological Fermi arcs. We kindly suggest leaving fur- ther theoretical analysis to future work. The rich evolution of the bulk linked loops and Seifert boundary modes under changes in symmetry will likely soon emerge as one of manyfascinating directions of research on linked loop quantum states.

Reviewer Reports on the Second Revision:

Referee #2 (Remarks to the Author):

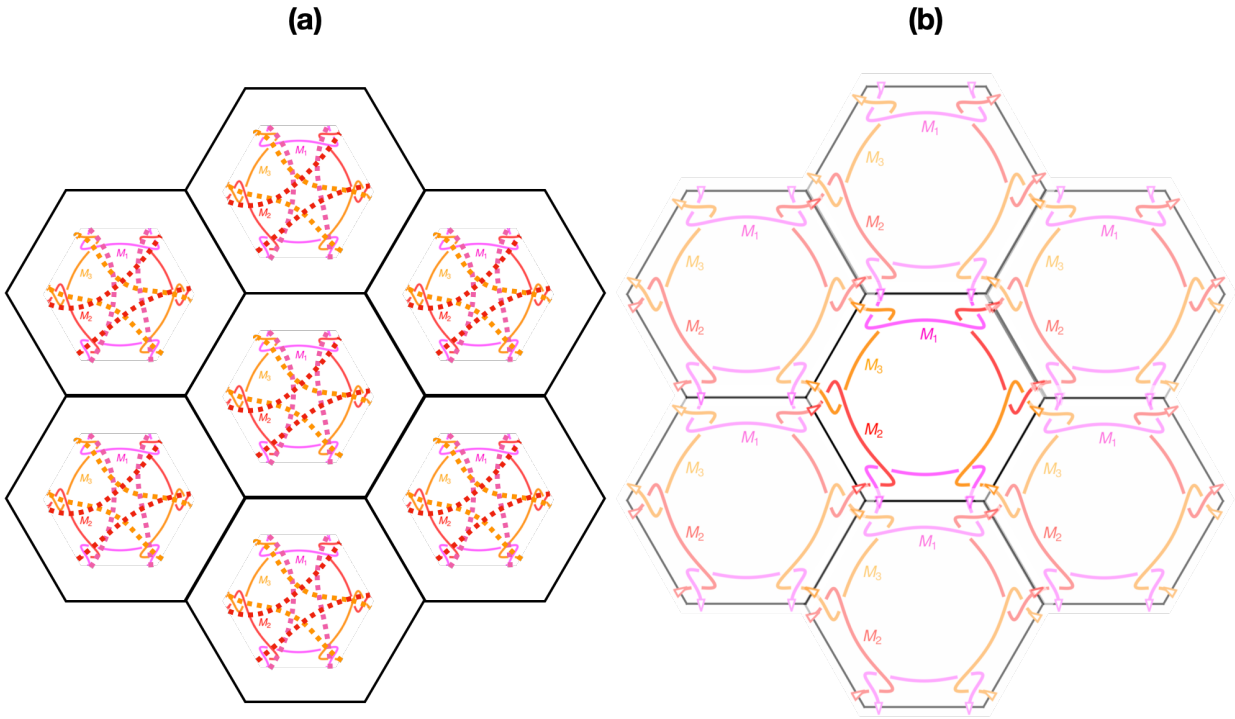

Fig. RR1, Sketch of two different knot structures in the extended BZ. These two knot structures are topologically distiguishable.One can't be smoothly transformed into the other one.

First of all, thanks the detailed replies to my questions. Fig R1 is quite useful to understand more details about the knot structure in the BZ. However, their interpretation of the linkingnumber (2,2,2) is still wrong.

"It is important to consider all loops in a single Brillouin zone (a reduced zone scheme). Considering fewer loops by ignoring physically-distinct link components, as is done in Ref.[20], does not correctly characterize the linking number. On the other hand, considering the whole of momentum space (an extended zone scheme) introduces redundant copies ofthe electronic states and is also incorrect."

If we don't consider the whole of momentum space, then, we can't distinguish two knot

structures that I made above. The knot structure in Fig. RR1(a) has linking number (2,2,2) just the same as what is proposed in this manuscript. However, this is topological different from theone shown in Fig.RR1(a) which is the nodal link case in this manuscript.

In order to characterize the linking number in the periodic systems, we should introduce othertopological numbers. Such a topological number is detailed discussed in this paper "The linkingnumber in systems with Periodic Boundary Conditions , E. Panagiotou, Journal of Computational Physics 300, 533 (2015) https://doi.org/10.1016/j.jcp.2015.07.058".

I hope the authors would carefully consider my suggestions.

Referee #3 (Remarks to the Author):

I thank the Authors for the extra experimental efforts done in order to answer my technical questions. The results shown in Figure R4 confirm the unique nature of the 3D Fermi surface of Co2MnGa that makes possible the observed link structure. This, together with the DFT calculations shown in the extended data Figure9, greatly helps the reader in understanding such a complex geometrical shape, and the linking number.

At the same time, the data of Figure R7 are useful to distinguish the spectral signature of the bulk states. Hence, I would encourage the Authors to include the results of Fig.R4 and Fig.R7 as additional extended data, since they provide valuable information. The author should also add, for completeness, a sketch of the 3D BZ indicating (similarly to what done in the extended data Figure 6) the momentum region accessed by the VUV data acquired with 63 eV and 54 eV photon energies. In this respect, the Authors should also state somewhere the value of the inner potential used to retrieve the kz values.

I believe that this manuscript constitutes a leap in the field of topological materials. The solid experimental evidences contained in this work open opportunities to further inspect the interplay between the electronic properties of solids and knot theory, something that was so far restricted to theory. I believe that in their reply to Referee 2, the Authors have substantiated in details the impact of their work.
Although few aspects, such as the origin and the topological nature of the surface state are still open points, those will certainly fuel an intense research activity in the field.

Author Rebuttals to Second Revision:

Re: *Nature* 2020-10-18080, Observation of a linked loop quantum state in a topological magnet

We thank the referees for taking the time to review our revised manuscript in a third round of review. In the following we respond to the remaining comments of the referees, point by point.

Report of Referee #2

Referee #2, Q1: First of all, thanks the detailed replies to my questions. Fig. R1 is quite useful to understand more details about the knot structure in the BZ. However, their interpretation of the linking number (2,2,2) is still wrong.

> "It is important to consider all loops in a single Brillouin zone (a reduced zone scheme). Considering fewer loops by ignoring physically-distinct link components, as is done in Ref. [20], does not correctly characterize the linking number. On the other hand, considering the whole of momentum space (an extended zone scheme) introduces redundant copies of the electronic states and is also incorrect."

If we don't consider the whole of momentum space, then, we can't distinguish two knot structures that I made above. The knot structure in Fig. RR1(a) has linking number (2,2,2) just the same as what is proposed in this manuscript. However, this is topological different from the one shown in Fig.RR1(a) which is the nodal link case in this manuscript.

Authors: We thank the referee for carefully reading our reply and for the additional comments. We maintain that examining a single Brillouin zone is necessary and sufficient to characterize the link. Intuitively, looking at the referee's sketch (Fig. RR1), it is clear that adding several copies of the Brillouin zone adds no information to the problem. Considering an extended zone scheme (the whole of momentum space) simply amounts to re-copying the same diagram multiple times.

More precisely, we can always distinguish two knot structures from a single Brillouin zone, because every link on the three-torus T^3 (a single Brillouin zone) corresponds to a unique periodic link in three-space R^3 (an extended zone scheme) [Ch. 1, Hatcher, *Algebraic Topology* (2001)]. To distinguish the link sketched by the referee (Fig. RR1a) from the quantum link we observe in Co_2MnGa (Fig. RR1b), note that the link in Fig. RR1a can be smoothly contracted to a small ball in a localized region of momentum space and is therefore not geometrically essential. On the other hand, the link in Fig. RR1b wraps around the Brillouin zone, cannot be smoothly deformed to a localized region, and is geometrically essential. This property distinguishes the two links and can be determined by examining the link structure in a single Brillouin zone.

Furthermore, any given mathematical invariant, such as the linking number, is typically shared by many topologically distinct knots or links [Ch. 1, Lickorish, *An Introduction to Knot Theory* (1997)]. We do not claim that the linking number (2,2,2) uniquely specifies the quantum link. Rather, we simply claim that the observed quantum link exhibits linking number (2,2,2). The link diagram (main text Fig. 4e) uniquely specifies the Co_2MnGa quantum link.

Following the referee's remark, we have emphasized in the revised manuscript that, "Since the Weyl loop link lives in the periodic momentum space of the crystal, it is necessary and sufficient to consider all link components in a single Brillouin zone." We have also noted that our "link diagram uniquely specifies the Co_2MnGa quantum link."

Referee #2, Q2: In order to characterize the linking number in the periodic systems, we should introduce other topological numbers. Such a topological number is detailed discussed in this paper [REDACTED]

I hope the authors would carefully consider my suggestions.

Authors: We thank the referee for the remark and the reference. The reference is from a very different field of research but is consistent with our analysis on the overlapping points. The reference does not mention condensed matter physics and is primarily concerned with "open chains" (curves which are not closed loops) in networks of polymers or "polyethylene melts". By contrast, in our manuscript we examine links of closed Weyl loops. At the same time, the linking number considered in the reference is the usual linking number (see Definition 2 or Section 4 of that work) and the reference does not propose any new or different topological numbers which could be relevant to our study. Regarding the usual linking number, the reference is consistent with our analysis of the Co_2MnGa quantum link.

Report of Referee #3

Referee #3, Q1: I thank the Authors for the extra experimental efforts done in order to answer my technical questions. The results shown in Fig. R4 confirm the unique nature of the 3D Fermi surface of Co_2MnGa that makes possible the observed link structure. This, together with the DFT calculations shown in the Extended Data Fig. 9, greatly helps the reader in understanding such a complex geometrical shape, and the linking number.

At the same time, the data of Fig. R7 are useful to distinguish the spectral signature of the bulk states. Hence, I would encourage the Authors to include the results of Fig. R4 and Fig. R7 as additional Extended Data, since they provide valuable information. The author should also add, for completeness, a sketch of the 3D BZ indicating (similarly to what done in the Extended Data Fig. 6) the momentum region accessed by the VUV data acquired with 63 eV and 54 eV photon energies. In this respect, the Authors should also state somewhere the value of the inner potential used to retrieve the k_z values.

Authors: We thank the referee for carefully examining our reply and revised manuscript. We have included Figs. R4 and R7 in the Supplementary Information as Figs. S1 and S2. Following the referee's suggestion, we have also added a Supplementary Information Fig. S4 indicating the momentum space slices captured by our VUV ARPES measurements (shown here as Fig. R1). We have also indicated the inner potential in the Methods, writing "The inner potential for converting $h\nu$ to k_z was determined to be $V_0 = 22$ eV".

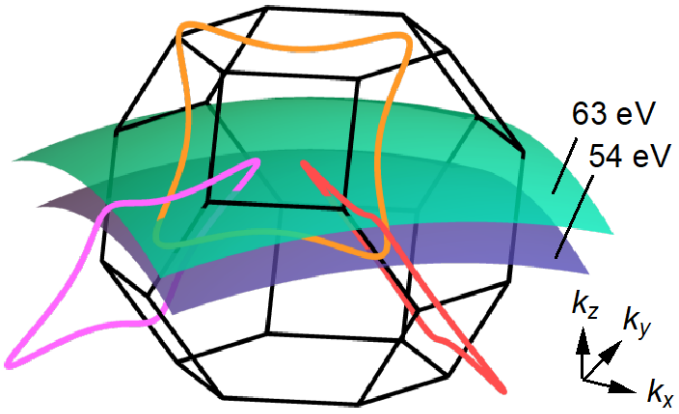

Figure R1. Fermi surfaces measured at VUV photon energies, (111) cleaving plane.

I believe that this manuscript constitutes a leap in the field of topological materials. The solid experimental evidence contained in this work open opportunities to further inspect the interplay between the electronic properties of solids and knot theory, something that was so far restricted to theory. I believe that in their reply to Referee 2, the Authors have substantiated in detail the impact of their work. Although few aspects, such as the origin and the topological nature of the surface state are still open points, those will certainly fuel an intense research activity in the field.

Authors: We thank the referee for enthusiastically supporting publication of our work.