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Peer Review File

Manuscript Title:

Experimental observation of non-Abelian topological charges and edge states

Editorial Notes:

Redactions – Mention of other journals

This document only contains reviewer comments, rebuttal and decision letters for versions considered at *Nature*. Mentions of the other journal have been redacted.

Reviewer Comments & Author Rebuttals

Reviewer Reports on the Initial Version:

Ref #1

In this work, Guo et al. investigate the recently proposed non-Abelian band topology that arises in one-dimensional systems with PT (parity and time-reversal) symmetry in the presence of multiple band gaps. In particular, they use transmission-line networks to emulate 1D systems characterized by the various elements of the underlying quaternion group. These 1D systems are then used by the authors to experimentally confirm a previously predicted bulk-boundary correspondence, and to establish a new bulk-domain wall correspondence for these one-dimensional topological bands.

Let me first put this work into a wider perspective. The non-Abelian band topology considered here was recently predicted to characterize gapped PT-symmetric systems in one dimension, and to capture momentum-space exchange of band nodes in two-dimensional PT-symmetric systems. The interplay of the non-Abelian topology with crystalline symmetry has led to fresh insights into the stability of nodal chains, braided Dirac points, and into nodal links; yet simple and tunable systems to investigate the non-Abelian topological charges are at present missing. From this perspective, the results and the experimental setting presented by Guo et al. are very timely, and certainly deserve publication. On the other hand, however, I would like to point out that there is nothing intrinsically “quantum” about this topology (as the authors implicitly admit when referring to line defects in nematic crystals), therefore I find the references by the authors to “quantum computation” misleading, and suggest to drop them. There is a major conceptual difference between non-Abelian topological order required for quantum computing and the non-Abelian band topology discussed here.

When considering the suitability of this work for a high-profile journal, one should regard the novelty brought by this work, and the impact it would have on the field. From this perspective, it appears to me that this work does not put forward a new theory [the theoretical discussion until Eq. (3) is mostly a review of Ref. 19, while the bulk-edge correspondence follows Supplementary Material of the same reference], nor a novel experimental technique [transmission-line networks have become a common tool of choice for tabletop emulation of band structures, and the introduction of complex hoppings through the internal structure of the “meta-atoms” has been also previously established e.g. by Ref. 30], nor a striking experimental result [the correspondence between theoretical spectrum of a (non-interacting) matrix Hamiltonian and its emulation in artificial systems is generally good in this class of experiments]. Therefore, I am of the opinion that this work does not constitute a paradigm shift and the very high standards expected by journal Nature.

On the other hand, as the work provides the (to my knowledge) very first experiment detection of the non-Abelian band topology of one-dimensional bands and of their bulk-edge correspondence, I can imagine it could possibly be suitable for one of the Nature sister journals (such as [REDACTED]), provided that the work is properly edited and supplemented (see concrete suggestions and questions listed below). What I see as a further value of this work is the illustrative way of presenting the 1D Hamiltonians using the eigenstate-frame sphere, and the investigation of bound states at domain walls, thus generalizing the previously introduced bulk-boundary correspondence for the non-Abelian topological charge. To meet these goals, the article would serve better if it were shortened to the “letter format”, with properly referencing and shortening the theoretical review (which presently constitutes the first half of the text), while moving to the main text certain details about the experiment.

In summary, I think the work (even after the appropriate revisions) does not meet the high standards of journal Nature. However, assuming the work is properly edited, it could be appropriate for a more topic-specific NPG journal.

Besides the general assessment summarized above, I list here several concrete comments and questions that the authors should consider before resubmitting their work.

1.) The authors write in the abstract, that they “propose the non-Abelian bulk-edge correspondence, where the edge states are found to be fully determined by non-Abelian topological charges”. However, this bulk-edge correspondence does not seem to encapsulate a particularly strong statement, namely, it is stated in the text that the edge state pertaining to charge j “can be anywhere between the 1st and 3rd band”, while the edge states emanating from charge -1 “are fickle”. Can the bulk-edge correspondence be transformed into a some stronger statement?

2.) Eq. (2) contains a general nearest-neighbor PT-symmetric Hamiltonian. The authors claim that by choosing the parameters properly, they can mimic all the flat-band cases in Fig.1b—e (do these correspond to Fig. S16 and S17 from Ref. 19?). However, a look at the Supplementary Material reveals that Fig.1(e) requires next-nearest-neighbor elements beyond the Hamiltonian in Eq. (2).

3.) Certain key specifications of the experimental setting are missing. Is a coaxial used as the transmission line? Furthermore, the authors claim below Eq. (2) that the constructed tight-binding model breaks both time-reversal and parity symmetry (while preserving the combined PT-symmetry). Could the authors specify where in their experimental setting is the time-reversal-breaking element? (Or do the authors mean that although the most general model described by Eq. (2) breaks P and T individually, the specific 1D models which they experimentally build preserve both symmetries?)

4.) Per the trick with the four internal nodes of each “meta-atom” (sublattice), there are in total 4x3 = 12 nodes per unit cell, which should result in 12 bands. However, only three bands that emulate the non-Abelian band topology with quaternion invariant are explicitly discussed and plotted. Could the authors comment/clarify what is the fate of the remaining (12-3) = 9 bands?

5.) Could the authors briefly comment how the bulk and edge spectra in Fig. 3 are measured? It appears to me, that to obtain the bulk spectrum through Fourier-transforming the measured voltages, a periodic boundary condition needs to be set up by closing the 1D system into a ring. In contrast, open boundary conditions should be present to detect the edge spectrum.

6.) It remains unclear to me after studying this manuscript to what extent are the topological phases “+i” and “-i” distinct from each other. Naively, I would expect that the overall sign cannot be meaningfully defined, since one can change the pi-rotation of the eigenstate-frame into a (-pi)-rotation by doing a discrete gauge transformation on the eigenstate-frame with an element of the

O(3) group. In other words, the action of the gauge group should split the elements of the quaternion group into conjugacy classes, with "+i" and "-i" corresponding to a single conjugacy class.

However, my conclusion seems be refuted by the data in Fig.S12(a), where a domain wall between phases "+i" and "-i" results in bound states reminiscent of the edge of phase -1 (=+i/-i). Could the authors clarify this issue? In fact, this is the only type of domain where the arising bound states cannot be explained using solely the quantized Berry phases in the individual band gaps. From this perspective, I would like to urge the authors to move this very interesting result from the Supplemental Material into Fig. 4 of the main text.

Ref #2

The non-Abelian quaternion topological charges in momentum space were firstly introduced in ref. 19 (Science 364, 1273, 2019) to characterize some novel line nodes. Some features of these non-Abelian quaternion topological charges have been demonstrated in the authors' recent work in ref. 22 (Phys. Rev. Lett. 125, 033901 2020), but those are indirect evidences. I believe the current work is indeed the first direct measurement on the non-Abelian quaternion topological charges, which certainly is very important. I have a few questions, which hopefully the authors can clarify.

1. In the case of a domain wall between +i and -i charges, if we follow Fig. 4c, then the first bandgap should have no edge state, and the second bandgap should have no edge state either (Zak phase of pi is equivalent to -pi). But according to the new bulk-edge correspondence proposed in the current work, the edge state should correspond to the case of charge -1, which does support edge states as shown in Fig. 2e. Could the authors clarify this issue?

2. In Fig. S12a, varying the probe position changes not only the edge state distribution, but also the number of edge states. Why is that?

Ref #3

In their manuscript "Experimental observation of non-Abelian topological charges and bulk-edge correspondence", Guo and collaborators investigate both theoretically and experimentally the emerging concept of non-Abelian topological charges in 1D reciprocal space, which was first kick started by Bzdusek and his colleagues last year. The authors present the mathematical analysis (homotopy group theory) as well as the first experimental observation of the quaternions in the 1D PT-symmetric transmission line network. In particular, the authors propose for the first time the non-Abelian bulk-edge correspondence, and provide the experimental evidences for their statement. All of these aspects are novel and very interesting, their experimental results are neat and timely, and the manuscript is well organized. I believe this work will be of significant interest to the community. Before recommending it for publication in Nature, the authors need to address the following comments and suggestions.

I find the mathematical model present to explain the essential idea of the paper is hard to read, I believe the readers who are not familiar with the language of homotopy group need more explanations from the authors. For example, how does the rotation matrix R(k) come? Why it has the form R(k)=exp((k+\pi)/2*Lx,y,z) for topological charges +i/+j/+k but has the different form R(k)=exp((k+\pi)*Lx,y,z) for charge -1? What's more, The discussion of isomorphism map between O(3) and S0(3) reads obscure, and the figure 1(f-i) do not provide me more insight than the figure 1(b-e) do, and I don't see the single curve is terminated at antipodal points in SO(3) sphere for charges +i/+j/+k. I suggest the authors either provide more physical intuition for these abstract concepts in the main text, and detail the according explanation in the supplementary materials if necessary, or give relevant references to their argument.

In the 3 bands model, the topological transition between different non-Abelian charges follows the order +i -> +j -> +k because of the bandgaps close and reopen. For Abelian case, the topological transition occurs between the trivial phase and topological phase. Does similar scenario occur for the non-Abelian case, for example, the quaternion charge transition from +i/+j+/k to +1? A broader question is, can the topological transition of quaternion charges occur in an arbitrary order?

The bulk quaternions are distinguished by their distinct behaviors on the so called "EigenS-Frame" sphere, which not only determine the number of edge states in the bandgap, but also decide the distribution of them over several bandgaps. To experimentally show the non-Abelian topological charges, the authors extract the eigenvectors from measured phases and fields and plot them on the "EigenS-Frame", and the experimental results and theoretical results match so well. Since eigenvectors are not gauge invariant, and therefore, in principle are not measurable quantities, how do the authors keep track of the right eigenstates (eigenstates might not be excited), and fix the gauge of the measured eigenvectors in the experiment?

Different from the edge states in 1D SSH model, the edge states for charge +j and -1 are not topologically stable in their energy since they are fickle and vary according to the details of the model. What about the edge states for charge +i/+k? Are they stable in their energy? In relation to this, are these edge states topologically robust again the disorder? The authors may include the study of this aspect either theoretically or experimentally.

The authors claim the non-Abelian bulk-edge correspondence is determined by \DeltaQ=Q_L/Q_R, although all edge states can be predicted from Zak phases of the respective bandgaps, or equivalently the phases \pi winding of the eigenstates. The authors provide some but not exclusive examples to show such relation indeed holds, but how can we assure this non-Abelian relation is generally true?

For the model implemented in the experiment, why the number of edge states is 4 for topological charge +j (Figure S4, (b))? I expect it is also 2 based on the flat band TBM in Figure 2(c).

The following questions are also very interesting and general, but they might not directly relate to the major points of this manuscript, therefore, the authors might or might not consider implementing them.

Are the quaternion charges unique for three bands model? Assume if an extra band is added into the system, will this band trivialize other bands like the case of fragile topology?

Since the non-Abelian charges are non-commutative, do they place the constraint on the admissible configurations of the 3 bands model?

Provided further studies addressing my comments are performed, the manuscript meets the criteria of Nature and therefore I recommend the acceptance.

Author Rebuttals to Initial Comments:

Referee #1 (Remarks to the Author):

Comment 1.1: In this work, Guo et al. investigate the recently proposed non-Abelian band topology that arises in one-dimensional systems with PT (parity and time-reversal) symmetry in the presence of multiple band gaps. In particular, they use transmission-line networks to emulate 1D systems characterized by the various elements of the underlying quaternion group. These 1D systems are then used by the authors to experimentally confirm a previously predicted bulk-boundary correspondence, and to establish a new bulk-domain wall correspondence for these one-dimensional topological bands.

Reply 1.1: We thank the referee for the careful reading and nice summary of our work.

Comment 1.2: Let me first put this work into a wider perspective. The non-Abelian band topology considered here was recently predicted to characterize gapped PT-symmetric systems in one dimension, and to capture momentum-space exchange of band nodes in two-dimensional PT-symmetric systems. The interplay of the non-Abelian topology with crystalline symmetry has led to fresh insights into the stability of nodal chains, braided Dirac points, and into nodal links; yet simple and tunable systems to investigate the non-Abelian topological charges are at present missing. From this perspective, the results and the experimental setting presented by Guo et al. are very timely, and certainly deserve publication. On the other hand, however, I would like to point out that there is nothing intrinsically "quantum" about this topology (as the authors implicitly admit when referring to line defects in nematic crystals), therefore I find the references by the authors to "quantum computation" misleading, and suggest to drop them. There is a major conceptual difference between non-Abelian topological order required for quantum computing and the non-Abelian band topology discussed here.

Reply 1.2: We thank the referee for his/her recognitions and positive assessments of our work.

We comply with the referee's suggestion and we have deleted the phrase "quantum computation" as well as the related reference in the revised text.

Comment 1.3: When considering the suitability of this work for a high-profile journal, one should regard the novelty brought by this work, and the impact it would have on the field. From this perspective, it appears to me that this work does not put forward a new theory [the theoretical discussion until Eq. (3) is mostly a review of Ref. 19, while the bulk-edge correspondence follows Supplementary Material of the same reference], nor a novel experimental technique [transmission-line networks have become a common tool of choice for tabletop emulation of band structures, and the introduction of complex hoppings through the internal structure of the "meta-atoms" has been also previously established e.g. by Ref. 30], nor a striking experimental result [the correspondence between theoretical spectrum of a (non-interacting) matrix Hamiltonian and its emulation in artificial systems is generally good in this class of experiments]. Therefore, I am of the opinion that this work does not constitute a paradigm shift and the very high standards expected by journal Nature. On the other hand, as the work provides the (to my knowledge) very first experiment detection of the non-Abelian band topology of one-dimensional bands and of their bulk-edge correspondence, I can imagine it could possibly be suitable for one of the Nature sister journals (such as [REDACTED]), provided that the work is properly edited and supplemented (see concrete suggestions and questions listed below). What I see as a further value of this work is the illustrative way of presenting the 1D Hamiltonians using the eigenstate-frame sphere, and the investigation of bound states at domain walls, thus generalizing the previously introduced bulk-boundary correspondence for the non-Abelian topological charge. To meet these goals, the article would serve better if it were shortened to the "letter format", with properly referencing and shortening the theoretical review (which presently constitutes the first half of the text), while moving to the main text certain details about the experiment. In summary, I think the work (even after the appropriate revisions) does not meet the high standards of journal Nature. However, assuming the work is properly edited, it could be appropriate for a more topic-specific NPG journal.

Reply 1.3: We thank the reviewer for considering that our work "provides the (to my knowledge) very first experiment detection of the non-Abelian band topology of one-dimensional bands". Furthermore, we also propose non-Abelian bulk-edge correspondence for the first time, and as the reviewer stated, our work directly generalizes "the previously introduced bulk-boundary correspondence for the non-Abelian topological charge". It has been well known that the celebrated Abelian bulk-edge correspondence serves as a corner stone for the field of topological physics. Therefore, we believe that the notion of non-Abelian bulk-edge correspondence will bring similar impact to the field.

We thank the referee for pointing out that "the article would serve better if it were shortened to the "letter format" to improve the presentation of our work. In the revised text (also to satisfy the length limit up to 2500 words), we have moved some parts of theoretical discussion into the supplementary materials and we have added certain details about the experiment in the main text.

1. We revised and moved the following paragraphs into the supplementary materials (Sec. I):

"It is worth noting that in general, the winding trajectories of eigenstates are not fixed on great circles. In contrast to which axis the states wind about, …These curves all thread a pair of antipodal points, so they are closed and not contractible, which is topologically guaranteed by $\pi_1(SO(3)) = \mathbb{Z}_2$.".

2. We moved Fig. 1f-i into the supplementary materials (Sec. I) as Fig. S1.
3. We revised and moved the following paragraph into the method section in supplementary materials (Sec. IX),

"In order to observe the non-Abelian topological charges experimentally, we implement the above tight binding model using transmission line networks with braiding connectivity. For a network connected by transmission lines, the wave function of each node satisfies the network equation[1,2]: … Thus, transmission line network offers an ideal platform to realize various tight binding models."

4. We supplemented the following details about the experiment (on page 6 in the revised main text).

"In the experiment, a network consisting of 13 periods was designed to characterize the non-Abelian topological charges corresponding to $+i$, $+j$, $+k$ and -1, respectively. There are three meta-atoms A, B and C in one unit-cell, and Fig. 3a is the photo of the network for the specific configuration of charge -1. The inset shows that there are four nodes represented by the cable connectors labelled 1, 2, 3, 4 in each meta-atom. As such, there are four allowed subspaces. Each subspace can be characterized by a pseudo angular momentum with $e^{i4\varphi_n} = 1\ (n = 1,2,3,4)$ [2], with $\varphi_1 = 0, \varphi_2 = \frac{\pi}{2}, \varphi_3 = \pi\ and\ \varphi_4 = -\frac{\pi}{2}$. In the experiment, we work with the subspace $\varphi_2 = \frac{\pi}{2}$ to realize our 3×3 real Hamiltonian (Eq. 2) and within this subspace, the eigenfunctions in the 4 nodes have relative phases of $(1, i, -1, -i)$. The meta-atoms were connected to the next unit cell by 2m-long coaxial cables (Model RG58C/U) to realize the complex hoppings by braiding (Fig. 3b, see SM, Sec. IX) [2,3]. To selectively excite the modes in the subspace $(1, i, -1, -i)$ realizing our PT-symmetric Hamiltonian, the AC signals are input to four nodes with a constant phase shift $\frac{\pi}{2}$ in a designated unit-cell. Both amplitude and phase of the voltage of each meta-atom are monitored by an oscilloscope (see detailed methods in SM, Sec. IX and Fig. S25)."

Comment 1.4: Besides the general assessment summarized above, I list here several concrete comments and questions that the authors should consider before resubmitting their work. 1.) The authors write in the abstract, that they "propose the non-Abelian bulk-edge correspondence, where the edge states are found to be fully determined by non-Abelian topological charges". However, this bulk-edge correspondence does not seem to encapsulate a particularly strong statement, namely, it is stated in the text that the edge state pertaining to charge j "can be anywhere between the 1st and 3rd band", while the edge states emanating from charge -1 "are fickle". Can the bulk-edge correspondence be transformed into a some stronger statement?

Reply 1.4: We thank the referee for the nice question.

We will first give a short answer and more detailed explanation will follow.

In the original text, there is a statement " the edge states can be anywhere between the 1st and 3rd bands for the charge of $\pm j$". This statement is correct and consistent with the bulk-edge correspondence. What we mean by the statement is that the "frame rotation bands" are the 1st and the 3rd bands, and hence the edge states with a hard boundary should exist in the bandgaps sandwiched by these two bands, meaning that the edge states can exist in both the bandgaps between 1st/2nd band and 2nd/3rd band. In the general situation, each bandgap will carry one edge state; but in the special configuration that the middle 2nd band is decoupled from the 1st and 3rd bands, the edge states can be "anywhere" in the big bandgap sandwiched between the 1st and 3rd bands. As the original statement, while correct, is difficult to understand; we removed it from the revised text.

The edge states emerging from charge -1 "are fickle", we mean the following: The charges $\pm i$, $\pm j$, $\pm k$ are topologically distinct, they cannot be transformed to one another without gap closing. However, all configurations as shown in Figs. R1a-c are classified as charge -1 and can be transformed to one another by system parameter tuning without gap closing. But, they still differ in details, which can affect how boundary states are formed as shown in Fig. R1d-f. The bulk-edge correspondence can predict those properties that are mandated by the topological principles, but not those properties that come from the "details". As the charge -1 is "richer in details" than charges $\pm i$, $\pm j$, $\pm k$, the formation of edge modes is more subtle, we hence say in the original text that "edge states emerging from charge -1 are fickle".

In the revised manuscript, we have rephrased the sentence (as shown below) to make the statement of non-Abelian bulk-edge correspondence more accurate,

"propose the non-Abelian bulk-edge correspondence, which provides a global view of the distribution of edge/domain-wall states."

In the revised supplementary materials, we have added a section (Sec. VII) to predict the positions of edge state for all charges, especially for charge -1.

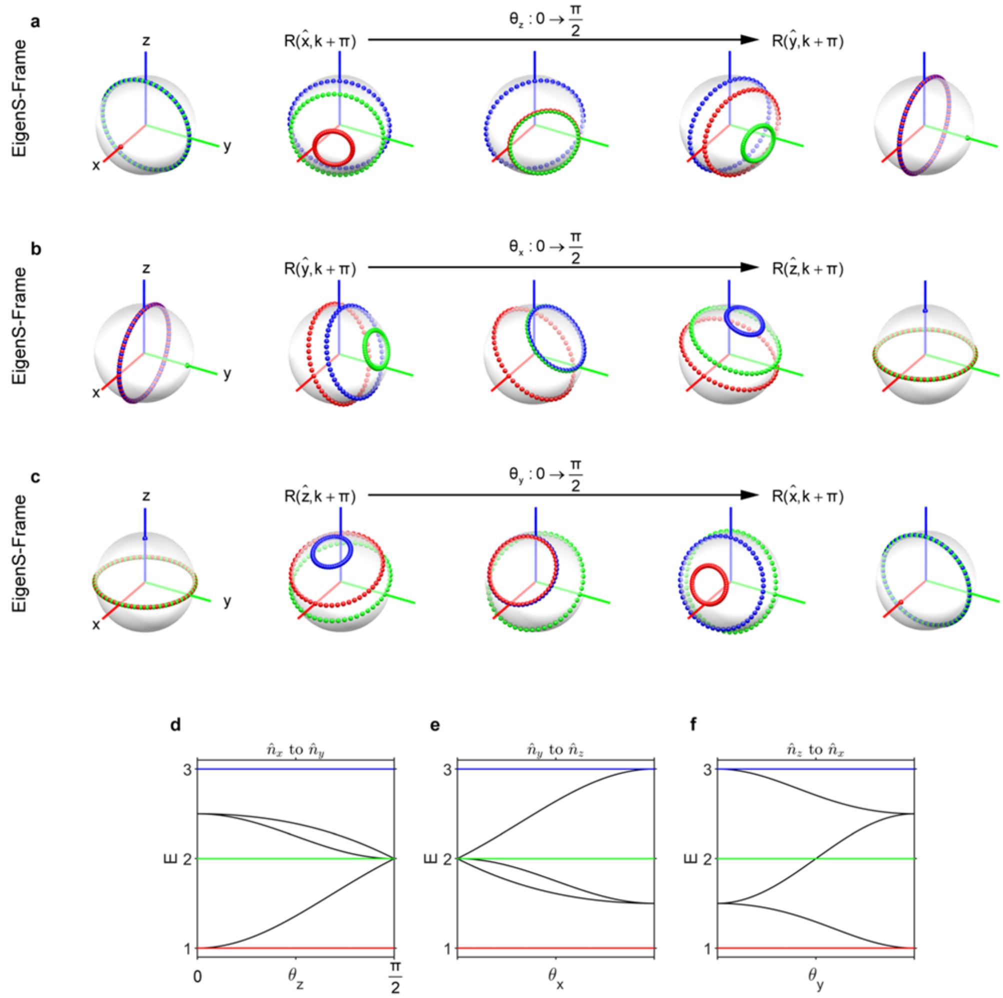

Fig. R1 (Figure S11) | Continuous transition parametrized by $\theta_{x,y,z}$ in the charge -1. a, The eigenstate frame evolving with rotating the rotation axis $\hat{x}$ to $\hat{y}$ as parametrized by $\theta_z: 0 \to \frac{\pi}{2}$. The rotation axis is defined as $\hat{n} = \cos\theta_z\,\hat{x} + \sin\theta_z\,\hat{y}$. b, Similar to (a) but for $\hat{y} \to \hat{z}$. The rotation axis is defined as $\hat{n} = \cos\theta_x \hat{y} + \sin\theta_x \hat{z}$. c, Similar to (a) but for $\hat{z} \to \hat{x}$. The rotation axis is defined as $\hat{n} = \cos\theta_y \hat{z} + \sin\theta_y \hat{x}$. Red/green/blue color indicates the eigenstate trajectory of the 1st /2nd /3rd band. d/e/f, Edge states (black lines) evolve with $\theta_{z/x/y}$ defined in (a/b/c). The 1st, 2nd and 3rd bulk bands are coloured in red, green and blue, respectively.

Detailed explanation:

In the revised text, we have added more information for the understanding and the prediction of the edge states, especially for the charge -1. As the complete argument is long, to make the reply well organized, we respectfully ask the referee to check the details in the supplementary materials, Sec. VI and VII, where we prove the non-Abelian bulk-edge correspondence and predict the edge states for different quaternion charges. In the following, we give the key arguments to show our ideas.

In an Abelian topological system, the bulk-edge correspondence can precisely predict the number of edge/domain-wall states as there is only one single bandgap under consideration. More generally in the non-Abelian topological system, the bulk-edge correspondence shows a global view of edge/domain-wall states, which includes both the number of edge/domain-wall states and the positions (bandgaps) they reside in.

For those edge/domain-wall states that can transform into each other by system parameter tuning while no topological phase transition happens in the bulk, we regard them as topologically equivalent. This is exemplified by the edge state configurations of charge -1 as shown in Fig. R1. Although these edge states change with parameter θ_i, they are topologically equivalent as they all belongs to charge -1. While the same number of edge states are sandwiched between the 1st and the 3rd bands in all the configurations, the details vary. It means we need a finer description beyond the current topological classification to precisely predict the edge state distributions in charge -1.

Our basic arguments are outlined (Fig. R2) in the following,

1. A topological phase transition does not occur without bandgap closing. In turn, the detail of bandgap closing determines the topological phase transition.
2. The bandgap closing determines the position (i.e. which bandgap) and number of edge/domain-wall states for systems. For example, the 2nd bandgap has to close when we tune system parameters to induce the transition between charges $+i$ and $+1$. We then expect the existence of edge states in the 2nd bandgap with a hard boundary.
3. The bandgap closing and the transformation of a specific 1D Hamiltonian labelled by quaternion charges to the trivial case with charge $+1$ can be visualized clearly by extending the 1D Hamiltonian onto a 2D plane, as we shall see below.
4. Topological phase transition from charge -1 to $+1$ is multiple-pathed, thus the edge states are "fickle".

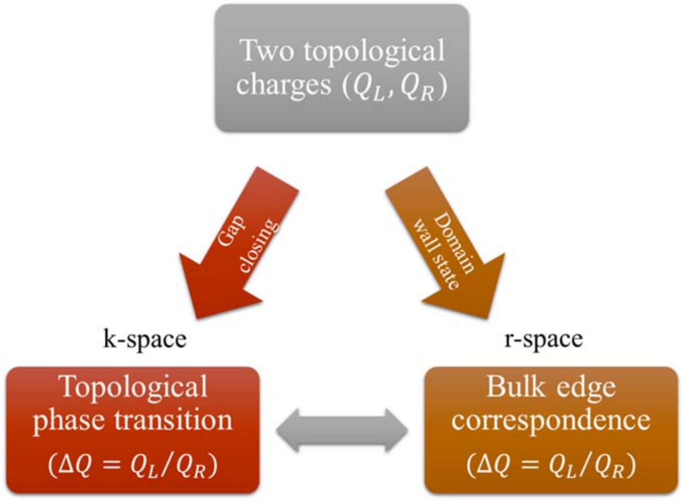

Fig. R2 (Figure S12) | Flow chart for proving bulk-edge correspondence

Arguments #1:

For Abelian topological phase transition, we only need to consider a single bandgap. Thus the bandgap closing happens there without any doubt.

For non-Abelian topological phase transition, there are multiple bandgaps. The topological transition requires that there must be bandgap closing during the process. However, the position and number of bandgap closing need more details. In our three-band system, either the 1st or 2nd bandgap may close, or both of them can close.

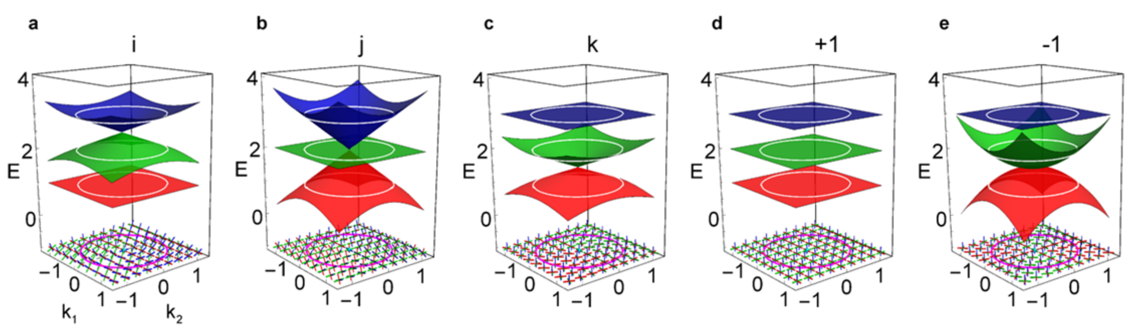

Fig. R3 (Figure S15) | Extension to 2D extended plane of the flat-band Hamiltonian (parameters are from Tab. S1). The non-Abelian charges are labelled in each panel (a-e). The white unit circle indicates the 1D Hamiltonian. The three bands are coloured in red, green and blue, respectively. The bottom plane indicates the eigenstate distribution.

Arguments #2 and #3:

First we extend the 1D Hamiltonian $H(k)$ (Eq. 2 or Eq. S2) onto a 2D extended plane,

$$H(k) = \overline{H}(\cos k, \sin k) \rightarrow \widetilde{H}(k_1, k_2)$$

where we applied the substitution $\cos k \rightarrow \rho \cos k = k_1$ *and* $\sin k \rightarrow \rho \sin k = k_2$ with $\rho \in [0,1]$. It is worth noting that the substitution preserves PT symmetry with $H(k_1, k_2) = H^*(k_1, k_2)$. The parameter ρ can be absorbed by those hopping parameters in Eq. 2 (or Eq. S2) in actual implementation.

The advantage of this extension is that the quaternion charge configuration and the edge state formation become visually obvious. Figure R3 shows the 2D band structure and eigenstate distribution for different non-Abelian topological charges, where the original Hamiltonian exactly locates on the unit circle $k_1^2 + k_2^2 = 1$. According to the definition of non-Abelian topological charges, there must be band degeneracy, i.e., Dirac cone, in the range $k_1^2 + k_2^2 < 1$ as one can see in Fig. R3a.

This can be understood in the following way. This 2D plane can be regarded as a cutting plane in the 3D space. According to the definition of non-Abelian topological charges[4], the unit circle here works like a closed loop encircling nodal lines in the 3D space. Thus, inside the unit circle ($k_1^2 + k_2^2 < 1$) there must be band degeneracy for those non-trivial topological charges ($\neq +1$). They cannot be removed unless a topological phase transition happens with the degeneracy point moving out of the unit circle.

Then we apply the Jackiw-Rebbi type argument[5] to show that there must be domain-wall state relating to the topological phase transition. In Fig. R4, we take the $+i$ charge as an example. First, by tuning system parameter, we continuously shrink the unit circle (black circle in Fig. R4a) into the magenta one, which still belongs to the charge $+i$ as long as the Dirac point lies within the circle. Secondly, when we further shrink the magenta circle as shown in Fig. R4b, topological phase transition happens and the 2nd bandgap closes. Finally, the degeneracy point of Dirac cone stands outside the magenta circle and the topological charge becomes $+1$ (Fig. R4c). Figure R4d-f give the corresponding 1D band structures along the polar angles of the three magenta circles. Around the transition point $\theta = \pi$, the PT-symmetric Hamiltonian can be parameterized as $H = \delta k_\theta \sigma_x - \delta m \sigma_z$ (Fig. R4d) and $H = \delta k_\theta \sigma_x + \delta m \sigma_z$ (Fig. R4f) with $0 < \delta m \ll 1$ being the mass term. The standard Jackiw-Rebbi argument claims that there must be one domain wall state between them two, described by $H = -i\partial_x \sigma_x + m(x)\sigma_z$ and $\lim_{x\rightarrow\pm\infty} m(x) = \pm 1$. When one consider the Dirac cone between the 1st and 3rd bands, the edge state in charge $+j$ (Fig. R3b) can be explained similarly. The arguments also work for the quadratic degeneracy as shown in Fig. R3e with charge -1. The difference for the charge -1 is that the degeneracy is quadratic, which splits into to two linear Dirac cones upon small perturbation and hence it carries two edge states (see Fig. R1e with $\theta_x \rightarrow \frac{\pi}{2}$).

From the above argument, we see that the distribution of edge states becomes apparent when we examine degeneracy in the extended 2D space as shown in Fig. R3. And this relation can be checked with comparing Fig. 2 (copied below as Fig. R5 for convenience) and Fig. R3. For example, in Fig. R3a, there is a single degeneracy between the 2nd and 3rd bands, then there exists an edge state (per edge) located in the 2nd bandgap as shown in Fig. 2b (Fig. R5b). The charge -1 in Fig. R3e and Fig. 2e (Fig. R5e) supports the similar argument. It is worth reiterating that the quadratic degeneracy supports two edge states per edge because they can split into two linear Dirac cones as mentioned above. Following the same way, more detailed explanation of edge states for charge -1 (corresponding to Figs. R1d-f) can be found in the revised supplementary materials, Sec. VII (Figs. S17-S19).

Finally, the number of extended 2D band degeneracy (as given by the number of linear Dirac cones) is equal to the number of edge states per edge. Moreover, the bandgap in which the edge states locate in is also implied by the position of degeneracy in the 2D extended space. We conclude that the gapless topological phase transition in k-space (bulk) implies the presence of topological edge/domain-wall state in r-space. More specifically, where there is a bandgap closing there is an edge/domain-wall state.

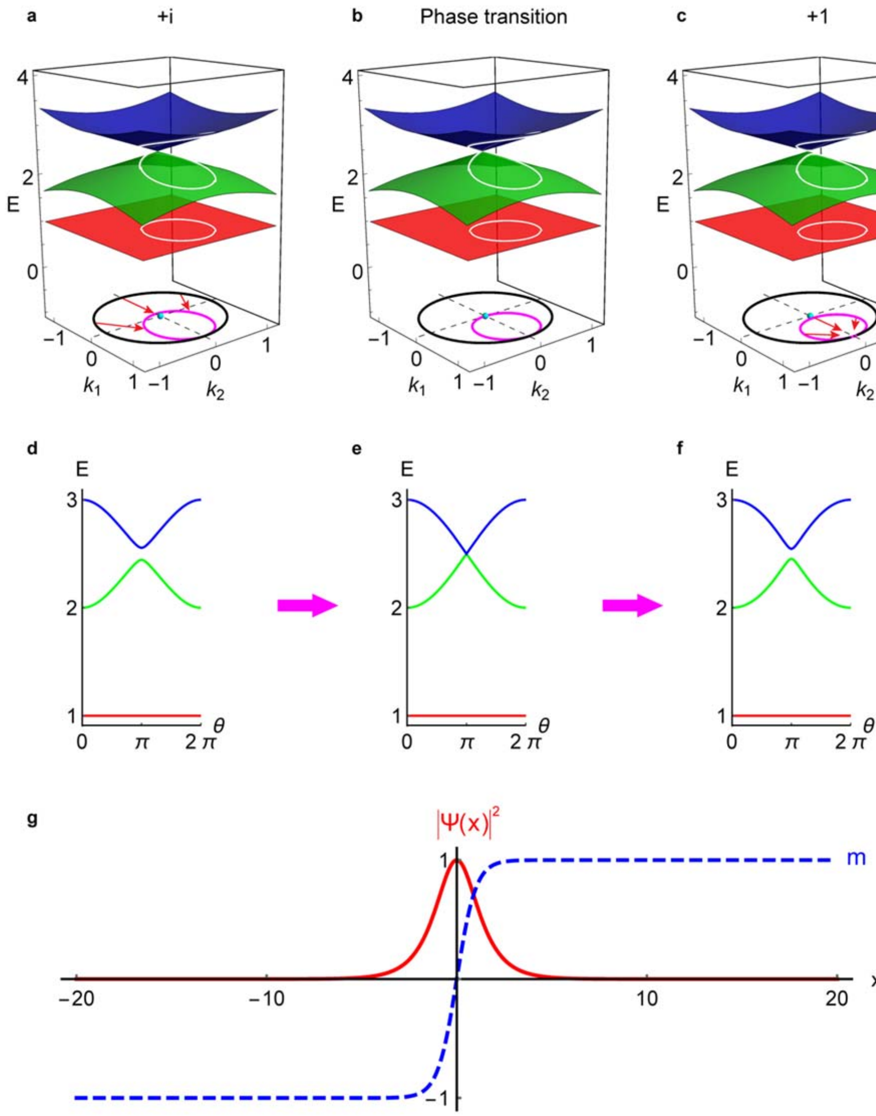

Fig. R4 (Figure S14) | Jackiw-Rebbi argument for “where there is a bandgap closing there is a domain-wall state”. a-c, Topological phase transition from charge $+i$ to $+1$ when continuously shrinking the magenta circle (white circle in each 2D band sheet) via system parameter tuning. d-f, The corresponding 1D band structure along the polar angle θ of each magenta circle. The gap-closing and gap reopening imply edge mode. g, Schematically localized domain-wall state between charges $+i$ and $+1$, corresponding to the topological phase transition (b and e).

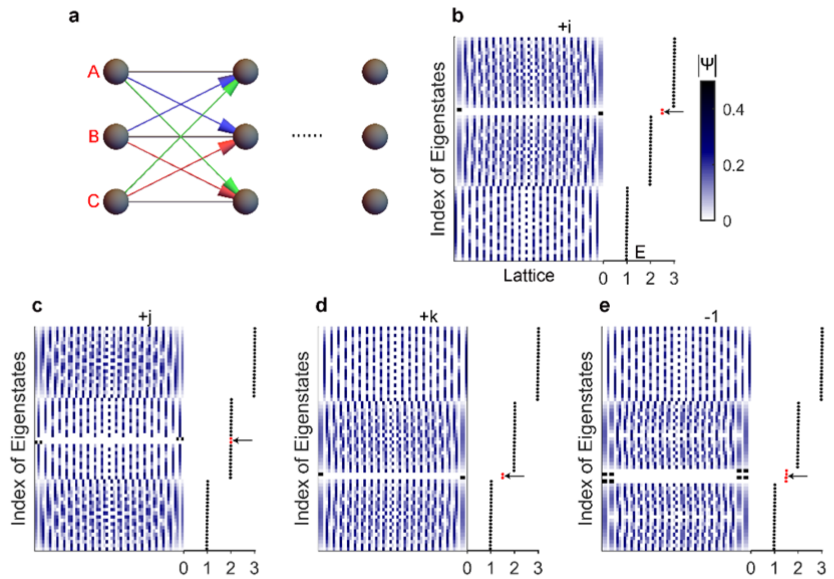

Fig. R5 (Fig. 2 in the main text) | Distribution of edge states in flat-band models.

Arguments #4:

However, the topological phase transition is more complicated in non-Abelian topological phases. For conventional topological systems characterized by Abelian charges, the transition from one topological state to another topological state involves a fixed sequential path (similar to nodes on a string or a loop as shown in Fig. R6a) and a fixed number of gap closing steps, and as such, the number of topological edge states is well defined. This is not the case for non-Abelian systems, where the transition from one topological phase to another one can take multiple distinct paths (like nodes in a network as shown in Fig. R6b). Along different paths, the topological phase transition will close the corresponding bandgap(s), which is path-dependent. Taking our system as an example, the gapless topological phase transition between charges -1 and $+1$ can lead to the closing of either the 1st bandgap or the 2nd bandgap, or even both them, depending on how we continuously tune system parameters. Thus, specifying charge -1 alone cannot determine the precise positions of edge/domain-wall states. Thus, the positions of edge states in charge -1 need more information of bulk bands (which depend on the details), which is not outside the scope of the non-Abelian bulk-edge correspondence. In this case, the existence of edge/domain-wall states is guaranteed by the topological charge, as there must be topological phase transition between charges -1 and $+1$ and thus (bandgap closings in the k-space) edge/domain-wall states in the real space; but the precise details depend on the structural details.

Very similar arguments also work for charges $\pm j$. The only difference is that for charges $\pm j$, both the 1st and 2nd bandgaps have to close simultaneously during the topological phase transition to charge $+1$. Therefore, one can expect edge states locating in the whole bandgap bounded by the top (3rd) and the bottom (1st) bands. If the 2nd band is fully decoupled from the other two bands, the whole bandgap supports a single edge state per edge (like a two-band model). In the general situation where the 2nd band couples to other bands, the 2nd band will separate the whole bandgap into two adjacent bandgaps, each of them support one edge state per edge. Again, the existence of edge states is topologically guaranteed.

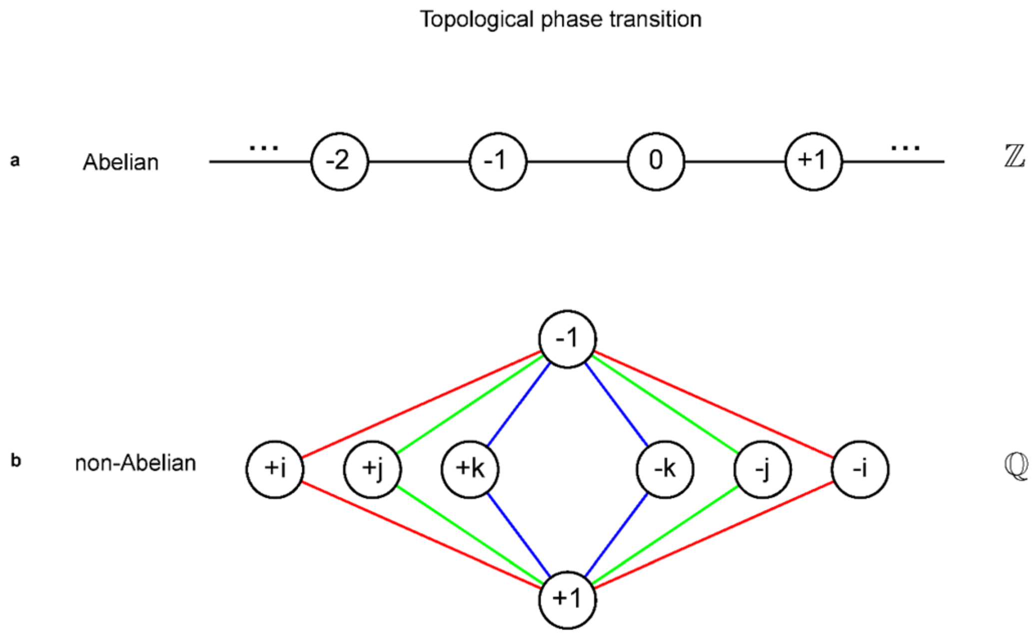

Fig. R6 (Figure S10) | Topological phase transition with Abelian (a) and non-Abelian (b) topological charges. b, Cycle graph of the quaternion group $\mathbb{Q}$. In the Abelian topological phase, the topological phase transition is single pathed. While in the non-Abelian topological phase, the topological phase transition is multiple-pathed. There are six paths that the system can be changed from charge -1 to $+1$, and the bandgap closing details are different in these paths.

Put it in a simple way, the complication of edge states in charges $-1/\pm j$ is due to the involvement of multiple bandgaps in non-Abelian topological phases. In the revised supplementary materials, we have added a section (Sec. VII) to predict the positions of edge state for all charges.

Overall, the details of edge/domain-wall states depend on additional detailed information of bulk bands, which is not outside the scope of the non-Abelian bulk-edge correspondence.

Comment 1.5: 2.) Eq. (2) contains a general nearest-neighbor PT-symmetric Hamiltonian. The authors claim that by choosing the parameters properly, they can mimic all the flat-band cases in Fig.1b—e (do these correspond to Fig. S16 and S17 from Ref. 19?). However, a look at the Supplementary Material reveals that Fig.1(e) requires next-nearest-neighbor elements beyond the Hamiltonian in Eq. (2).

Reply 1.5: Yes, the flat-band cases correspond to Figs. S16 and S17 from Ref. 19.

Fig. 1(e) does require next-nearest-neighbor hoppings beyond the Hamiltonian in Eq. (2) for the "flat band" model. Here, we want to use an ideal model with flat bands to describe the non-Abelian topological charges. However, in the practical experiment, we realized all the cases (including charge -1) with the general nearest-neighbor Hamiltonian, and the bands are not flat in the experimental configuration, which are nonetheless topologically equivalent to the ideal flat band mathematical models.

We have further stressed this in the revised text as following (on page 5),

"We first choose parameters (Tab. S1) that can mimic the flat-band cases as shown in Figs. 1b-e (Note: the panel (e) requires next-nearest-neighbour hoppings, see SM, Sec. III)."

Comment 1.6: 3.) Certain key specifications of the experimental setting are missing. Is a coaxial used as the transmission line? Furthermore, the authors claim below Eq. (2) that the constructed tight-binding model breaks both time-reversal and parity symmetry (while preserving the combined PT-symmetry). Could the authors specify where in their experimental setting is the time-reversal-breaking element? (Or do the authors mean that although the most general model described by Eq. (2) breaks P and T individually, the specific 1D models which they experimentally build preserve both symmetries?)

Reply 1.6: Yes, in the experiment we used coaxial cable as the transmission line.

We apologize for the lack of detailed information of the experimental setting in our original manuscript. In the revised text, we have supplemented those parts (on page 6) as following.

"In the experiment, a network consisting of 13 periods was designed to characterize the non-Abelian topological charges corresponding to $+i$, $+j$, $+k$ and -1, respectively. There are three meta-atoms A, B and C in one unit-cell, and Fig. 3a is the photo of the network for the specific configuration of charge -1. The inset shows that there are four nodes represented by the cable connectors labelled 1, 2, 3, 4 in each meta-atom. As such, there are four allowed subspaces. Each subspace can be characterized by a pseudo angular momentum with $e^{i4\varphi_n} = 1\ (n = 1,2,3,4)$ [2], with $\varphi_1 = 0, \varphi_2 = \frac{\pi}{2}, \varphi_3 = \pi\ and\ \varphi_4 = -\frac{\pi}{2}$. In the experiment, we work with the subspace $\varphi_2 = \frac{\pi}{2}$ to realize our 3×3 real Hamiltonian (Eq. 2) and within this subspace, the eigenfunctions in the 4 nodes have relative phases of $(1, i, -1, -i)$. The meta-atoms were connected to the next unit cell by 2m-long coaxial cables (Model RG58C/U) to realize the complex hoppings by braiding (Fig. 3b, see SM, Sec. IX) [2,3]. To selectively excite the modes in the subspace $(1, i, -1, -i)$ realizing our PT-symmetric Hamiltonian, the AC signals are input to four nodes with a constant phase shift $\frac{\pi}{2}$ in a designated unit-cell. Both amplitude and phase of the voltage of each meta-atom are monitored by an oscilloscope (see detailed methods in SM, Sec. IX and Fig. S25)."

In order to realize a full rotation of the three-band eigenstate frame, we start from the ideal flat-band configuration (Fig. 1b-e), where the Hamiltonian takes an explicit real form. When realizing the Hamiltonian in a tight-binding model, one has to introduce imaginary hopping parameters, which breaks time-reversal symmetry. Here the time-reversal and inversion symmetries are represented by the complex conjugate K and identity matrix under operation $k \to -k$, respectively. Obviously, each of them has been explicitly broken in Eq. 2 (or Eq. S2).

In the experimental setup, there is no time-reversal breaking element (i.e., magnetic field) involved in the system. Thus, the overall system is time-reversal invariant.

However, there are four nodes designed in each meta-atom. As such, there are four allowed subspaces. Each subspace can be characterized by a pseudo angular momentum with $e^{i4\varphi_n} = 1\ (n = 1,2,3,4)$, with $\varphi_1 = 0, \varphi_2 = \frac{\pi}{2}, \varphi_3 = \pi\ and\ \varphi_4 = -\frac{\pi}{2}$. We work with the subspace $\varphi_2 = \frac{\pi}{2}$ to realize our 3×3 Hamiltonian and within this subspace, the eigenfunctions in the 4 nodes have relative phases of $(1, i, -1, -i)$.

In the experiment, we specifically excite the four-nodes inside the meta-atom with a relative constant phase shift $\frac{\pi}{2}$ as $(1, i, -1, -i)$. The system of course has its time-reversal counterpart subspace $(1, -i, -1, i)$ (corresponding to φ_4), but there is no conversion between them (i.e. the system contains two block-diagonalized time-reversal counterparts, in a similar way as the Kane-Mele model). Thus, in the subspace corresponding to $\varphi_2 = \frac{\pi}{2}$, the time reversal symmetry is effectively broken, in the same way as that each spin space in the Kane-Mele model breaks the time-reversal symmetry.

Overall, both the tight-binding model and experiment setup break the time-reversal symmetry within the subspace we are considering.

We have added the above statement in the method section of revised supplementary materials (Sec. IX).

Comment 1.7: 4.) Per the trick with the four internal nodes of each “meta-atom” (sublattice), there are in total 4x3 = 12 nodes per unit cell, which should result in 12 bands. However, only three bands that emulate the non-Abelian band topology with quaternion invariant are explicitly discussed and plotted. Could the authors comment/clarify what is the fate of the remaining (12-3) = 9 bands?

Reply 1.7: Yes, the reviewer is correct that there should be a total of 12 bands. As mentioned in **Reply 1.6**, here we constrain the excitation of the four internal nodes by a constant phase step ($\frac{\pi}{2}$) between the neighboring nodes, then three bands are relevant to our investigation. In other words, our system can be block-diagonalized into the four disjoint subspaces, with each subspace containing three degrees of freedom. Here we only consider one subspace, which corresponds to our three-band system.

We have added the above statement in the method section of revised supplementary materials (Sec. IX).

Comment 1.8: 5.) Could the authors briefly comment how the bulk and edge spectra in Fig. 3 are measured? It appears to me, that to obtain the bulk spectrum through Fourier-transforming the measured voltages, a periodic boundary condition needs to be set up by closing the 1D system into a ring. In contrast, open boundary conditions should be present to detect the edge spectrum.

Reply 1.8: Yes, the bulk bands can be investigated by applying a periodic boundary condition via arranging the 1D system into a ring, as mentioned by the reviewer. On the other hand, the bulk bands together with edge states can be simultaneously studied by using a finite but long chain of lattices. In a Hermitian system, the periodic boundary condition and open boundary condition have the same bulk continuum as long as the systems contain a large enough number of unit cells.

In the experiment, we measured the bulk spectrum using an open chain containing 13 unit cells (and about 470 coaxial cables) and an open chain of this size gives essentially the same bulk spectrum as a ring configuration. In our measurement of bulk bands, the signal is injected from the middle position of an open chain that is very far away from the boundary so that the edge states cannot be excited. For the detection of edge states, the signal is injected at the position near the edge.

We have added the above statement in the method section of revised supplementary materials (Sec. IX).

Comment 1.9: 6.) It remains unclear to me after studying this manuscript to what extent are the topological phases “+i” and “-i” distinct from each other. Naively, I would expect that the overall sign cannot be meaningfully defined, since one can change the pi-rotation of the eigenstate-frame into a (-pi)-rotation by doing a discrete gauge transformation on the eigenstate-frame with an element of the O(3) group. In other words, the action of the gauge group should split the elements of the quaternion group into conjugacy classes, with “+i” and “-i” corresponding to a single conjugacy class.

Reply 1.9: Yes, the charges of $+i$ and $-i$ belong to the same conjugacy class. They are obtained in the fundamental homotopy group $\pi_1\left[\frac{O(3)}{O(1)^3}\right] \cong \pi_1\left[\frac{SO(3)}{D_2}\right] \cong \mathbb{Q} = (1, \pm i, \pm j, \pm k, -1)$. The quaternion group $\mathbb{Q}$ has five conjugacy classes in total and three of them contain two conjugate elements, i.e., $\pm i$, $\pm j$ and $\pm k$. In the Abelian topological charge group, such as $\mathbb{Z}$ and $\mathbb{Z}_2$, each element is a class by itself. However, in the non-Abelian topological charge group one class may contain several elements. Thus, conjugacy class is special to the non-Abelian topological phases. The study of conjugacy classes of non-Abelian groups is fundamental for the study of their structure. Elements of the same conjugacy class cannot be distinguished by using only the group structure and therefore share many properties. However, it does not mean that the two conjugate elements are the same. In the non-Abelian topological phases one cannot continuously transform charge $+i$ to $-i$ without gap closing.

In the revised supplementary materials, we have added the above discussion in Sec. I and the non-Abelian topological phase transition from $+i$ to $-i$ in Sec. IV (Fig. S9d, copied below as Fig. R7 for convenience).

In addition, two elements a and b of group G are conjugate, if there exists an element $g \in G$ such that $gag^{-1} = b$). An discrete group element in $O(3)$ does not belong to the quaternion group $\mathbb{Q}$, and thus it cannot make the conjugacy equivalence between charges $+i$ and $-i$.

Although two rotations $e^{\pm\frac{\phi}{2}L_x} \in O(3)$ are conjugate by $e^{-\frac{\phi}{2}L_x} = M_{y,z}e^{+\frac{\phi}{2}L_x}M_{y,z}^{-1}$ ($M_y = diag(1,-1,1)$ and $M_z = diag(1,1,-1) \in O(3)$ indicating mirror operations), $e^{\pm\frac{\phi}{2}L_x}$ are different from charges $\pm i$ by definition. The latter are the discrete elements of fundamental homotopy group $\mathbb{Q}$ representing non-Abelian topological phases while the former are continuous elements of $O(3)$.

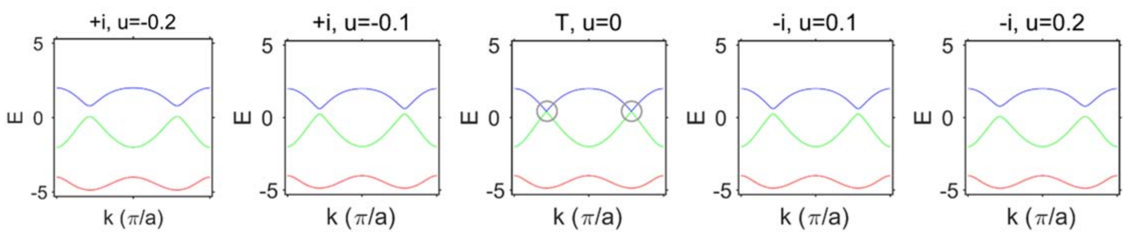

Fig. R7 (Figure S9d) | Topological phase transition (T) from charge $+i$ to $-i$ by changing one of the system parameters ($u = -0.2 \rightarrow 0.2$). Two linear crossings arise during the transition implies a change of charge $\Delta Q = i/-i = -1$. (Parameter setting, $s_{CC} = -4, v_{AA} = -1, v_{BB} = 1, v = 1$, the rest are all 0)

Comment 1.10: However, my conclusion seems be refuted by the data in Fig.S12(a), where a domain wall between phases “+i” and “-i” results in bound states reminiscent of the edge of phase -1 (=+i/-i). Could the authors clarify this issue? In fact, this is the only type of domain where the arising bound states cannot be explained using solely the quantized Berry phases in the individual band gaps. From this perspective, I would like to urge the authors to move this very interesting result from the Supplemental Material into Fig. 4 of the main text.

Reply 1.10: We thank the referee for the nice suggestion.

As mentioned in **Reply 1.9**, charges $+i$ and $-i$ are two distinct topological phases. There must exist two linear Dirac cones (between the 2nd and 3rd bands) during the transition from charge $+i$ to $-i$ (as shown in Fig. R7). Thus, the domain-wall between charges $+i$ and $-i$ supports two topological domain-wall states (in the 2nd bandgap), just like the edge states of charge -1 (Fig. S11d with $\theta_z \rightarrow 0$). On the other hand, according to our non-Abelian bulk-edge correspondence, the relation $\Delta Q = +i/-i = -1$ implies that the domain-wall of charge pair $(+i, -i)$ supports similar edge states of charge -1 with a hard boundary. Finally, the two arguments are consistent.

In the revised manuscript following the referee's kind suggestion, we have moved Fig. 12a-b into the main text as Fig. 4g-h.

Referee #2 (Remarks to the Author):

Comment 2.1: The non-Abelian quaternion topological charges in momentum space were firstly introduced in ref. 19 (Science 364, 1273, 2019) to characterize some novel line nodes. Some features of these non-Abelian quaternion topological charges have been demonstrated in the authors' recent work in ref. 22 (Phys. Rev. Lett. 125, 033901 2020), but those are indirect evidences. I believe the current work is indeed the first direct measurement on the non-Abelian quaternion topological charges, which certainly is very important. I have a few questions, which hopefully the authors can clarify.

Reply 2.1: We thank the referee for his/her recognitions and positive assessments of our work.

Comment 2.2: 1. In the case of a domain wall between +i and -i charges, if we follow Fig. 4c, then the first bandgap should have no edge state, and the second bandgap should have no edge state either (Zak phase of pi is equivalent to -pi). But according to the new bulk-edge correspondence proposed in the current work, the edge state should correspond to the case of charge -1, which does support edge states as shown in Fig. 2e. Could the authors clarify this issue?

Reply 2.2: We thank the referee for pointing out this concern.

For a single bandgap system (or we only focus on a single bandgap while ignoring other bandgaps, in other words other bandgaps may close at will), the topological phase protected by PT symmetry is $\mathbb{Z}_2$ classified. It means that the bandgap may have Zak phase taking values of 0 and $\pm\pi$, where $\pm\pi$ refer to the same phase with mod of 2π. Thus, no topological edge state is guaranteed to exist on the domain-wall between $+\pi$ and $-\pi$.

In the non-Abelian topological system with three bands separated by two bandgaps, if we label each band with Zak phase, there are $2^{n-1} = 4$ (the total number of band $n = 3$) possibilities. Because each band can take two values, 0 or $\pm\pi$ ($\pm\pi$ *are the same with mod of* 2π), and the sum of all Zak phases $\sum_{n=1}^{3} \phi_n = 0\ (mod\ 2\pi)$ imposes that only $n-1$ bands are independent. However, the non-Abelian topological charge $\mathbb{Q}$ has 5 conjugacy classes $(+1, \pm i, \pm j, \pm k, -1)$. Therefore, the class -1 goes beyond the Zak phase description[4], where we label it as 2π to make the distinction between it and the trivial class $+1$ (as elaborated by the '*' in the caption in Fig. 4). For other conjugacy classes, each has the unique Zak phase distribution. Between two different classes, the Zak phases $+\pi$ and $-\pi$ are topologically equivalent being consistent to the usual argument in the Abelian topological phases. But, in a single conjugacy class, we apply them two distinguish two distinct elements. For example, we label the 1st/2nd bandgap of charge $\pm i$ with $0/\pm\pi$ (as shown in Fig. 4c in the main text). It is due to that in each conjugacy class the two elements (i.e. $+i$ and $-i$) have a relative meaning, i.e., representing the two opposite rotations of the eigenstate frame when k running across the first Brillouin zone (k: $-\pi \rightarrow \pi$). It is worth stressing that although the two charges $+i$ and $-i$ are belong to one conjugacy class, they are still two topologically distinct charges as their transition has to induce bandgap closing. Furthermore, there are two linear Dirac cones during the transition (as shown in Fig. R8). Thus, the domain-wall between $+\pi$ and $-\pi$ of charges $+i$ and $-i$ supports two topological domain-wall states, just like the edge states of charge -1 (Fig. S11d with $\theta_z \rightarrow 0$). On the other hand, according to our non-Abelian bulk-edge correspondence, the relation $\Delta Q = +i/-i = -1$ implies that the domain-wall of charge pair $(+i, -i)$ supports similar edge states of charge -1 with a hard boundary. Finally, the two arguments are consistent.

In summary, it is different for a two-bandgap system, where the two bandgaps are tangled together. Zak phases $\pm\pi$ indicate two relatively different phases in one conjugacy class. Thus, the domain-wall between $+\pi$ and $-\pi$ supports topological states, just like the edge states of charge -1.

We have added above discussion in the revised supplementary materials, Sec. I.

In the main text, we further emphasized this as following (on page 8),

"As shown in Fig. 4c, two bandgaps are individually labelled by the corresponding Zak phases for each quaternion charge. The Zak phase for a single bandgap takes the value of 0 or π as the homotopy mapping $\pi_1\left(\frac{O(3)}{O(2)\times O(1)}\right) = \pi_1(\mathbb{R}P^2) = \mathbb{Z}_2$, which means one cannot distinguish $+\pi$ from $-\pi$. Here in the multiple bandgap system, we use $+\pi$ and $-\pi$ to make the distinction between two conjugate elements in one conjugacy class. For example, we label the 1st/2nd bandgap of charge $\pm i$ with $0/\pm\pi$ (as shown in Fig. 4c). It is due to that in each conjugacy class the two elements (i.e. $+i$ and $-i$) have a relative meaning, i.e., representing the two opposite rotations of the eigenstate frame when k running across the first Brillouin zone (k: $-\pi \rightarrow \pi$). Between two different classes, the Zak phases $+\pi$ and $-\pi$ are topologically equivalent being consistent to the usual argument in the Abelian topological phases. From above discussion, we see that the non-Abelian system (M_3) possesses a more refined topological structure than $\mathbb{R}P^2$. As the class -1 goes beyond the Zak phase description[4], where we label it as 2π to make the distinction between it and the trivial class $+1$ (as elaborated by the '*' in the caption of Fig. 4). The Zak phase of charge -1 is 2π only in some special cases, i.e., when one of band is fully decoupled and the rest two bands can be classified by $\mathbb{Z}$ for the PT symmetric system[4]."

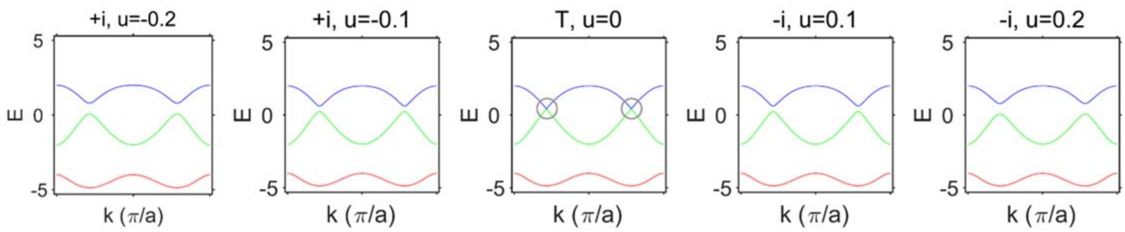

Fig. R8 (Figure S9d) | Topological phase transition (T) from charge $+i$ to $-i$ by changing one of the system parameters ($u = -0.2 \rightarrow 0.2$). Two linear crossings arise during the transition implies a change of charge $\Delta Q = i/-i = -1$. (Parameter setting, $s_{CC} = -4, v_{AA} = -1, v_{BB} = 1, v = 1$, the rest are all 0)

Comment 2.3: 2. In Fig. S12a, varying the probe position changes not only the edge state distribution, but also the number of edge states. Why is that?

Reply 2.3: We thank the referee for pointing this out.

This is due to the different spatial distributions of domain-wall states. In the revised supplementary materials, we have added the following figure (Fig. R9) to clearly address the issue. For the two domain-wall states (the 1st and 3rd in Fig. R9b), we need to probe them at the unit cell indicated by the orange triangle, while the last (the 2nd in Fig. R9b) one can only be probed at the position of gray triangle.

In short, the domain wall states can have nearly zero amplitude at some particular sites and the probe has to be moved to other positions to detect those states.

In the revised supplementary materials (Sec. VIII), we have added the above discussion.

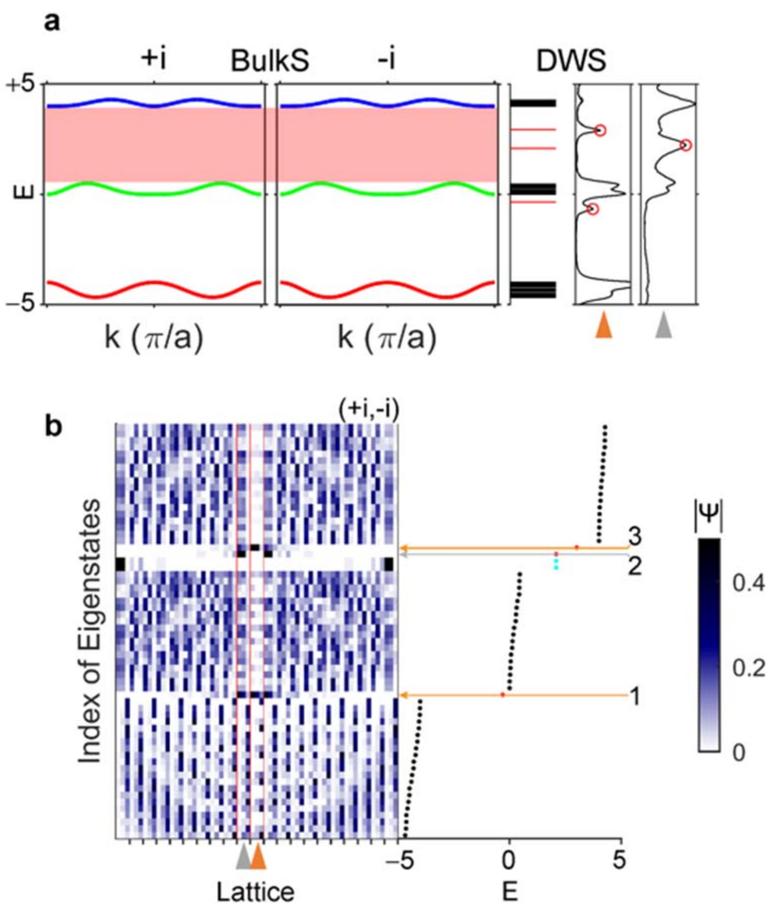

Fig. R9 (Figure S22) | Field distribution of domain-wall states between charges of $+i$ and $-i$. a, Bulk (BulkS) and domain-wall states (DWS). On the right side the two experimental panels correspond to different probing positions as shown in (b). b, Field distribution of the bulk and domain-wall states. Orange/gray triangle indicates the position to probe the (1st and 3rd)/2nd domain-wall states. We note that the domain-wall states can have nearly zero amplitude at some sites.

Referee #3 (Remarks to the Author):

Comment 3.1: In their manuscript "Experimental observation of non-Abelian topological charges and bulk-edge correspondence", Guo and collaborators investigate both theoretically and experimentally the emerging concept of non-Abelian topological charges in 1D reciprocal space, which was first kick started by Bzdusek and his colleagues last year. The authors present the mathematical analysis (homotopy group theory) as well as the first experimental observation of the quaternions in the 1D PT-symmetric transmission line network. In particular, the authors propose for the first time the non-Abelian bulk-edge correspondence, and provide the experimental evidences for their statement. All of these aspects are novel and very interesting, their experimental results are neat and timely, and the manuscript is well organized. I believe this work will be of significant interest to the community. Before recommending it for publication in Nature, the authors need to address the following comments and suggestions.

Reply 3.1: We thank the referee for the nice summary and positive assessments.

Comment 3.2: I find the mathematical model present to explain the essential idea of the paper is hard to read, I believe the readers who are not familiar with the language of homotopy group need more explanations from the authors. For example, how does the rotation matrix R(k) come? Why it has the form R(k)=exp((k+\pi)/2*Lx,y,z) for topological charges +i/+j/+k but has the different form R(k)=exp((k+\pi)*Lx,y,z) for charge -1? What's more, The discussion of isomorphism map between O(3) and SO(3) reads obscure, and the figure 1(f-i) do not provide me more insight than the figure 1(b-e) do, and I don't see the single curve is terminated at antipodal points in SO(3) sphere for charges +i/+j/+k. I suggest the authors either provide more physical intuition for these abstract concepts in the main text, and detail the according explanation in the supplementary materials if necessary, or give relevant references to their argument.

Reply 3.2: We thank the referee for the questions and suggestions. We apologize for the confusion in our presentation.

Here the rotation matrix is employed to build an ideal real Hamiltonian for the three-band system. Generally a Hermitian Hamiltonian can be constructed by $H(k)=\sum_{n=1}^{N}\lambda_n|u_k^n\rangle\langle u_k^n|$, where we assume no band degeneracy and thus $\lambda_1<\lambda_2<\cdots<\lambda_N$. Each eigenvalue λ_n corresponds to a single eigenstate $|u_k^n\rangle$, i.e. $H(k)|u_k^n\rangle=\lambda_n|u_k^n\rangle$. Furthermore, spectral theorem says that any Hermitian matrix can be written as $H(k)=U(k)\Lambda U(k)^\dagger$ ('†' combines both complex conjugate '∗' and transpose 'T') with $\Lambda=diag(\lambda_1,\lambda_2,\ldots,\lambda_N)$ being diagonal and real, where $U(k)$ is a unitary matrix consisting of (ordered) orthonormal eigenstates $|u_k^n\rangle$. In our PT symmetric system, the Hamiltonian can take real symmetric form and thus the unitary matrix $U(k)$ reduces to be real rotation matrix $R(k)$. On the other hand, we make $\lambda_n=n$ to further simplify the analytical calculation without changing the underlying topological arguments. Thus, the real symmetric Hamiltonian takes the simple form of $H(k)=R(k)diag(1,2,\ldots,N)R(k)^T$. When $k=-\pi\to\pi$ runs across the first Brillouin zone, $R(k)$ serves to rotate the eigenstate of each band from an initial state $|u_k^n\rangle=(0,\ldots,\underbrace{1}_{n},\ldots,0)$ with only the n^{th} entry being 1. For the non-Abelian charges $+i/+j/+k$, two of three eigenstates rotate π ($|u_k^n\rangle\to-|u_k^n\rangle$) when $k=-\pi\to\pi$ spans the 2π range, which can be expressed by $R(k)=\exp\left[\frac{(k+\pi)}{2}L_i\right]$ $(i=x,y,z)$ with $(L_i)_{jk}=-\epsilon_{ijk}$ and ϵ_{ijk} being the fully antisymmetric tensor. For the charge -1, two of three eigenstates rotate 2π ($|u_k^n\rangle\to|u_k^n\rangle$) when $k=-\pi\to\pi$ spans the 2π range, which gives $R(k)=\exp[(k+\pi)L_i]$ $(i=$

x, y, z). Thus, when $k = -\pi$ the rotation matrix is a $N \times N$ unit matrix $I_{N\times N}$, i.e., $R(k = -\pi) = \exp\left[\frac{(-\pi+\pi)}{2} L_i\right] = I_{N\times N}$ $(i = x, y, z)$. Actually, different non-Abelian topological charges will have different rotation matrix $R(k)$. In other words, the rotation matrix $R(k)$ carries the underlying topological characters and determines the non-Abelian topological charges.

In the revised manuscript, we have added the above details into supplementary materials (Sec. I) to make our presentation clearer.

In order to provide a more compact presentation (and also satisfy the length limit of <2500 words), we decide to move Fig. 1f-i and the associated discussions into the supplementary materials (Sec. I). In addition, in the same section, we added the following paragraph to make the arguments of the isomorphism $\frac{O(3)}{O(1)^3} \cong \frac{SO(3)}{D_2}$ clearer.

"The space of Hamiltonian can be written as $M_3 = \frac{O(3)}{O(1)^3} \cong \frac{SO(3)}{D_2}$ [4,6], where the first equality is obtained by an $O(3)$ rotation of the eigenstate frame. As flipping the sign of each eigenstates $|u_k^n\rangle \to -|u_k^n\rangle$ $(n = 1,2,3)$ leaves the Hamiltonian $H(k) = \sum_{n=1}^{N} \lambda_n |u_k^n\rangle\langle u_k^n|$ invariant, one then imposes the $O(1)^3$ quotient, where $O(1)^3 \cong D_{2h} \cong \mathbb{Z}_2^3$ is generated by three mutually perpendicular mirror symmetries. In the second equality, both groups have been replaced by their proper subgroups, i.e., $O(3) \to SO(3)$ and $D_{2h} \to D_2$, where the dihedral point group D_2 consists of the identity and three π- rotations around mutually perpendicular axes."

We also have added the following paragraph to further explain the antipodal points in the $SO(3)$ space.

"The special orthogonal group $SO(3)$ can be further parameterized as a solid sphere, wherein the normalized vector $\vec{n} = (n_x, n_y, n_z)$ indicates the rotation axis and radius is the rotation angle $\phi \in [0, \pi]$. It is worth noting that the antipodal pairs $(\vec{n}, \pi)$ and $(-\vec{n}, \pi)$ represent the same rotation and thus are identical. When k runs from $-\pi$ to π, $R(k)$ traces out a curve in the space of $SO(3)$. On the other hand, one can form the topological space X/H with identifying points of X which can be related by some element of H $(x \equiv xh)$, where $x \in X$ and $h \in H$. For the above coset space $\frac{SO(3)}{D_2}$, the D_2 quotient further generates each element $R(k) \in SO(3)$ to be four elements as $R(k) \circ g$ where $g \in D_2 = (I, C_{2x}, C_{2y}, C_{2z})$. They represent four equivalent rotations up to D_2 rotations. If $R(k) \in SO(3)$, then the trajectories of $R(k\!: -\pi \to \pi) \circ D_2$ represent the four curves in the $SO(3)$ parameter space. For different non-Abelian charges, the curves shown in Figs. S1a-d correspond to Figs. 1b-e in the main text, respectively. Although one single curve cannot connect a pair of antipodal points on the parameterized solid sphere of $SO(3)$, two curves connected together are terminated at the antipodal points as shown in Fig. S1. For example, the green and blue (red and black) curves in Fig. S1a for charge $+i$ thread a pair of antipodal points, they are hence closed and not contractible, which is topologically guaranteed by $\pi_1(SO(3)) = \mathbb{Z}_2$. However, for the charge of -1 as shown in Fig. S1d, one single curve connects a pair of antipodal points because when k running across the first Brillouin zone the rotation $R(k)$ goes from $(\vec{n}, 0) \rightarrow (\vec{n}, \pi) \rightarrow (\vec{n}, 2\pi) \equiv (\vec{n}, 0)$ as indicated by the black curve."

In the revised text, relevant references have been cited accordingly for all above details.

Comment 3.3: In the 3 bands model, the topological transition between different non-Abelian charges follows the order +i -> +j -> +k because of the bandgaps close and reopen. For Abelian case, the topological transition occurs between the trivial phase and topological phase. Does similar scenario occur for the non-Abelian case, for example, the quaternion charge transition from +i/+j+/k to +1? A broader question is, can the topological transition of quaternion charges occur in an arbitrary order?

Reply 3.3: We thank the referee for the insightful question.

Yes, the topological phase transitions can occur via different pathways, and there exist topological phase transitions from $+i/+j/+k$ to $+1$. Generally for the Abelian topological phase transitions, one only needs to focus on the single bandgap, whose closing-reopening process implies topological phase transitions. However, in the non-Abelian topological systems, there are multiple bandgaps involved, and bandgaps may close-reopen individually or simultaneously. For example, the topological phase transition from $+i/+j/+k$ to $+1$ requires closing-reopening of the 2nd/(1st and 2nd)/1st bandgap(s) as shown in Fig. R10. We also notice that the nontrivial phase transitions with charge change of $\Delta Q \neq -1$ have one Dirac like transitions (Figs. R10a-c) while the cases with $\Delta Q = -1$ imply two Dirac cones (Fig. R10d).

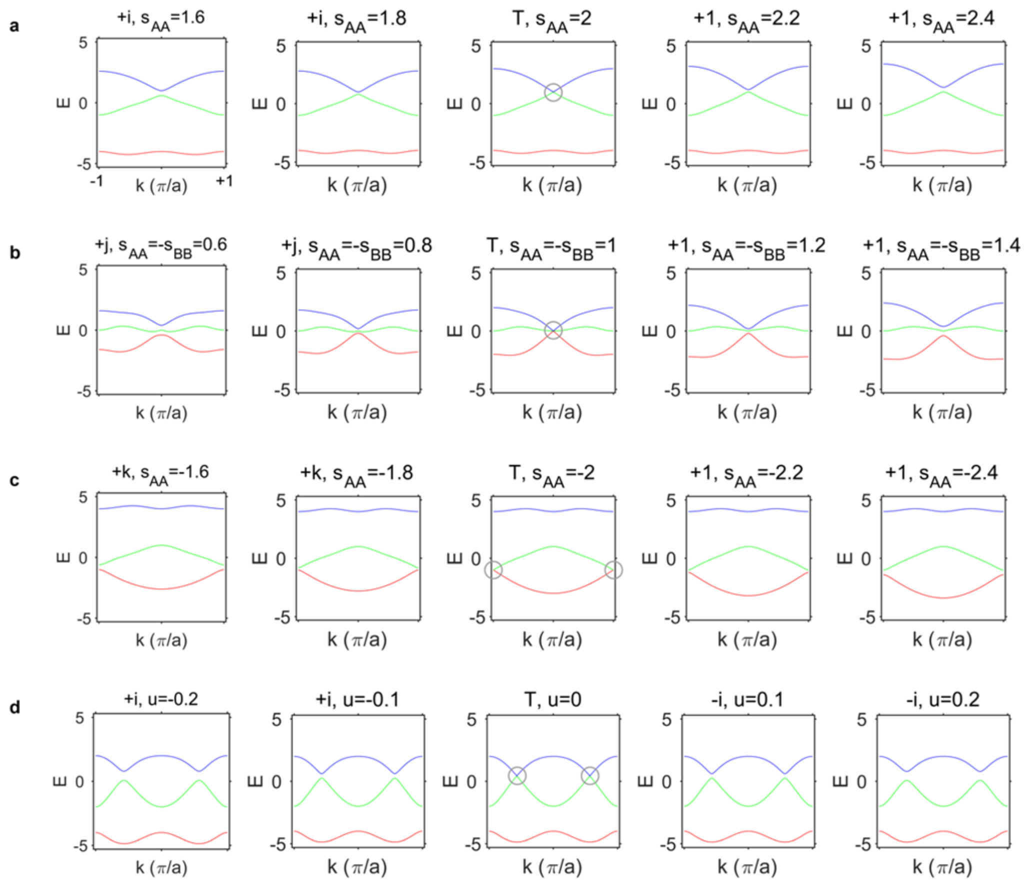

Fig. R10 (Figure S9) | Topological phase transition (T) between charges $+i$ to 1 (a), $+j$ to 1 (b), $+k$ to 1 (c) and $(+i)$ to $-i$ (d). Two linear crossings in (d) arise during the transition implies a change of charge $\Delta Q = i/-i = -1$. All others experience only one linear crossing. (Parameter setting, a: $s_{CC} = -4, v_{AA} = -\frac{1}{2}, v_{BB} = \frac{1}{2}, u = -\frac{1}{2}, v = \frac{1}{2}$; b: $v_{AA} = -\frac{1}{2}, v_{BB} = \frac{1}{2}, u = -\frac{1}{2}, v = \frac{1}{2}$; c: $s_{CC} = 4, v_{AA} = -\frac{1}{2}, v_{BB} = \frac{1}{2}, u = -\frac{1}{2}, v = \frac{1}{2}$; d: $s_{CC} = -4, v_{AA} = -1, v_{BB} = 1, v = 1$; the rest are all 0)

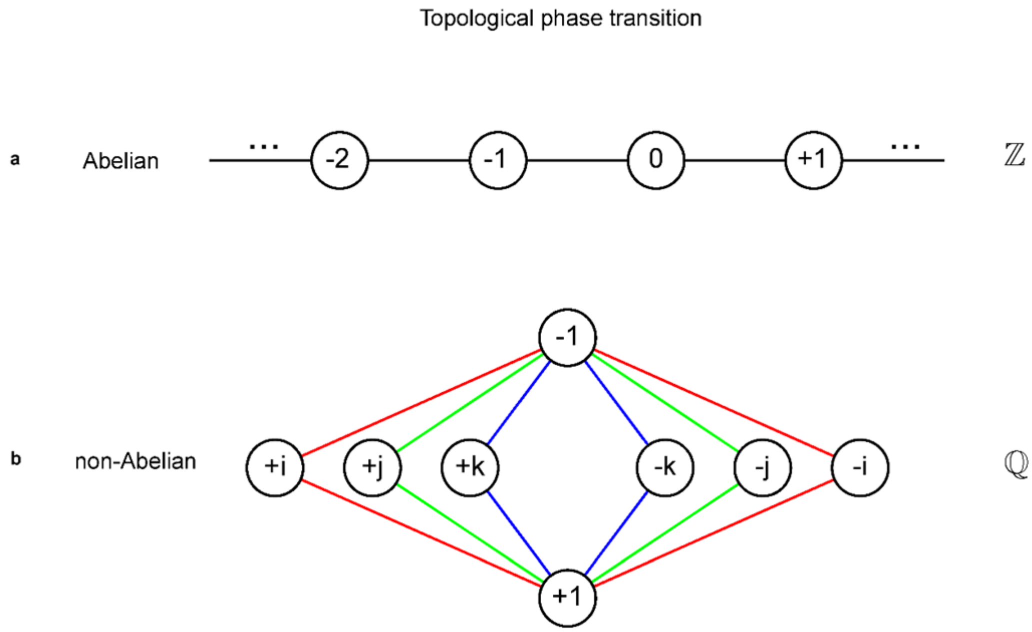

Fig. R11 (Figure S10) | Topological phase transition with Abelian (a) and non-Abelian (b) topological charges. b, Cycle graph of the quaternion group $\mathbb{Q}$. In the Abelian topological phase, the topological phase transition is single pathed. While in the non-Abelian topological phase, the topological phase transition is multiple-pathed. There are six paths that the system can be changed from charge -1 to $+1$, and the bandgap closing details are different in these paths.

More interestingly, the topological phase transition is more complicated in non-Abelian topological phases. For conventional topological systems characterized by Abelian charges, the transition from one topological state to another topological state involves a fixed sequential path (similar to nodes on a string or a loop as shown in Fig. R11a) and a fixed number of bandgap closing steps, and as such, the number of topological edge states is well defined. This is not the case for non-Abelian systems, where the transition from one topological phase to another one can take multiple distinct paths (like nodes in a network as shown in Fig. R11b). Along different paths, the topological phase transition will close the corresponding bandgap(s), which is path-dependent. For example, from charge -1 to $+1$ there are exhaustively six distinct paths as shown in Fig. R11b. The six paths are topologically equivalent, one cannot single out any one of them just from topological viewpoint. The gapless topological phase transition between charges -1 and $+1$ can lead to the closing of either the 1st bandgap or the 2nd bandgap, or even both them, depending on how we continuously tune system parameters. From this perspective we see that the gapless topological phase transitions between charges of -1 and $+1$ are not unique, and this additional complexity implies the fickle edge states.

We have added the above discussion in the revised supplementary materials, Sec. IV (Figs. S9-S10).

Comment 3.4: The bulk quaternions are distinguished by their distinct behaviors on the so called "EigenS-Frame" sphere, which not only determine the number of edge states in the bandgap, but also decide the distribution of them over several bandgaps. To experimentally show the non-Abelian topological charges, the authors extract the eigenvectors from measured phases and fields and plot them on the "EigenS-Frame", and the experimental results and theoretical results match so well. Since eigenvectors are not gauge invariant, and therefore, in principle are not measurable quantities, how do the authors keep track of the right eigenstates (eigenstates might not be excited), and fix the gauge of the measured eigenvectors in the experiment?

Reply 3.4: We thank the referee for the interesting question.

Yes, eigenvectors are not gauge invariant, it is hard to measure without imposing some extra conditions. In other words, one has to fix the gauge field first and then map them in the experiment. First we design the PT symmetric Hamiltonian (Eq. 2) to be real (via carefully introducing complex hoppings). Consequently, the eigenvectors are real as well. From this perspective, the only freedoms left are $\pm|u_k^n\rangle$. Finally, we impose the right-handed rule to the eigenstate frame, i.e., $(|u_k^1\rangle \times |u_k^2\rangle) \cdot |u_k^3\rangle > 0$ when plotting the eigenstate frame sphere.. The eigenstates in the momentum space (k-dependent) are Fourier-transformed from their distribution in the real space. In the experiment, to excite the eigenmodes better, the AC signals are input to meta-atoms A, B and C successively in a designated unit cell.

In the revised manuscript, we have added the above description in the method section (SM, Sec. IX).

Comment 3.5: Different from the edge states in 1D SSH model, the edge states for charge +j and -1 are not topologically stable in their energy since they are fickle and vary according to the details of the model. What about the edge states for charge +i/+k? Are they stable in their energy? In relation to this, are these edge states topologically robust against the disorder? The authors may include the study of this aspect either theoretically or experimentally.

Reply 3.5:

The edge states of charge $+i/+k$ always stably locate in the 2nd/1st bandgap. Following the referee's nice suggestion, we have calculated the disorder system as shown in Fig. R12a/b to show the robustness of the edge states against disorder for charge $+i/+k$, as long as both two bandgaps are not closed by the disorder.

We have added the discussion in the revised supplementary materials, Sec. III (Fig. S7).

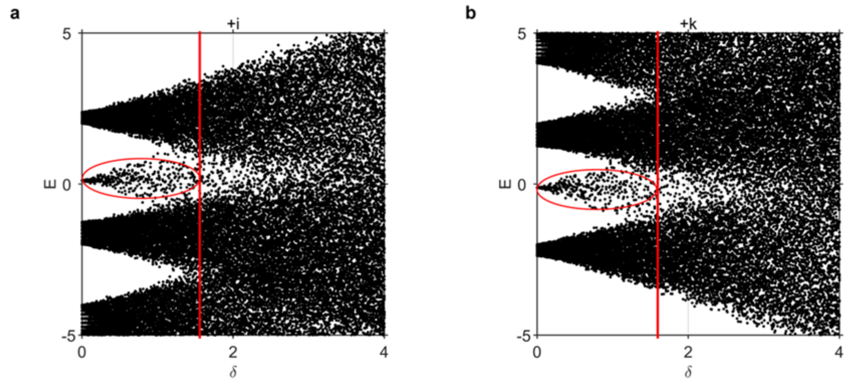

Fig. R12 (Figure S7) | Robustness of edge states against disorder for charges $+i$ (a) and $+k$ (b). The solid red lines indicate non-Abelian topological phase transitions (as long as one of two bandgaps is closed) with increasing the disorder strength δ added onto s_{AA}, s_{BB} and s_{CC}, i.e., $s_{AA,n} = s_{AA} + \epsilon_A(|\epsilon_A| < \delta)$ with n indicating the site number (Parameters are from Tab. S2). The red ellipses indicate the corresponding edge states.

Comment 3.6: The authors claim the non-Abelian bulk-edge correspondence is determined by \DeltaQ=Q_L/Q_R, although all edge states can be predicted from Zak phases of the respective bandgaps, or equivalently the phases \pi winding of the eigenstates. The authors provide some but not exclusive examples to show such relation indeed holds, but how can we assure this non-Abelian relation is generally true?

Reply 3.6: We thank the referee for the great question.

In the revised text we use the following three steps to prove the non-Abelian bulk edge correspondence.

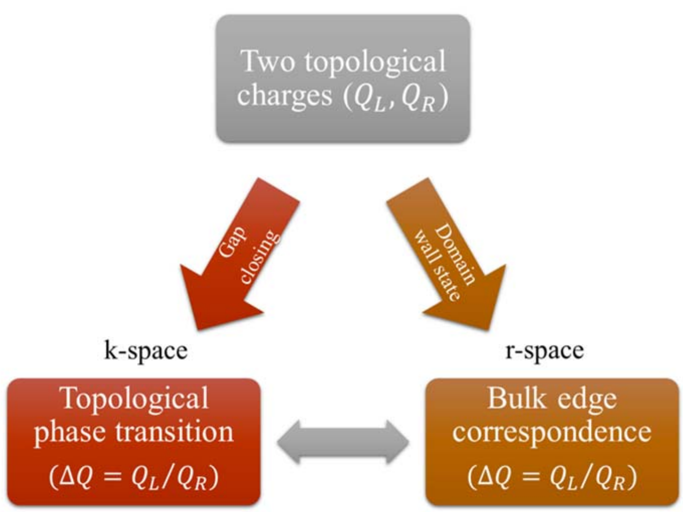

Fig. R13 (Figure S12) | Flow chart for proving bulk-edge correspondence

1. From the most general perspective, a topological phase transition between two distinct topological phases (described by two different topological charges) must be accompanied by bandgap closing and re-opening. It works for both Abelian and non-Abelian topological phases. (Fig. R13)
2. The topological phase transition between two distinct topological charges Q_L and Q_R is described by $\Delta Q = Q_L/Q_R$. In other words, the detail of bandgap closing is described by $\Delta Q = Q_L/Q_R$. (Fig. R14)
3. The closing bandgap in k-space implies the existence the domain wall state(s) in r-space when splicing the two topologically distinct samples together (especially in the 1D system). And thus the domain wall states are described by $\Delta Q = Q_L/Q_R$.

(Note that left inverse ($Q_R^{-1}Q_L$) and right inverse ($Q_L Q_R^{-1}$) show similar results finally, thus we write $\Delta Q = Q_L/Q_R$.)

Proof:

1. The first step is obvious.

For Abelian topological phase transition, we only need to consider a single bandgap. Thus the bandgap closing happens there without any doubt.

For non-Abelian topological phase transition, there are multiple bandgaps. The topological transition requires that there must be bandgap closing during the process. However, the position and number of bandgap closing need more details. In our three-band system, either the 1st or 2nd bandgap may close, or both of them can close.

■

2. The second step arises from the definition of non-Abelian topological charges.

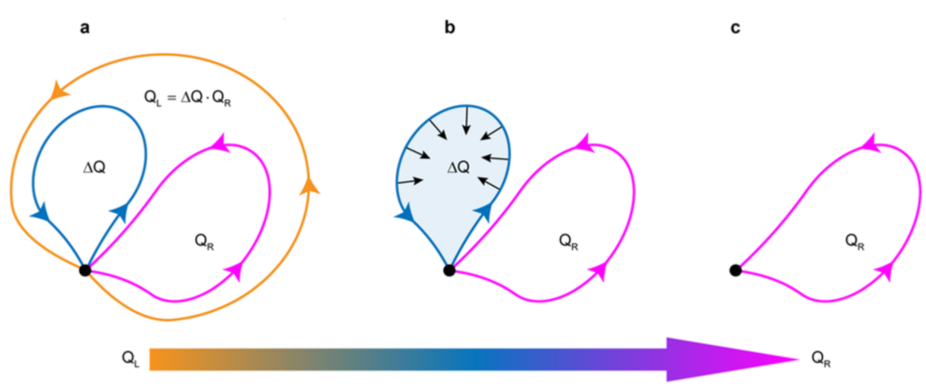

Fig. R14 (Figure S13) | The loop product in the fundamental homotopy group and topological phase transition from charge Q_L (a) to charge Q_R (c) with shrinking the blue loop charge ΔQ

(b) to the base point (black dot). **a,** The charge $Q_L = \Delta Q \cdot Q_R$ indicated by the loop product. **b,** Shrinking the charge ΔQ loop to be the base point to enable the topological phase transition from Q_L to Q_R. **c,** The charge Q_R after topological phase transition.

Definition: The set of all homotopy classes of loops in a space X under the loop product is the fundamental homotopy group $\pi_1(X)$.

For the 1D Hamiltonian when k runs from $-\pi$ to π, the Hamiltonian traces a loop in the order parameter space[4] $X = M_3 = \frac{O(3)}{O(1)^3}$. The quotient form of M_3 imposes that all three bands are fully gapped for any $k \in [-\pi, \pi]$. Then, each non-Abelian topological charge is well-defined via the above definition of fundamental homotopy group $\pi_1(M_3) = \mathbb{Q}$. All non-Abelian topological charges together construct the group satisfying all group axioms. Here, the group product is loop product between two homotopy classes by definition. For example, the group product $\Delta Q \cdot Q_R$ indicates that k first running from $-\pi$ to π obtains charge Q_R and then k running again with charge ΔQ finally reaches to $Q_L = \Delta Q \cdot Q_R$ ($Q_L, Q_R, \Delta Q \in \mathbb{Q}$). Obviously, the topological transition between charges Q_L and Q_R has to experience "k running from $-\pi$ to π with charge ΔQ". Finally, the continuous transition between the two charges Q_L and Q_R has to make the ΔQ loop shrink to one point (base point), which inevitably encounters bandgap closing as long as $Q_L \neq Q_R$ ($\Delta Q \neq +1$). In other words, the bandgap closing is characterized by the charge ΔQ. Thus, the topological phase transition is described by $\Delta Q = Q_L/Q_R$.

Basically, the relation of $\Delta Q = Q_L/Q_R$ works for all of topological charges, including both Abelian and non-Abelian, for systems with single and multiple bandgaps, respectively.

■

3. First we extend the 1D Hamiltonian $H(k)$ (Eq. 2 or Eq. S2) onto a 2D extended plane,

$$H(k) = \overline{H}(\cos k, \sin k) \rightarrow \widetilde{H}(k_1, k_2)$$

where we applied the substitution $\cos k \rightarrow \rho \cos k = k_1$ *and* $\sin k \rightarrow \rho \sin k = k_2$ with $\rho \in [0,1]$. It is worth noting that the substitution preserves PT symmetry with $H(k_1, k_2) = H^*(k_1, k_2)$. The parameter ρ can be absorbed by those hopping parameters in actual implementation.

The advantage of this extension is that the quaternion charge configuration and the edge state formation become visually obvious. Figure R15 shows the 2D band structure and eigenstate distribution for different non-Abelian topological charges, where the original Hamiltonian exactly locates on the unit circle $k_1^2 + k_2^2 = 1$. According to the definition of non-Abelian topological charges, there must be band degeneracy, i.e., Dirac cone, in the range $k_1^2 + k_2^2 < 1$ as one can see in Fig. R15a.

This can be understood in the following way. This 2D plane can be regarded as a cutting plane in the 3D space. According to the definition of non-Abelian topological charges[4], the unit circle here works like a closed loop encircling nodal lines in the 3D space. Thus, inside the unit circle ($k_1^2 + k_2^2 < 1$) there must be band degeneracies for those non-trivial topological charges ($\neq +1$). They cannot be removed unless a topological phase transition happens with the degeneracy point moving out of the unit circle.

Then we apply the Jackiw-Rebbi type argument[5] to show that there must be domain-wall state relating to the topological phase transition. In Fig. R16, we take the $+i$ charge as an example. First, by tuning system parameter, we continuously shrink the unit circle (black circle in Fig. R16a) into the magenta one, which still belongs to the charge $+i$ as long as the Dirac point lies within the circle. Secondly, when we further shrink the magenta circle as shown in Fig. R16b, topological phase transition happens and the 2nd bandgap closes. Finally, the degeneracy point of Dirac cone stands outside the magenta circle and the topological charge becomes $+1$ (Fig. R16c). Figures R16d-f give the corresponding 1D band structures along the polar angles of the three magenta circles. Around the transition point $\theta = \pi$, the PT-symmetric Hamiltonian can be parameterized as $H = \delta k_\theta \sigma_x - \delta m \sigma_z$ (Fig. R16d) and $H = \delta k_\theta \sigma_x + \delta m \sigma_z$ (Fig. R16f) with $0 < \delta m \ll 1$ being the mass term. The standard Jackiw-Rebbi argument claims that there must be one domain wall state between them two, described by $H = -i\partial_x \sigma_x + m(x)\sigma_z$ and $\lim_{x\to\pm\infty} m(x) = \pm 1$. When one consider the Dirac cone between the 1st and 3rd bands, the edge state in charge $+j$ (Fig. R15b) can be explained similarly. The arguments also work for the quadratic degeneracy as shown in Fig. R15e with charge -1. The difference for the charge -1 is that the degeneracy is quadratic, which splits into two linear Dirac cones upon small perturbation and hence it carries two edge modes.

From the above arguments, we see that the bandgap that the edge states locate in is implied by the position of degeneracy in the 2D extended space. Finally, the number of extended 2D band degeneracy (as given by the number of linear Dirac cones) is equal to the number of edge states per edge. Moreover, the bandgap in which the edge states locate in is also dictated by the position of degeneracy in the 2D extended space. We conclude that the gapless topological phase transition in k-space (bulk) implies the presence of topological edge/domain-wall state in r-space. More specifically, where there is a bandgap closing there is an edge/domain-wall state.

Therefore, the edge/domain-wall states are characterized by $\Delta Q = Q_L/Q_R$ as it fully describes the topological phase transition (Step 2).

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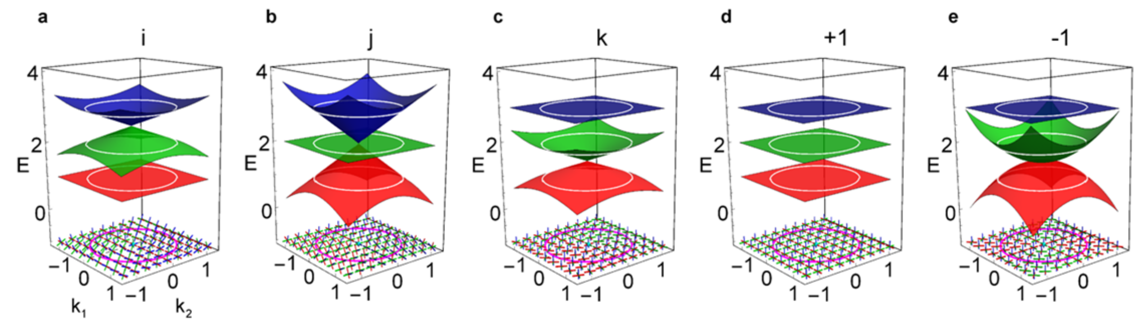

Fig. R15 (Figure S15) | Extension to 2D extended plane of the flat-band Hamiltonian (parameters are from Tab. S1). The non-Abelian charges are labelled in each panel (a-e). The white unit circle indicates the 1D Hamiltonian (Eq. 2 or Eq. S2). The three bands are coloured in red, green and blue, respectively. The bottom plane indicates the eigenstate distribution.

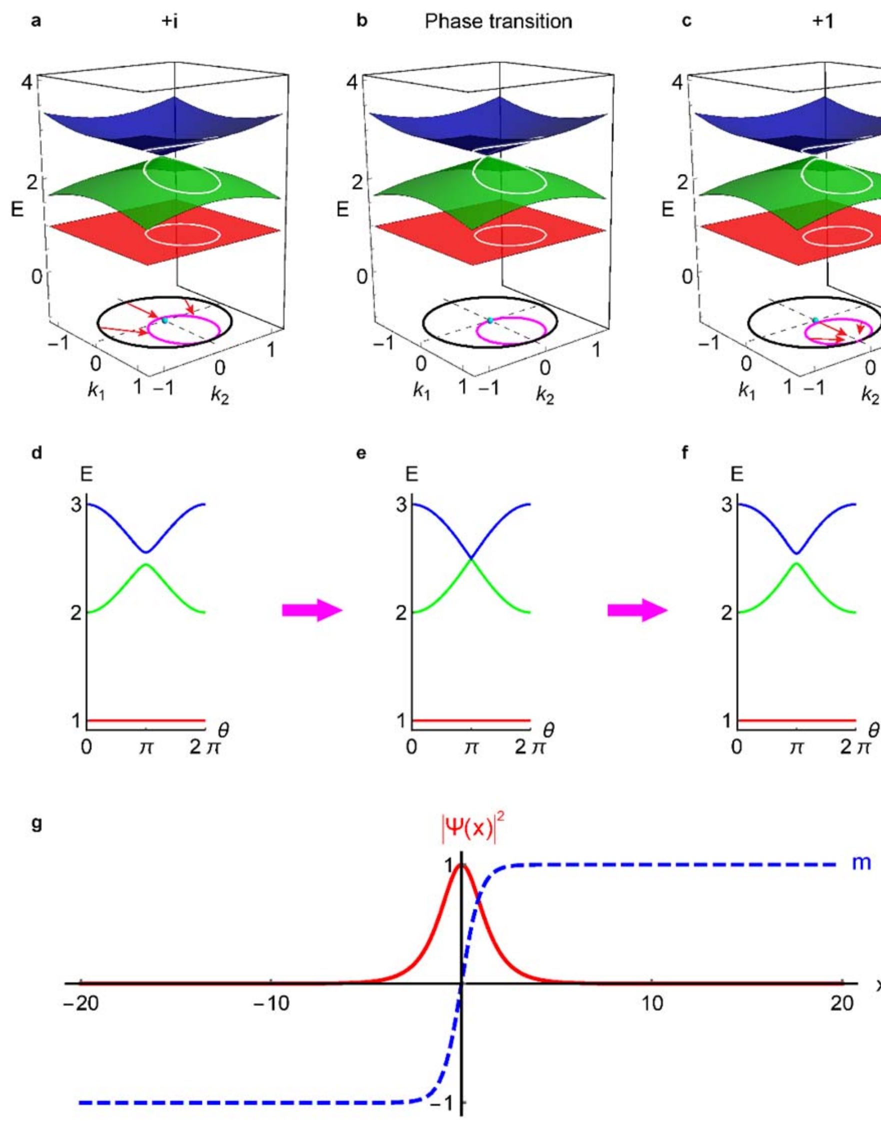

Fig. R16 (Figure S14) | Jackiw-Rebbi argument for "where there is a bandgap closing there is a domain-wall state". a-c, Topological phase transition from charge $+i$ to $+1$ when continuously shrinking the magenta circle (white circle in each 2D band sheet) via system parameter tuning. d-f, The corresponding 1D band structure along the polar angle θ of each magenta circle. The gap-closing and gap reopening imply edge mode. g, Schematically localized domain-wall state between charges $+i$ and $+1$, corresponding to the topological phase transition (b and e).

The above proof has been added as a new section (Sec. VI) in the revised supplementary materials.

We have also added another new section (Sec. VII) in the revised supplementary materials to further make the arguments with predicting edge states from the extended 2D plane.

Comment 3.7: For the model implemented in the experiment, why the number of edge states is 4 for topological charge +j (Figure S4, (b))? I expect it is also 2 based on the flat band TBM in Figure 2(c).

Reply 3.7: We thank the referee for pointing this out.

We divide all cases of charge $+j$ into two categories. The first one is a special case in which the 2nd band is fully decoupled from other two bands, and as such we can neglect the 2nd band and consider the whole bandgap carrying Zak phase of π and thus supporting 1 edge state per edge (2 edge states in total) accordingly. The other case is the more general case in which the 2nd band couples with the 1st and 3rd bands, and there are two bandgaps (respectively between 1st/2nd band and 2nd/3rd band), where each gap carries a Zak phase of π and thus supports 1 edge states per edge (in total of 4 edge states for two bandgaps and two edges).

Following the referee's suggestion, we show the transition between the two cases, i.e., from decoupling to coupling of the 2nd band (Fig. R17). Basically, we see one extra edge state (per edge) appears upon switch-on of the coupling. In Fig. R17b, we shift the 2nd band away from $E = 2$ to more explicitly show the phenomena.

We have added the above discussion in the revised supplementary materials (Fig. S6).

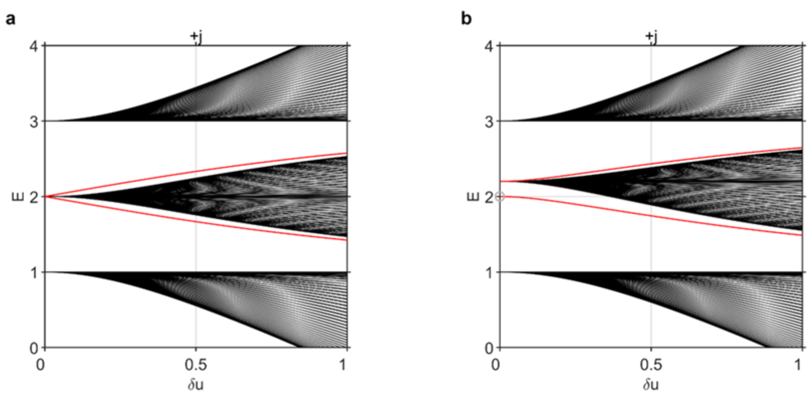

Fig. R17 (Figure S6) | Coupling of the 2nd band induces one more edge state (red line) per edge in the charge $+j$. a and b, Corresponding to $s_{BB} = 2$ and $s_{BB} = 2.2$, respectively. When $\delta u = 0$, the 2nd band is decoupled from the other two bands, the system can be regarded as a two-band system. There is only one edge state per edge as indicated by the grey circle in (b) (see Fig. 2c in the main text corresponding to (a)), while one more edge state (per edge) emerges from the middle (2nd) band when $\delta u > 0$ (switching on the coupling). (Parameter setting, $u \to u + \delta u$, the rest is from Tab. S1)

Comment 3.8: The following questions are also very interesting and general, but they might not directly relate to the major points of this manuscript, therefore, the authors might or might not consider implementing them.

Are the quaternion charges unique for three bands model? Assume if an extra band is added into the system, will this band trivialize other bands like the case of fragile topology?

Reply 3.8: We thank the referee for the interesting questions.

Here, we mainly focus on experimentally characterization of the non-Abelian topological phases in a three-band PT symmetric system, which has the smallest band number towards realizing non-Abelian topological phases. Involving extra bands into the system definitely deserves further investigations and may introduce novel physics as well.

We initially divide the questions into two categories: 1. The first one is that after involving the extra bands there is no degeneracy between two mutually adjacent bands, i.e., the eigenvalues $\lambda_1 < \lambda_2 < \cdots < \lambda_N$; 2. The second case is that after involving the extra bands the degeneracies between adjacent bands are permitted, i.e., $\lambda_1 < \lambda_2 < \cdots \leq \cdots < \lambda_N$.

The first category has been theoretically studied by Bzdušek and his colleagues (Science 365, 1273, (2019)) via generalizing the quaternion charge from $\mathbb{Q}$ to be $\bar{P}_N$, i.e., $\bar{P}_N = \bigcup_{n_i \in (0,1)} (\pm e_1^{n_1} e_2^{n_2} \ldots e_{N-1}^{n_{N-1}})$ $(2^N\ elements\ in\ total)$ with the Clifford algebra satisfying $e_i^2 = -1, e_i e_j = -e_j e_i, 1 \leq i \neq j \leq N-1$. Basically, the corresponding topological charges are non-Abelian and exhibit many intriguing properties.

The second category still remains an open question, the order space of Hamiltonian is different and may have more fine structures, i.e., $\frac{O(N)}{O(1)\ldots O(2)\ldots}$. It may exhibit the fragile topology as speculated by the referee. We will work on this interesting topic in the future.

Comment 3.9: Since the non-Abelian charges are non-commutative, do they place the constraint on the admissible configurations of the 3 bands model?

Reply 3.9: Yes, they do place some constraints on the admissible configurations of the three-band model. Particularly in the 3D, the PT symmetric nodal links formed by consecutive pairs of bands exhibit interesting braiding structures and the underlying topological features are described by the non-Abelian topological charges, such as quaternions[4].

Interestingly, on each cutting-plane in the 3D momentum space, 3D nodal lines reduce to 2D Dirac points and so the same admissible constraints can also be inherited by 2D Dirac points. The two Dirac points of honeycomb lattices carry opposite winding numbers and they can annihilate in pairs[7] due to the famous no-go theorem[8]. However, when multiple bands (n>2) are involved, the annihilation of Dirac points depends on the path they travel before merging. It implies the so-called "failure of no-go theorem" [9] and shows the non-Abelian braiding features.

In addition, in a 3D system, if the whole system does not possess PT symmetry but there exists C_2T-invariant plane, Weyl points can be braided on the C_2T plane[10] and the underlying non-Abelian constraints will determine the path-dependent pairwise-annihilation of Weyl points.

Comment 3.10: Provided further studies addressing my comments are performed, the manuscript meets the criteria of Nature and therefore I recommend the acceptance.

Reply 3.10: We genuinely thank the referee for the recommendation.

Reference:

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10 Bouhon, A. et al. Non-Abelian reciprocal braiding of Weyl points and its manifestation in ZrTe. Nature Physics, doi:10.1038/s41567-020-0967-9 (2020).

Reviewer Reports on the First Revision:

Ref #1

I have read the resubmitted version of the manuscript by Guo et al., titled “Experimental observation of non-Abelian topological charges and bulk-edge correspondence”, and also carefully studied the detailed reply to all the referees’ comments. Although I have read the largely supportive reports of the other two referees, I am still wondering why this particular work merits the high attention of journal Nature. It is admittedly an enthusiastic experience to emulate tight-binding models of novel topological bands in a table-top experiment with hundreds of coaxials, and eventually to observe that the spectra of an actual physical system match the theory. However, from the broader perspective, the authors’ results don’t push neither the theoretical understanding of the non-Abelian band topology, nor do they put forward novel experimental technique or overcome a major experimental challenge. I don’t want to be disrespectful, but one could say the authors manifested a great ability to practically implement matrix Hamiltonians, but what does one really learn? How is an experiment of a scientific value, if it does not really offer a possibility to disprove a theory (in this case the diagonalization of Hermitian matrices)? If the authors failed to find agreement with the theory, the explanation would most certainly be that their emulation of the matrix Hamiltonian was not precise enough. Therefore, while the experiment looks impressive, and it is the first particular such experiment to implement the non-Abelian band-topological models, my recommendation (which I fear to be overruled by the other two referees) remains to publish these results in a more specialized journal such as [REDACTED]. Irrespective of my critique above, let me comment on the actual experiment and analysis presented in this work, which applies regardless of the journal in which the results would eventually be disseminated. I find the reply to most referees’ comments satisfiable, and I appreciate the authors’ efforts with moving parts of the theoretical discussion to the Supplemental Material, significantly shortening the text while improving its clarity, and providing a more detailed information about the further details on the experimental setup. The analysis of experimental data looks solid and the text is well organized. The work is nearly ready for publication. There is, however, one major point about which I must be very strict. In all fairness, the authors cannot claim that they propose a non-Abelian bulk-edge correspondence (as they do in both the abstract and the introduction). While in their reply (and also in the amended version of the Supplemental Material) the authors go to great lengths to the make the appearance of a novel proof of the bulk-edge correspondence, their results cannot be taken as one; in fact the authors hardly prove anything on this question beyond what has already been established by previous works. Before I explain the problems with the “proof”, let me emphasize that this deficiency does not in any way spoil the value of the experimental efforts and data presented by this work. However, misleading advertisements and false claims in abstract would have adverse implications on the overall understanding of the authors’ results by the community, as well as for the interpretation of any follow- up studies. Therefore, I must insist that a milder formulation, such as “Furthermore, we experimentally observe the manifestation of the non-Abelian topological charges [at domain walls…]” would be far more appropriate in the abstract/intro. Let me explain why the authors’ proof and statement on this issue are very misleading. Due to the technicality and the various aspects of the "proof", I have divided my critique into three parts: 1.) When focusing on the case of conjugacy classes of "i/j/k", one is considering topological bands that are readily distinguished by the quantized Berry-Zak phases. The bulk-edge correspondence for non-trivial Berry-Zak phase is well understood: in general, it does not lead to in-gap edge state, because by shifting the energy of the edge state it can disappear inside the bulk energy band. When that happens, the edge develops a quantized (e/2) edge charge of the valence/conduction bands, which is taken as the universal signature of the non-trivial Berry-Zak phase. Equivalently, the filling anomaly is developed, and also taken to be a universal signature of the non-trivial Berry-Zak phase irrespective of the presence/absence of the topological edge mode [see e.g. the discussion in Sec. III of PRB 99, 245151 (2019)]. The signatures of the non-trivial Berry-Zak phase as summarized by the previous paragraph are at present well established. By applying this known correspondence to any selected energy gap (one gap at a time) of multi-gapped models, one readily deduces the completely analogous signatures of the conjugacy classes i/j/k beyond the discussion of the present work. In a sharp contrast, as the authors' analysis in the present work focuses too much on band structures close to a topological phase transition (i.e., near a gap closing), cf. Figs. S14-S19, their "proof" completely overlooks the possible absence of the edge mode in generic models with more complicated dispersion. In particular, it should in principle be possible for the topological edge states of the "+/-i" phase to drop all the way from the upper energy gap to the lower energy gap. (This is compatible with the known bulk-boundary correspondence for non-trivial Berry-Zak phase, and is not refuted by the authors' present analysis.) Note also that the conjectured bulk-boundary correspondence for these classes does not go beyond the one readily conjectured by Ref. [19]. 2.) The topological class "-1" is the only one here that really goes beyond the characterization with BerryZak phases. Therefore, here there is a possibility to discover some truly novel aspect of the bulkboundary correspondence. However, as the authors themselves admit in their reply: "As the charge −1 is "richer in details" than charges ±i, ±j, ±k, the formation of edge modes is more subtle, we hence say in the original text that "edge states emerging from charge -1 are fickle"" The authors present an argument based on an analogy with the Jackiw-Rebbi model, which suggests that the -1 charge should exhibit a pair of edge modes. Nevertheless, as visible in Fig. S11, this charge is found to be associated with three (rather than two) edge state in the studied models, unless the model parameters are fine-tuned to specific values (namely ones that correspond to a complete decoupling of the third band – cases which I find analyzed below Eq. (69) of Ref.19). The authors' simplistic analysis has troubles interpreting these features, and it also does not predict any actually robust signature (such as the bulk polarization and filling anomaly for the non-trivial Berry-Zak phase) which would characterize the -1 charge. Once again, there is simply not enough ground for the claim "we propose the non-Abelian bulk-edge correspondence" in the abstract and introduction. 3.) The authors seem to distinguish between the bulk-edge (two different band structures meeting at a domain wall?) and the bulk-boundary (vacuum on the other side of the edge?). I didn't find the distinction explicitly clarified in the text, and it thus took me a while to decipher in their reply the intended meaning of "Furthermore, we also propose non-Abelian bulk-edge correspondence for the first time, and as the reviewer stated, our work directly generalizes "the previously introduced bulk-boundary correspondence for the non-Abelian topological charge"". Perhaps "bulk-domain wall" would be a less confusing formulation. In either case, there is a subtle but important issue here that closely relates to one of my earlier questions. Namely, in my previous reply, I have asked how does one distinguish phases "+i" and "-i", which are related to each other by a gauge transformation. I respectfully disagree with the authors statement that “In the non-Abelian topological phases one cannot continuously transform charge +i to −i without gap closing” and with the paragraphs that follow this statement. To see the problem, consider the explicit (flat-band) model encoded with the SO(3) eigenframe $R(k)$ = (cos k 2 sin k 2 0 − sin k 2 cos k 2 0 0 0 1) which the authors take to corresponds to charge „+i“. By flipping in this expression the sign k → -k, one obtains the inverse matrix which encodes a Hamiltonian with charge “-i”. Now, imagine a system with a domain wall, and a band structure initially characterized by the R(k) given above on both sides of the domain wall (thus, initially, the domain wall is not “really” there as the system remains translationally invariant). Then, as a function of a tuning parameter p, one changes the flat-band Hamiltonian on the right side of the domain wall to $Q(k; p)$ = (1 0 0 0 cos p sin p 0 − sin p cos p) (cos k 2 sin k 2 0 − sin k 2 cos k 2 0 0 0 1) (1 0 0 0 cos p − sin p 0 + sin p cos p) while keeping the system on the left side of the domain wall unchanged. This is a continuous deformation that keeps the energy bands on both sides flat (and both energy gaps open), and which interpolates between what the authors would call “charge +i" at p=0 and “charge -i" at p=Pi [note that Q(k,Pi) = R(-k)]. The authors claim to investigate the domain wall between band structures with charges “+i" and “-i" (which is found to resemble end modes of charge “-1”). However, as the above example explicitly illustrates, this domain wall is continuously removable without closing the bulk energy gaps and while keeping the PT symmetry of each band structure. [An analogous issue stemming from the related non-Abelian properties of the Euler class in 2D PTsymmetric systems is discussed in Appendix D of PRB 102, 115135 (2020).] Therefore, when comparing two different band structures, one can only tell whether they are in the same or different conjugacy class. In contrast, the “absolute” charge difference ∆Q=QL/QR (the quantity purported to govern the “generalized bulk-edge correspondence”) seems to be meaningful only if the two models are relatable through a small (gap-closing) tuning of parameters, but not in general. [In particular, the explicit models considered by the authors’ simplistic analysis all produce the same matrix H(k=0). This choice misguides one to think that a canonical gauge/base point can always be found that leads to a unique element of the quaternion group, but it really is just a consequence of the overly simple form of the models considered by the authors.] These simple observations thus further undermine the authors’ claim that their work “generalizes “the previously introduced bulk-boundary correspondence for the non-Abelian topological charge””. I don’t find in this work any new and rigorous theoretical analysis concerning the non-Abelian bulk-boundary correspondence. For all these reasons, I have to urge the authors to relax their statement in the abstract and in the introduction (as well as at the appropriate places in the main text) by the weaker, but more appropriate, formulation such as the one I have explicitly suggested in blue earlier in my report. I cannot recommend this work for publication unless this serious issue is properly resolved by the authors. I have no further major comments on the manuscript – the authors have improved its quality and readability significantly following the various suggestions of myself and of the other referees. I have thought, however, that it might be appropriate to cite in this context the very recent theoretical work by Kang and Vafek “PRB 102, 035161 (2020)” which shows how non-Abelian braiding could be at the heart of certain strongly-correlated phases of twisted bilayer graphene at the magic angle.

Ref #2

I think the authors have done a great job in answering my questions and resolving my concerns. Although the non-Abelian bulk-edge correspondence is not mathematically proved, it is reasonably argued with convincing evidences. I have no more questions and recommend the acceptance.

Ref #3

I carefully read their response and the revised manuscript. Apparently the authors take significant efforts to implement the Reviewers' comments and suggestion, and address most of my comments in a satisfactory manner. I still have few comments with respect to their replies and revision, I believe it will help the readers to better understand their work if the authors clarify their statements furthermore.

Reply 3.3
When discussing the non-Abelian topological transition (Fig. R11), why there is no direct transition between the topological charges -1 and +1?

Reply 3.6
The authors propose a flow chart to give a proof about non-Abelian bulk edge correspondence, it's very nice to see the connection between the topological transition and charges relation from the perspective of fundamental homotopy group. However, I get confused about the argument 3, when they extend the 1D Hamiltonian in 2D space, why the 2D plane can be analogous to the cutting plane in the 3D space? This statement seems a speculation without a support. Besides, I don't see how using 2D plot to support the Jackiw-Rebbi argument for other topological transitions like i->k.

A technique question related to experimental characterization in the supplementary
If I understand correctly, the authors experimentally choose the subspace \phi_2=\pi/2 by exciting the four nodes inside the meta-atom with the corresponding phase shifts. Since they specifically excite one meta-atom, how do the authors justify the same subspace is selected for other meta-atoms?

Author Rebuttals to First Revision:

We moved the reply to Referee #1 in the final section as it is comparably lengthy.

Referee #2 (Remarks to the Author):

Comment 2.1: I think the authors have done a great job in answering my questions and resolving my concerns. Although the non-Abelian bulk-edge correspondence is not mathematically proved, it is reasonably argued with convincing evidences. I have no more questions and recommend the acceptance.

Reply 2.1: We thank the referee for his/her positive assessments of our work and reply, especially for considering our argument on the non-Abelian bulk-edge correspondence to be evidently convincing.

Referee #3 (Remarks to the Author):

Comment 3.1: I carefully read their response and the revised manuscript. Apparently the authors take significant efforts to implement the Reviewers' comments and suggestion, and address most of my comments in a satisfactory manner. I still have few comments with respect to their replies and revision, I believe it will help the readers to better understand their work if the authors clarify their statements furthermore.

Reply 3.1: We thank the referee for his/her recognitions and positive assessments of our work and reply. We have followed the referee's suggestions to further clarify several related statements as detailed below.

Comment 3.2: Reply 3.3-When discussing the non-Abelian topological transition (Fig. R11), why there is no direct transition between the topological charges -1 and +1?

Reply 3.2: We thank the referee for pointing this out. The graph shown in Fig. R11 (Fig. S10 in supplementary materials) is the "cycle graph" of quaternion group, which shows that each element ($\pm i, \pm j\ or \pm k$) multiplies itself gives -1. As the topological phase transition from charge $+1$ to -1 can only take one of $(\pm i)^2, (\pm j)^2\ and\ (\pm k)^2$ configurations, the direct transition should not be shown in the cycle graph according to the standard definition of a group cycle graph.

In the revised supplementary materials (Sec. IV and page 11), we have properly cited a mathematics book (see below) on the definition of the cycle graph of quaternion group as,

"where the transition from one topological phase to another one can take multiple distinct paths (like the nodes in a network as shown in Fig. S10b, which is the cycle graph of quaternion group[3]).".

Reference:

[3]Shanks, D. Solved and Unsolved Problems in Number Theory. 2nd edn, pp. 85 (Chelsea, 1978).

Comment 3.3: Reply 3.6-The authors propose a flow chart to give a proof about non-Abelian bulk edge correspondence, it's very nice to see the connection between the topological transition and charges relation from the perspective of fundamental homotopy group. However, I get confused about the argument 3, when they extend the 1D Hamiltonian in 2D space, why the 2D plane can be analogous to the cutting plane in the 3D space? This statement seems a speculation without a support. Besides, I don't see how using 2D plot to support the Jackiw-Rebbi argument for other topological transitions like i->k.

Reply 3.3: We thank the referee for recognizing our argument upon non-Abelian bulk edge correspondence.

To see the connection with 3D systems, we note that non-Abelian quaternion was first introduced to classify the disclination line defects in 3D biaxial nematic liquid crystals [Ref. 19: Wu, Q. et al., Science 365, 1273 (2019)]. If we cut a 2D plane through the disclination line, the line defect appears as a point singularity on the 2D cutting plane. The rotation of the principal axis of the biaxial molecule (see Fig. S8 in Ref. 19) around the point singularity gives a visually obvious distinction between various quaternion ($\pm i, \pm j, \pm k, -1\ldots$ etc) charges that can label the loops encircling the singularity.

The non-Abelian topological character of our 1D system is also more apparent when we extend our 1D Hamiltonian to 2D (so that the 1D Hamiltonian is a unit circle in the 2D space). In fact, the eigenvector rotations in the 2D plane (as shown in the bottom planes of the Fig. S15 to S19) are the same as the rotation of biaxial molecules in a 2D plane cutting through a disclination line defect of 3D biaxial nematic liquid crystals.

For the band structure of a PT symmetric system, each degeneracy in 2D k-space can also be promoted into a nodal line in 3D by adding a synthetic dimension [see for example, Fig. 1 in Ref. 23: Bouhon, A. et al., Nature Physics 16, 1137–1143(2020)], and the nodal line is a momentum space counterpart of a disclination line defect in 3D biaxial nematic liquid crystals. Conversely, the cutting plane of nodal lines in the 3D k-space corresponds to 2D degeneracies in the band structure.

It is worth noting "the 2D plane can be analogous to the cutting plane in the 3D space" is really for a better visualization and not a necessary condition in our argument (proof). We added it just for further understanding the extended 2D Hamiltonian (Eq. S5). In the revised supplementary materials (Sec. VI and page 16), we have deleted "the 2D plane can be analogous to the cutting plane in the 3D space" to avoid any misunderstanding caused.

Our argument for the non-Abelian bulk-edge correspondence is divided into three steps (Supplementary materials, Sec. VI):

Step 1. We simply state the well-accepted notion that there must be bandgap closing/re-opening during topological phase transition.

Step 2. For the transition from $Q_L \to Q_R$, we first find $\Delta Q = Q_L/Q_R$ from the homotopy group argument (Fig. S13), where ΔQ encodes the information of bandgap closing during the transition from $Q_L \to Q_R$.

Step 3. The domain-wall state formation between charges $Q_L and\ Q_R$ can be inferred from the edge state formation within the transition $\Delta Q \to +1$ (as ΔQ encodes all topological information within the transition $Q_L \to Q_R$). We then focus on the transition $\Delta Q \to +1$ and Jackiw-Rebbi argument to show the domain-wall states (between charges $Q_L and\ Q_R$), as an example depicted in Fig. S14.

For the domain-wall states between charges $+i$ and $+k$, first we have $\Delta Q = +i/+k = +j$, then we apply Jackiw-Rebbi argument onto the charge transition $+j \to +1$ in a similar manner (as what we have done in Fig. S14).

In the revised supplementary materials (Sec. VI and page 13) we added,

"The domain-wall state formation between charges $Q_L and\ Q_R$ can be inferred from the edge state formation within the transition $\Delta Q \to +1$ (as ΔQ encodes all topological information within the transition $Q_L \to Q_R$)."

Comment 3.4: A technique question related to experimental characterization in the supplementary. If I understand correctly, the authors experimentally choose the subspace

\phi_2=\pi/2 by exciting the four nodes inside the meta-atom with the corresponding phase shifts. Since they specifically excite one meta-atom, how do the authors justify the same subspace is selected for other meta-atoms?

Reply 3.4: We thank the referee for the question. The transmission line system is a linear system which can be block-diagonalized into 4 disjoint sub-spaces, each having 3 modes. Each subspace can be labeled by a pseudo angular momentum (or a fixed phase shift between nodes inside a meta-atom). In the experiment, we could specifically control the phase shift of the four nodes in one meta-atom to selectively excite one particular sub-space. When we excite one meta-atom by imposing the phase shift, the excitation only has projection on the eigenmodes of the sub-space (and is orthogonal to the modes in other sub-spaces) and hence only the three modes in that selected subspace are excited. In other words, each subspace corresponds to a pseudo angular momentum (as mentioned in the main text), and as long as the angular momentum is preserved, other subspaces are not excited.

In the experiment, we checked the results by making the projection of the measured data onto the specified subspace and we indeed found that the projection is nearly 100%, reassuring that the leakage into other subspaces (due to imperfections) is negligibly small.

In the revised supplementary materials (Sec. IX and page 24) we added the above discussion to further clarify our descriptions.

Referee #1 (Remarks to the Author):

Comment 1.1: I have read the resubmitted version of the manuscript by Guo et al., titled "Experimental observation of non-Abelian topological charges and bulk-edge correspondence", and also carefully studied the detailed reply to all the referees' comments.

Although I have read the largely supportive reports of the other two referees, I am still wondering why this particular work merits the high attention of journal Nature. It is admittedly an enthusiastic experience to emulate tight-binding models of novel topological bands in a table-top experiment with hundreds of coaxials, and eventually to observe that the spectra of an actual physical system match the theory. However, from the broader perspective, the authors' results don't push neither the theoretical understanding of the non-Abelian band topology, nor do they put forward novel experimental technique or overcome a major experimental challenge. I don't want to be disrespectful, but one could say the authors manifested a great ability to practically implement matrix Hamiltonians, but what does one really learn? How is an experiment of a scientific value, if it does not really offer a possibility to disprove a theory (in this case the diagonalization of Hermitian matrices)? If the authors failed to find agreement with the theory, the explanation would most certainly be that their emulation of the matrix Hamiltonian was not precise enough.

Therefore, while the experiment looks impressive, and it is the first particular such experiment to implement the non-Abelian band-topological models, my recommendation (which I fear to be overruled by the other two referees) remains to publish these results in a more specialized journal such as [REDACTED].

Irrespective of my critique above, let me comment on the actual experiment and analysis presented in this work, which applies regardless of the journal in which the results would eventually be disseminated.

Reply 1.1: We thank the reviewer think "the experiment looks impressive, and it is the first particular such experiment to implement the non-Abelian band-topological models".

We respectfully disagree with some other comments. In addition to being the first experiment observation of non-Abelian topological charges, we also generalize the bulk-edge correspondence for the non-Abelian topological charges. We note that in Ref. 19 (both main text and supplementary materials), there has been no mention of the "bulk edge correspondence". Our proposal shows that an elegant global relation exists, which not only unifies all the Abelian arguments, but also predicts those that cannot be explained by the Abelian arguments, such as the domain-wall states between charges $+i$ and $-i$.

We also would like to point out that the reviewer's suggestion of the gauge transformation in **Comment 1.7** generates Hamiltonians that violate the periodic boundary condition and hence no band structures can be associated with the Hamiltonians defined by such a transformation pathway. As such, it cannot be used to describe the transition between charges $+i$ and $-i$ without gap closing and re-opening. We will give detailed replies to each of the reviewer's comments as follows.

We thank the reviewer for bringing to our attention some interesting works, in particular the very recent paper by Jian Kang and Oskar Vafekk [PRB 102, 035161 (2020)], which relates non-Abelian braiding and strongly-correlated phases of twisted bilayer graphene at the magic angle.

We also thank the reviewer for all the critical comments to improve our presentation and make our work more rigorous.

Comment 1.2: I find the reply to most referees' comments satisfiable, and I appreciate the authors' efforts with moving parts of the theoretical discussion to the Supplemental Material, significantly shortening the text while improving its clarity, and providing a more detailed information about the further details on the experimental setup. The analysis of experimental data looks solid and the text is well organized. The work is nearly ready for publication.

Reply 1.2: We thank the reviewer for considering our reply to most referees' comments to be satisfactory, and for considering "The analysis of experimental data looks solid and the text is well organized. The work is nearly ready for publication".

Comment 1.3: There is, however, one major point about which I must be very strict. In all fairness, **the authors cannot claim that they propose a non-Abelian bulk-edge correspondence** (as they do in both the abstract and the introduction). While in their reply (and also in the amended version of the Supplemental Material) the authors go to great lengths to the make the appearance of a novel proof of the bulk-edge correspondence, their results cannot be taken as one; in fact the authors hardly prove anything on this question beyond what has already been established by previous works.

Reply 1.3: As we have mentioned in **Reply 1.1**, we generalized for the first time the bulk-edge correspondence for non-Abelian topological charges. Our argument starts from the definition of fundamental homotopy group and shows the relation of $\Delta Q = Q_L/Q_R$ (governing the topological phase transition) which is new and has not appeared in any form in prior literature. Then we show that a certain type of phase transition across a domain wall is always accompanied with band inversions within certain band gaps, and use Jackiw-Rebbi approach to relate the closing/re-opening of the bulk bandgap to the appearance of edge/domain-wall states. Our non-Abelian bulk-edge correspondence indeed predicts something that has never been discussed before, such as the domain-wall states between charges $+i$ and $-i$, which are confirmed by theoretical argument, tight-binding simulation and the experimental result.

Comment 1.4: Before I explain the problems with the "proof", let me emphasize that this deficiency does not in any way spoil the value of the experimental efforts and data presented by this work. However, misleading advertisements and false claims in abstract would have adverse implications on the overall understanding of the authors' results by the community, as well as for the interpretation of any follow-up studies. Therefore, I must insist that a milder formulation, such as "Furthermore, we experimentally observe the manifestation of the non-Abelian topological charges [at domain walls…]" would be far more appropriate in the abstract/intro.

Reply 1.4: We thank the reviewer for suggesting another way to state our findings.

It is a very nice suggestion to state our finding as a manifestation of the domain wall non-Abelian topological charges. We note that Q_L and Q_R are bulk topological quantities, and so if we label ΔQ as a domain wall non-Abelian topological charge (following the referee's opinion), the formula is then relating bulk topological characters (Q_L and Q_R) to a domain wall character ΔQ. We then do not see why this relationship is not a form of bulk-edge correspondence. If the term "edge" and "domain-wall" should not be used interchangeably (in our view, an edge is a domain-wall between a material and vacuum), we will use the "domain-wall" in appropriate places in the manuscript. In addition, for conventional Chern insulators, the literature does not call the difference of the bulk Chern numbers ($\Delta C = C_L - C_R$, which is the Abelian counterpart of our $\Delta Q = Q_L/Q_R$) the "Chern number of the domain-wall". Hence, calling $\Delta Q = Q_L/Q_R$ the non-Abelian topological charges at domain-wall in the abstract may make our paper look a bit unusual, which is the reason why we are reluctant to do so although we have no objection to this viewpoint. That said, we now removed the statement “propose a non-Abelian bulk-edge correspondence”. We now state that “the numerical and experimental results suggest that a non-Abelian bulk-edge correspondence exists”.

Comment 1.5: Let me explain why the authors’ proof and statement on this issue are very misleading. Due to the technicality and the various aspects of the “proof”, I have divided my critique into three parts:

1.)

When focusing on the case of conjugacy classes of “i/j/k”, one is considering topological bands that are readily distinguished by the quantized Berry-Zak phases. The bulk-edge correspondence for non-trivial Berry-Zak phase is well understood: in general, it does not lead to in-gap edge state, because by shifting the energy of the edge state it can disappear inside the bulk energy band. When that happens, the edge develops a quantized (e/2) edge charge of the valence/conduction bands, which is taken as the universal signature of the non-trivial Berry-Zak phase. Equivalently, the filling anomaly is developed, and also taken to be a universal signature of the non-trivial Berry-Zak phase irrespective of the presence/absence of the topological edge mode [see e.g. the discussion in Sec. III of PRB 99, 245151 (2019)].

The signatures of the non-trivial Berry-Zak phase as summarized by the previous paragraph are at present well established. By applying this known correspondence to any selected energy gap (one gap at a time) of multi-gapped models, one readily deduces the completely analogous signatures of the conjugacy classes i/j/k beyond the discussion of the present work.

In a sharp contrast, as the authors’ analysis in the present work focuses too much on band structures close to a topological phase transition (i.e., near a gap closing), cf. Figs. S14-S19, their “proof” completely overlooks the possible absence of the edge mode in generic models with more complicated dispersion. In particular, it should in principle be possible for the topological edge states of the “+/-i” phase to drop all the way from the upper energy gap to the lower energy gap. (This is compatible with the known bulk-boundary correspondence for non-trivial Berry-Zak phase, and is not refuted by the authors’ present analysis.)

Note also that the conjectured bulk-boundary correspondence for these classes does not go beyond the one readily conjectured by Ref. [19].

Reply 1.5:

We first focus on the key critical comment:

“In a sharp contrast, as the authors’ analysis in the present work focuses too much on band structures close to a topological phase transition (i.e., near a gap closing), cf. Figs. S14-S19, their “proof” completely overlooks the possible absence of the edge mode in generic models with more complicated dispersion.”

As long as there is no gap closing/re-opening during the parameter tuning process, the topology of the system remains the same and so it is sufficient to examine the topological phase transition near gap closing/re-opening point. Furthermore, the edge/domain-wall states around gap-closing can explicitly exhibit the topological properties of the corresponding bulk bands. Although they may merge into bulk bands with more complicated dispersions, their topological arguments remain the same as stated above by the referee.

We thank the reviewer for bringing to our notice the interesting reference. We agree with referee that for the $i/j/k$ configurations, the edge state formation can be explained using the Abelian approach (i.e., Berry-Zak phase) when we consider the band gaps one by one. But even for the $i/j/k$ cases, it has an advantage to use the concept of non-Abelian charge to characterize the bulk-edge correspondence, since the non-Abelian charge formula reveals that the edge/domain-wall states in ALL the bandgaps can be labelled with a single element as a whole which respects the rule of element quotient $\Delta Q = Q_L/Q_R$ in the quaternion group. For example, the domain-wall states between charges $+i$ and $+j$ can be globally described by charge $-k$. We also note that our non-Abelian charge formula is entirely consistent with the Abelian Zak phase approach, as already mentioned explicitly in the text and in Fig. 4. In addition, we would like to point out that not all of edge/domain-wall states in non-Abelian systems can be explained using the Abelian approach. For example, the domain-wall states between charges of $+i$ and $-i$ can be explained/predicted by our formula, but not the Abelian approach.

In our argument, we explicitly relate the appearance of edge/domain-wall states with the transition of non-Abelian topological phases across the edge/domain-wall, and also the distribution of the obstructions that have to encounter during the phase transition.

Comment 1.6:

2.)

The topological class “-1” is the only one here that really goes beyond the characterization with Berry- Zak phases. Therefore, here there is a possibility to discover some truly novel aspect of the bulk- boundary correspondence. However, as the authors themselves admit in their reply:

“As the charge −1 is "richer in details" than charges ±i, ±j, ±k, the formation of edge modes is more subtle, we hence say in the original text that "edge states emerging from charge -1 are fickle””

The authors present an argument based on an analogy with the Jackiw-Rebbi model, which suggests that the -1 charge should exhibit a pair of edge modes. Nevertheless, as visible in Fig. S11, this charge is found to be associated with three (rather than two) edge state in the studied models, unless the model parameters are fine-tuned to specific values (namely ones that correspond to a complete decoupling of the third band – cases which I find analyzed below Eq. (69) of Ref.19).

The authors' simplistic analysis has troubles interpreting these features, and it also does not predict any actually robust signature (such as the bulk polarization and filling anomaly for the non-trivial Berry-Zak phase) which would characterize the -1 charge. Once again, there is simply not enough ground for the claim "we propose the non-Abelian bulk-edge correspondence" in the abstract and introduction.

Reply 1.6: We respectfully disagree with the comment.

First, as admitted by the reviewer the charge -1 is the only one that really goes beyond the characterization with Berry-Zak phases. Therefore, the previous Abelian argument fails and so does the associated bulk polarization.

Without any topological phase transition, the edge states of charge -1 can be parametrized and evolve with an extra parameter (i.e. θ_i in Fig. S11). It is obviously a signature that belongs to the same topological phase. In other words, the all possible evolution of edge state globally characterizes the charge -1.

However, the evolution of edge states in charge -1 does not break our non-Abelian bulk-edge correspondence ($Q_L/Q_R = \Delta Q/+1$). The domain-wall states between charge pairs of $(+i, -i)$, $(+j, -j)$ and $(+k, -k)$ may locate in different bandgaps, which is consistent with what we see from edge states at the hard boundaries of a chain with charge -1.

Moreover, we have provided a prediction with extending the Hamiltonian onto a 2D space in the supplementary materials (Sec. VII). Wherein, we have also analysed the variation of edge states during the evolution. As the quadratic-double (linear-triple) degeneracy can split into two single Dirac cones with each supporting one edge state, therefore there are only two topological edge states even away from the mentioned completely decoupling cases. In the flat-band model, the third edge state plays an auxiliary role during the edge state evolution of charge -1.

As mentioned by the reviewer (Comment 1.5), generally, with shifting the energy of the edge state, it may merge into the bulk energy bands. However, the edge/domain-wall states around gap-closing can explicitly exhibit the topological properties of the corresponding bulk bands. We take the following case as an example. Without loss of generality, we select $\theta_z = \frac{\pi}{4}$ (Fig. R1a) and extend the 1D Hamiltonian onto a 2D plane (Eq. S5) as shown in Figs. R1b and c. We then consider two different loops. The one in Fig. R1b encircles the singularity (quadratic double degeneracy) and thus carries charge -1, while the other one in Fig. R1c is trivially contractible. Finally we calculate the domain-wall states between them two and find only two topological states locating on each domain wall as shown in Fig. R1d (there are two domain-walls there and thus four topological states in total). Thus, this example shows that the number of topological edge/domain-wall states can be rigorously determined by the non-Abelian bulk edge correspondence.

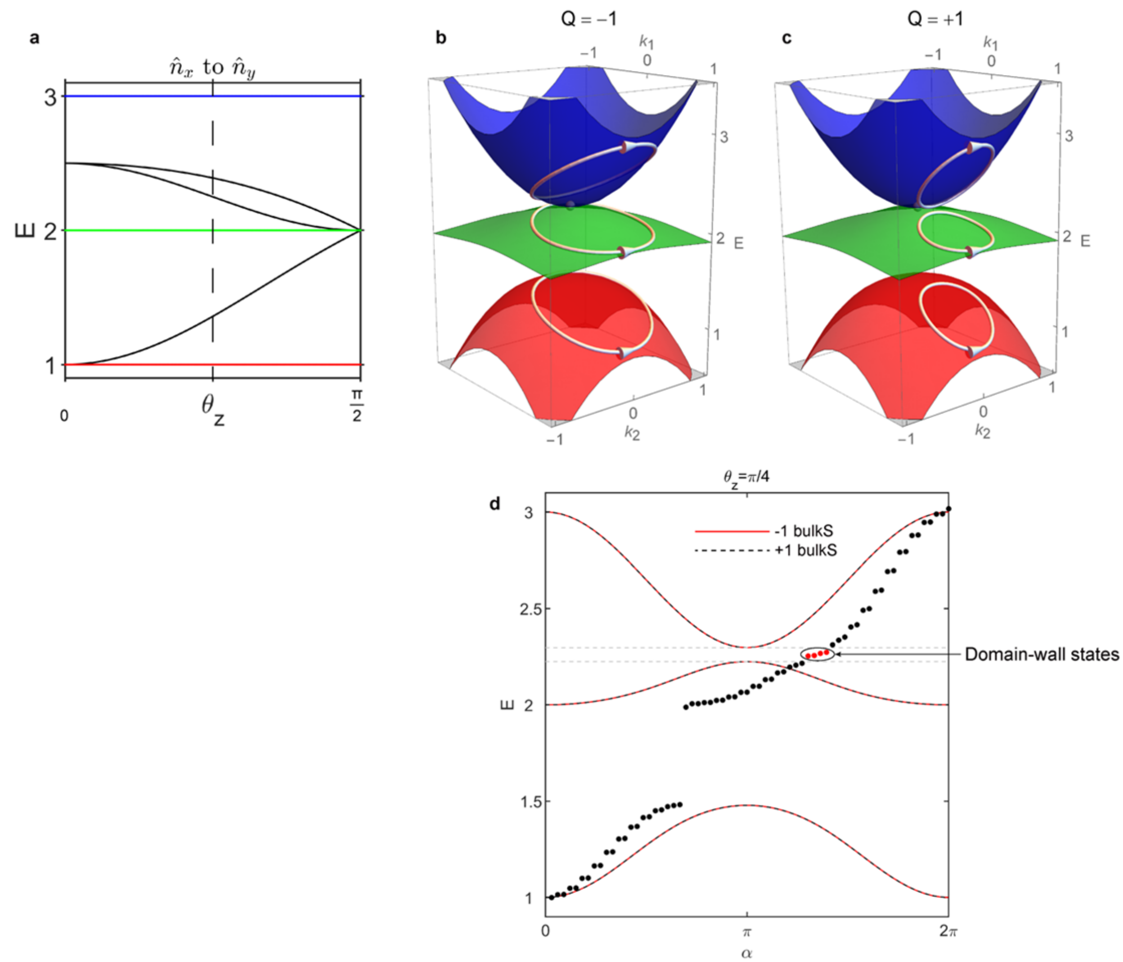

Fig. R1. Only two topological edge states supported by charge -1. **a,** The edge state evolution of flat-band model with charge -1 when $\theta_z = 0 \to \frac{\pi}{2}$ as defined in Fig. S11. **b/c,** At $\theta_z = \frac{\pi}{4}$ as indicated by the vertical dashed line in (a), we extend the 1D Hamiltonian onto a 2D plane (Eq. S5). We then consider two loops, where one (b) encircles the singularity (quadratic-double degeneracy) and thus carries charge -1, while the other (c) is trivial. **d,** On each domain-wall between the two cases (b and c) there are only two topological domain-wall states (In the simulation there are two domain-walls and thus four topological states in total). The circle-loops in (b) and (c) are defined as $(k_1, k_2) = \left(\frac{5}{8}\cos\alpha + \frac{3}{8}, \frac{5}{8}\sin\alpha\right)$ and $\left(\frac{3}{8}\cos\alpha + \frac{5}{8}, \frac{3}{8}\sin\alpha\right)$, respectively. The two sets of bulk bands are overlapped for $\alpha = 0 \to 2\pi$.

It is worth emphasizing that while the Abelian bulk edge correspondence can in general predict the number of edge states as there is only one single bandgap and the topological charge is a number, the non-Abelian bulk-edge correspondence provides a global perspective. One cannot simply move the Abelian arguments onto non-Abelian system without changing the concept. Certainly, the non-Abelian argument is more general and compatible with Abelian argument.

Comment 1.7:

3.)

The authors seem to distinguish between the bulk-edge (two different band structures meeting at a domain wall?) and the bulk-boundary (vacuum on the other side of the edge?). I didn't find the distinction explicitly clarified in the text, and it thus took me a while to decipher in their reply the intended meaning of "Furthermore, we also propose non-Abelian bulk-edge correspondence for the first time, and as the reviewer stated, our work directly generalizes "the previously introduced bulk-boundary correspondence for the non-Abelian topological charge"". Perhaps "bulk-domain wall" would be a less confusing formulation.

In either case, there is a subtle but important issue here that closely relates to one of my earlier questions.

Namely, in my previous reply, I have asked how does one distinguish phases "+i" and "-i", which are related to each other by a gauge transformation. I respectfully disagree with the authors statement that "In the non-Abelian topological phases one cannot continuously transform charge +i to −i without gap closing" and with the paragraphs that follow this statement.

To see the problem, consider the explicit (flat-band) model encoded with the SO(3) eigenframe

$$R(k) = \begin{pmatrix} \cos\frac{k}{2} & \sin\frac{k}{2} & 0 \\ -\sin\frac{k}{2} & \cos\frac{k}{2} & 0 \\ 0 & 0 & 1 \end{pmatrix}$$

which the authors take to corresponds to charge "+i". By flipping in this expression the sign k → -k, one obtains the inverse matrix which encodes a Hamiltonian with charge "-i".

Now, imagine a system with a domain wall, and a band structure initially characterized by the R(k) given above on both sides of the domain wall (thus, initially, the domain wall is not "really" there as the system remains translationally invariant). Then, as a function of a tuning parameter p, one changes the flat-band Hamiltonian on the right side of the domain wall to

$$Q(k;p)=\begin{pmatrix}1 & 0 & 0\\ 0 & \cos p & \sin p\\ 0 & -\sin p & \cos p\end{pmatrix}\begin{pmatrix}\cos\frac{k}{2} & \sin\frac{k}{2} & 0\\ -\sin\frac{k}{2} & \cos\frac{k}{2} & 0\\ 0 & 0 & 1\end{pmatrix}\begin{pmatrix}1 & 0 & 0\\ 0 & \cos p & -\sin p\\ 0 & +\sin p & \cos p\end{pmatrix}$$

while keeping the system on the left side of the domain wall unchanged. This is a continuous deformation that keeps the energy bands on both sides flat (and both energy gaps open), and which interpolates between what the authors would call "charge +i" at p=0 and "charge -i" at p=Pi [note that Q(k,Pi) = R(-k)]. The authors claim to investigate the domain wall between band structures with charges "+i" and "-i" (which is found to resemble end modes of charge "-1"). However, as the above example explicitly illustrates, this domain wall is continuously removable without closing the bulk energy gaps and while keeping the PT symmetry of each band structure.

Reply 1.7: We thank the reviewer for suggesting an interesting transformation that can transform between different quaternion elements ($+i$ and $-i$) in the same conjugacy class without gap-closing and re-opening. However, we notice that the proposed $Q(k;p)$ matrix is not applicable to crystalline systems because it generally leads to a Hamiltonian that is not periodic in the momentum k (except for a few special values of p at 0, π/2, and π), with details shown below. In addition, the rotation matrix $R(k)$ given above corresponds to charges $\pm k$ rather than $\pm i$.

If we construct the Hamiltonian following the reviewer's transformation, and at each k and p,

$$H(k;p)=Q(k;p)I_{1,2,3}Q^T(k;p)$$

where $I_{1,2,3}=diag(1,2,3)$, we note that $H(k;p)$ contains explicitly $\frac{k}{2}$ term which cannot satisfy the periodic boundary condition. For example, $H\left(k=-\pi,p=\frac{\pi}{4}\right)\neq H\left(k=\pi,p=\frac{\pi}{4}\right)$ with,

$$H\left(k=-\pi,p=\frac{\pi}{4}\right)=\begin{pmatrix}\frac{5}{2} & \frac{1}{2\sqrt{2}} & \frac{1}{2\sqrt{2}}\\ \frac{1}{2\sqrt{2}} & \frac{7}{4} & \frac{3}{4}\\ \frac{1}{2\sqrt{2}} & \frac{3}{4} & \frac{7}{4}\end{pmatrix}$$

$$H\left(k=\pi,p=\frac{\pi}{4}\right)=\begin{pmatrix}\frac{5}{2} & -\frac{1}{2\sqrt{2}} & -\frac{1}{2\sqrt{2}}\\ -\frac{1}{2\sqrt{2}} & \frac{7}{4} & \frac{3}{4}\\ -\frac{1}{2\sqrt{2}} & \frac{3}{4} & \frac{7}{4}\end{pmatrix}$$

The Hamiltonians at a general value of p does not have a band structure. And hence the above rotation cannot be used to realize the transformation between two elements in one conjugacy class.

In order to address the referee's concern, we can define transformations that can satisfy periodic boundary conditions by considering the rotation matrix such as, $\tilde{Q}(k;p) = P(p)R(k)$ with,

$$P(p) = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \cos p & \sin p \\ 0 & -\sin p & \cos p \end{pmatrix}$$

and construct the Hamiltonian for a general value of p as,

$$\widetilde{H}(k;p) = \tilde{Q}(k;p) I_{1,2,3} \tilde{Q}^T(k;p)$$

then the Hamiltonian is periodic for any value of $p \in [0, \pi]$. And at $p = 0$, $\widetilde{H}(k;p)$ has charge $-k$ while at $p = \pi$, $\widetilde{H}(k;p)$ has charge $+k$. In Fig. R2 we show the three eigenvectors calculated as $\widetilde{H}(k;p)|V_n\rangle = n|V_n\rangle$ with $n = 1,2,3$.

However, during the mapping (p: $0 \rightarrow \pi$) there exists no common basepoint. Basepoint is the point that the Hamiltonian keeps invariant when p takes different values, i.e., $\widetilde{H}(k_0, p = 0) = \widetilde{H}(k_0, p \in [0, \pi])$. For two loops to be homotopic they must share the same basepoint. Without the basepoint, even the fundamental homotopy group cannot be defined because there is no loop product (product of the fundamental homotopy group). Going back to our question, two conjugate elements in the same class cannot be continuously transformed into each other when there is a fixed basepoint.

We should emphasize that for all the cases of $+i/-i$ investigated in the paper, the two sides share a fixed basepoint (otherwise the + and – cannot be defined in the first place), so the domain-wall charge is always well defined in the paper. It is also worth noting that between two different classes, the basepoint can always be found. In general, the basepoint is implied when discussing homotopy group.

It is useful to note that all of the braiding features proposed in Ref. 19 depend on the presence of a basepoint; without the basepoint, all of related arguments would fail. Actually, the supplementary materials of Ref. 19 (although not in the main text) repeatedly mentioned basepoint.

We thank the reviewer's nice suggestion on the phrase of "bulk-domain wall". In the revised main text (on page 8) we have added following sentence to make the phrase clear,

"The term "bulk-edge" can be generalized to be "bulk-domain-wall" when considering both two charges $Q_L \neq 1$ and $Q_R \neq 1$ (instead, an edge is a domain-wall between a material and vacuum)."

We also stressed basepoint in the revised main text (on page 4) as following, "It is worth emphasizing that two conjugate elements (i.e. $\pm i$) can only be well-defined when they share a common basepoint[19]".

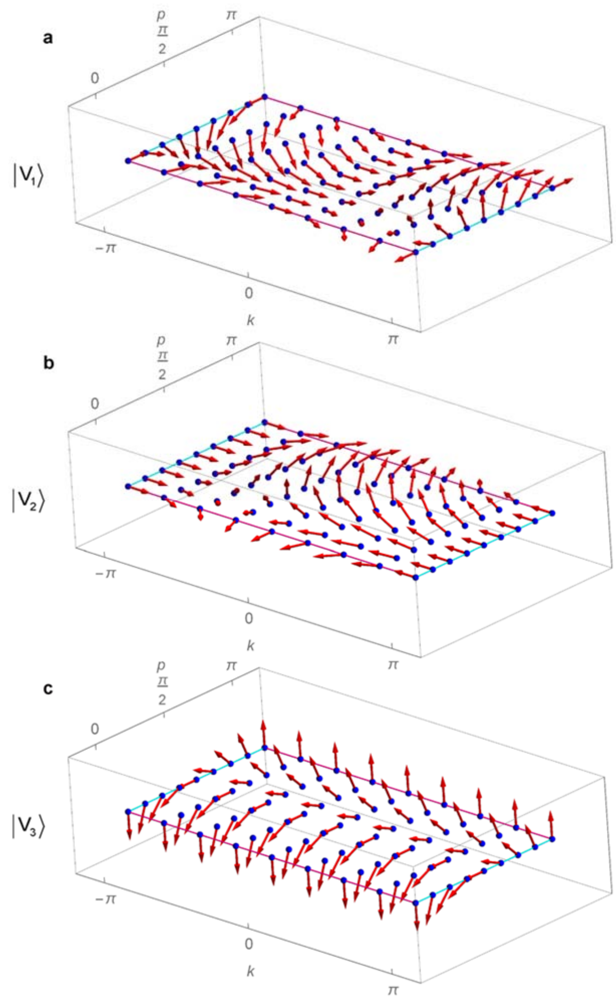

Fig. R2. Eigenvectors of $\widetilde{H}(k;p)$ for $k \in [-\pi, \pi]$ and $p \in [0, \pi]$. During the mapping $(p: 0 \to \pi)$ there is no common basepoint. Basepoint is the point that the Hamiltonian keeps identical when p taking different values, i.e., $\widetilde{H}(k_0, p = 0) = \widetilde{H}(k_0, p \in [0, \pi])$. The three eigenvectors are calculated as $\widetilde{H}(k;p)|V_n\rangle = n|V_n\rangle$ with $n = 1,2,3$.

Comment 1.8: [An analogous issue stemming from the related non-Abelian properties of the Euler class in 2D PT- symmetric systems is discussed in Appendix D of PRB 102, 115135 (2020).]

Reply 1.8: The Hamiltonian order-parameter space discussed in Appendix D of PRB 102, 115135 (2020) is $\mathbb{R}P^2$ (as explicitly shown in the section title: "APPENDIX D: EULER CLASS REVERSAL IN $\mathbb{R}P^2$"). The corresponding fundamental homotopy group $\pi_1\left(\mathbb{R}P^2 = \frac{O(3)}{O(1)\times O(2)}\right) = \mathbb{Z}_2$, which is an Abelian group and has two elements 0 and $\pm\pi$. It is well known that $+\pi$ and $-\pi$ are the same phases in the Abelian description and thus can be continuously transformed.

However, our order space is $M_3 = \frac{O(3)}{O(1)\times O(1)\times O(1)}$, which is different from $\mathbb{R}P^2$, and the fundamental homotopy group is $\pi_1(M_3) = \mathbb{Q}$, the non-Abelian quaternion charge.

Comment 1.9: Therefore, when comparing two different band structures, one can only tell whether they are in the same or different conjugacy class. In contrast, the "absolute" charge difference $\Delta Q = Q_L/Q_R$ (the quantity purported to govern the "generalized bulk-edge correspondence") seems to be meaningful only if the two models are relatable through a small (gap-closing) tuning of parameters, but not in general.

Reply 1.9: We respectfully disagree with the comment. We reiterate that the non-Abelian charges are characterized using based homotopy, not free homotopy. Our formula refers to quaternion group elements defined with respect to a basepoint (based homotopy), not conjugacy classes.

As already mentioned in **Reply 1.5,** the proposed formula $\Delta Q = Q_L/Q_R$ is good for describing the non-Abelian bulk-edge correspondence is general. If the reviewer agrees that the formula "seems to be meaningful only if the two models are relatable through a small (gap-closing) tuning of parameters", then continuous topological phase transitions without any gap closing will make it general.

Comment 1.10: [In particular, the explicit models considered by the authors' simplistic analysis all produce the same matrix H(k=0). This choice misguides one to think that a canonical gauge/base point can always be found that leads to a unique element of the quaternion group, but it really is just a consequence of the overly simple form of the models considered by the authors.]

Reply 1.10: As explained in our **Reply 1.7**, basepoint is the basic condition to define the fundamental homotopy group. Otherwise, the conjugate elements (i.e. $\pm i, \pm j$ or $\pm k$) cannot be defined. Our simple construction automatically satisfies the condition. In addition, basepoint is a topologically necessary condition, not a special "consequence" of our simple model.

Comment 1.11: These simple observations thus further undermine the authors' claim that their work "generalizes "the previously introduced bulk-boundary correspondence for the non-Abelian topological charge"". I don't find in this work any new and rigorous theoretical analysis concerning the non-Abelian bulk-boundary correspondence.

Reply 1.11: We respectfully disagree with the comment.

As replied above, the non-Abelian bulk-edge correspondence is for the first time proposed by us. And it has been verified by theoretical arguments, tight-binding simulations and practical experiments. Our theoretical argument starting from homotopy theory is rigorous and general.

Comment 1.12: For all these reasons, I have to urge the authors to relax their statement in the abstract and in the introduction (as well as at the appropriate places in the main text) by the weaker, but more appropriate, formulation such as the one I have explicitly suggested in blue earlier in my report. I cannot recommend this work for publication unless this serious issue is properly resolved by the authors.

Reply 1.12: As we mentioned in **Reply 1.4**, for the first time we proposed the non-Abelian bulk-edge correspondence, which is not only compatible with the Abelian argument, but also implies domain-wall states that cannot be explained by Abelian topological phases, i.e., domain-wall states between charges $+i$ and $-i$. Upon the insistence of the referee, we do remove the statement "propose a non-Abelian bulk-edge correspondence". We now state that "the numerical and experimental results suggest that a non-Abelian bulk-edge correspondence exists".

Comment 1.13: I have no further major comments on the manuscript – the authors have improved its quality and readability significantly following the various suggestions of myself and of the other referees. I have thought, however, that it might be appropriate to cite in this context the very recent theoretical work by Kang and Vafek "PRB 102, 035161 (2020)" which shows how non-Abelian braiding could be at the heart of certain strongly-correlated phases of twisted bilayer graphene at the magic angle.

Reply 1.13: We thank the reviewer for all above comments and suggestions.

We thank the reviewer for bringing us the interesting paper, in the revised main text we have properly cited it (on page 2 as Ref. 24).

Reviewer Reports on the Second Revision:

Ref #1

I hope this is the last round of the reviewing process, as the communication with the authors is becoming growingly tiresome. The whole experience can be summed up with the old joke about there being "some new and correct things" (but what is new is not correct, and what is correct is not new...)

But to give this whole report a constructive spin:

As explained in my report, I condition the acceptance of the manuscript by the authors dropping the phrase "bulk-edge correspondence" from the title as well as generally from the manuscript text. If this is thoroughly implemented, I will go along with the other two referees and recommend acceptance to Nature.

To explain the problem with this phrase: The first part of their theoretical considerations concerning the hard boundary is not new, but closely follows Sec. V.C of Ref.[19] -- I have now added to my report references to concrete equations and figures such that the equivalence of their analysis with this older work is completely transparent.

The second part of their theoretical considerations concerns domain walls between two topological insulators. As the authors admitted in their reply, their formula is meaningful only if the two insulators share a base point. In practice this means that the two Hamiltonians coincide at momentum k=0. But that is an unnatural condition of a very limited applicability.
(For another jovial comparison -- imagine that someone claims to prove the Riemann hypothesis, but then adds that "only by checking the first billion roots". That is not a good type of mathematical proof. Similarly, the authors' assumption on coinciding Hamiltonians at k=0 is unnatural in the context of topological band theory.)

In contrast, the experimental data obtained by the authors are interesting and can be disseminated in journal Nature. The limited heuristics with ΔQ that the authors develop appears sufficient for the particular simple insulators emulated in their experiment, and can be kept as such. So the bulk of the manuscript can remain unchanged. However, for reasons as outlined (and detailed in the report), this heuristic method cannot be called bulk-edge correspondence.

Ref #2

N/A

Ref #3

The authors successfully address all my comments. In addition, I'm happy to see their statement about the "proof of non-Abelian bulk-edge correspondence" is modified appropriately throughout the manuscript with respect to the criticism from other Reviewers. Therefore, I recommend its publication on Nature.

Author Rebuttals to Second Revision:

Referee #3 (Remarks to the Author):

Comment 3.1: The authors successfully address all my comments. In addition, I'm happy to see their statement about the "proof of non-Abelian bulk-edge correspondence" is modified appropriately throughout the manuscript with respect to the criticism from other Reviewers. Therefore, I recommend its publication on Nature.

Reply 3.1: We thank the referee for his/her positive assessments and for the recommendation of our work to publish on Nature.

Referee #1 (Remarks to the Author):

Comment 1.1: I thank the authors for their detailed reply to my report.

It is clear from the other referee reports that there seems to be enough interest in publishing these experimental data in some form. At this stage I am not going to further argue whether the very high-profile journal Nature is an appropriate platform to disseminate these results. (With all respect, my opinion on this did not change since my previous communication.)

More importantly, (1) I still don't see any substantial theoretical or mathematical result in this work on top of those already presented by Ref. [19], and (2) I find several claims concerning the purported generalization of bulk-edge correspondence too misleading. In this spirit, the comments appended below are organized into two parts following, addressing in sequence the issues (1) and (2).

At the end of the report I try to be constructive and propose some concrete changes that should be implemented to make this work appropriate for publication.

Reply 1.1: We thank the referee for the constructive and concrete suggestions. In the revised manuscripts, we have accepted them all.

Comment 1.2: (1) The novelty compared to Ref. [19].

The authors write in their reply "we also generalize the bulk-edge correspondence for the non-Abelian topological charges. We note that in Ref. 19 (both main text and supplementary materials), there has been no mention of the "bulk edge correspondence"." I find this statement unreasonable.

While it is true that the concrete formulation "bulk-edge correspondence" is not used by the cited work, that precise discussion is contained in Sec. V.C of its supplemental material, titled "Topological edge modes in 1D systems". The cited work presents elementary flat-band Hamiltonians in Eqs. (66—70) which represent the 8 possible elements of the quaternion group. Their corresponding edge states are plotted in Fig. S-16 and S-17 therein.

I remark that the Hamiltonians defined through R(k) on line 90 resp. 106 of the work by Guo et al. are exactly the flat-band Hamiltonians in Eq. (67) resp. (69) from the work by Wu et al. (with an unimportant shift of momentum by pi).

The present work adopts these flat-band Hamiltonians, and explicitly expands the matrices (with coefficients of expansion tabulated in Table S1). In the main text the authors "choose parameters (Tab. S1) that can mimic the flat-band cases", but this translates to simply adopting the models of Ref. [19]. Thus the analysis of "hard boundaries" in Fig. 2, which is used by the present authors to motivate or illustrate the bulk-boundary correspondence, does really not convey any novel information beyond the "topological edge modes" analysis of these same models in Figs. S16-17 from Ref. [19].

[For the experimental realization the authors opt to implement a different version of the 1D Hamiltonians. I didn't find the computation of topological invariants of these models in neither the main text nor the supplement. – See comment in square brackets further below.]

In the second part of their work, the authors claim to generalize the result of Ref. [19] from "hard edges" to "domain walls". While the presented experiment is valuable, interesting and should be published in some form, I argue below that the presented theoretical analysis is mathematically problematic and cannot be claimed as a new mathematical/theoretical result.

Reply 1.2: (Note: Ref. [19] has been re-ordered to be Ref. [16] during the revision)

Our Hamiltonians are indeed similar to those in Ref. [19], and it is purposely chosen so because this class of models is much easier to comprehend and appreciate the non-Abelian topological properties. As the first experimental realization of the non-Abelian topological charges, it is important to choose systems that are experimentally feasible to characterize and the models in Ref. [19] have all these nice properties.

We note that the authors of Ref. [19] analyzed edge states using the usual Zak phase argument, which is an Abelian approach. Our work (using $\Delta Q = Q_L/Q_R$) shows a non-Abelian way, which had not been mentioned in Ref. [19].

Thus, the main novelty compared with Ref. [19] includes: 1. First time experimental characterization of non-Abelian topological charges and edge/domain-wall states; 2. Analysis of edge/domain-wall states from a non-Abelian perspective.

The computation of the non-Abelian topological invariants has been added in the revised Supplementary Information (Section I, page 6). Furthermore, the corresponding codes will be uploaded onto a public data repository.

Comment 1.3: (2) validity and practical value of the proposed bulk-to-domain-wall correspondence.

The authors write in the paper "The well-known Abelian bulk-edge correspondence, given by the difference of two integers (Fig. 4a, $\Delta N = N_L - N_R \in \mathbb{Z}$) becomes inadequate for describing the global edge state configuration in non-Abelian topological systems." The same could be said about Z_2-classified topological insulators, where one has to "generalize" the displayed formula by taking the difference mod 2. Generally, one needs to combine group elements following the group composition law – which in the case of the quaternion group is multiplication. Therefore the authors do nothing particularly special when, as they write, they "propose a non-Abelian bulk-edge correspondence [..] which states that the topology of the domain-wall is characterized by the relation of $\Delta Q = Q_L/Q_R$."

To restate from my previous report, the problem is that this naive "generalization" does not work, because the 1D phases are not characterized by a unique element of the quaternion group but by a conjugacy class of elements. Put plainly, one cannot distinguish +i from -i; they together belong to topological phase {±i}. Per the theory proposed by the referees, the domain wall is characterized by ΔQ ={±i}/{±i} = +1 or -1, i.e. the "trivial" (+1) domain-wall configuration can be continuously evolved into the "non-trivial" (-1) configuration. The quantity that the authors propose to describe the domain wall states is mathematically not well defined.

As for the continuous deformation from +i to -i, I sincerely apologize for the typo in my previous report, where I wrote one matrix too much in the relevant formula. The authors correctly "notice that the proposed $Q(k;p)$ matrix is not applicable to crystalline systems because it generally leads to a Hamiltonian that is not periodic in the momentum k", and properly undo my mistake by considering matrix $\tilde{Q}(k;p) = P(p)R(k)$. The authors then confirm my statement that the sign of the invariants i/j/k can be flipped by such continuous deformations of the Hamiltonian.

The way the authors resolve the problem is by adding to the introductory part of the manuscript "It is worth emphasizing that two conjugate elements (i.e. ±i) can only be well-defined when they share a common basepoint."

While adopting this simplifying assumption appears sufficient for the analysis of the particular models that the authors implement in their experimental setup (and I would like to urge the authors to restate this assumption in the context of Fig. 4), such an assumption is simply not true for a generic model in this symmetry class. (The authors effectively limit their attention to Hamiltonians that all coincide at momentum k=0, which is a very artificial assumption.) Since the notion of bulk-boundary correspondence is reserved for topologically robust features – ones that are invariant unless there is a topological phase transition – the characterization by the mathematically ambiguous quantity $\Delta Q = Q_L/Q_R$ cannot be designated as such.

Reply 1.3: With a fixed basepoint, charge +i cannot be continuously changed to be -i. We think the referee agrees with us on this issue. This is a necessary condition for defining all the non-Abelian topological charges in a quaternion group (c.f. Ref. [16]). Thus, the relation of $\Delta Q = Q_L/Q_R$ always works with the condition there is a fixed basepoint. In the revised manuscript, we have restated this in the context of Fig. 4 (on page 7).

All of the (non-interaction) topological classifications originate from homotopy group. For Abelian topological charges, each element constitutes a class by itself. Arbitrary two elements cannot share a common conjugate class. Therefore, when identifying the homotopy element for a Hamiltonian, fixing a basepoint is not a necessary condition. In other words, based homotopy and free homotopy will give the same topological classification for Abelian cases.

However, the basepoint is crucial for non-Abelian topological charges, where two mutually conjugate elements belong to the same class. The basepoint serves to distinguish them from each other. Otherwise, the non-Abelian topological charges cannot construct a group.

Different from the Abelian cases, the based homotopy and free homotopy lead to two different ways to classify the non-Abelian topological phases, both of which we think are interesting and deserve in-depth studies.

Comment 1.4: [Concerning the sign ambiguity, the sign also crucially depends on the choice of gauge beyond just the existence of the base point. For example, in Fig.1(b) the authors characterize phase +i (-i) as encoding a rotation of green-and-blue eigenstates by pi in the positive (negative) direction around the red eigenstate. But eigenstates have an overall sign ambiguity (the gauge degree of freedom) – by inverting the orientation of the red eigenstates for an initial point k in BZ, the rotation in positive direction becomes a rotation in the negative direction, and vice versa (thus flipping the sign of +/-i). If the authors worry about flipping the handedness of the frame, one could flip the orientation of both red and blue eigenstates simultaneously, which also changes the direction of the rotation while preserving the handedness. To remove this ambiguity, the authors should comment on the choice of the gauge for the models analyzed in Fig.4.]

Reply 1.4: We thank the referee for raising this interesting question.

Given a common basepoint, one cannot continuously change charge +i to be -i by applying different gauges.

The order-parameter space of Hamiltonian of the three-band model can be written as $M_3 = \frac{O(3)}{O(1)^3} \cong \frac{SO(3)}{D_2}$, where the first equality is obtained by an $O(3)$ rotation of the eigenstate frame. As flipping the sign of each eigenstates $|u_k^n\rangle \to -|u_k^n\rangle$ $(n = 1,2,3)$ leaves the Hamiltonian $H(k) = \sum_{n=1}^{N} \lambda_n |u_k^n\rangle\langle u_k^n|$ invariant, one then imposes the $O(1)^3$ quotient, where $O(1)^3 \cong D_{2h} \cong \mathbb{Z}_2^3$ is generated by three mutually perpendicular mirror symmetries. In the second equality, both groups have been replaced by their proper subgroups, i.e., $O(3) \to SO(3)$ and $D_{2h} \to D_2$, where the dihedral point group D_2 consists of the identity and three π rotations around three mutually perpendicular axes.

Note that when $M_3 = \frac{O(3)}{O(1)^3}$, one can operate a $O(3)$ rotation on the eigenstate frame, then the $O(1)^3$ quotient means that flipping the sign of each eigenstate $|u_k^n\rangle \to -|u_k^n\rangle$ $(n = 1,2,3)$ leaves the Hamiltonian invariant. The eigenstate frame does not have handedness.

On the other hand, when $M_3 = \frac{SO(3)}{D_2}$, one can operates a $SO(3)$ rotation operator on the eigenstate frame, then D_2 quotient means that rotating the eigenstate frame with π around one axis leaves the Hamiltonian invariant. Now the eigenstate frame does have handedness (either right or left). And since D_2 rotation is proper, it leaves the handedness of frame intact.

The two different order-parameter spaces are isomorphic. In the calculation of non-Abelian topological charges we used the later one (c.f. Ref. [16]), which conveniently enables the lifting of $SO(3)$ onto its double cover $Spin(3) \cong SU(2)$ (see details in the Supplementary Information Section I). In Fig. 1b we plot the eigenstate frame with right-handedness [i.e. $(|1\rangle \times |2\rangle) \cdot |3\rangle > 0$]. The eigenstate frame with further D_2 gauge freedom keeps the handedness unchanged as mentioned above. The other D_2 gauge choices are given in Fig. S1a.

The charges +i and -i depend on the sign convention, just like the left/right circular polarizations of light depends on whether we look into or away from the source. However, as long as we have agreed on what is +i, it is impossible to continuously change it to -i under a gauge transformation. No matter how one applies the gauge of D_2, the chirality of rotation of eigenstate frame from $k = 0$ to $k = 2\pi$ will keep invariant. For example, we consider the gauge transformation proposed by the referee. If one flips the orientations of both red and blue eigenstates simultaneously, the right handedness of the eigenstate frame still preserves (see Fig. R1b) [Note that the red-green-blue vectors form a right-handed triad in both the left and right panels]. The sense of rotation remains unchanged as well (see Fig. R1b). In other words, both cases correspond to the rotation of $\exp(\theta L_x)$ with $\theta = 0 \rightarrow \pi$.

In summary, the non-Abelian topological charges cannot be changed with applying different gauges provided that a common basepoint is fixed.

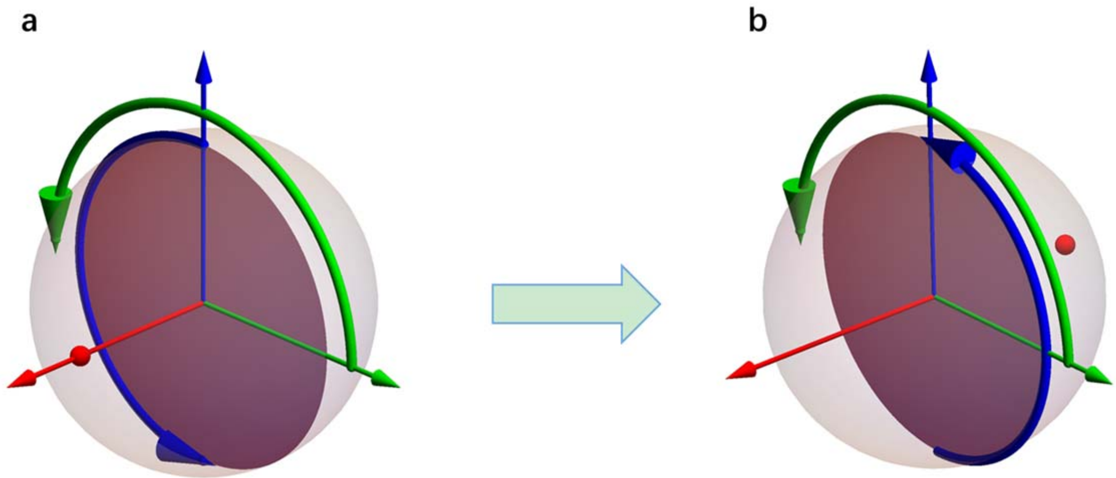

Fig. R1. (a) The copy of Fig. 1b. (b) One flips the orientations of both red and blue eigenstates simultaneously. The right handedness of the eigenstate frame still preserves. Obviously, the rotation direction of eigenstate frame for $k = 0 \rightarrow 2\pi$ remains unchanged as well. Both (a) and (b) correspond to charge +i.

Comment 1.5:

Given the mature stage of the refereeing process, let me try to wrap up with a few suggestions how to bring the manuscript into a form not containing misleading statements that could be misinterpreted by the community. Only if these suggestions are thoroughly implemented I can consider recommending this work for publication in Nature.

First, the authors should altogether avoid using the phrase “bulk-<<something>> correspondence” (whether with “edge”, or “boundary”, or “domain wall”, etc.) for the presented discussion, because such wording suggest a robust one-to-one relation between topological phases and edge states. The authors do not find such a relation; and the stipulated assumption “which only holds for models that share a common base point” is plainly unnatural in the context of topological phases. Furthermore, as it is possible that an actual and robust one-to-one correspondence will be discovered for these topological phases in the future, the terminology of the current work just contributes to confusion.

I thus suggest the authors explicitly reveal the non-uniqueness of the topological invariant in the main text, pointing out that a domain-wall can be characterized by ΔQ in the presented models only thanks to the existence of a joint base point – the existence of which is not guaranteed in general. Then it is appropriate to discuss the role of ΔQ in the interpretation of the experimental data. However, instead of speaking about “correspondence” between ΔQ and edge/domain-wall states, weaker formulation should be found; for example, that “the value of quotient $\Delta Q = 1$ suggests/indicates such and such edge modes”. For consistency, make sure the modified terminology is implemented both in the main text and in the supplement.

Let me add that multiple researchers in the community might have wrong intuition about the non-Abelian invariant, thus I believe there is an inherent value if the authors explicitly add to the main text a comment along the discussed lines, i.e. why the phrase “bulk-edge correspondence” should be avoided in the context. They may also comment in the conclusion that a robust bulk-edge correspondence which remains meaningful in the absence of base point remains an open question for the future theoretical work. This all would help misinterpretation of the authors’ results, and alert others to try to find one.

Finally, the proposed changes in terminology culminate in a problem with the manuscript title, in which the phrase “bulk-edge correspondence” is featured prominently. I have to urge the authors to propose a more appropriate title where the word “correspondence” is dropped. One straightforward option that comes to my mind would be “Experimental observation of edge/boundary states/modes in non-Abelian topological insulators”. Of course, the authors have the freedom to come with other ideas.

I apologize if the authors find my requests too demanding, but as they thrive to have their results published in journal Nature which receives an especially wide attention, I find it particularly important to have their results reported scientifically accurately. If my comments are thoroughly implemented, I will find no reason to further extend the refereeing process and the work would be suitable for publication.

Reply 1.5: We thank the referee for these suggestions. We have made the following revisions.

1. In the revised manuscripts (including both the main text and Supplementary Information), we have completely avoided using the phrase of “non-Abelian bulk-<<something>> correspondence”.
2. When discussing the role of ΔQ in the interpretation of the experimental data, we added the following sentence on page 7,
   “Here we would like to emphasize that a domain-wall can be characterized by ΔQ in the presented models only thanks to the existence of a joint basepoint.”

3. We have adopted the referee's suggestion and used the sentence "the topology of the domain-wall is indicated by ΔQ ..." on page 7.
4. We have also accepted the referee's suggestion and added the following sentences in the discussion section,
   "In this work we avoid using the phrase of "non-Abelian bulk-edge correspondence" because the non-Abelian quotient relation $\Delta Q=Q_L/Q_R$ inherently depends on the existence of a basepoint. A robust bulk-edge correspondence which is meaningful even in the absence of basepoint remains an open question for the future investigations, such as only considering the conjugacy classes in the fundamental homotopy group or equivalently free homotopy classes of free loops."
5. We have changed the title to be "Experimental observation of non-Abelian topological charges and edge states".