Responses to Referee Dániel Varjas:

We thank Dr. Dániel Varjas for carefully reading our manuscript and judging our paper as "clearly written, good presentation", "the results are sound", "discusses the timely problem of amorphous topological phases, and more broadly, topological phases protected by non-crystalline spatial symmetries" and "novel use of "momentum-space" invariants derived from twisted periodic boundary conditions with more k components than spatial dimensions.". The referee's main concerns are "the novelty of the results compared to the earlier works on quasicrystals" and "the relevance of the results to physical amorphous materials". In the following response, we have addressed these concerns very carefully and made changes as requested.

Strengths:
1) Clearly written, good presentation.
2) Discusses the timely problem of amorphous topological phases, and more broadly, topological phases protected by non-crystalline spatial symmetries.
3) Novel use of " momentum-space" invariants derived from twisted periodic boundary conditions with more k components than spatial dimensions.

Reply: We thank the referee for the positive comments of our work.

Weaknesses 1: My main concern is the novelty of the results compared to the earlier works on quasicrystals (refs 28 and 29), and the relevance of the results to physical amorphous materials. The p-fold rotational symmetries (particularly p=8, 10, 12) arise naturally in quasicrystals, but seem artificial in an amorphous system. Amorphous materials are typically isotropic, possessing continuous rotation symmetry on average, lacking any preferred directions. ln my view, the systems studied here are better described as highly disordered quasicrystals - or perhaps an amorphous layer on a quasicrystal substrate - inheriting the discrete rotational symmetry in the hopping Hamiltonian, but the atomic positions completely disordered. Do the authors agree with this view? Can the authors suggest other physical setups that these models can describe? I believe that these considerations should be discussed at some point of the manuscript to make the context of the research clear to the reader, and motivate the relevance of the results to physically realizable amorphous materials.

Reply: We thank the referee for the insightful comment. Below are our detailed responses to the referee's concerns.

For the novelty of the results compared to the earlier works on quasicrystals (Refs. [30,31], previous Refs. [28,29]), we acknowledge that we drew inspiration from quasicrystals, especially the excellent work Ref. [30] in the definition of the $\mathbb{Z}_2$ topological invariant. Nevertheless, our work is far from a straightforward generalization of the quasicrystal case in both the nature of amorphous matter and the construction of *two* topological invariants.

Previous works show the existence of higher-order topological phase supporting eight corner modes in quasicrystals. The phase is protected by the eight-fold rotational symmetry. A $\mathbb{Z}_2$

topological invariant is defined based on the C_8M symmetry. In addition, prior to our paper, it was still unclear whether the quadrupole moment could be extended to characterize the higher-order topology in quasicrystals without crystalline counterparts. As the referee must know, amorphous systems are short of the *exact* rotational symmetry protecting the higher-order phases in quasicrystals so that one cannot define the $\mathbb{Z}_2$ topological invariant. Therefore, it is not obvious to deduce whether higher-order topological phases can exist in amorphous systems from the previous results in quasicrystals. In fact, the higher-order topological phases in amorphous systems are protected by the *average* rotational symmetry, different from the *exact* symmetry in quasicrystals.

Furthermore, to the best of our knowledge, whether all $4n$-fold (n is a positive integer) rotational symmetries can arise in quasicrystals remains unclear. For instance, there has been no quasicrystals constructed with sixteen-fold or twenty-fold rotational symmetry [Generalized Dynamics of Soft-Matter Quasicrystals: Mathematical Models, Solutions and Applications (Springer, Singapore, 2022), 2nd ed]. In comparison, amorphous materials can possess any $4n$-fold average rotational symmetry, and we can explore richer higher-order topological phases with arbitrary $4n$ corner modes which cannot be realized in quasicrystals.

For the $\mathbb{Z}_2$ topological invariant, in quasicrystals, the effective Hamiltonian is constructed based on the plane wave in the 4D Brillouin zone. However, amorphous lattices cannot be generated from a higher-dimensional space with translational symmetry by a cut-and-project method, thus the construction method of the effective Hamiltonian for quasicrystals are no longer applicable. To fix the problem, we introduce unconventional twisted boundary conditions to construct the effective Hamiltonian in $\boldsymbol{k}$ space. This helps us build the $\mathbb{Z}_2$ topological invariant.

Another important contribution of our work is the introduction of how to calculate the quadrupole moment in topological phases without crystalline counterparts in amorphous systems. In fact, such a topological invariant is also applicable to quasicrystals (please see arXiv:2307.14974). Compared with the $\mathbb{Z}_2$ index introduced in Ref. [30], the generalized quadrupole moment can also be extended to characterize the chiral and helical hinge modes in 3D quasicrystals (arXiv:2307.14974). For example, one can use the winding number of the generalized quadrupole moment to characterize the chiral mode of 3D second-order topological insulators in quasicrystals without crystalline counterparts. We therefore believe that the generalized quadrupole moment introduced here will play an important role in the study of higher-order topology in non-crystalline systems.

Finally, we would like to emphasize that while both the higher-order topological phases in amorphous and quasicrystal lattices can support corner modes lacking crystalline equivalents, they have fundamentally distinct properties. In the amorphous case, the topological phase is gapless with a vanishing bulk energy gap, and the bulk states near zero energy are spatially localized corresponding to an Anderson insulator. However, in the quasicrystal case, the topological phase is gapped with a finite bulk energy gap corresponding to a band insulator.

Because of the above points, we believe that our work is novel and meets the acceptance criteria of SciPost Physics.

For the relevance of the results to physical amorphous materials, we agree with the referee that ideal amorphous materials are typically isotropic. However, in experiments, anisotropy can be introduced in the growing process by applying magnetic field [J. Appl. Phys. 106, 023918 (2009)] or through interface interactions [Phys. Rev. Lett. 100, 117201 (2008)]. As the referee suggests, using quasicrystals as the substrate is a good way to realize the C_pT symmetry in our model. For general $p = 4n$ without quasicrystal counterparts, the C_pT symmetry might be realized by doping magnetic impurities. Ideally, one can dope magnetic impurities with spin up in one $1/p$ sector and spin-down impurities in its adjacent sectors. Other sectors are doped in the similar manner such that the $1/p$ sectors have magnetic moments in alternating up and down directions. In this way, the average continuous rotational symmetry of amorphous materials is broken while the average C_pT symmetry is respected. Another approach is to apply local magnetic fields under the substrate with alternating directions for each $1/p$ sector during the growing process of amorphous materials. We understand that realizing the phase in experiments is a significant challenge. However, it is theoretically possible.

We therefore believe that our work is novel and is far from a straightforward generalization of Refs. [30,31] and it is also relevant to physical amorphous materials. We have added the discussion on the relevance to physical amorphous materials in the revised manuscript and hope that these responses will satisfy the referee and convince the referee to recommend publication of our work in SciPost Physics.

Weaknesses 2: What guarantees that there are no gapless modes at the boundary of the segments after restoring the rotational symmetry when constructing the Hamiltonians H j, or when applying periodic boundary conditions? As described in the text, the amorphous structure is not completely uncorrelated, rather it is a random set of hard disks. ls this constraint on the minimal interatomic distance obeyed by the symmetrized systems? Based on the description of the construction, it is not, which may result in additional sub-gap states at the gluing interfaces in the symmetrized and periodic boundary condition systems. I ask the authors to clarify these questions and demonstrate that no new subgap physics arises at the gluing interfaces, for example by examining the spatial localization of the low-energy states that appear at large system sizes (fig 3c inset).

Reply: We thank the referee for the nice question. We agree with the referee that when we restore the rotational symmetry, the new system may not satisfy the constraint on the minimal interatomic distance. As a result, the sub-gap states arise in the symmetry restored system as shown in the following figure (a), where the configuration averaged density distribution of two low-energy states near zero energy under periodic boundary conditions (PBCs) is plotted. However, we would like to clarify the following two points:

1. We only use the symmetry restored system to calculate the topological invariants shown in Fig. 3(a). For all the other results, we consider random lattices without restoring the symmetry. For example, in Fig. 3(c), we calculate the energy gap for random lattices without restoring the symmetry so that no sub-gap physics arises from the gluing operations. This can be clearly seen in the following figure (b) where the configuration averaged density distribution of two low-energy states near zero energy for the system under PBCs is plotted. Thus, the gapless behavior arises from the bulk states.

2. To calculate the topological invariant in Fig. 3(a), we consider restoring the rotational symmetry. Thus, the topological invariant χ can be constructed. For the quadrupole moment, the rotational symmetry is not required. One can directly calculate the quadrupole moment for the original system without restoring the symmetry. We find that the quadrupole moment exhibits nonzero values in the topological region and its value increases toward 0.5 with system size (see the following figure (c)), similar to the quadrupole moment calculated for the symmetry restored system. In addition, the calculated topological phase transition points agree well with the results of the energy gap and localization properties. All these results suggest that despite the presence of introduced sub-gap states, their existence does not affect the calculation of topological invariants.
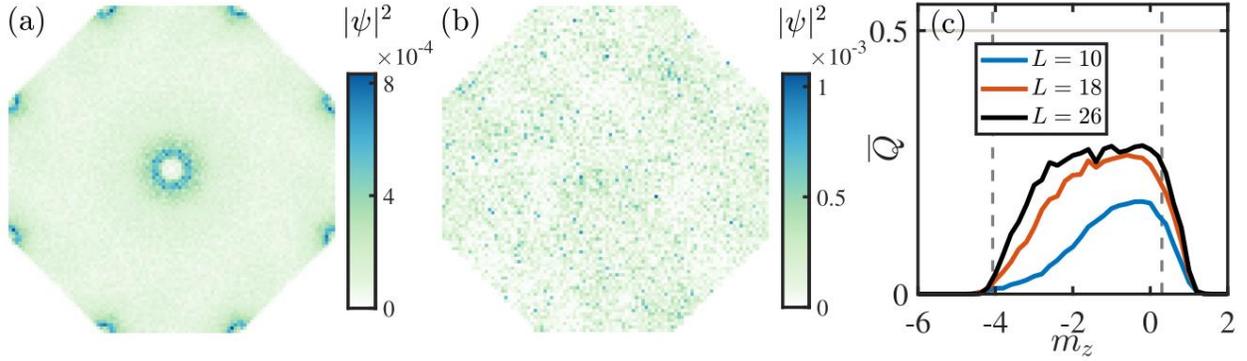
We have added the three figures in the Appendix C to show the existence of mid-gap states and provide arguments that their existence does not qualitatively affect the calculation of topological invariants there.

Weaknesses 3: Why does the invariant chi_n not depend on n? The manuscript only presents numerical evidence for chi_1=chi_3. I would expect a similar scenario as in ref 28 SM, when additional symmetries forcing a vanishing Chern-number are responsible for this equality, and is not forced by protecting symmetries. It would be worth to check whether this is the case, or at least comment on the possibility.

Reply: We thank the referee for the very interesting question. In the following, we will show that the equality $\chi_1 = \chi_3$ should not be forced by additional symmetries.

According to Phys. Rev. B 82, 184525 (2010), for a Hamiltonian with particle-hole symmetry, there exists a relation $e^{i\pi C} = \Pi_{K_0}\nu(K_0)$, where C is the Chern number and $\nu(K_0)$ is the sign of the Pfaffian of the anti-symmetrized Hamiltonian at particle-hole symmetry momenta K_0. In Ref. [30], the equality is generalized to the quasicrystal case with C_8 rotational symmetry, reading $e^{i\pi C} = \Pi_n \nu_n$, where $\nu_n = \nu_{n,0}/\nu_{n,\Pi}$ with $\nu_{n,0}$ ($\nu_{n,\Pi}$) being the sign of the Pfaffian of the anti-symmetrized Hamiltonian restricted in the eigenspace of $C_8 M$ operator with eigenvalue $\omega_n = e^{in\pi/8}$ for $n = [\pm 1, \pm 3, \pm 5, \pm 7]$ at the high-symmetry momentum 0 (Π). Due to the zero Chern number of the effective quasicrystalline Hamiltonian, $\Pi_n \nu_n = 1$. Besides, the topology protected by C_4 symmetry is trivial, so $\chi_1 = \chi_7$ and $\chi_3 = \chi_5$ for $\chi_n = (1 - \nu_n)/2$,

which also ensures $\Pi_n \nu_n = 1$. Similarly, in our amorphous model, we also have $\chi_1 = \chi_7$ and $\chi_3 = \chi_5$, as discussed in Appendix B.

However, for the quasicrystal model in Ref. [30], $\chi_1 \neq \chi_3$. As an indicator of the non-trivial higher-order topology, $\chi_1 = \chi_7 = 1$. In contrast, $\chi_3 = \chi_5 = 0$ because the subspaces with $C_8 M$ eigenvalues $e^{\pm i 3\pi/8}$ and $e^{\pm i 5\pi/8}$ are empty, which is straightforward given the matrix representation of the $C_8 M$ operator $U_{C_8 M} = \sigma_z e^{-i\pi\tau_z/8} = \text{diag}\left(e^{-\frac{i\pi}{8}}, e^{\frac{i\pi}{8}}, e^{\frac{i7\pi}{8}}, e^{-\frac{i7\pi}{8}}\right)$ in the basis of the onsite internal degrees of freedom in Ref. [30]. For the amorphous model in our paper, numerical results also demonstrate $\chi_1 \neq \chi_3$ near the phase transition point in a finite system. The transition points of χ_1 and χ_3 as a function of m_z are different, as shown in the following figure. The discrepancy implies that $\chi_1 = \chi_3$ is not protected by any symmetry. However, the difference between the two transition points Δm_z exhibits a power-law decay with the increase of the lattice size L as shown in the figure below, suggesting that $\chi_1 = \chi_3$ in the thermodynamic limit. Perhaps this equality in the thermodynamic limit is protected by some emerging symmetry in an infinite system, which indeed deserves future investigations.
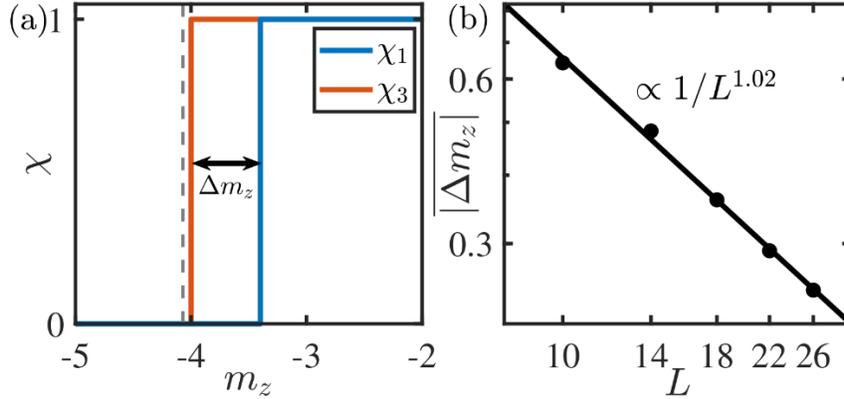
We have added the figure of χ_1 and χ_3 as a function of m_z for a typical random configuration in the revised manuscript.

Weaknesses 4: Why is it necessary (or advantageous) to distort the system to calculate the quadrupole moment? Is the quantity measured this way really the quadrupole moment, or perhaps some higher moment? It is really unclear to me how a procedure like this manifestly breaking the protecting rotational symmetries, can extract a topological invariant.

Reply: We thank the referee for the question. To be short, if we do not perform the site transformation, then we always obtain zero quadrupole moment in our case with $p = 8$.

Specifically, similar to the polarization, the quadrupole moment is a widely used topological invariant to characterize the quadrupole topological insulator. The higher-order topology of these insulators can be protected by chiral symmetry or particle-hole symmetry as we have previously proved that chiral symmetry (or particle-hole symmetry) protects the quantization of the quadrupole moment (see Refs. [62,63]). Similar to the Berry phase, the quadrupole moment is a $\mathbb{Z}_2$ topological invariant. For a quadrupole insulator on square lattices, when there are odd

number of corner modes, the quadrupole moment is equal to 0.5. However, in our case with $p = 8$, there are two corner modes in each quarter in a topological phase, which may explain why the traditional quadrupole moment vanishes. To obtain the reliable topological invariant to characterize the topology of the system, we propose the site transformation while keeping the bulk wave functions unchanged. Such a method indeed gives the correct topological characterization of the system.

In fact, in our another paper (arXiv:2307.14974), we apply the method to calculate the generalized quadrupole moment of a quasicrystalline system. We find that it can correctly characterize the topology of the 2D quasicrystal, similar to the $\mathbb{Z}_2$ index based on the rotational symmetry. In the paper, we also generalize the quadrupole moment to characterize the chiral and helical hinge modes in 3D quasicrystals based on the site transformations. For example, one can use the winding number of the generalized quadrupole moment to characterize the chiral mode of 3D second-order topological insulators in quasicrystals without crystalline counterparts. We therefore believe that the generalized quadrupole moment introduced here will play an important role in the study of higher-order topology in non-crystalline systems.

For the physical meaning of the quadrupole moment, let us consider the limiting case of the Benalcazar-Bernevig-Hughes model where the intracell hoppings are zero (see the following figure (a)). In this case, the quadrupole moment is equal to 0.5, and the Wannier center is localized at the position of A. However, in the general case, it remains an open question whether the connection of the nontrivial quadrupole moment to the position of Wannier center is always true. In the non-crystalline case, this connection becomes even more elusive. In our case, this quantity may represent a hidden moment structure. We admit that we do not know its exact nature, but it does serve as a topological invariant. This problem is indeed interesting and merits further study.
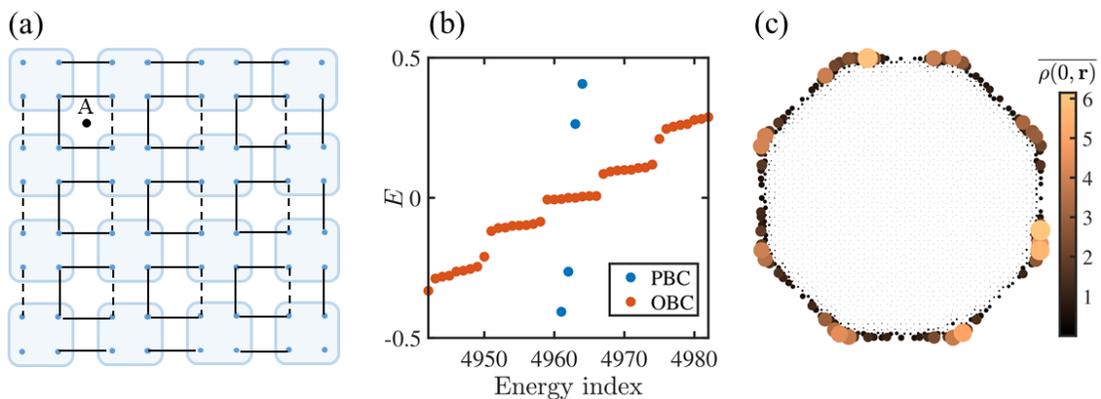
We also would like to point out that the rotational symmetry is not an indispensable ingredient for the quadrupole insulator. Without it, a system can still exhibit a quadrupole moment of 0.5 with zero-energy corner modes as long as chiral or particle-hole symmetry persists. To clarify our viewpoint, we consider a 2D quasicrystal with eight-fold rotational symmetry and add some structural disorder to explicitly break the symmetry. We find that this system still has eight zero-energy corner modes [see the above figure (b) for the energy spectrum and (c) for the local DOS at zero energy] and its generalized quadrupole moment is still equal to 0.5. We understand that

the rotational symmetry is important as it ensures that the local DOS at zero energy also respects this symmetry. In the amorphous case, the average rotational symmetry plays the role.

We have added some discussions to show that the generalized quadrupole moment can also be used to identify the higher-order topology of 2D and 3D quasicrystals in the revised manuscript.

Report: The manuscript studies topological phases of amorphous systems protected by chiral and average p-fold (p even) rotation symmetries. The analysis is carried out using a tight-binding model on a random graph, using "momentum-space" invariants derived from twisted periodic boundary conditions, quadrupole moment, and spectral signatures, the manuscript is well written, and the results are sound.

In my opinion the manuscript is an interesting, but fairly straightforward generalization of earlier work on quasicrystalline topological phases with added disorder (see point 1of the Weaknesses section for details).Hence l recommend moving the manuscript to SciPost Physics Core, and publication with minor clarifications: or would request the authors to further support the novelty of the work, and motivate why the paper meets the acceptance criteria of SciPost Physics in light of the questions raised.

Reply: We thank the referee for the nice summary of our work, the positive comment that "The manuscript is well written, and the results are sound" and the critical comments and constructive suggestions. We have follow the referee's suggestion to clarify the novelty of our work (see our reply to Weakness 1) and hope that this considerably improved version will satisfy the referee and convince him/her to recommend publication of this work in SciPost Physics.

Requested changes 1:
Minor change requests below, for major questions see the Weaknesses section.

The hopping Hamiltonian in eqn.1 for p=8 is (up to a basis transformation, and inclusion of bond-length-dependent prefactors) is identical to that of ref (28), and the generalization of the last term for cases with p other than 8 was presented in ref(29), l ask the authors to make this clear by citations when introducing the Hamiltonian.

Reply: We thank the referee for the nice suggestion. We have followed the suggestion to add the sentence that "This Hamiltonian can be derived through a unitary transformation of the Hamiltonian introduced in Refs. [30,31]." in the revised manuscript.

Requested changes 2: The following statement is unclear to me: "Note that near m_z 0.3 there appears an intermediate region with the coexistence of topologically nontrivial and trivial samples where the gapless bulk states are extended as shown in Fig. 3(d)." Should clarify the text or fig. 3 to make this clear.

Reply: We thank the referee for the comment. We have modified the sentence to "Note that near $m_z \approx 0.3$, $\bar{\chi}$ lies between 0 and 1 and does not converge to 0 or 1 as L increases, implying the existence of an intermediate region with the coexistence of topologically nontrivial and trivial samples. In this region, the gapless bulk states are extended as shown in Fig. 3(d)."

Requested changes 3: Should mention in the caption what the vertical dashed grey lines in fig. 3 denote.

Reply: We thank the referee for the nice suggestion. We have followed the suggestion to add the sentence in the revised caption of Fig. 3 that "The higher-order topological phase is separated from other phases by the vertical dashed grey lines".

Requested changes 4: "high-symmetric momenta" should be, following standard terminology, "high-symmetry momenta".

Reply: We thank the referee for the nice suggestion. We have followed the suggestion to change "high-symmetric momenta" to "high-symmetry momenta" in the revised manuscript.