Dear Editor,

We are in receipt of the assessment of our manuscript “Towards a theory of surface orbital magnetization” by Daniel Seleznev and David Vanderbilt, with manuscript number BX14243 and thank the referees for their helpful comments. We hereby resubmit our revised manuscript publication in Physical Review B. Referee 1 has given a positive evaluation of our work and recommended publishing in PRB, while Referee 2 preferred to withhold a recommendation to publish pending our response to their concerns and questions. As you can see in our response to the manuscript, we have fully addressed all concerns and queries of the referees. We look forward to hearing your response.

Yours sincerely,
Daniel Seleznev and David Vanderbilt

Referee comments are in blue Italics; our responses are in black Roman. In our responses to the referees, when quoting specific portions of the revised manuscript, we highlight revisions with red font. The detailed list of changes follows after our response to Referee 2. In the final section of the reply, we also list all changes made to the manuscript that are not based on referee comments.

In addition to the reply, we also attach a marked up version of the manuscript where all changes are highlighted in red font, while deleted sentences or paragraphs are indicated with a strikeout.

Reply to Referee 1

The manuscript BX14243 attempts to develop the theory for surface orbital magnetization. The authors identify the symmetries which allow for the definition of surface orbital magnetization as precisely those symmetries that quantize axion coupling. Additionally, even when the surface orbital magnetization is not well defined, the authors show that calculations in two corresponding slab geometries can give the correct value of the corresponding hinge current using their proposed marker.

The questions posed in the manuscript are very relevant and the results are interesting. Furthermore, the manuscript is well written, it was a pleasure reading it. I recommend the manuscript for publication in Phys. Rev. B. Below I list a few questions for the authors.

We thank the referee for the very positive comments. We address the referee's questions.

1) Since the 3 local markers studied in the manuscript do not show anticipated "shift freedom", should one conclude that there might be more local markers (satisfying all the conditions listed in the manuscript)? If so, can you exclude the possibility that for certain systems none of the markers studied in this work gives the correct value of the hinge current, i.e. one rather has to use a different local marker?

The referee is correct to point out the possibility that other markers may satisfy the constraints described in the paper. The constraints we describe in Secs. IV A 1-4 of the manuscript initially reduce the list of acceptable markers down to the $\mathcal{M}^{\mathcal{C},\mathcal{C}}(\mathbf{r})$ and $\mathcal{M}^{\mathcal{E},\mathcal{E}}(\mathbf{r})$ or any weighted average of these. However, only one specific weighted average, namely the $\mathcal{M}^{\text{lin}}(\mathbf{r})$ marker, is found to consistently predict the hinge currents. Thus, we are left with only one candidate marker, so that we do not have the means to address the shift freedom question directly.

We believe that our search for markers began in a well motivated way from the expressions derived by Bianco and Resta, but we cannot rule out the possibility that another line of approach could lead to one or more markers that also pass our tests and correctly predict the hinge currents for the models we tested. We also cannot discount the possibility that some of these markers would fail for another model system not considered here. This is why we emphasize the need for new formal arguments at the end of the Summary of our paper.

2) Can you comment if the local current density and correspondingly the local conductivity tensor are measurable quantities?

We take it as self-evident that the local current density $\mathbf{j}(\mathbf{r})$ is measurable *in principle* as the curl of the magnetic field that it generates. Admittedly, it may be difficult *in practice* to measure such local fields with sufficient spatial resolution. Scanning (nanoSQUID or magnetic force) probes might resolve the macroscopic hinge currents, but could probably not be expected to resolve atomic-scale fields. Magnetic impurity substitutions or interstitials or trapped muons might give information at some locations. Magnetic neutron scattering might give such information on $\mathbf{j}(\mathbf{r})$ inside the bulk unit cell, but the cross section is presumably much too weak to allow surface sensitivity. However, we do not believe that these practical difficulties should stand in the way of a theoretical discussion that takes $\mathbf{j}(\mathbf{r})$ and its field derivatives as physical observables.

In particular, is the expression in Eq. (45) unique? What is the relation to the local current operator discussed in J. Stat. Phys., 177, 717 (2019)?

In a continuum setting, the current density operator in Eq. (45) is broadly accepted as the unique standard definition in the single-particle context. In the discrete tight-binding setting, things become more complicated. In the paragraph below Eq. (B6) of Appendix B of the manuscript, we note that "Since the current is assumed to flow uniformly along the straight-line bond connecting the sites, it makes no difference if we treat it as distributed uniformly along the length, concentrated at the center, or partitioned between the two end points of the bond". We pick the latter choice, which assigns half of the current on the bond to each end point comprising the bond, but our results for the macroscopic hinge currents would be unchanged had we selected the other choices. The local current operator in J. Stat. Phys., 177, 717 (2019) amounts to assuming the current on the bond is concentrated at the center of the bond.

Reply to Referee 2

The manuscript represents an important contribution to the theory of the surface orbital magnetization. That their theory is incomplete should not impede publication in PRB; theirs is a self-contained and technically assured investigation of certain hypotheses that will guide future formulations of a more complete theory.

I am not recommending immediate publication, because the presentation of their results can be substantially improved. The greatest difficulty in comprehending this paper is that a few important premises/postulate are implicit and left to the expert's imagination, while a non-expert will likely flounder. I have also identified one error, which may be an instructive error that calls into question the premises of the paper. Readers will benefit substantially if the authors make their premise/postulate explicit and admit their premise is questionable. Certainly, it was not the intent of the authors to present a one-sided story; the authors admit to finding themselves in an "ambiguous position" with regard to the main conclusions of their findings. However, it is questionable if most readers will appreciate what exactly is ambiguous about the authors' position.

We thank the referee for carefully reading the manuscript and for the constructive feedback. We address the referee's comments below.

Points for improvement:

1. Dangers in using "well-defined" in the first major claim

It is claimed in the abstract (and throughout the paper) that the surface orbital magnetization (SOM) is not "well-defined" in certain low-symmetry contexts. "Well-defined" means many different things to many people; an object may be "well-defined" within one theory but "ill-defined" in another; moreover, there are multiple ways to be "ill-defined".

The authors would do well to avoid vague descriptions and say directly what they mean, namely, to replace "well-defined" with "uniquely defined/determined within classical electromagnetism theory". (Possible short hand: "classically determined/defined".) The recommended emphasis on "unique determination" tells the reader precisely what goes wrong in low-symmetry contexts; the recommended emphasis on "classical electromagnetism theory" manifests one of the authors' implicit premises/biases, namely that

2. Implicit bias/premise: classical theory is equally capable of uniquely determining the SOM as the as-yet-unestablished quantum theory.

This premise/bias should be made explicit, rather than left to the reader's imagination. After all, the premise is questionable. Classical theory constrains quantum theory, but quantum theory is ultimately more fundamental and has the potential to uniquely determine objects that have no unique meaning in classical theory.

The authors' implicit premise actually runs counter to the spirit/philosophy of earlier works by one of the authors (David Vanderbilt) on the bulk orbital magnetization (BOM). The BOM is not uniquely defined in classical theory but is uniquely defined in the quantum theory that David played a role in developing [PRL 95, 137205 (2005) and follow-up works].

In the face of the earlier success with the BOM, the authors' premise for the SOM comes as *a surprise to the referee, and potentially to many readers as well. However, because this premise is never stated explicitly, the dichotomy between the BOM and the SOM generates a great deal of confusion.*

The first two comments by the referee are closely related, so we address them together. We agree with the referee, and have edited the manuscript accordingly. The edits pertaining to these points can be found in the abstract as well as in Secs. I, II, before the start of III A, in III C, as well as in VI and VII. Throughout, we emphasize that a classical knowledge of the microscopic current distribution in the bulk, at the surfaces, and at the hinge is generally sufficient to only determine the differences of magnetizations of two surfaces sharing a hinge, but not the individual values of the surface magnetizations. However, as our symmetry analysis in Sec. III then demonstrates, in specific high symmetry cases (namely axion-odd systems) this classical knowledge does turn out to be sufficient to uniquely determine the surface magnetization. In the rest of the manuscript, then, our use of the term "uniquely defined within the classical theory" refers to this particular combined knowledge of the current distribution in the bulk, at the surfaces, and at the hinges. In this context, if the information of the current distribution is lacking at the hinges, then the surface magnetizations and their differences cannot be assigned unique values. We highlight this issue at the end of Sec. III C, and point out that we subsequently turn to a quantum-mechanical treatment via the local marker formulation to try to resolve this problem. These edits now reflect the spirit of the theory of bulk orbital magnetization that the referee mentions. As the full list of changes in relation to these points by the referee is extensive, we list all of them in detail at the end of our reply.

3. Implicit hypothesis: SOM being uniquely determined in the classical theory <=> existence of a unique local marker in the quantum theory

<=> means "if and only if". This hypothesis is closely related to the implicit premise in point 2, and is the precise logical link between the two halves of the paper. It is therefore a tragedy that the hypothesis is never stated explicitly. The result is that the current manuscript reads like two disconnected halves.

Some of the authors' conclusions are mystifying if the reader does not fully appreciate the implications of <=>. In particular, the self-professed "ambiguous position" is only ambiguous when viewed as the authors' unsuccessful attempt to prove <=>. Indeed, in trying unsuccessfully to prove <=>, the authors spent considerable effort in finding distinct local markers that reproduce the same hinge-current observable. The motivation for this considerable effort would be mysterious to most readers.

A third reason for stating the hypothesis directly: only by making a conjecture explicit can the authors tempt a reader to prove or disprove the conjecture.

4. It should be stated explicitly that the authors' findings do not rule out a more fundamental quantum theory where a unique local marker exists independent of symmetry.

This follows from the authors' failure to prove the hypothesis in point 3. Because the authors do not admit to the possibility of a fundamental quantum theory (where a unique local marker exists independent of symmetry), the authors' exposition is one-sided, and biases the reader to not even *consider this logical possibility. Such bias may stunt future attempts to formulate a complete theory of the SOM.*

As the referee's points 3 and 4 are closely related, we address them together. In the revised manuscript, we now emphasize that without the proper classical knowledge, it is not immediately clear how to compute the surface magnetization and that we turn to the quantum-mechanical local magnetization markers in an attempt to resolve this issue. We subsequently describe our hypotheses regarding the local markers at the end of Sec. IV A 5, where we have replaced the second to last paragraph with the following:

"Having formulated our list of markers, we are faced with several questions regarding their behavior. First, it remains to be seen which, if any, of the markers will correctly predict the hinge current. For each of the two surfaces forming a hinge, identical markers will be used to compute their respective magnetizations, which in turn will be substituted into Eq. (2) to yield the hinge current. In such tests, it is of interest to check whether there is a dependence on the presence of axion-odd vs. axion-even symmetries. Recall that at the level of classical electromagnetism, only differences of the magnetizations of surface facets sharing a hinge are generally well defined. As an exception, when axion-odd symmetries are present, the $M_\perp$ values can be determined from a knowledge of the hinge currents. We should then like to see whether the quantum marker-based theory is also better behaved when axion-odd symmetries are present, and whether, in the absence of such symmetries, the shift freedom of the classical framework reappears at the quantum level. This would be signaled by the existence of multiple markers correctly predicting the hinge currents but differing by a constant shift for all facets".

Other edits, found in Secs. VI and VII, also address our hypotheses in light of our numerical studies done in Sec. V. We list all other edits at the end of this document.

5. The authors' hypothesis for the SOM (in point three) is incorrect for a closely-related quantity: the SOM compressibility.

SOM compressibility = the derivative of the SOM with respect to the chemical potential. The SOM compressibility has a uniquely-defined local marker (sometimes called the local Chern marker). Yet the SOM compressibility is not associated to any uniquely defined current observable in the classical theory. This is a violation of <=> in point 3, if "SOM" in point 3 is replaced by "SOM compressibility".

What I am saying contradicts the authors' vague claim: "We can argue that there is no such ambiguity for the local Chern marker ... This analysis puts the definition of the local Chern marker $C(r)$ on a relatively firm footing." The vague descripton of "relative firm footing" has no place in this paper, and is gravely misleading. When the authors' argument is understood precisely, it is actually an argument for the absence of any uniquely-defined current observable, owing to the "cross-gap" term.

This error should be corrected. The significance of this error should also be pointed out, namely that as a matter of principle, uniquely-defined local markers (of certain quantities) exist in the quan-

tum theory that are not associated with any current observable in the classical theory. This throws some doubt on the authors' hypothesis in point 3, but it is a doubt that should be manifested to the reader.

6. The manuscript lacks an explicit comparison with Zhu-Alexandradinata, Phys. Rev. B 103, 014417 (2021)

The authors allude to Zhu-Alexandradinata as the only other work on the SOM: "To date, however, there has been very little discussion of surface orbital magnetization in the literature [Zhu-Alexandradinata], and a proper theory of orbital magnetization at the surface of a bulk material has yet to be developed."

This glancing reference suggests unflatteringly that Zhu-Alexandradinata is not a "proper theory" of the SOM. Rather than this veiled criticism, it is much more helpful for the community if criticisms are offered directly and precisely. It is worth stating in the manuscript that (a) Zhu-Alexandradinata assumed (without rigorous justification) that the Bianco-Resta local marker can be applied to define a SOM, in contention with the main findings of Seleznev-Vanderbilt, (b) the major claims of Zhu-Alexandradinata are based on the SOM compressibility and are not in contention with Seleznev-Vanderbilt.

Points 5 and 6 both reference the topic of the compressibility, so we group them together. In response to the referee's comments, we have deleted the last two paragraphs immediately preceding Sec. IV A 1 and replaced them with the following, where we mention that the compressibility is related to the uniquely defined Chern marker and geometric component of the surface AHC, but it is not the full surface anomalous Hall response:

"As we will see in the following subsections, there is no such ambiguity with respect to cyclic permutation of operators for the local Chern marker $C(\mathbf{r})$, which is therefore well defined at a specific $\mathbf{r}$ in the context of a marker-based theory. As a result of Eq. (16), this implies that $\mathcal{M}_{\mathrm{C}}(\mathbf{r})$ has a linear dependence on the chemical potential μ as it is scanned across the gap. This observation allowed Zhu et al. [19] to introduce a uniquely defined "surface magnetic compressibility" $dM_\perp/d\mu$ in terms of the presence of a net coarse-grained Chern-marker concentration at the surface. We note in passing that the Chern marker is closely related to the local anomalous Hall conductivity, i.e., the antisymmetric part of the surface conductivity tensor $\sigma_{ij}(\mathbf{r}) = \partial j_i(\mathbf{r})/\partial E_j$ describing the first-order response of the local current density $\mathbf{j}(\mathbf{r})$ to a homogeneous macroscopic electric field $\mathbf{E}$. However, as shown by Rauch et al. [11], the two are not identical, since the local anomalous Hall conductivity also contains a non-geometric or "cross-gap" term that is not captured by the Chern marker.

In the following subsections, we discuss several physical requirements that we impose on the markers, allowing us to reduce the number of candidates to just a few that can be regarded as physically acceptable".

The above paragraphs also serve as a more explicit description of the work by Zhu et al.. We also add an additional paragraph to Sec. VI that connects their work to ours:

"We now also comment in more detail on the connection of this work to that of Zhu et al. [19]. The authors of that work also introduced the notion of surface orbital magnetization, and similarly used the local marker formulation of orbital magnetism to compute it. Their primary focus, as we mentioned just before Sec. IV A 1, was to explore the "facet magnetic compressibility" $dM_\perp/d\mu$ that is directly proportional to the geometric component of the surface AHC, especially in the context of links to higher-order topology. We agree with their conclusion that this compressibility is uniquely determined. On the other hand, where they assumed that the Bianco-Resta $\mathcal{M}^{C,C}$ marker could be applied to define surface magnetization, our findings indicate that this assumption was problematic. We also note that they did not explore the issue of a possible shift freedom of the surface magnetization".

Appendix of shorter remarks:

a) "However, there is no a priori reason to believe that the individual values of facet magnetizations are well defined. We must therefore understand whether, or under what circumstances, we can resolve this ambiguity" There is no a priori reason within the classical theory.

As this remark is essentially a summary of the more detailed points made by the referee above, we refer to our responses to those points.

b) The use of "nonsymmorphic" is confusing. Firstly, the authors believe "nonsymmorphic" means to involve a fractional translation; however, nonsymmorphic space groups don't always contain symmetry elements with fractional translations. Secondly, the authors further confuse the issue by introducing "n-symmorphic" vs "n-nonsymmorphic".

We agree with the referee that it is not usually advisable to refer to a symmetry operation as symmorphic or nonsymmorphic, since in many cases this can depend on the setting. The terms are usually reserved to describe space groups. Here, however, having restricted the discussion to operations that preserve a selected axis $\hat{\mathbf{n}}$, it is convenient to classify those that do, or do not, involve a fractional translation along $\hat{\mathbf{n}}$. We still think that describing these as "$\hat{\mathbf{n}}$-symmorphic" and "$\hat{\mathbf{n}}$-nonsymmorphic" is a reasonable compromise in this context, but we are now careful to define these terms more clearly on first use by including the following clarification in the first paragraph of Sec. III A:

"We further classify operations that preserve $\hat{\mathbf{n}}$ as either symmorphic or nonsymmorphic along that direction, where by the latter term we mean operations that explicitly feature a fractional translation along $\hat{\mathbf{n}}$, such as a screw or glide mirror operation".

c) Regarding: "The application of these principles to our case suggests that, for a crystal having a generic nonzero axion coupling and a common branch choice of the quantum on all surfaces, the $M_\perp$ values of all facets will undergo a common constant shift in response to a global shift of the Fermi level in the gap. This is already a kind of a realization of the "shift freedom" discussed in

Sec." This point should be discarded or elaborated. It is not at clear to me what is the relation with the previously-defined "shift freedom". In particular, $M_\perp$ being dependent on the Fermi level does not make $M_\perp$ non-uniquely defined.

We agree with the referee and have removed that entire paragraph (second to last paragraph in Sec. III C) in the revised manuscript.

d) "We can argue that there is no such ambiguity for the local Chern marker..." It is worth clarifying that the "ambiguity" refers to the process of cyclic permutation, and also worth clarifying that the invariance of the chern marker under cyclic permutation has been proved in previous literature. Some citation is appropriate.

We have implemented the referee's suggestion and have added a paragraph to the end of Sec. IV A 1 discussing the Chern marker's invariance under cyclic permutation of operators. We also cite the relevant literature (Reference [11]):

"As for the Chern-marker contribution, similar considerations imply that $\mathcal{M}_{\mathrm{C}}(\mathbf{r})$ should also be written in terms of X and Y operators. Eq. (15) then leads to $\mathcal{M}_{\mathrm{C}}(\mathbf{r}) = -(2\mu e/\hbar)\mathrm{Im}\langle \mathbf{r}|X^\dagger Y|\mathbf{r}\rangle = (2\mu e/\hbar)\mathrm{Im}\langle \mathbf{r}|Y^\dagger X|\mathbf{r}\rangle$. Further algebra demonstrates that these expressions are identical with $\mathcal{M}_{\mathrm{C}}(\mathbf{r}) = (2\mu e/\hbar)\mathrm{Im}\langle \mathbf{r}|XY^\dagger|\mathbf{r}\rangle = -(2\mu e/\hbar)\mathrm{Im}\langle \mathbf{r}|YX^\dagger|\mathbf{r}\rangle$ (see Eq. (A14) of the Appendix of Ref. [11]). Thus, we are left with a unique expression for $\mathcal{M}_{\mathrm{C}}(\mathbf{r})$ as well as the local Chern marker".

In addition, we have added short comments at the ends of subsections IV.A.2-3 pointing out that the local Chern marker passes the reasonableness tests considered there as well.

e) "However, this does not necessarily imply that M(r) vanishes everywhere, only that its unit cell average vanishes." I believe the correct formulation by Bianco-Resta is that the macroscopic average of M(r) vanishes. It is hard to believe the much stronger claim that the "unit-cell average vanishes", if by "unit-cell" the authors mean a primitive unit cell. A similar question mark is raised for the author's definition in Eq (42).

We are puzzled by this comment. The full quote at the beginning of Sec. IV.B is "Recall that we assume that the system has enough symmetry to force the macroscopic magnetization to vanish in the bulk. However, this does not necessarily imply that $\mathcal{M}(\mathbf{r})$ vanishes everywhere, only that its unit cell average vanishes" So, we are talking only about a crystallite whose bulk magnetic space group precludes any net magnetization, and only about unit cells deep in the bulk interior. In this case the unit-cell average of $\mathbf{M}(\mathbf{r})$ vanishes by assumption, and since the markers are constructed to reproduce the bulk orbital magnetization when integrated over a bulk unit cell, the unit-cell average of $\mathcal{M}(\mathbf{r})$ must vanish as well. Perhaps the referee had some other context in mind, but without more guidance we do not know what more to say here. For added clarity, in the manuscript we have edited the second sentence of the cited quote to become "However, this does not necessarily imply that $\mathcal{M}(\mathbf{r})$ vanishes everywhere, only that its bulk unit cell average vanishes".

List of all edits in response to referee comments

Here we list all edits to the manuscript in response to the referees. We list those edits that were not explicitly cited in our responses above.

Referee 1

No changes needed.

Referee 2

Changes made in response to points 1 and 2

- In the first sentence of the abstract we now have "The theory of bulk orbital magnetization has been formulated both in reciprocal space based on Berry curvature and related quantities, and in real space in terms of the spatial average of a quantum mechanical local marker". Furthermore, we later specify that "By performing a symmetry analysis, we find that only crystals exhibiting a pseudoscalar symmetry admit well-defined magnetizations at their surfaces at the classical level. We then explore the possibility of computing surface magnetization via a coarse-graining procedure applied to a quantum local marker".

- In Sec. I, we delete the paragraph that begins as "Even from a phenomenological point of view...", and instead add two consecutive paragraphs that begin with "Even at a classical level, another difficulty arises" and "With the introduction of the quantum description...", respectively. Furthermore, we edit the paragraph starting with "In this work..." to now read

  "In this paper, we first investigate the role of symmetry and show that certain classes of symmetries do allow for the surface magnetization to be uniquely extracted from a knowledge of hinge currents, even at the classical level. We then turn to the quantum problem and explore..."

- In the second to last paragraph of Sec. I, we edit the sentence "We show that this single relation is insufficient to ascribe an unambiguous value of..." to "We show that within the framework of a classical theory, this single relation is insufficient to ascribe a uniquely defined value of...".

- In the second sentence of the first paragraph of Sec. II, we change "bound current" to "macroscopic bound current". We then split the paragraph into two, with the second paragraph beginning with "If instead we consider...". To the end of the new first paragraph, we also add two sentences: "At the classical level, $M_\perp$ can be determined from the combined knowledge of the microscopic current density deep in the bulk and at the edge. Without the added knowledge of the current density at the edge, however, $M_\perp$ can be determined only from a knowledge of the quantum-mechanical bulk Bloch eigenstates".

- In the first sentence of the new second paragraph of Sec. II we add "If instead we consider the surface of a 3D bulk system (with vanishing bulk magnetization),...". Additionally, to the same paragraph we add the sentence "Hence a classical knowledge of the microscopic current distribution in the bulk, at the surfaces, and at the hinge is sufficient to uniquely determine the difference of $M_\perp$ values across facets sharing a hinge, but not the $M_\perp$ values individually" that precedes the sentence ending with Eq. (2).

- In the paragraph starting with "For the entirety of this paper..." in Sec. II, we edit the last two sentences to be "Combined with Eq. (2), this assumption indicates that *differences* between magnetizations on neighboring surface facets are also observables, in addition to being classically well defined in the sense described earlier. However, there is no *a priori* reason to believe that the individual values of facet magnetizations are either observables or are uniquely defined in the context of classical theory". We then break off the rest of the paragraph to become a new paragraph. In between these two paragraphs, we then add a new paragraph...

- In Sec. II we add a paragraph that is now the third to last in the section, which begins with "As mentioned in the Introduction..." explaining the classical origin of the shift freedom arising from a change of gauge.

- We change the first two sentences of the first paragraph of Sec. III to become "The goal of this section is to establish under what circumstances we can specify a uniquely defined magnetization on a given surface facet of a 3D crystallite in the context of classical theory. By this, we mean whether a classical knowledge of the microscopic current distribution in the bulk, at the surfaces, and at the hinges is sufficient to assign a unique value of $M_\perp$ to a given surface facet".

- To the beginning of the first sentence of the first paragraph of Sec. III C we append "Within a classical context...".

- In the second paragraph of Sec. III C, we add a sentence after the first: "The facet magnetizations could then be determined from a knowledge of the hinge currents in these geometries".

- In the last paragraph of Sec. III C we insert into the second sentence "... and classical theory...". In the last sentence of the paragraph we also insert "... quantum-mechanical...".

- In the first paragraph of Sec. VII we replace the sentence starting with "We have demonstrated that..." as well the next sentence with two sentences: "We have demonstrated that in a general classical context, the knowledge of the macroscopic currents residing on a hinge formed by two surface facets is sufficient to determine only the difference of the magnetizations of the two facets. The said macroscopic hinge currents are the physical observables corresponding to the presence of surface orbital magnetization, and this fact indicates that differences of surface orbital magnetization are observables as well"

<u>Changes made in response to points 3 and 4</u>

- We replace the entire first paragraph of Sec. VI with "The results presented above help us to address the main questions motivating this work. First, does the introduction of quantum mechanics in the form of a marker-based theory provide a prescription for computing $M_\perp$ directly for a given facet, in such a way that hinge currents at adjoining facets are correctly predicted? Is this equally true in the axion-odd and axion-even cases? Second, if multiple markers succeed in doing so, does the quantum theory inherit the shift freedom of the classical theory? The most simplistic scenario would be one in which all of the physically acceptable markers identified at the end of Sec. IV A yield identical $M_\perp$ values for any given facet over all models, and correctly predict the hinge currents where facets meet".

- The first two sentences of the second paragraph of Sec. VI are replaced by "Focusing first on the axion-odd case, we found that our tests were consistent with this simplistic scenario for the alternating Haldane and two-layer square-plaquette models".

- We replace the paragraph in Sec. VI beginning with "Notably, our findings provide no..." to be "Notably, our findings provide no evidence for the possible marker shift freedom of the surface magnetizations within the quantum-mechanical marker-based theory. In fact, since only the $\mathcal{M}^{\text{lin}}$ marker always yielded the correct hinge currents, we do not have multiple marker candidates as would be be needed to allow us to test this proposition"

- In the paragraph of Sec. VI starting with "We find ourselves, then, in a somewhat ambiguous position", we add a sentence prior to the last one mentioning "It also cannot yet be ruled out that M^{lin} does not pass all tests in models beyond those considered in this paper".

- In the first paragraph of Sec. VII we also edit the sentence starting with "We further expanded on the local marker formulation..." to become "To develop the theory of surface magnetization in a quantum context, we further expanded on the local marker formulation..."

Additional edits to the manuscript

- In the fourth paragraph of Sec. I, we have made the following alterations in the sentence: "To date, however, there has been very little discussion of surface orbital magnetization in the literature [18, 19], and a complete theory of orbital magnetization at the surface of a bulk material has yet to be developed". The new Reference 19 now appears in the list of citations as well.

- In the sixth paragraph of Sec. I, we have made the following change to the last sentence: "Another way to frame the problem is to note that quantum-mechanical expressions are available for neither the local polarization density $\mathbf{P}(\mathbf{r})$ nor the local orbital magnetization density $\mathbf{M}(\mathbf{r})$.

- In the paragraph immediately after Eq. (2) in Sec. II we have added a new Reference [20]: "... such as a step or domain wall [20]..."

- We add a new paragraph detailing the rotational covariance of the Chern marker contribution to the end of Sec. IV A 2:

  "For the Chern marker contribution of Eqs. (15-16), it is easy to see that for arbitrary rotations θ about $\hat{\mathbf{z}}$ we have $\mathcal{M}_{\mathrm{C}}(\mathbf{r}) = -(2\mu e/\hbar)\mathrm{Im}\langle\mathbf{r}|X^{\dagger}Y|\mathbf{r}\rangle \rightarrow (2\mu e/\hbar)\mathrm{Im}\langle\mathbf{r}|Y^{\dagger}X|\mathbf{r}\rangle = \mathcal{M}_{\mathrm{C}}(\mathbf{r})$. We may then define a vector marker $\boldsymbol{\mathcal{M}}_{\mathrm{C}}$ analogously to $\boldsymbol{\mathcal{M}}_{\mathrm{LC}}$, and the former is found to exhibit rotational covariance for the same reasons as the latter"

- We add a new paragraph discussing the transformation of the Chern marker contribution under $H \rightarrow -H$ transformations to the end of Sec. IV A 3:

  "We note that for the Chern marker contribution of Eqs.(15-16), under such a transformation $C(\mathbf{r}) \rightarrow -C(\mathbf{r})$, while $\mu \rightarrow -\mu$. Therefore $\mathcal{M}_{\mathrm{C}}(\mathbf{r})$ is left invariant."

- We replace the first sentence of the first paragraph of Sec. IV A 5 with

  "The considerations of Secs. IV A 1-IV A 3 have reduced the list of acceptable markers to the two given in Eqs. (27-28), augmented by the uniquely defined Chern-marker contribution $\mathcal{M}_{\mathrm{C}}(\mathbf{r})$".

  Additionally, later in the paragraph we add the specification "... while $\mathcal{M}^{\mathcal{E},\mathcal{E}}$ of Eq. (28)...". In the second paragraph, we change "To end the discussion of the markers, we also note..." to "We also note..." and subsequently merge the second paragraph with the first.

- In Sec. VI, we replace the first sentence of the paragraph beginning with "To summarize this discussion, when..." with "To summarize our discussion of the numerical results, when...", and also altered "...we found that the only cases in which markers disagree, with only the $\mathcal{M}^{\mathrm{lin}}$ marker correctly predicting...".

- We correct a typo in Sec. VI in the paragraph starting with "We find ourselves, then, in a somewhat ambiguous position". In the second sentence of the paragraph $\mathcal{M}^{CC}$ is corrected to $\mathcal{M}^{C,C}$.

- In Sec. VI, we have added a comment highlighted in red regarding the application of the local marker to metallic systems:

  "We note, however, that prior numerical tests performed on TB models of metals at $T = 0$ suggest that the local marker is able to reproduce the bulk magnetization of Eq. (1) within open boundary conditions, but exhibits slower convergence with sample size than in the insulating case [34]".

  We have also added a new Reference [34] citing this work.

- We have added a note at the end of Sec. VII regarding the appearance of a preprint on arXiv by Gliozzi, Lin and Hughes (new Reference [36]):

  "**Note added:** After submission of this paper, we became aware of a subsequent preprint by Gliozzi, Lin and Hughes [36] that also considers, though from a different perspective, issues related to surface orbital magnetization, hinge currents, and bulk magnetic quadrupole moments".