We appreciate Referee 1's generally positive report on our manuscript. Below we reply to the issues raised by him/her. The referee's statements are in blue. Our replies are in black.

The paper illuminates on the discussion of the subtleties of the quantised magneto-electric effect in 3D topological insulators. However, at the same time, it tries to advocate the view, somewhat in between the lines, that a measurement of this effect is not that significant or important. Also it seems to bypass the question on whether the single isolated 1/2 quantised Hall conductivity in the surface of the TI can be measured.

We certainly did not mean to imply that this is not that important or significant. Indeed we have spent the last several years of our career working to measure exactly this effect! And we even wrote the present manuscript because we thought the subject was so important and interesting. The point of our manuscript was 1) to provide a alternative language to discuss the effect and 2) to make important analogies to related physical effects to improve people's understanding.

In this revised version we have added some additional discussion on the significance of the effect. Moreover near the end of the manuscript we have added some text on how the isolated ½ quantized response can be measured directly. We now write:

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"The interpretation of all these experiments in terms of quantized formal ME
response rests on the fact that symmetry constrains the bulk axion angle to be an
odd integer times $pi$ in the bulk.    Therefore, the observation of a quantum
Hall odd integer sum of top and bottom surfaces can be interpreted as a half-
integer quantum Hall effect of a single surface.   However, it is still desirable
to isolate the half-integer Hall conductance of a single surface.   It may be
possible to do this through performing a THz reflection experiment off of a single
surface directly.   This would be a completely model free measurement of the
formal ME lattice in much the same fashion as the measurement of a single end
charge establishes the formal polarization lattice of a 1D chain.  Although in
principle possible, such an experiment has not yet been performed.   It will
require thick single insulating crystals."
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The paper is a valuable addition to the discussion of the subtleties surrounding the magneto-electric effect in topological insulators. I found the discussion very instructive and at a level that can help communicate these issues to a broad audience.

We appreciate the referee's positive comments.

In spite of the conceptual value of their discussion, the authors seem to suggest that there is not much value in pursuing the measurement of the quantised magneto-electric effect, and that this is basically a closed matter given that they have already measured the quantised Faraday rotation. There is no doubt that this effect is intimately related to the Faraday rotation effect described by the authors. But even though they are intimately related it is healthy not to blur completely their identities (think of the zero resistance and the perfect diamagnetism of a superconductor which are also intimately related to each other and come hand-in-hand in the superconducting state yet it is important to distinguish them).

As we mention above, we didn't mean to give the impression that we don't think there is much value in further experiments. Indeed, one of our principle goals was to make clear exactly the relationship between the quantum Faraday effect and a true magnetoelectric effect. As also mentioned above we have added text explaining what experiments we believe still need to be done.

More importantly, in my opinion no measurement to this date has been able to detect the isolated 1/2 quantised conductance expected at the surface of the TI (under the right symmetry breaking conditions to gap the surfaces). This is the essentially anomalous feature of a single surface of the 3D TI (which cannot be mimicked by any stand alone 2D band insulator). All measurements to this date basically contain additive contributions of the two surfaces making the result strictly non-anomalous. This a key aspect of the discussion that seems to have been mostly overlooked in the paper.

We agree that we did not discuss much what other experiments need to be done, although we did discuss that the quantum Faraday effect measures the sum of surfaces. And it was only because of inversion symmetry in the bulk of Bi2Se3 that we could infer a half-quantized surface conductance in our measurements. But it is indeed better to measure it directly as we now say explicitly in added text. We also agree that no experiment has measured an isolated single surface. We now say so explicitly.

Requested changes

1) In Eq(11) they mean "Re" instead of "Im"?

This was a typo. We have corrected it.

2) The argument that leads to Eq.(16) overlooks one important fact: that threading one flux per surface unit cell is an extremely large perturbation, so adiabaticity is far from guaranteed. In fact it is known that when half-flux quantum is threaded per unit cell strong topological insulators develop a kind of 1D metallic wire along the flux tube, in the form of a pair of gapless conter-propagating 1D gapless modes that penetrates into the bulk (see e.g. Phys. Rev. B 82, 041104(R) (2010)). Perhaps a safer argument can be made by assuming an enlarged unit cell. Can the authors should clarify or remove these arguments?

This is an argument that we borrowed from the papers of Essin, Moore, and Vanderbilt. And it is important to point out that it only is included here to give an alternative motivation for the scale of the magnetoelectric susceptibility quantum e^2/h. There can be other interesting effects of applying magnetic flux in these materials as the referee points out. But it does not invalidate the reasoning that e^2/h is the quantum. We don't see any problem with our formulation that "the smallest magnetic field that can be applied without destroying the periodicity of a crystalline system is one flux quantum per surface unit cell (e/Acell).". We believe this is correct. We think that a full investigation of how the half-flux quantum effects discussed in Phys. Rev. B 82, 041104(R) (2010) relate to what is discussed by Essin et al. is very important, but beyond the scope of our manuscript.

3) The authors state: "An effective magnetoelectric susceptibility can only be defined in ... where the net Hall response is zero." (notice missing word ...="systems"). This is an important point, and I kind of see why (my view: if it is non-zero the system might have net charge accumulation from Streda formula after threading magnetic field, also surface cannot be fully insulating and hence charge might flow). But the authors should explain why this makes the effective magnetoelectric susceptibility ill defined.

Our argument is physical. If a finite size (say rectangular slab shaped) object is to have a bulk magnetization, then it needs to be able to have Hall currents flowing in equal magnitude but opposite directions on opposing surfaces to not have charge accumulation anywhere. If there is a net Hall current then there will be continuous charge accumulation, there cannot be a continuous surface current, and there cannot be a dc magnetization.

4) The authors state: "The hybrid Wannier function representation makes explicit the fact that one cannot create Wannier functions in such a topological systems despite the fact that the eigenstates of Hamiltonian have the Bloch form." I guess the authors mean that one cannot create Wannier functions that are localized and strictly respect the symmetry? (see e.g. Phys. Rev. B 83, 035108 (2011), Phys. Rev. B 93, 035453 (2016)).

Yes, this is what we meant. (Incidentally we did mention symmetries in the sentence immediately below, but in regards to the 1D polarization and connection to the atomic limit). This is a crucial point that we thank the referee for having us emphasize. We have changed the passage to read "The hybrid Wannier function representation makes explicit the fact that one cannot create Wannier functions in such topological systems that respect all symmetries, despite the fact that the eigenstates of Hamiltonian have the Bloch form." And now we cite the papers that the referee points out.

5) Related limitations to the measurement of the apparent monopole at the surface of a TI described here were also discussed in Phys. Rev. Lett. 111, 016801 (2013).

This was a very important and relevant paper we had missed! We now write "Pesin and MacDonald treated the related problem of the effective magnetic monopole induced near the surface of a TI in the presence of finite longitudinal conductance due to the presence of a suddenly introduced external electric charge \cite{pesin2013topological}. In a very related fashion to the above they found that finite longitudinal conductivity introduces certain dynamical constraints on seeing the topological magnetoelectric effect."

6) The authors write: "Again by way of analogy with the 1D chain, this suggests a way of looking at inversion symmetric insulators as overlapping e^2/h and -e^2/h layers. As shown in Fig. 8, one can conceive of conventional insulators as being materials these conducting layers are centered on top of each other and spatially overlap and cancel, whereas a TI is where layers of them are displaced from each other by half a unit cell, giving ½ e^2/h on the surface." How literal should this picture be taken? e.g. how is time reversal supposed to act in this hypothetical system of displaced quantum Hall layers? or, are the authors then imagining a system with large breaking of TRS throughout the bulk? if so, how to think then about TR invariant TI's?

It is a cartoon, but aspects of this picture should be taken quite literally. For instance, as we point out it is precisely the situation for the trivial insulator in many layered transition metal dichalcogenides where there are K and K' valleys that each host a Chern insulator with opposite Hall conductance. Since each of these valleys are time reversed versions of each other the system is globally time reversal symmetric. Each valley breaks TRS "strongly" but the system is globally TRS. Similarly, for the TI case, for the bulk of the material the QH layers cancel giving no net macroscopic signal of TRS (macroscopically similar to the TMD case in the bulk. Irrespective of the microscopics and TRS breaking of any subsystems the bulk is (or can be) globally TRS. However… we would also point out that in most of the manuscript (and in our experiments), we have considered a situation where the quantized response is protected by inversion symmetry in the bulk and a magnetic field is actually applied. So the issue of TRS or not in the bulk is not directly relevant.

7) In view of the comments in report above, the authors might want to revisit/rephrase statements such as:

"Although the development of systems that realize this configuration is very important from a materials perspective, we do not believe it warrants any particular consideration as anything special or fundamental. Both scenarios have the same formal ME susceptibility. As shown in Fig. 10, the two configurations should just be considered as different experimental conditions and realize fundamentally the same thing."

"However, as is hopefully clear from this discussion there is no intrinsic difference from one scenario the other. They are all just different demonstrations of the same underlying physics and both experiments are measures of the formal ME susceptibility."

We can revise the statements that the referee highlights, but from our current thinking we believe that what we wrote are accurate and precise representations of the physics we have emphasized. Systems which show an effective ME susceptibility (which must break T and P) are NOT fundamentally different in their underlying physics than systems that don't break T and P and cannot have a ME susceptibility defined. They do exhibit different phenomena, but they both have the same formal ME susceptibilities. This is the point we want to emphasize.