Reviewers' comments:

Reviewer #1 (Remarks to the Author):

In this work, the authors addressed the magneto-elastic coupling in Ising spin ice, focusing on the stability of phases with finite density of monopoles.
Firstly, they examine the effect of oxygen (O) displacement and clarify how it affects the magnetic coupling in terms of Goodenough-Kanaomori-Anderson rules.
This consideration leads to the effective model including both spin and O displacement degrees of freedom, where the role of magneto-elastic coupling is shown to stabilize monopoles.
From the Monte Carlo simulation of this effective model, the authors show the stability of monopole liquid, when the direct magnetic couplings is absent between spins.
They compare the result with analytical solution of the model which integrates out the O displacements and found complete agreement between the results.
Furthermore, the authors supplement the model with monopole-monopole interactions and found the crystal phase of single monopoles is stabilized at low temperatures with disordered spin configurations.
The disorder of spins results in the correlated disordered electric moment distributions, originated from O displacements, suggesting the correlated fluctuations of dipolar electric and magnetic moments.
Finally, the authors succeed in reproducing the double-layered monopole phase observed in $Tb_2Ti_2O_7$ under the [110] magnetic field, with the help of magnetostriction effects and monopole-monopole interactions.

The importance of lattice degrees of freedom is well established in the area of frustrated magnetism.
Nevertheless, so far coupling to lattice has not been explored sufficiently in spin ice, so far.
The authors addresses this problem, starting with the basic model, and obtain nontrivial results, which may shed light on the origin of field-induced ordering in $Tb_2Ti_2O_7$.
The analysis is sound and may serve as a model case for the analysis of magneto-elastic coupling of other pyrochlore-based frustrated magnets.
For this reason, I think this work deserves publication in Communications Physics.
Before publication, however, I recommend addressing the following points.

[1] On the definition of monopolar charge
In Section III, the authors introduce monopolar charge and their interactions in Eq. 6.
I wonder how the monopolar charge Q is defined. More specifically, is Q simply defined as a summation of Ising spin or does it contain sign factor, according to the orientation of tetrahedron ?
This distinction is important, as it affects the interpretation of like charge attraction or repulsion.

[2] On the neutron signature of oxygen displacements
In the end of section IV, the authors mention possible detection of oxygen displacements as the fluctuation of electric dipole moments through an electron beam or X-ray diffraction.
This prediction is interesting, however, I wonder if the displacement is large enough to be observable.
It would be helpful to readers, if (rough) estimates are given to the order of typical displacement length.

[3] Possible origin of inter-monopole interaction in $Tb_2Ti_2O_7$
The authors raise inter-monopolar interaction as one necessary ingredients to reproduce double-layered monopole phase in $Tb_2Ti_2O_7$ on the basis of their scenario.
Is their any independent evidence to support the presence of such interactions ?
For example, is there any experimental result available that indicates the relevance of further neighbor interactions ?

Reviewer #2 (Remarks to the Author):

This manuscript studied the coupling between the spin degrees of freedom and the lattice degrees of freedom on the pyrochlore lattice, and the spins are designed in proximate to the spin ice regime. The physics and related phenomena are commonly existed and relevant for various rare-earth based pyrochlore magnets. Moreover, there are very limited theoretical works about this physics. The only one that I am aware of is the work by Daniel Khomskii. Thus, I think this current work is a valuable contribution to the field and community. The microscopic model about the coupling is reasonable, and the analysis is quite standard. I also suggest authors to include "Experimental Identification of Electric Dipoles Induced by Magnetic Monopoles in Tb2Ti2O7" in PhysRevLett.124.087601 (2020).
In conclusion, I recommend this work for publication.

Reviewer #3 (Remarks to the Author):

In this paper the authors consider the effects of a magnetoelastic coupling on the Ising model in the pyrochlore lattice, which describes spin ice materials. The starting point is a classical model that accounts for the displacement of the oxygen ions located in the center of every tetrahedra. The authors then note that "integrating out" the vibration degree of freedom gives rise to a four-spin interaction that had been studied in Ref. [29] and is known to stabilize what they call a "monopole liquid" phase. The latter is a massively degenerate state with a finite density of magnetic monopoles. Using classical Monte Carlo methods, they show that additional perturbations can then stabilize different monopole crystal phases, such as the zincblende structure at zero magnetic field and the double layered crystal for a magnetic field along the 110 direction. The discussion about monopole order is accompanied by an analysis of the associated lattice distortion due to the magnetoelastic coupling. The authors claim that their simple model can account for some experimental observations in the compound Tb2Ti2O7, and propose that pinch points in the structure factor, characteristic of Coulomb phases, could also be observed by scattering off the oxygen ions.

This work represents an interesting contribution to the understanding of the rich phase diagram of spin ice materials, particularly of Tb2Ti2O7. The numerical methods are standard and have been employed by the same authors in several previous publications. The original results in this paper are connected with the presence of a fluctuating lattice degree of freedom, which the authors say provide "strong physical foundations" for Hamiltonians that had been studied before but seemed somewhat artificial. Indeed, their model is able to capture the spontaneous symmetry breaking in the zinc blend monopole crystal. However, I would like the authors to clarify some points before I can make a recommendation:

1. The authors claim their model has strong physical bases, but it's important to acknowledge the limitations of the classical model. For instance, is it reasonable to describe the vibrations as classical harmonic oscillators? The displacement of the oxygen ions must be related to some optical phonon mode, which probably has an activation energy much higher than the exchange coupling in these materials, so this degree of freedom would be frozen at the low temperatures where magnetism comes into play.

2. The giant magnetostriction reported in Tb2Ti2O7 (Ref. [46]) is related to coupling to transverse acoustic phonons. What is the connection with the vibration modes considered in the model of Eq. (2)?

3. The authors state that they focus on the strong coupling limit. Is there a critical coupling for the stabilization of the monopole liquid? Is it possible to estimate the energy scales involved and compare them to realistic parameters?

4. It would be helpful if the authors could comment on the microscopic mechanism by which the magnetic field can generate the exchange anisotropy in Eq. (7), beyond the statement that this is allowed by symmetry.

Authors answers to Reviewers' comments:

Reviewer #1 (Remarks to the Authors):

"In this work, the authors addressed the magneto-elastic coupling in Ising spin ice, focusing on the stability of phases with finite density of monopoles. Firstly, they examine the effect of oxygen (O) displacement and clarify how it affects the magnetic coupling in terms of Goodenough-Kanaomori-Anderson rules. This consideration leads to the effective model including both spin and O displacement degrees of freedom, where the role of magneto-elastic coupling is shown to stabilize monopoles. From the Monte Carlo simulation of this effective model, the authors show the stability of monopole liquid, when the direct magnetic couplings is absent between spins. They compare the result with analytical solution of the model which integrates out the O displacements and found complete agreement between the results. Furthermore, the authors supplement the model with monopole-monopole interactions and found the crystal phase of single monopoles is stabilized at low temperatures with disordered spin configurations. The disorder of spins results in the correlated disordered electric moment distributions, originated from O displacements, suggesting the correlated fluctuations of dipolar electric and magnetic moments. Finally, the authors succeed in reproducing the double-layered monopole phase observed in $Tb_2Ti_2O_7$ under the [110] magnetic field, with the help of magnetostriction effects and monopole-monopole interactions.

The importance of lattice degrees of freedom is well established in the area of frustrated magnetism. Nevertheless, so far coupling to lattice has not been explored sufficiently in spin ice, so far. The authors addresses this problem, starting with the basic model, and obtain nontrivial results, which may shed light on the origin of field-induced ordering in $Tb_2Ti_2O_7$. The analysis is sound and may serve as a model case for the analysis of magneto-elastic coupling of other pyrochlore-based frustrated magnets. For this reason, I think this work deserves publication in Communications Physics. Before publication, however, I recommend addressing the following points."

We thank the Referee for the careful reading of our work and recommendation. In what follows we address her/his comments in detail.

1) "On the definition of monopolar charge.
In Section III, the authors introduce monopolar charge and their interactions in Eq. 6. I wonder how the monopolar charge Q is defined. More specifically, is Q simply defined as a summation of Ising spin or does it contain sign factor, according to the orientation of tetrahedron? This

distinction is important, as it affects the interpretation of like charge attraction or repulsion."

In all cases (tetrahedra up or down) the definition of the magnetic charge is given by Q/2*(number spins in-number spins out). In order to make this point completely clear, we have expanded a sentence in Subsection II.A, where we describe the model, so as to say this explicitly:

"One can group the different spin configurations of a single tetrahedron into sets, using the net entrant spin flux as a label that defines the magnetic charge [12] of that tetrahedron. The same definition is valid for up or down tetrahedra.."

2)"On the neutron signature of oxygen displacements.
In the end of section IV, the authors mention possible detection of oxygen displacements as the fluctuation of electric dipole moments through an electron beam or X-ray diffraction. This prediction is interesting, however, I wonder if the displacement is large enough to be observable. It would be helpful to readers, if (rough) estimates are given to the order of typical displacement length."

After the prediction by Khomskii, there has been some experimental evidence of the presence of an electric dipolar moment associated to magnetic monopoles in spin ice materials. However, to our knowledge no direct measurement of the actual displacement has been provided. A previous theoretical work in $Tb_2Ti_2O_7$ (by Jaubert and Moessner, Ref. [54] in the text) estimates this displacement within the picometer range, similar to that observed in multiferroic materials. New methods based on the traditional XRD have been recently proposed and used to observe distortions within this range (Ref. [98], now included in the manuscript). To our knowledge, these methods have not yet been applied to the particular point raised by the referee.

To address this, we have now modified the end of the section dealing with this effect, adding the following sentence:

"The chances to observe the effect depends critically on the magnitude of the O-2 displacement. We will discuss this in Section III, where we also summarize the structure factor results for this and two other phases."

The Discussion section now has the following paragraph, commenting the experimental possibilities and providing a rough estimation of the main parameters for some important materials:

"The ability to detect effects related to the electric dipole on monopoles depends on its magnitude. While there are indications of the presence of such dipoles in $Tb_2Ti_2O_7$and $Dy_2Ti_2O_7$ [60, 98], the displacement δr has not been measured. Within our model, we can obtain it through J_{ml} (Eq. 4), provided we know the coupling constant \tilde{\alpha} and the spring constant K. A rough estimation based on the experimental and numerical results obtained in Refs. [48, 99–101] gives a small J_{ml} for $Dy_2Ti_2O_7$and $Ho_2Ti_2O_7$, on the order of 10 mK. In turn, this leads to δr≈0.1 pm for these canonical spin ices. On the other hand, Jaubert and Moessner [54] estimate a bigger δr for Tb2Ti2O7 (within the picometer range), similar to that observed in multiferroic materials. While this is still considerably small, new methods based on traditional XRD have been very recently proposed and used to observe distortions within this range in a strontium titanate oxyde [102]. The chances of a direct observation of the magnetoelastic phenomena we propose can increase if the efforts are first concentrated in compounds with a big coupling between magnetic and lattice degrees of freedom, starting with monopole crystals. The double monopole layer in Tb2Ti2O7 could then be an excellent starting point."

3) "Possible origin of inter-monopole interaction in Tb2Ti2O7.
The authors raise inter-monopolar interaction as one necessary ingredient to reproduce double-layered monopole phase in Tb2Ti2O7 on the basis of their scenario. Is there any independent evidence to support the presence of such interactions ? For example, is there any experimental result available that indicates the relevance of further neighbor interactions ?"

We are not aware of any evidence in this respect; the only possible indication we noticed is the presence of half moons in neutron diffuse scattering, which coincides with our observing them in the monopole liquid with like-attraction between monopoles. This is mentioned in the text, but since it is but a weak suggestion, we have tried to put little emphasis on it.

In spite of this, we think it is important to consider that further neighbour interactions is only one of the alternatives we mention in the the text. As shown earlier (Ref. [54] in the text) electric dipolar interactions (that should be present once the monopoles are stabilized by our proposed mechanism) would tend to favour their arrangement in a double layer. In this case, the term we propose is an effective way to take this interaction into account.

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Reviewer #2 (Remarks to the Authors):

"This manuscript studied the coupling between the spin degrees of freedom and the lattice degrees of freedom on the pyrochlore lattice, and the spins are designed in proximate to the spin ice regime. The physics and related phenomena are commonly existed and relevant for various rare-earth based pyrochlore magnets. Moreover, there are very limited theoretical works about this physics. The only one that I am aware of is the work by Daniel Khomskii. Thus, I think this current work is a valuable contribution to the field and community. The microscopic model about the coupling is reasonable, and the analysis is quite standard. I also suggest authors to include "Experimental Identification of Electric Dipoles Induced by Magnetic Monopoles in Tb2Ti2O7" in PhysRevLett.124.087601 (2020).

In conclusion, I recommend this work for publication."

We thank the referee for his/her recommendation, and also for calling our attention to this reference. We have now included it in the manuscript (Ref. [98] in the text).

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Reviewer #3 (Remarks to the Authors):

"This work represents an interesting contribution to the understanding of the rich phase diagram of spin ice materials, particularly of Tb2Ti2O7. The numerical methods are standard and have been employed by the same authors in several previous publications. The original results in this paper are connected with the presence of a fluctuating lattice degree of freedom, which the authors say provide “strong physical foundations” for Hamiltonians that had been studied before but seemed somewhat artificial. Indeed, their model is able to capture the spontaneous symmetry breaking in the zinc blend monopole crystal. However, I would like the authors to clarify some points before I can make a recommendation."

We thank the referee for her/his comments and input on our work. We answer below the questions and doubts posed.

1) "The authors claim their model has strong physical bases, but it’s important to acknowledge the limitations of the classical model. For instance, is it reasonable to describe the vibrations as classical harmonic oscillators? The displacement of the oxygen ions must be related to some optical phonon mode, which probably has an activation energy much higher than the exchange coupling in these materials, so this degree of freedom would be frozen at the low temperatures where magnetism comes into play."

We thank the referee for this observation. Our previous version did acknowledge to a certain degree some of the limitations of our classical description, but we agree that the reference to "strong" physical foundation or bases might have been a little too strong and needed to be toned down, something we have done in this version.

Regarding the need of optical (high energy) phonons at temperatures quite below the Einstein or Debye temperature of the system (for example, in Tb2Ti2O7): we think it is important to note all the consequences of the coupling between structural and magnetic degrees of freedom. Without coupling, the deformation of the O-lattice would certainly imply an energy bigger than the thermal energy [k_B T] at low temperature; however, the interaction with the magnetic lattice can reduce the combined energy scale to zero. Our classical model for O distortion can be understood as a mean to probe this instability in a minimalist way, that can be subject to a better theoretical description at a later stage.

In addition to toning down our claims of a "strong physical basis" we have also modified a previous comment in this respect (now in the Discussion section) that reads:

"Remarkably, notwithstanding its simplicity, the MeSI model provides a unified framework that explains the zero field ground state measured in Tb2Ti2O7 [55] and the double-layered monopole crystal at moderate fields [25]; this includes an improved version for the distortion on the O-2-lattice proposed previously [25].The classical treatment of distortions at low temperatures (which is unrealistic) can be understood as a simplistic way to probe the magnetoelastic instabilities of this system."

2)"The giant magnetostriction reported in Tb2Ti2O7 (Ref. [46]) is related to coupling to transverse acoustic phonons. What is the connection with the vibration modes considered in the model of Eq. (2)?"

In truth, our approach takes as an experimental fact the presence of giant magnetostriction in Tb2Ti2O7. Jaubert and Moessner suggest its possible role in helping to stabilize the double layered structure (Ref. [54] in the text): we took their suggestion, used symmetry arguments to evaluate its effect on the lattice, and explored its consequences within the framework of our model. In short, we have not explored the possibility of a direct connection between the MESI model and the giant magnetostriction in Tb2Ti2O7. There is a sentence within the relevant section that might have led to some confusion in this respect; it said:

"(...) Alternatively we will now inquire on the effect of h on the lattice structure through the magnetoelastic coupling \tilde{\alpha}."

We have now modified it to read:

"(...) Alternatively, we will now inquire on the effect of h on the lattice structure, and then of that on the spin lattice through the magnetoelastic coupling \tilde{\alpha}. "

3) "The authors state that they focus on the strong coupling limit. Is there a critical coupling for the stabilization of the monopole liquid? Is it possible to estimate the energy scales involved and compare them to realistic parameters?"

There is indeed a critical condition for the stabilization of the monopole liquid. We mention the "strong coupling limit" because we mostly concentrate on a regime where this condition is fulfilled. In terms of the effective constant J_{ml} and the nearest neighbours effective exchange constant J_0 this condition can be written as J_0 < J_{ml} for positive J_0, and J_{ml} > -3J_0 for negative J_0. If the first condition is not fulfilled, this implies that a spin ice-like phase is the stable one (this is what happens in Tb2Ti2O7 at low fields, within the framework of our model). For J_{ml} < -3J_0, a "double monopole crystal", with all-in/all-out spins, is the stable phase. This is now explained in Section IIB, in the following way:

"For a strong magnetoelastic coupling, the first term (with a four-spin product) stabilizes a Monopole Liquid at low temperatures [29]. It is easy to check that the range of stability is given by J_0 < J_{ml} for positive J_0 (which otherwise corresponds to a spin ice phase), and J_{ml} > -3J_0 for negative J_0 (usually leading to a double monopole crystal)."

Regarding the second point, we have made a rough estimation of J_{ml} for Dy2Ti2O7 and Ho2Ti2O7, obtaining (as expected) a very small value (of the order of 10 mK). However J_{ml} is expected to be bigger in other materials. We have included a paragraph mentioning this (and other aspects mentioned by the First referee) within the Discussion Section. It reads:

"The ability to detect effects related to the electric dipole on monopoles depend critically on its magnitude. While there are indications of the presence of such electric dipoles in Tb2Ti2O7 and Dy2Ti2O7 [60, 98], the displacement δr has not been measured. Within our model, we can obtain it through J_{ml} (Eq. 4), provided we know the coupling constant \tilde{\alpha} and the elastic constant K. A rough estimation based on the experimental and numerical results obtained in Refs. [48, 99–101] gives a small J_{ml} for Dy2Ti2O7and Ho2Ti2O7, on the order of 10 mK. In turn, this leads to δr≈0.1 pm for these canonical spin ices. On the other hand, Jaubert and Moessner [54] estimate a bigger δr for Tb2Ti2O7 (within the picometer range), near that observed in multiferroic materials. While this is still considerably small, new methods based on traditional XRD have been very recently proposed and used to observe distortions within this range in a strontium titanate oxyde [102]. The chances of a direct observation of the magnetoelastic phenomena we propose can increase if the efforts are first concentrated in compounds with a big coupling between magnetic and lattice degrees of freedom, starting with monopole crystals. The double monopole layer in Tb2Ti2O7could then be an excellent starting point."

4) "It would be helpful if the authors could comment on the microscopic mechanism by which the magnetic field can generate the exchange anisotropy in Eq. (7), beyond the statement that this is allowed by symmetry."

We thank the referee for this suggestion, which we have taken. We have now added a sentence before the aforementioned statement; it mentions briefly the microscopic mechanism, and only then there is an appeal to symmetry. It reads:

"Rare-earth ions usually have associated a very strong spin-orbit coupling; through it, the torque acting on spins can affect the orbital angular momentum, and then the lattice. Based on symmetry (...)"

REVIEWERS' COMMENTS:

Reviewer #1 (Remarks to the Author):

The authors answered all the questions in a satisfactory way and made suitable modifications to the manuscript.
Now I would like to recommend the publication of the article as it is.

Reviewer #3 (Remarks to the Author):

The authors have provided thorough and persuading answers to my questions. I recommend the paper for publication.