University of Groningen

Ferroelectricity in spiral magnets

Mostovoy, M

Published in:
Physical Review Letters

DOI:
[10.1103/PhysRevLett.96.067601](https://doi.org/10.1103/PhysRevLett.96.067601)

IMPORTANT NOTE: You are advised to consult the publisher's version (publisher's PDF) if you wish to cite from it. Please check the document version below.

Document Version
Publisher's PDF, also known as Version of record

Publication date:
2006

[Link to publication in University of Groningen/UMCG research database](#)

Citation for published version (APA):

Mostovoy, M. (2006). Ferroelectricity in spiral magnets. *Physical Review Letters*, 96(6), art. - 067601. [067601]. <https://doi.org/10.1103/PhysRevLett.96.067601>

Copyright

Other than for strictly personal use, it is not permitted to download or to forward/distribute the text or part of it without the consent of the author(s) and/or copyright holder(s), unless the work is under an open content license (like Creative Commons).

The publication may also be distributed here under the terms of Article 25fa of the Dutch Copyright Act, indicated by the "Taverne" license. More information can be found on the University of Groningen website: <https://www.rug.nl/library/open-access/self-archiving-pure/taverne-amendment>.

Take-down policy

If you believe that this document breaches copyright please contact us providing details, and we will remove access to the work immediately and investigate your claim.

Downloaded from the University of Groningen/UMCG research database (Pure): <http://www.rug.nl/research/portal>. For technical reasons the number of authors shown on this cover page is limited to 10 maximum.

Mostovoy Replies: In the preceding Comment [1] Kenzelmann and Harris (KH) argue against the conclusion made in [2] that spiral magnets are in general ferroelectric. This conclusion is, however, supported by the recent discovery of many new multiferroic spiral magnets with very different crystal structures, such as CoCr_2O_4 , MnWO_4 , LiCu_2O_2 , and LiCuVO_4 [3–6].

Furthermore, KH argue that the continuum theory outlined in [2] leads to misleading predictions about the magnetically induced electric polarization. To prove their point, they consider a hypothetical spin configuration with the $\Gamma_3 \times \Gamma_4$ symmetry [see Fig. 1(c) of the original version of their Comment [7]], and argue that the results of the continuum theory are incompatible with crystal symmetries. While one cannot deny the importance of symmetry considerations, the arguments of KH are themselves very misleading. They incorrectly assert that for this spin configuration “the spiral theory” would predict electric polarization along the c axis.

The continuum model [2] is based on the assumption that the spin state can be described by a single magnetization vector. For TbMnO_3 , where the wave vector of the magnetic spiral is along the b axis and spins are rotating in the bc plane, it predicts electric polarization $\mathbf{P}$ along the c axis, in agreement with experiment. The hypothetical magnetic structure of KH is of a different kind, as it is made of spirals rotating in opposite directions, which is why the net polarization along the c axis is zero.

The model of Ref. [2] can be easily extended to include these more general magnetic orders. For more than one magnetic ion per unit cell one can introduce several independent magnetic order parameters, which increases the number of possible magnetoelectric coupling terms. For TbMnO_3 magnetic structures in question can be described by four magnetic order parameters: $\mathbf{L}_1 = \mathbf{S}_1 + \mathbf{S}_2 + \mathbf{S}_3 + \mathbf{S}_4$, $\mathbf{L}_2 = \mathbf{S}_1 + \mathbf{S}_2 - \mathbf{S}_3 - \mathbf{S}_4$, $\mathbf{L}_3 = \mathbf{S}_1 - \mathbf{S}_2 + \mathbf{S}_3 - \mathbf{S}_4$, and $\mathbf{L}_4 = \mathbf{S}_1 - \mathbf{S}_2 - \mathbf{S}_3 + \mathbf{S}_4$, where $\mathbf{S}_i$ are 4 Mn sites in the $Pbnm$ unit cell. The observed structure is described by a single vector order parameter $\mathbf{L}_2$. As discussed in [2], the magnetoelectric coupling linear in the gradient of the magnetic order parameter (Lifshitz invariant) allowed by symmetries has the form $P^c(L_2^b \partial_y L_2^c) = P^c(L_2^b \partial_y L_2^c - L_2^c \partial_y L_2^b)$, which gives rise to magnetically induced P^c . The hypothetical configuration of KH is described by pairs of two different order parameters, (L_2^b, L_4^c) and (L_3^b, L_1^c) . The terms $L_2^b \partial_y L_4^c$ and $L_3^b \partial_y L_1^c$ do not transform like any of the components of $\mathbf{P}$, so that the induced polarization is zero. Since $\mathbf{L}_4$ is obtained from $\mathbf{L}_2$ by inverting the sign of spins on the sites 2 and 4 (the same holds for $\mathbf{L}_1$ and $\mathbf{L}_3$) the structures described by two different order parameters correspond to counterrotating spirals: the direction of spin rotation on the sites 1 and 3 is opposite to that on the sites 2 and 4.

The point is, however, that this hypothetical spin configuration is very artificial, as it is difficult to imagine a system where interactions between spins would favor counterrotating spirals. The average interaction between counterrotating spirals is zero, while for corotating spirals some energy can be gained by properly adjusting their relative phases. This is the reason why the simple model of Ref. [2] with a single order parameter successfully describes thermodynamics and magnetoelectric properties of many multiferroics.

This model was never meant to describe all multiferroic materials, in particular, those where electric polarization is induced by commensurate spin and charge (or structural) modulations, which is believed to be the case for RMn_2O_5 [8]. The form of magnetoelectric coupling and the resulting anomalies of polarization and dielectric constant at the ferroelectric transition in these systems are different from those in spiral magnets. Similarly, in $\text{RbFe}(\text{MoO}_4)_2$ polarization is induced by the staggered vector spin chirality that matches an alternating structural modulation (inequivalence of “up” and “down” triangles in ab layers). The incommensurate helical spin structure resulting from a relative rotation between spins in neighboring ab layers seems to be unrelated to ferroelectricity. This is consistent with Eq. (4) of Ref. [2] that gives zero polarization for a helix, in which spins rotate around the propagation vector (the c axis in this case), contrary to the statement made in the Comment.

Maxim Mostovoy

Zernike Institute for Advanced Materials
University of Groningen
Nijenborgh 4, Groningen 9747 AG, The Netherlands

Received 4 December 2006; published 26 February 2008

DOI: [10.1103/PhysRevLett.100.089702](https://doi.org/10.1103/PhysRevLett.100.089702)

PACS numbers: 77.80.Fm, 75.30.Fv, 75.60.Ch

- [1] M. Kenzelmann and A.B. Harris, preceding Comment, Phys. Rev. Lett. **100**, 089701 (2008).
- [2] M. Mostovoy, Phys. Rev. Lett. **96**, 067601 (2006).
- [3] Y. Yamasaki, S. Miyasaka, Y. Kaneko, J.-P. He, T. Arima, and Y. Tokura, Phys. Rev. Lett. **96**, 207204 (2006).
- [4] K. Taniguchi, N. Abe, T. Takenobu, Y. Iwasa, and T. Arima, Phys. Rev. Lett. **97**, 097203 (2006).
- [5] S. Park, Y.J. Choi, C.L. Zhang, and S.-W. Cheong, Phys. Rev. Lett. **98**, 057601 (2007).
- [6] Y. Naito *et al.*, J. Phys. Soc. Jpn. **76**, 023708 (2007).
- [7] M. Kenzelmann and A.B. Harris, arXiv:cond-mat/0610471.
- [8] S.-W. Cheong and M. Mostovoy, Nat. Mater. **6**, 13 (2007). The origin of electric polarization in this material is, however, controversial: the bc spiral propagating in the c direction, which can account for the polarization along the b axis, was reported in H. Kimura *et al.*, J. Phys. Soc. Jpn. **76**, 074706 (2007).