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$_2$O$_4$, MnWO$_4$, LiCu$_2$O$_2$, 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 $\Gamma_1 \times \Gamma_4$ 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 $P$ along the $c$ axis, in agreement with experiment. The hypothetical magnetic structure of KH is of a different kind, as it is made of $L_1$ and $L_2$ 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: $L_1 = S_1 + S_2 + S_3 + S_4$, $L_2 = S_1 + S_2 - S_3 - S_4$, $L_3 = S_1 - S_2 + S_3 - S_4$, and $L_4 = S_1 - S_2 - S_3 + S_4$, where $S_i$ are 4 Mn sites in the $Pbnm$ unit cell. The observed structure is described by a single vector order parameter $L_2$. As discussed in [2], the magnetoelectric coupling linear in the gradient of the magnetic order parameter ($L_2$) is given by the form $P_i = \alpha \partial L_k \sigma_{k,i} L_2$, which gives rise to magnetostrictive polarization $P$.

The hypothetical configuration of KH is described by pairs of two different order parameters, $(L^b_2, L^a_2)$ and $(L^b_2, L^c_2)$. The terms $L^b_2 \partial_i L^a_4$ and $L^b_2 \partial_i L^c_4$ do not transform like any of the components of $P$, so that the induced polarization is zero. Since $L_4$ is obtained from $L_1$ by inverting the sign of spins on the sites 2 and 4 (the same holds for $L_2$ and $L_3$) 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 co-rotating 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$_2$O$_4$ [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 RbFe(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.

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