Comment on "Ferroelectricity in Spiral Magnets"

There is much interest in materials that show a strong coupling between magnetic and electric degrees of freedom. Recently Mostovoy [1] has presented a theory based on symmetry arguments that leads to quite general claims which we show here are not justified. The two main conclusions which we find to be untrue are (i) the claim that the symmetry of the unit cell is not important and (ii) the implication that the stated model represents a universal phenomenological description of ferroelectricity induced by incommensurate magnetic order.

We take the example of the ferroelectric phase of $TbMnO_3$, where the magnetic structure is incommensurately modulated along the crystallographic *b* axis, and contains Mn^{3+} moments along the *b* axis and *c* axis that belong to two different irreducible representations Γ_3 and Γ_2, respectively [2]. The symmetry of the magnetic structure, described by the direct product of Γ_3 and Γ_2, breaks inversion symmetry and the mirror plane m_{ab}, thereby allowing a ferroelectric polarization only for $\mathbf{P} \parallel \mathbf{c}$ [2]. Similar conclusions can be drawn about other materials [3–5].

Now consider a hypothetical magnetic structure for $TbMnO_3$ that is a spiral with Mn^{3+} moments still in the crystallographic *bc* plane, but with the spin component along the *c* axis belonging to Γ_4 instead Γ_2. This structure is also a *bc*-polarized spiral structure, but the *b* and *c* component have different parallel or antiparallel arrangements between nearest neighbors. The symmetry of this structure is given by the direct product of Γ_3 and Γ_4. Since the relevant magnetic structure is odd under both mirror planes m_{ab} and m_{bc} and even under the twofold rotation 2_b, it cannot support a polar axis: the ferroelectric moment is zero even though inversion symmetry is broken.

In contrast, Mostovoy's theory [1] is based on the stated premise that "incommensurate spin-density-wave states are largely insensitive to details of crystal structure and can be described by a continuum field theory of the Ginzburg-Landau type", and Eq. 5 of his Letter wrongly predicts a ferroelectric polarization along the *c* axis for a *bc*-polarized spiral structure that belongs to Γ_3 and Γ_4. We argue that this wrong conclusion is reached because Mostovoy ignores the symmetry of the "incommensurate spin-density-wave" in this case. To correct this problem inevitably requires taking proper account of the symmetry of the unit cell. This was done previously [2,3] in a systematic way using the crystal structure to analyze the trilinear coupling, $M(\mathbf{q})M(-\mathbf{q})P$, of the incommensurate magnetization $M(\mathbf{q})$ to the spontaneous polarization P. Also, if one discusses why the spontaneous polarization is absent in the incommensurate state which is nearly linearly polarized, representation analysis guarantees that small transverse spin components (potentially precursors to the formation of a magnetic spiral) do not allow ferroelectricity.

The spiral formulation of Mostovoy does not capture the physics of systems in the family of YMn_2O_5, where ferroelectricity can be induced by a coplanar magnetic structure [6]. Whatever the magnetic structure of YMn_2O_5 may be, it is subject to a trilinear magnetoelectric interaction whose symmetry leads to a phenomenological explanation for magnetically induced ferroelectricity [5]. Another example where Mostovoy's approach completely fails is $RbFe(MoO_4)_2$ [7]. This material is a particularly simple magnet whose unit cell contains a single magnetic ion. The magnetic order at zero-field is a simple spiral that propagates along the *c* axis, with a 120° degree angle between nearest-neighbor in the same plane. Electric polarization is observed along the *c* axis, perpendicular to the spiral plane, and is completely unexpected in Mostovoy's approach that predicts the ferroelectric polarization to be in the plane containing the rotating magnetic moments.

An additional virtue of dealing with the symmetry of representations [2–4] is that one sees immediately that perturbations within the representations, such as magnetic components in addition to either the nonferroelectric collinear structure or to the ferroelectric spiral, do not change the symmetry. Case in point is Cs_2CuCl_4 that adopts an incommensurate order of two counterrotating spirals at low temperatures described by one irreducible representation [8] and is thus not expected to be ferroelectric.

In conclusion, we do not agree with Mostovoy's approach to explain multiferroic behavior because it ignores details of the magnetic structures. We present two experimental examples for which Mostovoy's theory fails. These examples unambiguously show that the symmetry of the crystal lattice and of the magnetic order play a crucial role in a phenomenological description of magneto-electric coupling [2,3], and that the continuum symmetry approach that Mostovoy proposes leads to misleading predictions.

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