

THz Magnetolectric Atomic Rotations in the Chiral Compound  $\text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14}$ L. Chaix,<sup>\*</sup> S. de Brion,<sup>†</sup> F. Lévy-Bertrand, V. Simonet, R. Ballou, B. Canals, and P. Lejay  
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We have determined the terahertz spectrum of the chiral langasite  $\text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14}$  by means of synchrotron-radiation measurements. Two excitations are revealed that are shown to have a different nature. The first one, purely magnetic, is observed at low temperature in the magnetically ordered phase and is assigned to a magnon. The second one persists far into the paramagnetic phase and exhibits both an electric and a magnetic activity at slightly different energies. This magnetolectric excitation is interpreted in terms of atomic rotations and requires a helical electric polarization.

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The electric-field control of spins and the converse magnetic-field control of electric dipoles inspire a number of hybrid technologies and motivate fundamental research on multiferroics and magnetoelectric materials [1]. These magnetoelectric couplings produce striking phenomena both at the static and at the dynamical level. A prominent example is the electric-charge dressing of magnons, resulting in the so-called electromagnons, that has been demonstrated experimentally in multiferroics [2,3]. This dressing enables the electric-field control of spin waves and produces enhanced optical responses with potential applications in photonics and magnonics [4]. Here we report experimental evidence of the dual phenomenon, which are atomic vibrations dressed with currents, hence magnetically active, in the terahertz (THz) spectrum of the chiral compound  $\text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14}$ . Based on symmetry arguments, we propose that this new type of magnetolectric excitation is associated with structural rotations; their unexpected magnetoelectric activity is triggered by a spontaneous helical polarization in the paramagnetic phase of the system.

When chirality meets magnetic order, unusual magnetic and electric properties often appear, encouraging the search for novel multiferroic and magnetoelectric phenomena. The highest magnetically induced polarization observed so far has been reported in  $\text{CaMn}_7\text{O}_{12}$  [5] where, interestingly, axial distortions of the lattice play a key role in generating ferroelectricity out of the chiral magnetic structure of the system [6,7]. We hereafter focus on the  $\text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14}$  langasite, which crystallizes within the noncentrosymmetric space group  $P321$  [8] thus displaying *structural* chirality (see Fig. 1). The remarkable magnetic properties of this system are due to the  $\text{Fe}^{3+}$  ions. These form triangles stacked along the  $c$  axis and compose a triangular lattice

in the  $(a, b)$  plane (see Fig. 1). The magnetic exchanges along the  $c$  axis are mediated by twisted Fe-O-O-Fe paths. Below the Néel temperature  $T_N = 27$  K, the system develops a chiral magnetic order with a  $120^\circ$  spin structure within the Fe triangles that rotates along the  $c$  axis and forms magnetic helices [9]. In addition, below  $T_N$ , the system shows a static magnetoelectric effect [10] and chiral spin-wave excitations [11–13]. A weak electric polarization along the  $c$  axis has also been reported [14], although our own measurements show no uniform polarization either along the  $c$  axis nor perpendicular to it.

We have studied  $\text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14}$  by means of synchrotron-radiation measurements in the THz regime. This technique is particularly well suited to investigate magnetolectric excitations [15] because (i) the THz energy

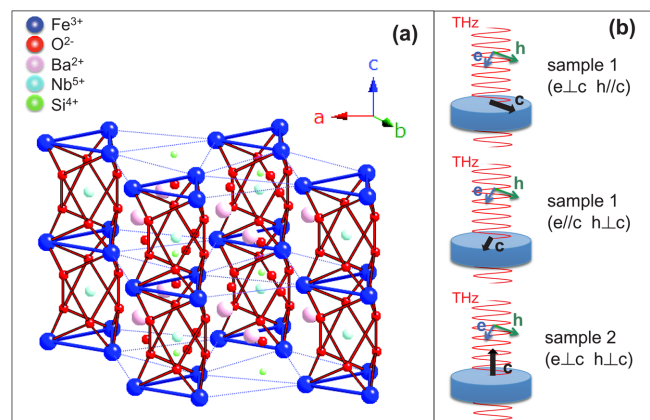


FIG. 1 (color online). (a)  $\text{Ba}_3\text{NbFe}_3\text{Si}_2\text{O}_{14}$  langasite crystallographic structure. (b) Relative orientation of the electric  $\mathbf{e}$  and magnetic  $\mathbf{h}$  fields of the THz wave with respect to the crystal  $c$  axis for samples 1 and 2.

range coincides with that of magnons and phonons and (ii) the THz wave delivered by the synchrotron radiation (98% linearly polarized) carries both an electric and a magnetic field, so that rotating the crystal with respect to the wave polarization direction allows the unambiguous determination of the excitation fields. We have used these selection rules and the temperature dependence of the excitations to elucidate their nature and the mechanisms at play in the dynamics. This is further ascertained by comparison with inelastic neutron scattering measurements.

Two plaquettes of millimeter size were cut from the same single crystal [9] with a thickness of 450 and 300  $\mu\text{m}$ ; the  $\mathbf{c}$  axis was oriented either in plane or out of plane in order to probe all possible geometries of the THz electric and magnetic fields as regards the crystal  $\mathbf{c}$  axis: ( $\mathbf{e} \parallel \mathbf{c} \mathbf{h} \perp \mathbf{c}$ ) and ( $\mathbf{e} \perp \mathbf{c} \mathbf{h} \parallel \mathbf{c}$ ) for one crystal, ( $\mathbf{e} \perp \mathbf{c} \mathbf{h} \perp \mathbf{c}$ ) for the other crystal (see Fig. 1). THz absorption spectra were obtained in the energy range 8–60  $\text{cm}^{-1}$  (0.24–1.8 THz) by measuring the transmission at the AILES beam line of Synchrotron SOLEIL [16]. A Bruker IFS125 interferometer equipped with a pulse tube cryostat was used, combined with a Helium pumped bolometer. The 10–60  $\text{cm}^{-1}$  energy range was explored at a resolution of 0.5  $\text{cm}^{-1}$  using a 6  $\mu\text{m}$  thick silicon-mylar multilayered beam splitter (BMS). The 8–18  $\text{cm}^{-1}$  energy range was explored with more sensitivity by using a 125  $\mu\text{m}$  thick mylar BMS.

The THz spectra measured at various temperatures from 300 to 6 K (for more details, see Supplemental Material [17]) reveal an absorption roughly three times larger for  $\mathbf{e} \parallel \mathbf{c}$  than for  $\mathbf{e} \perp \mathbf{c}$ , meaning that the dielectric response of the material is considerably stronger for this direction of the THz electric field. This additional absorption arises from (multi)phonons excited by  $\mathbf{e} \parallel \mathbf{c}$ , which contribute as a broad background centered at 40  $\text{cm}^{-1}$  growing with increasing temperature, as expected for thermally populated phonons.

Because the magnetic contribution is weak compared to the instrument and phononic background, two different methods were used to enhance the magnetoelectric response of the samples. For the 6  $\mu\text{m}$  BMS measurements, we used the same procedure as in Ref. [18]: we estimated the extrapolated absorption at a given temperature  $T_0$ ,  $\alpha(T_0)$ , as the common signal to the  $\alpha(T_0) - \alpha(T)$  curves for all measurements temperatures  $T$ . A linear background was then subtracted. On the other hand, for the 125  $\mu\text{m}$  BMS measurements, the sample at 45 K was used as a common reference. Interference patterns arising from coherent reflections on the disk surfaces, when present, were removed using a band block FFT filter. The extrapolated absorption for all the different combinations of the THz electric field  $\mathbf{e}$  and magnetic field  $\mathbf{h}$  alignment is displayed on Fig. 2.

The most striking feature is visible for ( $\mathbf{e} \parallel \mathbf{c} \mathbf{h} \perp \mathbf{c}$ ): a large asymmetric absorption is clearly observed that results

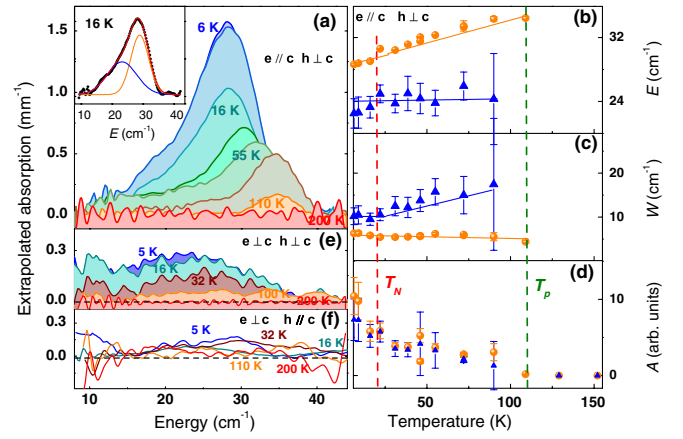


FIG. 2 (color online). THz excitations probed with the 6  $\mu\text{m}$  BMS. (a) Extrapolated absorption spectra for ( $\mathbf{e} \parallel \mathbf{c} \mathbf{h} \perp \mathbf{c}$ ) at different temperatures. Inset: Gaussian fits. (b), (c), (d) Temperature dependence of the fits position, width, and area. (e), (f) Same as (a) for the other THz polarizations.

from two Gaussian contributions [see insert of Fig. 2(a)]. At 16 K, for instance, the two peaks are found at 23 and 29  $\text{cm}^{-1}$  with a respective width of 10 and 5  $\text{cm}^{-1}$ . Rotating the electric field perpendicular to  $\mathbf{c}$  leaves with a single broad absorption centered around 25  $\text{cm}^{-1}$  [see Fig. 2(e)]. When an additional rotation is performed and the magnetic field lies parallel to  $\mathbf{c}$  no clear absorption is observed [see Fig. 2(f)]. These measurements evidence two excitations quite close in energy,  $E_1$  and  $E_2$ , which persist in the paramagnetic phase up to 100 K, that is, almost four times higher than  $T_N$  (see Fig. 2). This is a strong indication that they have a common origin not related to the long range magnetic order although they lie in the magnon energy range. It leads us to introduce a new characteristic temperature in the system,  $T_p \approx 100$  K.

The overall behavior of these excitations presents some discrepancies: the energy of excitation  $E_1$  remains constant while for  $E_2$  it increases at high temperature. The lifetime (inverse of the absorption peak width) starts to decrease above  $T_N$  for excitation  $E_1$  while it remains constant for  $E_2$ . The most remarkable difference concerns their selection rules:  $E_1$  is activated by the magnetic field  $\mathbf{h} \perp \mathbf{c}$ ,  $E_2$  by the electric field  $\mathbf{e} \parallel \mathbf{c}$  [see Figs. 2(a), 2(e), and 2(f)]. Note that both  $E_1$  and  $E_2$  are clearly seen in Fig. 2(a),  $E_1$  is still visible in Fig. 2(e), and no feature is detectable in Fig. 2(f).

Another weaker and even sharper excitation  $E_3$  is found at lower energy for ( $\mathbf{e} \perp \mathbf{c} \mathbf{h} \parallel \mathbf{c}$ ), using the more sensitive 125  $\mu\text{m}$  BMS, resulting from two close Gaussian contributions. At 10 K, they are observed at 12.9 and 14.1  $\text{cm}^{-1}$ , both with a width of 0.8  $\text{cm}^{-1}$  [see Fig. 3(a)]. This excitation shifts to lower energy and broadens as the temperature is increased to finally disappear at 24 K, just below  $T_N$ . Contrary to the previous excitations, the temperature dependence of this excitation is characteristic of a magnon.

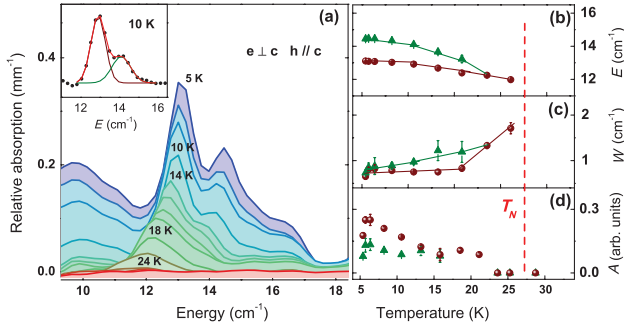


FIG. 3 (color online). THz excitations probed with the 125  $\mu\text{m}$  BMS. (a) Relative absorption spectra,  $\alpha(T)-\alpha(45\text{ K})$  ( $\mathbf{e} \perp \mathbf{c}$   $\mathbf{h} \parallel \mathbf{c}$ ) at different temperatures. Inset: Gaussian fits. (b), (c), (d) Temperature dependence of the fits position, width, and area.

This was confirmed from the comparison with the magnon spectra obtained through inelastic neutron scattering [11–13,19]. The THz measurements probe the first Brillouin zone center (0, 0, 0) where no neutron measurements are available. We therefore compared them to neutron spectra in other zone centers. Selected measurements performed on the time-of-flight IN5 and on the triple-axis IN12 spectrometers at the Institut Laue-Langevin are shown in Fig. 4. Two main branches emerge from the magnetic satellites at  $\pm\tau$  with  $\tau = 1/7$  [20]. The lower energy branch is associated with the dynamical correlation function of spin components along the  $\mathbf{c}$  axis. The upper energy branch is associated with those in the triangle plane. An intense signal is observed around 13  $\text{cm}^{-1}$  at the zone center, mainly from the lower branch [see Fig. 4(b)]. The tail of this signal and the less intense neutron signal up to 25  $\text{cm}^{-1}$  are attributed to the upper branch. The first neutron peak at 13  $\text{cm}^{-1}$  perfectly agrees with the weak excitation  $E3$  observed in the THz measurements in terms of energy and spin component involved (along the  $\mathbf{c}$  axis, hence excitable by  $\mathbf{h} \parallel \mathbf{c}$ ). The other magnonic signals

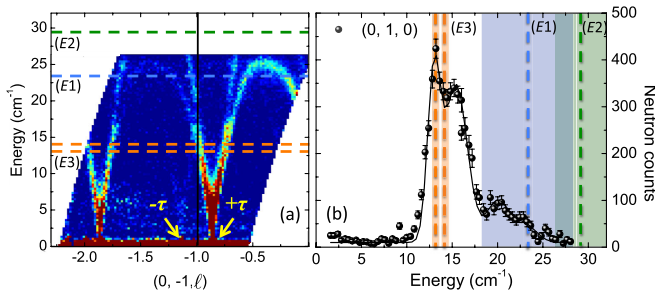


FIG. 4 (color online). Magnons probed by inelastic neutron scattering. (a) Magnons dispersion along the (0,  $-1$ ,  $\ell$ ) direction in the reciprocal space. The black line indicates the Brillouin zone center. (b) Energy scan at the reciprocal lattice zone center (0, 1, 0). The colored dashed lines give the energy position of excitations  $E1$ ,  $E2$ , and  $E3$  seen in THz measurements while the colored rectangles indicate their width.

[involving spin components in the (a, b) plane] are not clearly distinguished in the THz measurements for  $\mathbf{h} \perp \mathbf{c}$ , presumably because the 10 times stronger excitations  $E1$  and  $E2$  mask everything [see Figs. 2(a) and 2(e)]. These remarkable larger THz excitations  $E1$  and  $E2$  are clearly absent from the neutron data of Fig. 4 and also from Refs. [11,12,19].

In multiferroics, magnons can be electroactive at particular points of the Brillouin zone (related to the spiral wave vector or at the Brillouin zone edges) [2,3,21] and these so-called electromagnons can persist in the nonmultiferroic, yet magnetically ordered phases of these compounds [2,3,22]. In contrast, for the langasite compound, the THz excitations  $E1$  and  $E2$  persist up to 100 K  $\approx 4T_N$  when no well-defined magnetic excitation exists anymore [23]. Thus, whereas the THz excitation  $E3$  at 13  $\text{cm}^{-1}$  can be unambiguously identified as a conventional magnon, the magnonic origin of the THz excitations  $E1$  and  $E2$  can be discarded.

These excitations are more likely connected to the lattice [24], although their relatively low energies and the unusual behavior described above is different from that of standard phonons. The natural candidates to explain these features are axial distortions of the lattice, whose role has been recently highlighted in this class of materials [6,25,26]. These are rotations  $R_z$  about the  $\mathbf{c}$  axis whose interaction with the electric and magnetic fields of the external radiation is described by the effective Lagrangian:

$$L_{\text{int}} = \frac{\alpha}{\omega_{P_z}^2} [e_z + g(\mathbf{P}_{\perp} \times \dot{\mathbf{h}}_{\perp})_z] R_z. \quad (1)$$

This interaction results from the linear coupling between rotations and polar displacements allowed by the  $P321$  space-group symmetry of our system (see Supplemental Material for details on the derivation of this Lagrangian [17]). Here  $\alpha$  represents the strength of this coupling,  $\mathbf{P}$  is the electric polarization, with  $\omega_{P_z}$  being its characteristic frequency (or energy), and  $g$  is a gyromagnetic factor.

The excitation of  $R_z$  by the electric-field component of the radiation observed in our THz measurements is described by the first term in Eq. (1). This follows immediately from the aforementioned linear coupling between  $R_z$  and  $P_z$ . The additional response of  $R_z$  to the magnetic field is due to the second term in Eq. (1). This response, in contrast, is far more subtle. It requires a finite, static polarization perpendicular to the  $\mathbf{c}$  axis,  $\mathbf{P}_{\perp} \neq 0$ , to be operative. At room temperature, no perpendicular polarization is allowed by the symmetry. However, at low temperatures, recent neutron scattering results are not compatible with a perfect  $120^\circ$  spin structure, implying the loss of the threefold symmetry axis [23]. This  $P321 \rightarrow C2$  symmetry breaking implies the appearance of a spontaneous polarization perpendicular to the  $\mathbf{c}$  axis  $\mathbf{P}_{\perp}$  that, in turn, activates the coupling of  $R_z$  with the magnetic field in

Eq. (1). This is in perfect agreement with the gradual appearance of the magnetoactive excitation  $E1$  that we observe at  $24 \text{ cm}^{-1}$  below  $100 \text{ K}$  (see Fig. 2). The concomitant appearance of the electroactive excitation  $E2$  at  $100 \text{ K}$  can be explained as due to the increase of the overall effective coupling  $\alpha/\omega_{P_z}^2$  at the onset of the new phase. This increase is likely a by-product of the softening of  $\omega_{P_z}$  due to its dependence on  $\mathbf{P}_\perp$ . In fact, we observe a similar softening in the energy of the electroactive excitation itself that is in tune with this hypothesis [see Fig. 2(b)].

Finally, we note that in our spectra the energy of the electroactive and magnetoactive excitations are slightly different. This difference suggests that the spontaneous polarization that appears below  $100 \text{ K}$  is modulated with a finite wave vector  $Q$ . Thus, according to Eq. (1), the electric field couples to  $q = 0$  uniform rotations while the magnetic field does to  $q = \pm Q$  rotations whose resonant energy is lower (see Fig. 5). The detailed Ginzburg-Landau analysis of the  $P321 \rightarrow C2$  transition confirms this possibility. The perpendicular polarization  $\mathbf{P}_\perp$  can be seen as the order parameter driving the  $P321 \rightarrow C2$  transition. The Ginzburg-Landau free energy then can be written in terms of  $(P_x, P_y) = (\rho \cos\theta, \rho \sin\theta)$  as:

$$F = \frac{a}{2}\rho^2 + \gamma\rho^3 \cos 3\theta + \frac{b}{4}\rho^4 - \lambda\rho^2(\partial_z\theta) + \frac{c}{2}[(\nabla\rho)^2 + \rho^2(\nabla\theta)^2]. \quad (2)$$

Here, as is customary,  $a$  is assumed to be the only coefficient that depends on the temperature and changes sign as  $a = a'(T - T_0)$ , where  $T_0$  can be seen as the nominal transition temperature. If the space variations of the order parameter are ignored, then the cubic term in the free energy implies rather a first-order transition at  $T'_0 = T_0 + 2\gamma^2/(a'b)$ . The presence of the Lifshitz invariant  $-\lambda\rho^2(\partial_z\theta)$ , however, leads to a second-order transition at higher temperature, which sets up helical spontaneous polarization  $(P_x, P_y) = (P_0 \cos Qz, P_0 \sin Qz)$  at  $T_p = T_0 + \lambda^2/(a'c)$  (see Fig. 5). Here  $P_0^2 = |T - T_p|/(a'b)$  and  $Q = \lambda/c$ . This will actually be the case if  $2\gamma^2/b < \lambda^2/c$  (that is, if the expected discontinuity at the first-order transition is relatively weak). This type of helical polarization has been discussed theoretically in the context of incommensurate phases, although no experimental realization has been reported so far [27]. Here  $T_p$  is different from the direct  $P321 \rightarrow C2$  transition temperature, which is avoided due to the formation of the intermediate helical state. The  $C2$  symmetry is obtained when the helix becomes commensurate. This is expected to take place at a lower temperature, which may well coincide with the magnetic transition temperature.

The essential ingredients that give rise to the magneto-electric excitations in our system are also present in the newly reported ferroaxial multiferroic  $\text{CaMn}_7\text{O}_{12}$  and related compounds [5–7,26]. There, the linear coupling

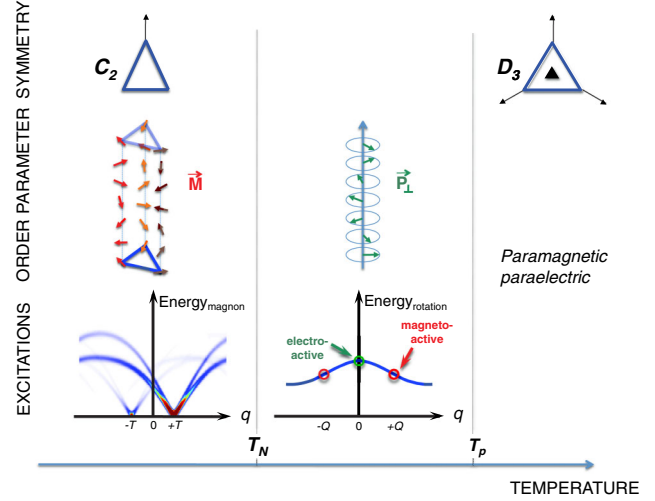


FIG. 5 (color online). Temperature evolution of the electric and magnetic phases. From top to bottom: symmetry, order parameter, and corresponding excitations. From right to left (decreasing temperature):  $T > T_p$  paramagnetic and paraelectric state,  $T_p > T > T_N$  helicoidal state of polarization,  $T_N > T$  additional helicoidal magnetic state. The  $P321$  to  $C2$  symmetry breaking is schematized by the loss of the  $C_3$  axis in the  $\text{Fe}^{3+}$  triangles (from equilateral to isosceles). The curvature of the dispersion curve  $E_{\text{rot}}(q)$  is deduced from the relative energies of the magnetic and electric active peaks  $E1$  and  $E2$ .

between the electric polarization and macroscopic structural rotations [6,26], is preceded by a structural helix due to the incommensurate modulation of the atomic bonds [7], in  $\text{CaMn}_7\text{O}_{12}$  at least. However a chiral magnetic order is necessary to activate the polar-axial coupling via the trilinear invariant  $\sigma P_z R_z$ , where  $\sigma$  represents the chirality of the magnetic structure. In the langasites, the lattice itself is chiral, so that the polar-axial coupling is “switched on” already in the paramagnetic phase. Thus, while structural rotations in both these systems can be expected to have similar magnetoelectric responses, they can reveal different key aspects associated to structural vs magnetic chirality.

In summary, our THz investigation of the langasite dynamical properties has revealed a new kind of excitation, associated with atomic vibrations, which is both electric and magnetic active, at slightly different energies. These findings demonstrate that not only magnons but also atomic vibrations can acquire a magnetoelectric character, thus opening new routes to carry and process information. This striking magnetoelectric activity has been related to the bilinear coupling between the atomic rotations allowed by the chiral symmetry of our system and a perpendicular electric polarization. The predicted helical polar order may itself exhibit original properties such as dielectric chirality and sets off new magnetoelectric functionalities that can be common among symmetry-equivalent systems.

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