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Single-crystal neutron diffraction study of hexagonal multiferroic YbMnO₃ under a magnetic field

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We report a single-crystal neutron diffraction study of the magnetic structure of the multiferroic compound YbMnO₃, a member of the hexagonal manganite family, in zero field and under a magnetic field applied along the c axis. We propose a scenario for the zero-field magnetic ordering and for the field-induced magnetic reorientation of the Mn atom and of the two Yb atoms on distinct crystallographic sites, compatible with the macroscopic measurements, as well as with previous powder neutron diffraction experiments and results from other techniques (optical second-harmonic generation and Mössbauer spectroscopy). Our study should contribute to settling some debated issues regarding the magnetic properties of this material as part of a broader investigation of the entire hexagonal RMnO₃ (R = Dy, Ho, Er, Tm, Yb, Lu, Y) family.

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I. INTRODUCTION

The hexagonal h-RMnO₃ multiferroic compounds (with $R = \text{Dy, Ho, Er, Tm, Yb, Lu, Y}$) have been the subject of an abundant amount of literature since their discovery in 1963 [1]. This interest is due to their exotic static and dynamical behaviors ascribed to the combination of ferroelectricity and magnetic frustration. The latter arises from the intraplane triangular arrangement of antiferromagnetically interacting Mn$^{3+}$ magnetic ions, weakly coupled along the c axis, as shown in Fig. 1. Unlike multiferroics, where the ferroelectricity is induced by the magnetic order, as in the orthorhombic $\text{RMnO}_3$ compounds (with larger rare-earth ions), h-$\text{RMnO}_3$ oxides become ferroelectric at much higher temperatures (around 1000 K) than their magnetic transition (Néel temperature $T_N$ below 100 K). In h-$\text{YMnO}_3$, ferroelectricity was found to be connected to the buckling of the layered MnO₃ polyhedra, displacements of the Y ions, and the trimerization of the Mn lattice associated with strong magnetoelastic effects [2–9]. Although the exact mechanism at the origin of the ferroelectricity has been debated, it is assumed to be identical in all members of the family, which are described in the hexagonal space group $P6_3cm$ (number 185) at low temperature.

The magnetism is a complex issue in itself in this class of materials. Mn$^{3+}$ ions occupy the Wyckoff site $6c$, forming triangular layers of Mn$^{3+}$ in the $(a, b)$ planes. Between them, $R^{3+}$ occupies two different crystallographic Wyckoff sites, $4b$ and $2a$. While decreasing the temperature, first the Mn$^{3+}$ order magnetically at $T_N$ due to superexchange antiferromagnetic interactions in a 120° magnetic structure, characterized by a $k = (0, 0, 0)$ propagation vector [1], and then $R^{3+}$ ($4b$) are polarized in the molecular field of the Mn, while $R^{3+}$ ($2a$) are believed to order at much lower temperature from their mutual interactions [10]. Additional spin reorientations occur at intermediate temperatures for some members of the family (e.g., Ho, Sc). Finally, metamagnetic processes are frequently observed under magnetic field. Magnetoelastic coupling is also evidenced by strong dielectric anomalies visible at each magnetic transition [11]. The major tools that have been used to determine the various magnetic configurations of h-$\text{RMnO}_3$ are neutron diffraction and optical second-harmonic generation (SHG). They often, but not always, agree. For the unpolarized neutron scattering method, difficulties arise from the indetermination among different possible magnetic structures (homometric pairs) [12], whereas the SHG technique has difficulty distinguishing different magnetic sublattices. In addition, neutron scattering has often been performed only on polycrystalline samples, which is usually not sufficient to determine field-induced magnetic structures.

In this paper, we concentrate on YbMnO₃, which was reported to undergo a magnetic transition below $T_N \sim 80$ K with a propagation vector $k = (0, 0, 0)$ [10,13]. Powder neutron diffraction [14] and SHG [15,16] agree on the high-temperature magnetic ordering. However, the interpretation of the low-temperature magnetic configuration, in particular...
when Yb (2a) atoms are expected to play a role, is not clear, and neither are the exact mutual orientations of the three magnetic ions under magnetic field. Moreover, hysteretic effects, the coexistence of competing phases even in zero field or a spin reorientation at intermediate temperature, have also been reported [16–18], justifying the necessity of a single-crystal neutron diffraction experiment on this composition.

II. SYNTHESIS AND EXPERIMENTAL DETAILS

The platelike single crystals of YbMnO₃, with the hexagonal c axis perpendicular to the surface and a thickness of about 0.5 mm, were grown using the flux method as described by Yen et al. [19]. The magnetization M measurements were performed in a commercial superconducting quantum interference device magnetometer and in a physical property measurement system from Quantum Design. Electric polarization P was measured along the crystallographic c axis using a Keithley 6517A electrometer. Electrical contacts were attached by Ag paint to the two parallel surfaces of the c-cut sample. The measurements were performed for both zero and nonzero (±1 kV/mm) applied electric field regimes. These have not revealed a significant difference, implying the absence of Ohmic currents. Note that the latter were observed in YMnO₃ at temperatures above 230 K and were accompanied by a negative magnetoresistive effect [20]. No preliminary poling was carried out to polarize the sample possessing a ferroelectric domain structure. Neutron diffraction measurements in zero magnetic field were performed on the CEA-CRG D15 and D23 single-crystal diffractometers at the Institut Laue-Langevin (ILL; wavelengths λ = 1.173 and 1.27 Å, respectively) in the four-circle mode using a standard orange cryostat. The CEA-CRG D23 diffractometer at the ILL was also used for the measurements under magnetic field using a lifting-arm detector and a vertical cryomagnet. The zero-field measurements on D15 and D23 were determined to be consistent, and only results from D23 are presented in the following.

III. IRREDUCIBLE REPRESENTATION ANALYSIS

Let us recall that group theory and representation analysis [21] reveal that the representation of the magnetic structure for Mn₃⁺ and Yb₃⁺ (4b) involve six possible irreducible representations (IRs), denoted as Γᵢ (i = 1, 2, ..., 6), which are compatible with the P̅6₃cm space group and the propagation vector k = (0, 0, 0) [22]. Four of these, Γ₁ to Γ₄, are unidimensional and are depicted in Fig. 2, with the last two being two-dimensional. For the Yb₃⁺ (2a) site, only the Γ₂, Γ₃, Γ₅, and Γ₆ irreducible representations are allowed.

Concerning the Mn₃⁺ site, the magnetic moments are constrained in the (a, b) plane for Γ₁ and Γ₄, while an out-of-plane ferromagnetic component is allowed for Γ₂ and an out-of-plane antiferromagnetic component is allowed for Γ₃. Among these configurations, those with symmetries Γ₁ and Γ₃ correspond to homometric pairs, and so do Γ₂ and Γ₄. This means that these pairs are hardly distinguishable (almost identical intensities of the magnetic Bragg reflections) by neutron diffraction when the Mn₃⁺ coordinate xᵣₐₐₜ is close to 1/3. For the two-dimensional representations Γ₅ and Γ₆, the Fourier components of the magnetic moments are written as a linear combination of six basis functions. The corresponding magnetic structures are reported in Ref. [22]. For Γ₅, there are four magnetic modes with the magnetic moments in the (a, b) plane, displaying either a ferromagnetic or a 120° arrangement. The coupling is ferromagnetic between the planes. The two other modes are discarded since they concern nonequal moments along the c axis. Similar solutions are found for Γ₆ but with an antiferromagnetic coupling of the in-plane magnetic structures along the c axis. Concerning the Yb₃⁺(4b)
and Yb$^{3+}$($2a$) sites, the one-dimensional IRs correspond to magnetic moments along the $c$ axis. Importantly, only the $\Gamma_2$ configuration allows a ferromagnetic component along the $c$ axis for the three sites (see Fig. 2).

IV. RESULTS

A. Magnetization and electric polarization

Figure 3(a) depicts the temperature $T$ variation of the dc magnetic susceptibility $\chi$ along the $c$ axis in a magnetic field of 1 kOe along the $c$ direction ($\chi_c$) and within the $(a, b)$ plane ($\chi_{ab}$), recorded on cooling. An anisotropic response is seen, apparently below the Néel temperature, with the $\chi_c/\chi_{ab}$ ratio value reaching $\sim 15$ at 2 K. The data well replicate the results reported in previous studies [10,13,14,19,23]. The anomaly near 80 K ($T_N$) in the $\chi_c$ data [inset of Fig. 3(a)] corresponds to the ordering of Mn$^{3+}$ magnetic moments, as reported earlier. The sharp increase in $\chi_c$ below about 5 K [Fig. 3(b)] signals the onset of long-range ordering of the Yb$^{3+}$ ($2a$) moments. The differences in the zero-field- and field-cooled thermomagnetic curves below about 4 K reflect the existence of net magnetic moment and coercivity.

The field dependence of the magnetization for various temperatures is shown in Figs. 3(c) and 3(d) for fields applied perpendicular and parallel to the $c$ axis, respectively, and is again consistent with previous reports [10,19,24–26]. No anomaly or hysteretic behavior is measured for the magnetic field applied along the $(a, b)$ plane. This is at variance with the behavior observed for the other orientation of the field where two steplike features are visible in the magnetization curve at low temperature. The low-field magnetization step is observed only for temperatures below about 5 K. As seen on the 2 K curve [Fig. 3(e)], the magnetization reaches $\sim 0.4 \mu_B/\text{f.u.}$ almost instantly after the magnetic field is increased (as soon as the applied field reaches $H_{c1} \sim 300$ Oe, which means that the internal field is even smaller). The magnetization step value and the magnetization hysteresis seen in Fig. 3(e) point to ferrimagnetic alignment between Yb$^{3+}$ ($4b$) and Yb$^{3+}$ ($2a$) moments. The second steplike anomaly appears near $H_{c2} = 30$ kOe for the 2 K curve. At this field, a jump in magnetization takes place, and it increases with about 1.2 $\mu_B/\text{f.u.}$ above its low-field step value of $\sim 0.4 \mu_B/\text{f.u.}$ and becomes closer to the $(a, b)$ plane magnetization. This steplike feature actually reveals a field-induced spin reorientation that is addressed through our neutron diffraction measurements presented below. It smears out with increasing temperature along with the shift of $H_{c2}$ towards higher field.

The field dependence of the electric polarization change $\Delta P_c(H)$ along the $c$ axis is presented in Fig. 4 for several temperatures. The magnetic field was applied along the same direction. A sharp jump in the polarization is observed at $\sim 30$ kOe at the lowest temperatures. It is clearly correlated to the field-induced magnetic phase transition since its threshold field coincides with the second steplike anomaly in the magnetization curves at $H_{c2}$. The polarization anomaly smears out with increasing temperature. Its field dependence changes character between $T_c \sim 4$ and 6 K, revealing a broad maximum in $\Delta P_c(H)$ at higher temperatures. These modifications are probably associated with a change in the nature of the field-induced transition above and below the Yb$^{3+}$ ($2a$) ordering at $T_c$. 

FIG. 3. (a) The dc magnetic susceptibility $\chi$ along the $c$ axis and in the $(a, b)$ plane. Measurements were performed in a magnetic field of 1 kOe on cooling. Inset: $d\chi_c/dT$ vs $T$ to show the transition at $T_N = 80$ K. (b) $\chi(T)$ measured with a field of 100 Oe along the $c$ axis after field-cooling and zero-field-cooling procedures, showing the low-temperature anomaly associated with the Yb$^{3+}$ ($2a$) ordering. Magnetization $M$ vs applied magnetic field $H$ for $H$ in the $(a, b)$ plane (c) and along $c$ (d). (e) The low-field $M(H)$ curve at 2 K for $H \parallel c$. The data are not corrected for the demagnetizing field effect.

FIG. 4. Change in electric polarization $\Delta P_c$ along the $c$ axis as a function of magnetic field $H$ applied along the same direction (measured in an applied electric field of 1 kV/mm).
TABLE I. Results of the refinement of the single-crystal neutron diffraction Bragg peak intensities recorded above (100 K) and below (30, 13, and 2 K) the transition temperature \((T_N = 80 \text{ K})\) under the zero-magnetic-field condition. \(R_F, R_p,\) and \(R_{RF}\) are the agreement factors of the fits \([27]\), \(M\) is the magnetic moment, and \(x_{\text{Mn}}\) is the \(x\) coordinate of \(\text{Mn}^{3+}\).

<table>
<thead>
<tr>
<th>Temperature (K)</th>
<th>(x_{\text{Mn}})</th>
<th>Ion/IR</th>
<th>(M) (units of (\mu_B))</th>
</tr>
</thead>
<tbody>
<tr>
<td>100</td>
<td>0.3345(20)</td>
<td>(\text{Mn}^{3+}/\Gamma_4)</td>
<td>3.32(3)</td>
</tr>
<tr>
<td>100 (30)</td>
<td>0.3343(14)</td>
<td>(\text{Yb}^{3+} (4b)/\Gamma_4)</td>
<td>0.81(5)</td>
</tr>
<tr>
<td>13</td>
<td>0.3350(15)</td>
<td>(\text{Mn}^{3+}/\Gamma_4)</td>
<td>3.41(4)</td>
</tr>
<tr>
<td>2</td>
<td>0.3349(15)</td>
<td>(\text{Yb}^{3+} (4b)/\Gamma_4)</td>
<td>1.15(5)</td>
</tr>
</tbody>
</table>

B. Single-crystal neutron diffraction

1. In zero magnetic field

About 700 Bragg reflections were recorded at 100, 30, 13, and 2 K [the magnetic Bragg peaks associated with the \(k = (0, 0, 0)\) propagation vector rise above the nuclear ones]. All the refinements were performed using the FULLPROF SUITE software package \([27]\), and we used the formalism of Becker-Coppens to refine the anisotropic extinction \([28]\). The refined atomic positions of the 100 K data (above \(T_N\)) were found to be very similar to the ones reported from x-ray diffraction at room temperature \([29,30]\). In particular, the \(z\) coordinate of \(\text{Mn}^{3+}\) is almost zero and is thus kept to zero for the lower-temperature refinements. The \(x\) coordinate of \(\text{Mn}^{3+}\) \(x_{\text{Mn}}\) is very close to 1/3 (see Table I), but it was allowed to vary in the lower-temperature refinements as its variation is believed to be intrinsically linked to the selection of the magnetic order \([31]\).

To get deeper insight into the contribution of the different sublattices to the magnetic order in zero field, we chose to study the thermal evolution of some characteristic site-specific reflections (see Fig. 5). We followed the \((1, 0, 1)\) and \((1, 0, 3)\) reflections, which are forbidden in the \(P6_2cm\) space group. As can be seen, they start to become nonzero only below \(T_N\), reflecting their magnetic character, and have a temperature dependence compatible with that of a magnetic order parameter. Under \(\Gamma_2\) and \(\Gamma_4\) symmetry, those two reflections are entirely dependent on the ordering of \(\text{Mn}^{3+}\) magnetic moments. Therefore, the rise of these reflections below \(T_N\) is consistent with the coincidence of the Néel temperature with the magnetic ordering of \(\text{Mn}^{3+}\) in one of these two IRs. In contrast, the \((-1, 0, 0)\) reflection shown in Fig. 5(e) depends, for its magnetic component, only on the \(\text{Yb}^{3+}\) magnetic moments for \(\Gamma_2\) (2a and 4b sites) and \(\Gamma_4\) (only the 4b site). The intensity of this reflection increases very slowly when decreasing the temperature down to \(\sim 20–30 \text{ K}\), below which it shows a sharp increment. Such behavior is consistent with the magnetic ordering of \(\text{Yb}^{3+} (4b)\) in the \(\text{Mn}^{3+}\) molecular field \([14]\). Note that, as the transition at \(T_N\) is second order, the two \(\text{Mn}^{3+}\) and \(\text{Yb}^{3+} (4b)\) sublattices must order with the same IR (\(\Gamma_2\) or \(\Gamma_4\)) since they are coupled. No anomaly can be seen in \((-1, 0, 0)\) below \(T_N\). Two other characteristic reflections, viz., \((1,0,2)\) and \((-2, -1, 0)\), combine the features of the previous two kinds of reflections, thus involving magnetic contributions from both \(\text{Mn}^{3+}\) and \(\text{Yb}^{3+}\).

We checked the information deduced from the temperature dependence of selected reflections through the refinement of all the Bragg reflections below \(T_N\). To obtain the best refinements, all the possible one-dimensional IRs predicted for \(\text{Mn}^{3+}\) and \(\text{Yb}^{3+}\) were tested. In addition to the magnitude of the magnetic moments, the \(x\) coordinate of the \(\text{Mn}^{3+}\) ions was also refined. At 30 and 13 K, the best refinements with

![FIG. 5. Neutron counts at the peak maximum versus temperature for three types of Bragg reflections. They are site specific for the magnetic arrangements corresponding to \(\Gamma_2\) and \(\Gamma_4\): (a) and (b) \((1, 0, 3)\) and \((1, 0, 1)\) contributed solely by ordered \(\text{Mn}^{3+}\), (c) and (d) \((1, 0, 2)\) and \((-2, -1, 0)\) contributed by all \(\text{Mn}^{3+}\), \(\text{Yb}^{3+} (4b)\), and \(\text{Yb}^{3+} (2a)\), and (e) \((-1, 0, 0)\) contributed solely by \(\text{Yb}^{3+} (4b)\) and \(\text{Yb}^{3+} (2a)\). Note that the \(\Gamma_4\) configuration is not allowed by symmetry for the \(\text{Yb}^{3+} (2a)\) magnetic order.](image-url)
factors and magnetic configurations for the three sites are indicated.

The agreement factor is only slightly better for the $\Gamma_4$ solution for all sites versus the solution with $\Gamma_4$ for Mn$^{3+}$ and Yb$^{3+}$ (4$b$) and $\Gamma_2$ for Yb$^{3+}$ (2$a$), the former is more consistent with the magnetization data, yielding almost $0.5 \mu_B$/f.u., as shown in Fig. 3(d). Indeed, the total ferromagnetic component deduced from the neutron refinement for the $\Gamma_2$ solution is found to be equal to $0.68(8) \mu_B$/f.u., instead of $0.06(3) \mu_B$ obtained for the $\Gamma_4$/ $\Gamma_2$ solution. We have also tested the possibility of an in-plane configuration of Yb$^{3+}$ (2$a$) magnetization that had been suggested [14] but found worse agreement with our data. The results of the fits are shown in Fig. 6, with the associated magnetic configuration given in Fig. 7, and the details of the important refined parameters are given in Table I.

2. Under magnetic field

Figure 8 depicts the magnetic field dependence of characteristic reflections measured at 1.5 and 4.5 K, i.e., below and close to the transition $T_c$, with a field applied along the $c$ axis. The results are identical: The $\Gamma_4$ and $\Gamma_2$ for Mn$^{3+}$/Yb$^{3+}$ (4$b$) (homometric Mn$^{3+}$ IR), with an enhancement of the Yb$^{3+}$ (4$b$) moment from 30 to 13 K and no magnetic moment on the Yb$^{3+}$ (2$a$) sites. However, in the $\Gamma_2$ magnetic configuration, the Yb$^{3+}$ (4$b$) are ferromagnetically coupled. From the neutron refinement, they should give rise to a ferromagnetic contribution amounting to $\sim 0.7 \mu_B$ at 30 K and $\sim 0.9 \mu_B$ at 13 K, which is not observed in the magnetization measurements. A small bifurcation of the field-cooled and zero-field-cooled susceptibilities has been reported [13] below $T_N$, but it cannot be accounted for by such a large magnetization and is rather due to field-polarized defective magnetic moments, for instance, in the domain walls [19]. Our finding thus points to the $\Gamma_4$ magnetic configuration for Mn$^{3+}$ and Yb$^{3+}$ (4$b$) (see Figs. 6 and 7), which is consistent with previous powder neutron diffraction [14] and SHG results [15]. The good quality of the fits obtained using $\Gamma_4$/ $\Gamma_2$ configurations allowed us to limit our analysis to solutions corresponding to one-dimensional IRs over the more complex two-dimensional ones.

Mössbauer and far-infrared spectroscopies have proven the ordering of the Yb$^{3+}$ (2$a$) below $\sim 5$ K [14,32,33]. Moreover, the onset of a ferromagnetic component along the $c$ axis is associated with this ordering, which suggests that the Yb$^{3+}$ (2$a$) moments order in the $\Gamma_2$ IR. The last uncertainty concerns the ordering of Mn$^{3+}$ and Yb$^{3+}$ (4$b$) below $T_c$. Do they remain in the $\Gamma_4$ IR, or do they reorient due to a coupling with the Yb$^{3+}$ (2$a$) in the $\Gamma_2$ IR? We checked both possibilities: Although the agreement factor is only slightly better for the $\Gamma_4$ solution for all sites versus the solution with $\Gamma_4$ for Mn$^{3+}$ and Yb$^{3+}$ (4$b$) and $\Gamma_2$ for Yb$^{3+}$ (2$a$), the former is more consistent with the magnetization data, yielding almost $0.5 \mu_B$/f.u., as shown in Fig. 3(d). Indeed, the total ferromagnetic component deduced from the neutron refinement for the $\Gamma_2$ solution is found to be equal to $0.68(8) \mu_B$/f.u., instead of $0.06(3) \mu_B$ obtained for the $\Gamma_4$/ $\Gamma_2$ solution. We have also tested the possibility of an in-plane configuration of Yb$^{3+}$ (2$a$) magnetization that had been suggested [14] but found worse agreement with our data. The results of the fits are shown in Fig. 6, with the associated magnetic configuration given in Fig. 7, and the details of the important refined parameters are given in Table I.
FIG. 7. Magnetic structures (and corresponding IR) refined in zero magnetic field at (a) 13 K and (b) 2 K. (c) Magnetic structure inferred from the neutron and magnetization data in a 60-kOe field at 2 K. The Mn$^{3+}$ moments are represented in red, the Yb$^{3+}$ (4b) in moments are blue, and the Yb$^{3+}$ (2a) moments are in yellow.

$= 13.7% / 15.4% / 18.7%$ and $13.6% / 14.5% / 24.3%$, respectively. At 60 kOe, i.e., above $H_{c2}$, the same configuration does not hold well any longer, as evidenced by worse agreement factors ($R_F / R_{F}^{2} / R_{F,w} = 24.2% / 17.9% / 43.8%$). Those improve significantly ($R_F / R_{F}^{2} / R_{F,w} = 12.7% / 11.8% / 25%$) by reversing the Yb$^{3+}$ (2a) magnetic moments and making them parallel to the Yb$^{3+}$ (4b) ones (see bottom panel of Fig. 6), hence validating the model. It should be noted that the strong decrease in the (1, 0, 0) Bragg reflection above $H_{c2}$, as depicted in Fig. 8, is obtained within this model (where all the Yb$^{3+}$ magnetic moments are aligned along the field) due to the destructive interference between the two Yb$^{3+}$ sublattices.

At 10 K ($T_e < T < T_N$) the same set of reflections was recorded at field values of 4 T and at 8 T, i.e., below and above the critical field corresponding to $H_{c2}$ for this temperature [see Fig. 3(c)]. Again, a full refinement is impossible, but the data unambiguously show that the Yb$^{3+}$ (4b) and Yb$^{3+}$ (2a) moments order in the $\Gamma_2$ IR and are aligned parallel to each other and to the field for both values of the field. It is impossible to determine whether the Mn$^{3+}$ magnetic moments are in $\Gamma_1$ or $\Gamma_2$. At 8 T, the data are compatible with the same configuration as the one inferred from the data at 2 K and 6 T which corresponds to the full polarization of the two Yb$^{3+}$ sites. Note that the low-field phase with both Yb$^{3+}$ moments antiparallel to each other does not seem to exist at 10 K.

C. Summary and discussion

In summary, we have achieved a good description of the magnetic properties of the h-YbMnO$_3$ compound through our single-crystal neutron diffraction study. We have found that the Mn$^{3+}$ magnetic moments order below $T_N = 80$ K, polarizing the Yb$^{3+}$ (4b) moments, whose ordered component strongly increases below 20–30 K. Both Mn$^{3+}$ and Yb$^{3+}$ (4b) moments are described with respect to $\Gamma_4$ in this temperature region. Below 5 K, the Yb$^{3+}$ (2a) moments order in $\Gamma_2$, dragging Mn$^{3+}$ and Yb$^{3+}$ (4b) into a new kind of magnetic configuration also corresponding to $\Gamma_2$. It consists of a ferromagnetic arrangement of the Yb$^{3+}$ (2a) and Yb$^{3+}$ (4b) moments along c in a way that the Yb$^{3+}$ (2a) and Yb$^{3+}$ (4b) magnetic sublattices are antiparallel to each other. There happens also to be an important spin reorientation of the in-plane Mn$^{3+}$ moments by 90°, as well as an additional change in its interlayer coupling from antiferromagnetic to ferromagnetic. Under a magnetic field applied along
the c axis, our magnetization and neutron diffraction data are compatible with a spin flip of the Yb$^{3+}$ (2a) moments that become aligned with the field and with the Yb$^{3+}$ (4b) magnetic moments. This correlates well with the observed steplike change in the electric polarization accompanying the field-induced magnetic transition. These polarization changes could originate from small alterations in the atomic/electronic positions due to spin-lattice coupling together with a change in Yb-Mn and Yb-Yb magnetic interactions.

The H-T phase diagram described above is consistent with the SHG results and very similar to the one reported for the h-ErMnO$_3$ compound [34]. It has been rationalized phenomenologically through the Landau theory of phase transition [35]. An important microscopic parameter seems to be the Mn-rare-earth coupling. In the present study, it is evidenced through the polarization of the Yb$^{3+}$ (4b) moments by the Mn$^{3+}$ ones and through the reorientation of the Mn$^{3+}$ moments triggered by the Yb$^{3+}$ (2a) magnetic ordering. This coupling also has signatures in the dynamical properties of YbMnO$_3$ [33,36,37], whereas it confers its electroactivity to a magnon in ErMnO$_3$ [38]. A puzzling issue remains in the mutual orthogonal orientation of the Mn$^{3+}$ and Yb$^{3+}$ magnetic moments which excludes a coupling mechanism by isotropic exchange interactions and calls for subtler mechanisms. The deviation of the $x_{Mn}$ coordinate from 1/3 has also been proposed as an important parameter in the selection of the Mn$^{3+}$ magnetic configuration, either by triggering the sign of the interlayer Mn$^{3+}$ magnetic effective interaction [31] or by determining the orientation of Mn$^{3+}$ within the (a, b) plane through spin-lattice coupling [39]. In our study, this parameter does not seem to vary significantly with the temperature, impeding a definite conclusion on this issue.

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