Interface effects in combined soft/hard ferrite permanent magnets

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Abstract

We address the detailed description of the magnetic properties of the (001) and (111) interfaces between the hard CoFe₂O₄ and soft NiFe₂O₄ spinel ferrites, used at bicomponent systems for permanent magnet applications. The similarity between the electronic properties, magnetic order and lattice structure of both oxides allows to isolate the effect of ideal interface formation on the magnetism of the combined system based on density functional theory. While the magnetic moments and long-range magnetic order preserve bulk-like values, the magnetic anisotropy is reduced by the presence of the interface, and depends on the specific termination. Partial inversion of CoFe₂O₄ is also relevant, as it enhances the net magnetization and alters both the magnitude and spatial dependence of the magnetic anisotropy.

Keywords: CoFe₂O₄, NiFe₂O₄, interface, density functional theory, magnetic anisotropy

1. Introduction

The combination of hard and soft magnetic materials, characterized respectively by their large and weak magnetic anisotropies, enables interesting applications in a wide diversity of areas: permanent magnets, magnetic recording media, microwave absorption, ferrofluids or biomedical solutions [1-4]. The magnetic energy product of permanent magnets, the key parameter that determines their efficiency, depends on the saturation magnetic polarization and the coercivity, which in turn originate from the magnetic anisotropy and the ferromagnetic ordering. Combining hard and soft magnets is intended to retain the high saturation magnetization of the soft material and the high coercivity of the hard one, in order to overcome the magnetic energy product of the individual counterparts.

Different studies indicate that it is possible to fabricate nanostructures where this goal is achieved [2,5-7], even though the details of the preparation procedure that determine the shape, size and morphology of the nanostructures are crucial. In addition to these features, also those at the interface between both materials (structural quality, lattice matching, strain, magnetic coupling between components) play an important role in the final properties of the system. From the experimental point of view, access to the interface properties is difficult, and often failure in the magnetic response emerges from defects at the interface. However, there are hints that complex interface interactions can be tuned to control the measured magnetic response of composite nanoparticles [8]. A detailed analysis of the interface properties needs to be addressed, and first principles methods are particularly suitable for this purpose.

Here, based on density functional theory, we study the magnetic properties at the interface between two inverse spinel ferrites, hard CoFe₂O₄ (CFO) and soft NiFe₂O₄ (NFO), that are currently used for permanent magnet applications. Both materials are attractive for different technological solutions, due to their abundance and low cost, together with their high chemical, magnetic and thermal stabilities, and a low electrical conductivity that reduces eddy current losses.
They share the same crystal structure with close lattice parameters, reducing mismatch and strain effects, and thus providing an ideal model to explore intrinsic features linked to the formation of the interface. These ferrites also share the same ferrimagnetic order, with tetrahedral and octahedral cation sublattices coupled antiferromagnetically, preserving high values of the local magnetic moments. Also their electronic features are quite similar, with insulating band gaps and akin orbital distributions. From a theoretical point of view, both materials have been widely studied, and the optimum conditions to simulate their properties in *ab initio* frameworks are well known [9-12].

Core/shell CFO/NFO nanoparticles of high structural quality have already been prepared by hydrothermal methods [13]. The magnetic measurements evidence a coherent interface and good exchange coupling between both components, while the saturation magnetization and the magnetic anisotropy of the entire system lie between those of the individual materials. As expected from the relative stability between the inverse and direct spinel structures of the bulk counterparts [14], NFO is completely inverted, while some Co cations occupy tetrahedral sites. This is relevant as CFO in the direct spinel lattice behaves as a soft magnet. In our study we simulate the ideal interfaces between CFO and NFO, trying to isolate the effect of interface formation on the magnetic properties of the system. We explore the different terminations that can occur at interfaces along the high symmetry directions [001] and [111], addressing their relative stability, magnetization and magnetic anisotropy. Our results indicate that even ideal interface formation has a controversial impact on the magnetic properties. Both magnetic moments and magnetic order follow bulk-like features, though the presence of Co at tetrahedral cation sites enhances the net magnetization. On the other hand, the magnetic anisotropy energy (MAE) of the combined system is weakened, and the differences between normal and in-plane interface directions strongly depend on the specific termination. The paper is organized as follows: in section 2 we summarize the conditions of our calculations, in section 3 we present the results, separating information about the magnetization and magnetic order (section 3.1), the relative stability of the different interfaces (section 3.2) and the magnetic anisotropy (section 3.3). We end with the main conclusions of our study in section 4.

2. Theoretical method

We have performed *ab initio* simulations based on the density functional theory (DFT) using the Vienna Ab-Initio Simulation Package (VASP) [15-16]. The Perdew-Burke-Ernzerhof parametrization revised for solids (PBEsol) has been chosen as the Generalized Gradient Approximation (GGA) for the exchange-correlation functional, implementing Hubbard type GGA+U corrections under the Dudarev approach with U = 4 eV at the d bands of the three cations: Fe, Co and Ni. The projector-augmented wave method (PAW) has been used to describe the core contributions, using an energy cutoff of 500 eV for the plane wave basis set and sampling the Brillouin zone with a (7 × 7 × 3) k-mesh. Electronic relaxations were carried out self-consistently until convergence in energy of 10⁻⁵ eV was reached. The local magnetic moments have been obtained after Bader charge analysis [17].

The spin-orbit coupling has been included for calculations of the magnetic anisotropy [18]. The MAE is derived directly from the difference between *ab initio* total energies corresponding to configurations with a different orientation of the net magnetization. To determine the detailed dependence of the total energy on this orientation, we explore the stepwise canting of the magnetization following the magnetic constraints method [19], including configurations with the magnetization along the relevant symmetry directions of the systems considered (bulk easy axis, interface normal and in-plane directions). We have checked the independence of our results on the specific choice of the initial configuration, as explained in the Supplementary Information S1.

![Figure 1. Unit cells of the CFO/NFO interfaces simulated along the [001] and [111] directions, showing the orientation of the unit cell axes a, b, c.](attachment:image.png)

We have studied the ideal interfaces between CFO and NFO along two high symmetry directions [001] and [111], as shown in Figure 1. CFO and NFO have close lattice parameters, that lead to an in-plane mismatch <0.5%. This is well below the threshold to introduce any noticeable strain effect in the electronic, optical or magnetic properties of any of the oxides [9-10,20-22]. We have considered the case in which NFO adopts the in-plane CFO lattice (that is, a_{NFO} =
aCFO and bNFO = bCFO), optimizing the corresponding cNFO from bulk calculations of NFO at fixed in-plane CFO lattice parameters. We have also restricted to uniform distributions of Co and Ni, as this is known to be the most stable solution for both oxides [14]. In all cases, the atoms around the interface have been allowed to relax until the forces were below 0.004 eV/Å. This includes two Fe planes on each side of the (001) interface and two planes (one formed by Fe and one by O) on each side of the (111) interface, as can be seen in Figure 2.

Along the [001] direction, a natural interface emerges at the plane defined by the tetrahedral Fe cations. We have considered this situation using a minimum unit cell with 56 atoms, that corresponds to 8 atomic layers of each oxide, and the CFO lattice parameters, a = b = 5.93 Å, and values of cCFO = 8.38 Å and cNFO = 8.21 Å. Increasing the unit cell to 84 atoms the interface properties do not change and the conclusions reported here remain valid. Along the [111] direction we have employed a unit cell with 84 atoms, 6 planes for each oxide, again choosing the in-plane lattice parameters of CFO, a = b = 5.94 Å, and values of cCFO = 14.40 Å and cNFO = 14.25 Å. Here, as shown in figure 2 b), the choice of the terminations that match at the interface is not unique. The three inequivalent interfaces that emerge can be understood regarding the inverse spinel structure as an ABC stack of oxygen layers, each one with a different environment of cation neighbors.

The relative stability of the different interfaces has been estimated from the work of separation (Wsep), defined as the energy needed to break the interface and separate the two oxides [23]. We have calculated it from the energy difference between the isolated oxides, obtained introducing a vacuum region of 10 Å in our slabs at the interface between the two materials, and the bicomponent system after relaxing the atoms around the interface, normalized to the surface area of the unit cell. Thus, positive values of Wsep correspond to stable interfaces.

Figure 2. Interfaces considered along a) (001) and b) (111) orientations. The blue boxes indicate the interface region where atoms are allowed to relax. FeA (FeB) distinguishes tetrahedral (octahedral) Fe sites.

A final remark is deserved about the partial conversion of CFO to the direct spinel lattice. In good agreement with the experimental evidence, our calculations provide the stability of the inverse spinel structure for both CFO and NFO, with a reduction in the total energy of each oxide relative to the direct lattice of 0.54 eV/f.u. and 5.18 eV/f.u. respectively, where f.u. accounts for formula unit. It is expected that only a limited amount of Co atoms will occupy tetrahedral positions. In our interfaces, we have modelled the extreme situation where the entire CFO is considered as a direct spinel structure, with all Co atoms replacing FeA. This provides a limiting case that maximizes the effect of inversion of CFO. The equilibrium lattice parameters of bulk CFO under the direct spinel structure (a = b = 5.94 Å and c = 8.40 Å using a (001) cell), are close to those of the inverse spinel. Thus, we do not expect any strain effect induced by the incomplete inversion of CFO, and we have used the inverse spinel lattice parameters in our calculations.

3. Results and discussion

3.1. Magnetic order and magnetization

Interface formation alters the bonds between atoms, modifying both the local magnetic moments and the balance of magnetic energies. As a result, it may lead to changes in the magnetic order and the net magnetization of the system. We have addressed these issues first by simulating different magnetic configurations at the interfaces under study. We explore possible deviations from the bulk ferrimagnetic order at both oxides, and relate them to the exchange constants determined from mapping our ab initio total energies to a Heisenberg Hamiltonian [24], as explained in detail in the Supplementary information S2. It is worth mentioning that some of the proposed configurations do not even converge to a self-consistent ground state within our accuracy limits, providing a hint of their instability. Our final results indicate that in all cases the bulk-like ferrimagnetic order is preserved. As an example, table I compiles the energy difference between the ferrimagnetic ground state and the ferromagnetic configuration, comparing the isolated bulk oxides to the interface systems under study. The quoted values for (111) interfaces correspond to the average of the different terminations; the ferromagnetic case did not converge to a ground state at all cases, but differences between the converged solutions remain below ±0.05 meV/Fe. All these results indicate a good exchange coupling between both oxides, that favours avoidance of spring magnets. The partial inversion of CFO will not change this
scenario, as the same ferrimagnetic order is stabilized at bulk CFO under the direct spinel structure.

<table>
<thead>
<tr>
<th>System</th>
<th>NFO</th>
<th>CFO</th>
<th>(001)</th>
<th>(111)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inverse</td>
<td>0.87</td>
<td>0.75</td>
<td>0.82</td>
<td>0.85</td>
</tr>
<tr>
<td>Direct</td>
<td>-</td>
<td>0.24</td>
<td>0.68</td>
<td>0.65</td>
</tr>
</tbody>
</table>

Table I. Energy difference in eV/f.u. between ferromagnetism and the ferrimagnetic ground state for bulk NFO, bulk CFO and the NFO/CFO (001) and (111) interfaces, both under the inverse and direct spinel structures of CFO.

Regarding the exchange constants between the different magnetic sublattices ($J_{ij}$), the magnetic configurations necessary to determine them in the bulk forms of CFO and NFO are described in the Supplementary material S2. The number of configurations required in the case of the bicomponent systems is much larger even at the (001) interfaces, due to their reduced symmetry. Only the magnetic exchange between Fe$_A$ and Fe$_B$ sublattices can be determined from the configurations used at the bulk, allowing us to compare the interface value, $J_{FeA-FeB} = 2.8$ meV/µ$_B^2$, to those obtained at the bulk forms of both CFO and NFO: $J_{FeA-FeB} = 2.5$ meV/µ$_B^2$.

As an additional proof of the minor influence of the interface on the exchange interactions, in figure 3 we compare the energies of configurations c0-c3 at the bulk and the (001) interface. It is clear that the energy barriers between configurations follow identical trends in all cases. Moreover, the interface values are approximately the average of the bulk ones, evidencing that the energy contributions to the magnetic exchange interactions are hardly affected by the presence of the interface. As explained in the Supplementary material, the other relevant terms taking part in the determination of the $J_{ij}$ are the sublattice magnetizations. Our calculations indicate that, for each specific configuration, similar values of the local magnetic moments are obtained at the bulk and interface systems.

<table>
<thead>
<tr>
<th>System</th>
<th>M</th>
<th>Fe$_A$</th>
<th>Fe$_B$</th>
<th>Co</th>
<th>Ni</th>
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<tr>
<td>CFO$_{bulk}$</td>
<td>3</td>
<td>4.06</td>
<td>4.17</td>
<td>2.65</td>
<td>-</td>
</tr>
<tr>
<td>NFO$_{bulk}$</td>
<td>2</td>
<td>4.00</td>
<td>4.12</td>
<td>-</td>
<td>1.60</td>
</tr>
<tr>
<td>(001)</td>
<td>5</td>
<td>4.07</td>
<td>4.17</td>
<td>2.66</td>
<td>1.62</td>
</tr>
<tr>
<td>(111)a</td>
<td>5</td>
<td>4.06</td>
<td>4.18</td>
<td>2.64</td>
<td>1.61</td>
</tr>
<tr>
<td>(111)b</td>
<td>5</td>
<td>4.07</td>
<td>4.18</td>
<td>2.65</td>
<td>1.60</td>
</tr>
<tr>
<td>(111)c</td>
<td>5</td>
<td>4.07</td>
<td>4.18</td>
<td>2.65</td>
<td>1.62</td>
</tr>
<tr>
<td>CFO$_{bulk}$</td>
<td>7</td>
<td>-</td>
<td>4.19-4.32</td>
<td>2.59</td>
<td>-</td>
</tr>
<tr>
<td>(001)$'$</td>
<td>8.5</td>
<td>4.07</td>
<td>4.03-4.19</td>
<td>2.54</td>
<td>1.63</td>
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<tr>
<td>(111)a$'$</td>
<td>8.8</td>
<td>-</td>
<td>4.20-4.22</td>
<td>2.60</td>
<td>1.64</td>
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<tr>
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<td>4.06</td>
<td>4.17-4.19</td>
<td>-</td>
<td>1.61</td>
</tr>
<tr>
<td>(111)c$'$</td>
<td>8.8</td>
<td>-</td>
<td>4.23</td>
<td>2.59</td>
<td>1.64</td>
</tr>
</tbody>
</table>

Table II. Net magnetization of our slabs (M, in µ$_B$/f.u.) and absolute value of the magnetic moment (in µ$_B$) of the interface cations (i.e. those included in the blue boxes of figure 2), for the systems under study. The upper (lower) part of the table refers to the inverse (direct) CFO form.

In the following we will thus restrict to the bulk-like ferrimagnetic order of the oxides. Table II provides the total magnetization and the cation magnetic moments corresponding to all interfaces under study. Both the inverse and direct forms of CFO are considered, distinguished labelling with a prime systems containing direct spinel CFO. As a reference, also bulk values are provided. Though not
shown in the Table, the Ni moments are not modified far from the interface, even though NFO adopts the in-plane CFO lattice. Regarding now the upper part of the Table, referred to full inverse spinel structures, the Co and Ni moments hardly show any variation with respect to the bulk, while interface Fe moments are closer to the values at CFO. Still, all interface systems preserve magnetizations that correspond to the sum of the individual bulk components, evidencing that interface formation does not have any impact on it.

Larger variations occur when we consider the direct spinel CFO. As ferrimagnetism in the spinel ferrites emerges from the antiferromagnetic coupling between octahedral and tetrahedral cations, replacing Fe\textsubscript{A} by Co leads to a significant enhancement of the net magnetization. Similarly, the Fe\textsubscript{B} moments increase, and show a noticeable dispersion of values. These features observed in the bulk form of direct CFO are also evident at the interfaces. In general, the interface Ni moments tend to slightly increase, but a local enhancement may arise from partial CFO inversion due to the replacement of Fe by Co at tetrahedral sites while preserving the ferrimagnetic order.

3.2. Stability

The relative stability of the (111) interfaces under study can be determined from direct comparison of their total energies, as their unit cells share the same composition. This serves to identify case (111)c as the most stable solution, with the (111)a and (111)b ground states higher in energy by 7.18 meV/f.u. and 7.27 meV/f.u., respectively. When direct CFO is considered, the energy differences are enhanced and their relative order is modified: (111)a’ becomes the most stable case by 1.9 meV/f.u. over (111)b’ and by 23.53 meV/f.u. over (111)c’. On the other hand, regarding the relative stability between direct and inverse spinel structures, the interface does not introduce any significant difference with respect to the bulk, with energy barriers that oscillate between 0.55 eV/f.u. and 0.49 eV/f.u. Specifically, the energy barrier between (111)c and (111)a’, which are the most stable interfaces for inverse and direct CFO respectively, is 0.50 eV/f.u. All this supports that the interface will not favor neither inhibit the occupation of tetrahedral positions by Co atoms.

To compare now all orientations, we resort to the work of separation, shown in Figure 4 also including the direct CFO lattice for all cases. The positive value of $W_{\text{sep}}$ reflects the stability of all systems, and again identifies the (111)c termination as the most stable situation. Mutual differences between the (111) terminations are slight, and much less than referred to the (001) orientation, as expected from their close-packed structure. When the direct CFO spinel is considered, case (111)c’ does not follow the trend of variation obtained from direct comparison of the total energies, although the order of magnitude of the relative stabilities is low and similar under both methods. In summary, the main conclusion from this study is that a coexistence of all the interfaces considered cannot be discarded in most experimental systems where no specific orientation is favoured, as magnetic powders and nanoparticle solutions.

3.3. Magnetic anisotropy energy

Experimentally, the bulk forms of both CFO and NFO have cubic symmetry, related to a random distribution of cations at the octahedral sites. In our model we are attached to a specific atomic arrangement, selected because it corresponds to the lowest energy configuration and it properly describes the short-range order measured for NFO [14]. As a result, the cubic symmetry is lowered, with consequences on the magnetic anisotropy. Previous studies [9,10] have shown that, while the effect is negligible for NFO, the calculated magnetoelastic response of CFO depends on the details of the structure. Considering different Co distributions at the interfaces under study is a formidable computational task, out of the scope of the present manuscript. Our results represent a first approach to explore
the global effect of interface formation on the magnetic anisotropy of the combined system. Even though they are necessarily restricted to our choice of a specific model, they are still representative, as the variations introduced by different configurations are expected to lie within the experimental uncertainty [10].

The bulk easy axis (EA) of CFO has been reported to lie along the [001] direction [25], thus it coincides with the out-of-plane orientation at (001) interfaces. According to our self-consistent calculations, the bulk MAE, determined from accurate total energy differences of configurations with different orientation of the magnetization, corresponds to values of $\text{MAE}_{\text{CFO}} = 0.6 \text{ meV/Fe}$ and $\text{MAE}_{\text{NFO}} = 0.001 \text{ meV/Fe}$ (which is close to our resolution). They have been obtained comparing the three cubic symmetry axes [001], [010], [100], and the [110] and [111] directions of the bulk unit cell, and both of them identify the magnetic EA as [001]. The actual EA of bulk NFO has been identified along [111] [26], our result being a consequence of the restriction to a fixed cation distribution; however, we correctly determine the order of magnitude of the MAE, that is negligible as compared to that of CFO. All results are normalized to the number of Fe atoms to ease comparison between systems with different chemical species. This is just a normalization, that still includes the contribution from all atoms in each structure. It is suitable as the number of iron atoms per unit cell keeps the same ratio throughout all the structures considered.

Figure 5. Rotation paths followed in Figure 6, for: a) CFO bulk, and b) the NFO/CFO (111)a,b,c. interfaces. In both cases, the magnetization vector is contained in the depicted planes. At the interface, the [111] and [-110] axis coincide with the normal and in-plane directions, respectively.

We can also explore the evolution of the MAE for gradual rotations of the magnetization departing from the bulk EA. Following the path indicated in figure 5a, this has been done for bulk CFO under the magnetic constraints method, and the results are shown in figure 6a. The MAE increases smoothly as the magnetization rotates from the bulk EA to the plane normal to it, where it reaches the same maximum value obtained for the other in-plane high symmetry axis, [010] and [100]. This supports a low magnetic anisotropy in the plane normal to the EA at bulk CFO in our model without cubic symmetry.
Figure 6. Evolution of the MAE under stepwise rotation of the magnetization following the path shown in figure 5 for a) bulk CFO, departing from the bulk easy axis at $\theta = 0^\circ$; b) the CFO/NFO (111)a,b,c interfaces, departing at $\theta = 0^\circ$ from the interface normal. As a reference, the values of $\theta$ corresponding to relevant symmetry directions are indicated by dashed lines.

The presence of the interface introduces new relevant symmetry directions that depend on the specific termination. At the (001) CFO/NFO system, the interface normal coincides with the CFO bulk EA. If we determine the MAE from direct energy differences between situations where the magnetization is aligned either to the interface normal or to any in-plane direction, we obtain a value of 0.22 meV/Fe. This is close to a simple additive effect from both oxide components, that would correspond to 0.3 meV/Fe, the presence of the interface only weakening slightly the anisotropy.

Figure 7. a) In-plane rotation path of the magnetization and b) resulting MAE, for the (111)c interface. The energy zero is set with the magnetization aligned to [-101].

The situation is different for (111) terminations. Here, besides the interface normal and in-plane directions, we need to consider the bulk easy axis as a relevant reference. Figure 6b shows the evolution of the MAE at the (111) interfaces as the magnetization rotates from the interface normal to an in-plane direction, following the path indicated in figure 5b. In all cases the bulk EA is preserved, with a MAE around 0.3 meV/Fe when comparing the same crystal directions than for the (001) interface (namely, [111] and [-110]). This seems to reflect again a simple additive effect of the bulk contributions, but without any further anisotropy weakening. Figure 6b also allows to compare the difference between situations where the magnetization is aligned either in-plane or normal to the interface. Here significant variations emerge between the different (111) terminations: while for (111)c aligning the magnetization to the normal increases the MAE by 0.1 eV/atom over the bulk EA, this MAE doubles in the case of the (111)a interface. This effect is related to the choice of the specific in-plane direction, as we will explain now.
Figure 7 shows the MAE when comparing different in-plane directions at the (111)c interface, referred to the in-plane orientation that minimizes the energy. The maximum variations are similar to those encountered between the interface normal and the [-110] axis at figure 6b. A similar situation arises at the other (111) terminations. Always the bulk EA provides the lowest energy.

All these results indicate that interface formation, even for these ideal interfaces lacking of mismatch and defects, tends to reduce the MAE of the bicomponent system as compared to the isolated hard material, a drawback to improve the energy product of a permanent magnet based on the multicomponent system. But in addition, in samples where multiple terminations coexist, the rotation of the magnetization along a fixed path (i.e. from normal to in-plane directions) faces different energy barriers depending on the termination, complicating the interpretation of experimental results. As interface formation does not introduce any significant changes neither in the local charges nor in the magnetic moments, the effect seems to be related to crystal-field effects beyond nearest neighbors, that are also behind the influence of the particular cation distribution on the magnetic anisotropy.

Finally, we have considered the influence of partial inversion of CFO and strain in the MAE. Bulk CFO under the direct spinel structure (CFO') is a soft magnet, so that introduction of direct CFO at the interface systems is expected to reduce their magnetic anisotropy. We have calculated the bulk MAE of CFO' following the same procedure that for the inverse spinel. According to our results, the largest energy variations obtained upon different orientations of the magnetization lead to a MAE_{CFO'} = 0.02 meV/Fe. The evolution of the MAE following the path shown in figure 6a is similar for CFO', though spanning an energy range thirty times lower. The [001] direction is identified as the magnetic easy axis, but the MAE range is close to our limiting resolution. When replacing CFO by CFO' in our interface systems, we have considered gradual rotations of the magnetization from out-of-plane to in-plane directions, exploring different in-plane orientations. The resulting MAE is very low, close to our accuracy limit, indicating that interface formation does not alter the magnetic softness induced by inversion of CFO.

Regarding strain effects, due to the excellent lattice matching of both oxides, we do not expect any noticeable influence of strain in our CFO/NFO systems. However, in our calculations NFO adopts the CFO lattice, while depending on the growth conditions, the reverse situation may be found. Taking into account the large magnetostriction of CFO, even this slight mismatch might be important [10]. We have done a preliminar study to address how adopting the NFO lattice parameters modifies the MAE of bulk CFO. The results provide a MAE similar to that obtained under the CFO bulk lattice, 0.61 meV/Fe, that reduces slightly to 0.58 meV/Fe after optimization of the c lattice parameter while keeping b_{NFO}, b_{CFO}. In both cases the bulk easy axis does not change. The slight reduction of the MAE when strain relief is allowed matches the expected trend from the magnetoelastic coefficient of CFO under constriction mismatch [10]. However, the low effect confirms the validity of our approach to neglect the impact of strain in the systems under study.

4. Conclusions

A detailed study of the magnetic properties at ideal CFO/NFO interfaces has been presented, including effects induced by the conversion of CFO to the direct spinel structure. As expected, close-packed structures are more stable, though interfaces along different orientations may coexist. The bulk magnetic order is preserved, without influence neither from strain effects in the expected limits induced by mixing NFO and CFO, nor from partial conversion of CFO to the direct lattice. Similarly, ideal interface formation does not modify the magnetization, that corresponds to the sum of the individual bulk components. Actually, partial inversion of CFO has a much larger impact on the magnetization, significantly enhancing it. Nevertheless, the presence of the interface does not seem to influence the ratio of Co atoms at tetrahedral sites.

A different scenario emerges regarding the magnetic anisotropy. The presence of the interface tends to weaken the net MAE of the bicomponent system, due to an additive effect of contributions from the soft and hard materials. Additionally, the MAE is affected by the particular terminations involved. While the bulk easy axis is preserved, the cost to rotate the magnetization is significantly reduced with respect to bulk CFO, and depends on the specific termination. Partial inversion of CFO further weakens the MAE.

Our study can be considered as a departing point to investigate more realistic interfaces including the presence of defects (disorder, inter-diffusion, vacancies, etc.). Though our choice of the particular materials allows to design ideal interfaces, strains and non-uniform cation distributions may contribute in most experimental situations. In particular, the large magnetostriction of CFO makes its magnetoelastic response sensitive to even moderate strains, and highly dependent on the specific cation arrangement. We hope that our study will motivate further research exploring these aspects.

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References