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A study of the magnetic structure of YFe_4Al_8

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Abstract

A study of the magnetic structure of YFe_4Al_8 is presented, based on the reported magnetic structure, as given by a neutron diffraction study; cycloid, with moments in the a - b plane and with two Fe sublattices with a 140° phase difference between them. Calculations were performed, using density functional theory, for cycloids with $\mathbf{q} = \frac{2\pi}{a}(\tau, \tau, 0)$, with τ varying between 0 and 1. The calculated magnetic structure agrees with the structure deduced from neutron diffraction results, however with a simpler description. The YFe_4Al_8 magnetic structure can be described by a single Fe lattice with a cycloid propagation vector $\mathbf{q} = \frac{2\pi}{a}(\tau, \tau, 0)$ with $\tau = 0.865$; there is no need for a second Fe lattice with an arbitrary phase difference. We show that the same description can be applied to DyFe_4Al_8 and HoFe_4Al_8 .

1. Introduction

The $\text{MFe}_x\text{Al}_{12-x}$ ($M = \text{rare earth or actinide}$) compounds crystallize in a relatively simple crystalline structure over a large composition range (x), which has allowed a systematic study of the system, with particular interest for the ordered compounds MFe_4Al_8 . The crystal structure is body centred tetragonal, a ThMn_{12} -type structure which belongs to the $I4/mmm$ symmetry group. The rare earth or actinide atom occupies the $2a$ sites, at the origin and in the centre of the unit cell. For the 1:4:8 stoichiometry, the Fe atoms fully occupy the $8f$ sites at the corners of a rectangular prism centred on the $2a$ sites. The Al atoms occupy the $8j$ and $8i$ sites.

Studies of polycrystalline samples of the rare earth series in the late 1970s determined antiferromagnetic interactions for the Fe sublattice with a range of ordering temperatures

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between 90 and 200 K [1, 2]. The ordering temperatures of the magnetic rare earth atoms were lower than 50 K.

In the case of YFe_4Al_8 , Y is non-magnetic and therefore the Fe moments form the only magnetic lattice. The magnetic transition temperature for YFe_4Al_8 was reported as 185 K [1, 2] and 94 K [3]. Polycrystalline samples of REFe_4Al_8 ($\text{RE} = \text{La}, \text{Ce}, \text{Lu}, \text{Y}$ [4] and Tb [5]) were studied by Mössbauer spectroscopy and neutron diffraction. The compounds were shown to order between 130 and 200 K with distinct types of spiral structures requiring two sets of wavevectors for their description. The Fe moments were reported to be close to $2 \mu_{\text{B}}$ per atom.

According to previous results, the Fe moments in the Y compounds order in a spin spiral with the propagation vector along [110] [4, 6]. The spin spiral lowers the symmetry of the REFe_4Al_8 system, since the propagation vector $\mathbf{q} = \frac{2\pi}{a}(\tau, \tau, 0)$ is not invariant under a tetragonal fourfold rotation, consequently the eight Fe atoms in the unit cell, which would be equivalent in the absence of the spin spiral, are split into two sublattices with the same propagation vector but a non-zero phase difference [7]. YFe_4Al_8 was found to order at around 185 K, with the Fe moments forming a cycloid in the a - b plane with propagation vector $\mathbf{q} = \frac{2\pi}{a}(0.135, 0.135, 0)$ [4]: the phase difference between the two Fe sublattices was given as $\pm 140^\circ$, with a magnetic moment of $2.1 \mu_{\text{B}}$.

Single crystals of REFe_4Al_8 were also studied. Neutron diffraction and magnetic x-ray diffraction measurements for DyFe_4Al_8 and HoFe_4Al_8 determined a cycloid Fe lattice with a wavevector along the [110] direction and an ordering temperature of 175 K; the reported magnetic moment was $1 \mu_{\text{B}}/\text{Fe}$ in the a - b plane [8]. The Dy and Ho sublattices were found to order around 50 and 80 K respectively, with a magnetic structure that follows the Fe cycloid. The propagation vectors are similar to the YFe_4Al_8 wavevector, $\mathbf{q} = \frac{2\pi}{a}(0.133, 0.133, 0)$ for DyFe_4Al_8 and $\mathbf{q} = \frac{2\pi}{a}(0.142, 0.142, 0)$ for HoFe_4Al_8 .

Measurements on YFe_4Al_8 single crystals, grown with bulk charges 1:4:8, showed an ordering temperature of 100 K, which is much smaller than the 185 K determined previously for the polycrystalline samples [2, 4]. A later study of nominal YFe_4Al_8 and LuFe_4Al_8 single crystals confirmed that the samples are richer in Fe than expected [6].

Another Mössbauer spectroscopy study of YFe_4Al_8 and ErFe_4Al_8 confirmed the gradual ordering of the Fe lattice [10]. The low-temperature spectra are consistent with the 1:4:8 stoichiometry. It is suggested that the rare earth lattice has a negligible effect on the Fe sublattice, as the temperature behaviour and the hyperfine field are the same for both compounds. The magnetic moment, estimated by comparison with the hyperfine field value for pure Fe, is given as $0.7 \mu_{\text{B}}/\text{Fe}$.

The reported experimental values for the Fe magnetic moment vary from $0.7 \mu_{\text{B}}/\text{Fe}$, determined by Mössbauer spectroscopy [10], to $2.1 \mu_{\text{B}}/\text{Fe}$, measured by neutron diffraction [4]. Both values are far from the values determined for similar compounds: $1.3 \mu_{\text{B}}$ for LuFe_4Al_8 [6], for example. Although the reported magnetic moments of Fe vary over a large range of values, within each report the Fe moments for different compounds are similar: for REFe_4Al_8 (with $\text{RE} = \text{La}, \text{Ce}, \text{Y}$ and Lu) the values reported range from 1.8 to $2.1 \mu_{\text{B}}$ [4], for the Dy and Ho compounds the values are in the range 1.0 – $1.3 \mu_{\text{B}}$ [6, 8], and another study claims a value of $0.7 \mu_{\text{B}}$ [10] for YFe_4Al_8 and ErFe_4Al_8 .

The differences in the magnetic properties of compounds with close but differing compositions could explain the disagreement between previous results, however there is still no unified view about the properties of these compounds. Therefore an analysis of the magnetic structure of YFe_4Al_8 , with a calculation of the magnetic moments and a detailed study of the spin spiral, was performed. The starting point was the magnetic structure reported by the neutron diffraction study [4] (Fe moments in the a - b plane) and the analysis was carried out for the cycloid structure with propagation vectors $\mathbf{q} = \frac{2\pi}{a}(\tau, \tau, 0)$ with $0 \leq \tau \leq 1$.

2. Calculation

2.1. Method

The calculations, based on density functional theory, with the von Barth–Hedin parameterization for the local density approximation, were performed using the augmented spherical wave method modified to account for the spin spiral structures, as described in [11–13].

The magnetic moment and density of states were determined by self-consistent calculations, with YFe_4Al_8 constrained to several magnetic configurations. The number of reciprocal lattice points in the irreducible part of the Brillouin zone was $10 \times 10 \times 10$, which corresponds to 555 inequivalent points. Convergence with respect to the number of reciprocal points was confirmed by some calculations with 4207 k -points. The variation of the magnetic moment is less than $0.001 \mu_{\text{B}}/\text{atom}$. Since the calculation of the energy is more sensitive to the number of points in the reciprocal space, the total energy of each magnetic configuration was determined using the magnetic force theorem (MFT) [14] with 1000 reciprocal lattice points in the Brillouin zone. The eigenvalue sum was calculated at 0 K and at 100 K, the latter using the Fermi-Dirac distribution and calculating the exact chemical potential; these results differ by less than 0.05 mRyd and do not change the positions of the maxima and minima. The value used in the calculations was $T = 100$ K. The MFT was further tested using two series of calculations with different charge densities which resulted in similar energy curves, with the maxima and minima in the same positions, with deviations smaller than 0.06 mRyd.

2.2. Magnetic moments

The density of states and magnetic moment were calculated self-consistently for the non-magnetic and for several magnetic configurations.

In figure 1 the density of states for the non-magnetic state of YFe_4Al_8 is plotted. The total density of states at E_{F} is 270 sta/f.u., and the major contribution (48 sta/Fe) is from the Fe atoms. The Fe 3d states show a narrow maximum exactly at E_{F} and a further maximum, wider than the first, at 0.12 Ry below E_{F} . Figure 2 shows the density of states for the ferromagnetic configuration. A comparison of figures 1 and 2 shows that the main effect of allowing the formation of magnetic moments is the splitting of the Fe states: one would therefore expect to find the majority of the magnetic moment on the Fe sites.

The magnetic moment was calculated self-consistently for cycloids with the moments in the a - b plane and the propagation vector along the [110] direction, given by $\mathbf{q} = \frac{2\pi}{a}(\tau, \tau, 0)$, with τ varying between 0 and 1. As seen in figure 3, the Fe magnetic moments are similar for all magnetic structures, varying between 1.23 and $1.30 \mu_{\text{B}}$, and the values for the two sublattices always coincide. The Y moment is lower than $0.1 \mu_{\text{B}}/\text{Y}$ and the Al is essentially non-magnetic. One can conclude that the orientation of the magnetic lattice has little influence on the magnitude of the individual moments. The reported experimental values for the Fe moment in YFe_4Al_8 range from 0.7 [10] to $2.1 \mu_{\text{B}}$ [4], which makes difficult a meaningful comparison with the calculated result. The calculated value is, however, similar to the reported Fe moment of $1 \mu_{\text{B}}$ for DyFe_4Al_8 and HoFe_4Al_8 [8] or $1.3 \mu_{\text{B}}$ for LuFe_4Al_8 [6].

2.3. Magnetic structure

Symmetry analysis leads to the conclusion that there are two Fe sublattices [7] in compounds of this type. Although the two Fe sublattices are not related by any symmetry operation, they have the same symmetry operations (the neighbourhood of the two sublattices is the same),

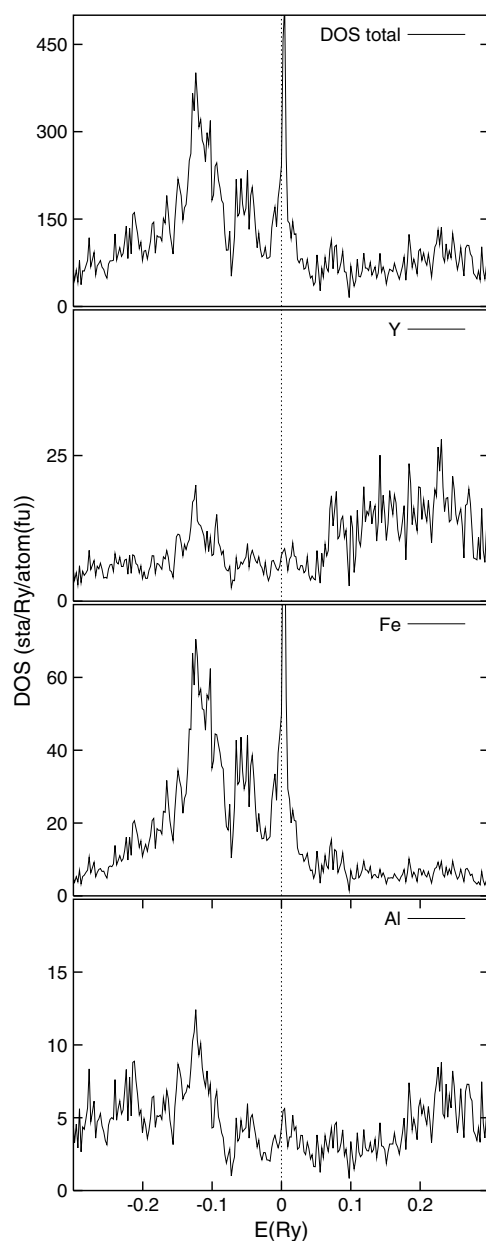


Figure 1. Total and partial density of states of the paramagnetic state of YFe_4Al_8 . $E = 0$ corresponds to the Fermi Level.

therefore the two iron sublattices are identical, with the same magnetic moments and magnetic structure. This is in agreement with Mössbauer spectroscopy measurements, since only one value of hyperfine field is detected [9], indicating that all the Fe atoms have the same magnetic and structural surroundings.

In order to analyse the two Fe sublattices, we label four different Fe positions (see figure 4): site **1**, at position $\mathbf{r}_1 = a(0.25, 0.25, 0.25)$, and site **2**, $\mathbf{r}_2 = a(0.75, 0.75, 0.25)$, belong to the

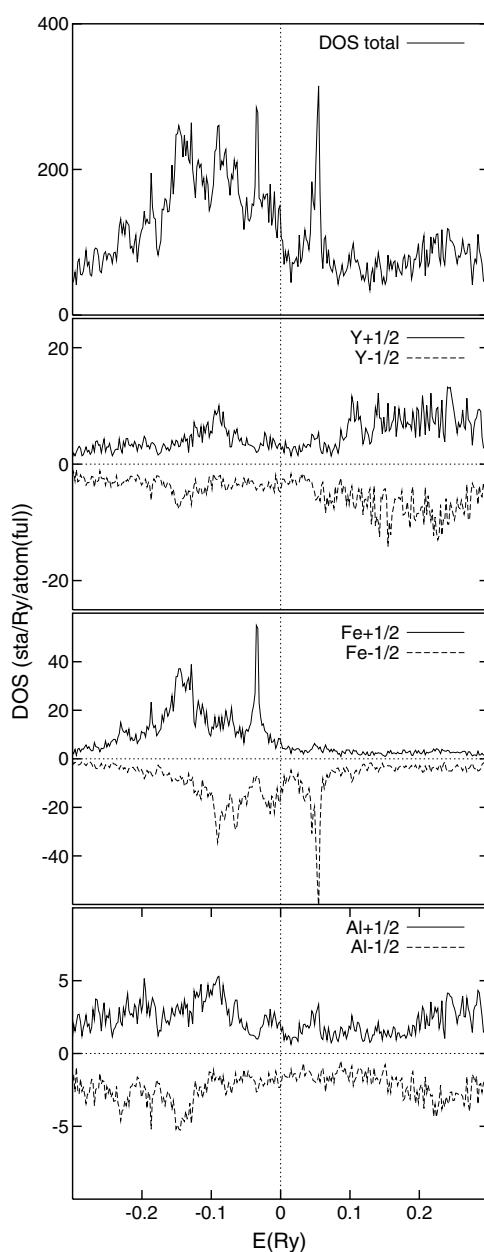


Figure 2. Total and partial density of states of the constrained ferromagnetic state of YFe_4Al_8 .

first magnetic sublattice; the sites **3**, $\mathbf{r}_3 = (0.25, 0.75, 0.25)$, and **4**, $\mathbf{r}_4 = (0.75, 0.25, 0.25)$, constitute the second sublattice. The atoms along the same line parallel to \mathbf{c} belong to the same sublattice. The relative angle between the moments on sites **1** and **2**, belonging to the same sublattice, will be called α and is given by the spin spiral: $\alpha = \mathbf{q} \cdot (\mathbf{r}_2 - \mathbf{r}_1)$. The angle between the two sublattices will be identified by the angle between the moments on the sites **1** and **3**, which will be referred to as β . In some calculations, we fix β to the value that corresponds to

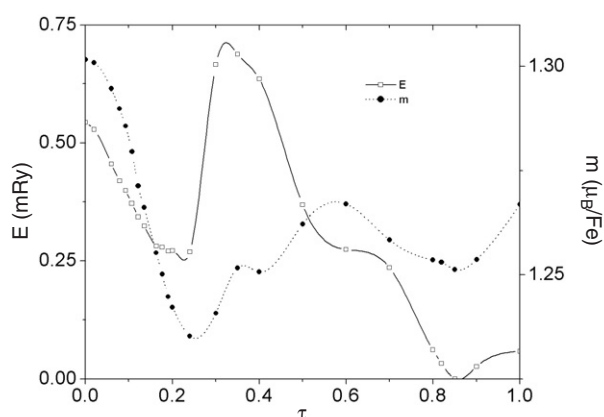


Figure 3. Calculated energy variation (open boxes, left-hand scale) and Fe magnetic moment (filled circles, right-hand scale), for magnetic cycloids with $\mathbf{q} = \frac{2\pi}{a}(\tau, \tau, 0)$

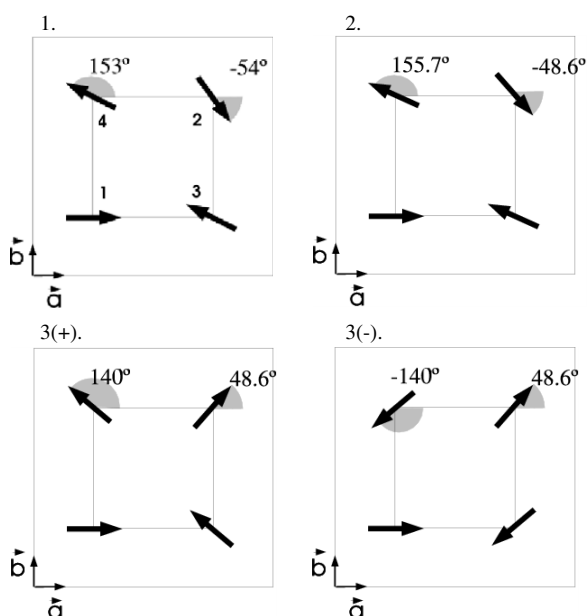


Figure 4. Magnetic configurations referred to in table 1. The first figure labels the four Fe atoms, as referred to in the text. The bottom two figures refer to $+140^\circ$ and -140° , respectively.

a spin spiral considering all the Fe atoms as belonging to one sublattice, which means that β is determined by the propagation vector, $\beta = \mathbf{q} \cdot (\mathbf{r}_3 - \mathbf{r}_1)$. In other calculations, we have considered two Fe sublattices and treated β as a parameter.

The energies for cycloids with different propagation vectors, calculated using the magnetic force theorem, are plotted in figure 3. All structures have propagation vectors along the [110] direction, $\mathbf{q} = \frac{2\pi}{a}(\tau, \tau, 0)$. The energy differences were calculated relative to the antiferromagnetic configuration, which corresponds to the wavevector $\mathbf{q} = \frac{2\pi}{a}(1, 1, 0)$. The angle between Fe sublattices, β , was fixed as if the four Fe atoms belonged to the same lattice.

The differences in the total energies are of the order of 0.1 mRyd. There is a local minimum for τ around 0.2, close to the wavevector determined for the cycloid by neutron diffraction for

Table 1. Comparison of the values of τ , α and β for calculations with one sublattice (rows 1 and 2) with the experimental values (row 3). These four possibilities are illustrated in figure 4.

	τ	α	β	Comment
1	0.85	-54°	153°	Energy minima from calculations
2	0.865	-48.6°	156°	Using $1 - \tau_{\text{experimental}}$
3	0.135(2)	48.6°	$\pm 140(10)^\circ$	Structure described in [4]

Table 2. Summary of two sublattice calculations, with the intersublattice angle β constrained (calculations i), (ii), (iii)) and free (calculation (iv)).

	τ	α (deg)	β (deg)	ΔE (mRyd)	Comment
(i)	0.135	48.6	24.3	0	One sublattice
(ii)	0.135	48.6	140	+0.51	[4]
(iii)	0.135	48.6	-140	-0.10	[4]
(iv)	0.135	48.6	-157	-0.16	Self-consistent

YFe_{4.4}Al_{7.6} [6]. The lowest energy configuration corresponds to the cycloid with wavevector $\mathbf{q} = \frac{2\pi}{a}(0.85, 0.85, 0)$. Our value $\tau = 0.85$ is far from the experimental value of 0.135 determined by neutron diffraction [4], however a more careful look shows similarities between the calculated and the experimentally determined structures.

The magnetic structure with wavevector $\mathbf{q} = \frac{2\pi}{a}(0.85, 0.85, 0)$ is shown in figure 4-1. The angle between the moments on the positions **1** and **2** is $\alpha_1 = \frac{2\pi}{a}(0.85, 0.85, 0) \cdot (\mathbf{r}_2 - \mathbf{r}_1) = -54^\circ$, close to the angle determined by neutron diffraction as $\alpha_2 = \frac{2\pi}{a}(0.135, 0.135, 0) \cdot (\mathbf{r}_2 - \mathbf{r}_1) = 48.6^\circ$. The opposite sign of the angles corresponds to a rotation in opposite directions, which are equivalent since the [110] and $[\bar{1}\bar{1}0]$ propagation directions are equivalent. The angles of rotation would be exactly the same if, instead of $\tau = 0.85$, we had $\tau = 0.865 = 1 - 0.135$ (see table 1).

According to the magnetic structure described in [4], the Fe atoms in positions **1** and **3** belong to two different sublattices. The reported angular difference that results from the fit of neutron diffraction data between the two sites is $\beta_{\text{neutron}} = \pm 140(10)^\circ$ and is not related to the reported propagation vector. Now considering our wavevector $\mathbf{q} = \frac{2\pi}{a}(0.85, 0.85, 0)$ and only one Fe lattice, $\beta_{\text{calculation}} = \frac{2\pi}{a}(0.85, 0.85, 0) \cdot (\mathbf{r}_3 - \mathbf{r}_1) = 153^\circ$. The calculated phase difference is close to the value determined by neutron diffraction, and furthermore is determined by the propagation vector \mathbf{q} , without the need for a second Fe sublattice.

In order to confirm the conclusions about the number of sublattices needed to describe the magnetic structure of YFe₄Al₈, we performed four calculations with propagation vector $\mathbf{q} = \frac{2\pi}{a}(0.135, 0.135, 0)$ and (i) one Fe lattice with $\beta_i = \frac{2\pi}{a}(0.135, 0.135, 0) \cdot (\mathbf{r}_3 - \mathbf{r}_1) = 24.3$; two Fe lattices with an angular deviation of (ii) $\beta_{ii} = 140^\circ$, and (iii) $\beta_{iii} = -140^\circ$, corresponding to the two possibilities reported in the neutron diffraction study [4]; (iv) two Fe lattices with all the moment directions calculated self-consistently. The results are summarized in table 2.

It must be noted that calculations (i), (ii) and (iii) are constrained calculations. In calculations (ii) and (iii), the spin density is not diagonal, which means that the moments would tend to change their orientations, if allowed. In calculations (i) (constrained) and (iv) (free), the spin density is diagonal and the spin orientations are therefore stable.

From the comparison of the first three calculations, we conclude that configuration (iii) has the lowest energy. Configuration (iii), with two Fe sublattices with $\mathbf{q} = \frac{2\pi}{a}(0.135, 0.135, 0)$, $\alpha_{iii} = 48.6^\circ$ and $\beta_{iii} = -140^\circ$, is the most similar to the calculated ground-state structure:

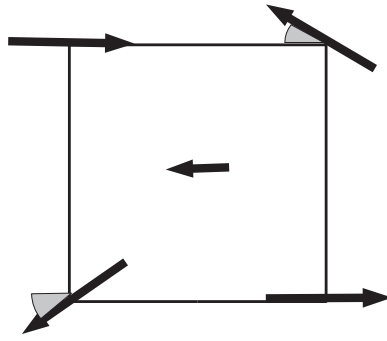


Figure 5. Relation between the directions of the Fe moments (in the corners) and the Y (in the middle), represented in the a - b plane.

one Fe lattice with propagation vector $\mathbf{q} = \frac{2\pi}{a}(0.85, 0.85, 0)$, which determines $\alpha_1 = -54^\circ$ and $\beta_1 = 153^\circ$ (see table 1). The self-consistent orientations from calculation (iv), $\alpha_{iv} = 48.6^\circ$ and $\beta_{iv} = 157^\circ$, confirm this idea, since the calculated angles are close to those expected from the $\mathbf{q} = \frac{2\pi}{a}(0.85, 0.85, 0)$ structure. In all the calculations, the Fe moment was found to be the same for both of the sublattices.

The Y moment, although small, also follows the spin spiral, with the moment antiparallel to the Fe moments along the direction perpendicular to the propagation vector. The Fe moments closer to Y along the propagation vector direction have a symmetric angular deviation (see figure 5). Therefore, this configuration can be seen as minimizing the Heisenberg interaction energy between Fe and Y. To test if the Y moment was responsible for establishing a relation between the two Fe sublattices, we performed two series of calculations with different orientations of the Y moment, both with a spin spiral with propagation vector $\mathbf{q} = \frac{2\pi}{a}(0.85, 0.85, 0)$: the first with the Y moment aligned following the Fe cycloid and the second with the Y moment rotated by 90° . The orientation of the second Fe sublattice was varied by δ , in a way that $\beta = \mathbf{q} \cdot \mathbf{r} + \delta$. In both calculations, the energy minimum corresponds to $\delta = 0$ and is symmetric, indicating that the Y orientation is irrelevant in the determination of the relative orientation of the two Fe lattices.

3. Discussion

The theoretical study of YFe_4Al_8 consisted of self-consistent calculations of the density of states and magnetic moments and the comparison of the total energy of different magnetic configurations, calculated with the use of the force theorem.

The magnetic moment is similar for all the studied magnetic configurations, around $1.3 \mu_B$ per atom. The similarity of the calculated values for the Fe moment with the experimental values for similar compounds, in contrast to the variety of values reported for YFe_4Al_8 , leads us to believe that the self-consistently calculated Fe moment has a reasonable value. A similar calculation for the UFe_4Al_8 compound predicts an Fe moment of $1.4 \mu_B/\text{Fe}$ [15], which compares with the $1.1 \mu_B/\text{Fe}$ determined by neutron diffraction measurements [16].

From the comparison of the total energy of several cycloid structures with the wavevector along the [110] direction, we were able to determine the lowest energy configuration. The magnetic structure consists of a cycloid with a propagation vector $\mathbf{q} = \frac{2\pi}{a}(0.85, 0.85, 0)$. The value $\tau = 0.85$ is very different from the neutron diffraction value, $\tau = 0.135$ [4], however our reinterpretation of the magnetic structure by considering only one Fe sublattice ($\beta = \mathbf{q} \cdot (\mathbf{r}_{3-r_1})$),

with $\mathbf{q} = \frac{2\pi}{a}(0.85, 0.85, 0)$, leads to an angular relation between the Fe moments similar to that proposed to fit neutron diffraction data. In this way, there is no need for a second Fe sublattice to describe the magnetic structure. Furthermore, the orientations calculated self-consistently for $\mathbf{q} = \frac{2\pi}{a}(0.135, 0.135, 0)$ confirm this reinterpretation, since they converge to essentially the same result as that given by the $\mathbf{q} = \frac{2\pi}{a}(0.85, 0.85, 0)$ structure, and have a lower energy than a calculation performed with the experimental values of $\tau = 0.135$ and $\beta = -140^\circ$, which we find to be unstable.

The same reasoning can be applied to other compounds which were previously described as having two magnetic Fe sublattices. DyFe₄Al₈ and HoFe₄Al₈ also have a cycloid magnetic structure with a wavevector $\mathbf{q} = \frac{2\pi}{a}(0.133, 0.133, 0)$ for DyFe₄Al₈ and $\mathbf{q} = \frac{2\pi}{a}(0.142, 0.142, 0)$ for HoFe₄Al₈ [8]. The Fe magnetic moment is 1 μ_B for both compounds. The propagation vectors are similar to the YFe₄Al₈ wavevector. For DyFe₄Al₈ and HoFe₄Al₈, the experimental angle between the two Fe sublattices is $\beta = \tau\pi + \pi$. This same angle can be obtained if the experimental value of $\mathbf{q} = \frac{2\pi}{a}(\tau, \tau, 0)$ is substituted by $\mathbf{q} = \frac{2\pi}{a}(\tau', \tau', 0)$, where $\tau' = (1 - \tau)$. With the latter wavevector, the angles between the Fe within the same sublattice are maintained and the angles between the two Fe sublattices come naturally from the relation $\theta = \mathbf{q} \cdot \Delta\mathbf{r}$. Within the same sublattice the angular rotation is given by $\alpha = 2\pi\tau' \cdot (1/2, 1/2, 0) = 2\pi - 2\pi\tau = -2\pi\tau$, and between sublattices the angular rotation is given by $\beta = 2\pi\tau' \cdot (1/2, 0, 0) = \pi - \pi\tau = -(\pi\tau + \pi)$.

The description of the YFe₄Al₈ magnetic structure with only one Fe lattice is not in agreement with the previous symmetry analysis of the system [6, 7]. According to these studies, the spin spiral lowers the symmetry of the REFe₄Al₈ system, since the propagation vector is not invariant under a tetragonal fourfold rotation. As a consequence, the four Fe atoms in the formula unit, which would be equivalent in the absence of the spin spiral, are split into two sublattices: the directions of the moments in different sublattices are not related, there is an arbitrary phase difference that can not be predicted by symmetry arguments, and the two sublattices could have different moments [7]. However, a further analysis shows that the two different Fe sublattices have *exactly* the same symmetry, therefore, although there is no symmetry operation that links the two sublattices, they should be identical. The neutron diffraction data is fitted with two Fe sublattices with the same atomic moment [4], and Mössbauer spectroscopy measurements are fitted with just one value of hyperfine field [9], indicating that all the Fe atoms have the same magnetic and structural surroundings. Moreover, our calculations predict an angle β that corresponds to the angle given by the spin spiral with all the four Fe belonging to the same lattice. This is also the case for HoFe₄Al₈ and DyFe₄Al₈, for which neutron diffraction measurements determined an angular difference $\beta = \tau\pi + \pi$ (equivalent to $\beta = \tau\pi$, as explained above). The fact that these three compounds can be described using only one Fe lattice suggests that the value determined for the phase difference is not arbitrary. Indeed, the calculations show that the alignment of the two Fe sublattices, as if they belonged to a single sublattice, is stable as the spin matrix is diagonal (that is, the local spin orientation is stable).

To justify the apparent coupling between the two Fe sublattices, one should first consider the spin rotation of a Bloch function between two identical Fe sites, for example site **1**, $\mathbf{r}_1 = a(0.25, 0.25, 0.25)$ and $\mathbf{r}_{1'}$ = $a(1.25, 0.25, 0.25)$. Going from one site to the other corresponds to a rotation $e^{i\mathbf{q} \cdot \Delta\mathbf{r}}$, where $\Delta\mathbf{r} = a(1, 0, 0)$, which is just the definition of a spin spiral. The two 'different' Fe sites 1 and 4 (at $\mathbf{r}_1 = a(0.25, 0.25, 0.25)$ and $\mathbf{r}_4 = a(0.75, 0.25, 0.25)$) are not linked by symmetry operations but are linked by hybridization: there is a permanent exchange of electrons between them. The spin rotation upon passing from atom 1 to 4 and then from 4 to 1' must be simply the spin rotation from 1 to 1', that is $e^{i\mathbf{q} \cdot \Delta\mathbf{r}}$. As $\Delta\mathbf{r}_{1-4}$ and $\Delta\mathbf{r}_{4-1'}$ are

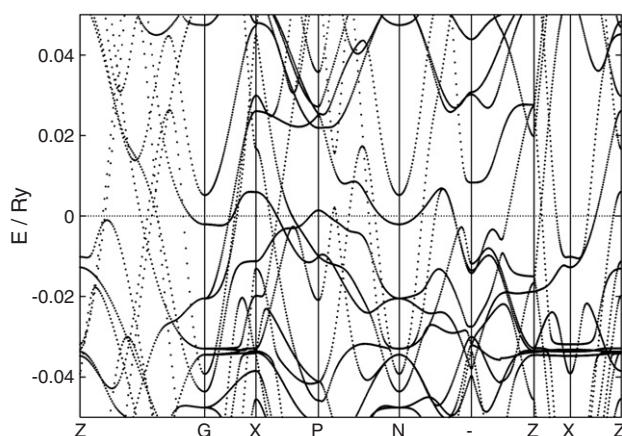


Figure 6. Ferromagnetic band structure near the Fermi level. The Fermi level is taken as the energy zero.

identical, it follows that the spin rotation from 1 to 4 must be half of that from 1 to 1', which is exactly the value that corresponds to a *single* Fe sublattice containing both Fe atoms 1 and 4.

An alternative way to analyse this situation is the following. Let us start by considering an Fe atom, belonging to one sublattice; this atom will, in the a - b plane, be surrounded by four Fe atoms that belong to the 'other' sublattice. If the angle between the two sublattices is not that given by the spin spiral, then the angle between each of the four atoms and the moment of the atom they surround is not the same. These different angles lead to different hybridization and energy interactions. However, one expects that the Fe atom would hybridize in the same way with the four equivalent atoms that surround it. To keep the four atoms that surround the central Fe atom identical, the angle between the Fe moments that minimizes the energy interaction should be the same for the four atoms. But this is true only if the phase difference between the two sublattices is zero, meaning that the angle between all the Fe moments is given by $\mathbf{q} \cdot \Delta\mathbf{r}$.

What remains to justify is the actual value of $\tau = 0.85$. In simpler compounds (with few atoms), an analysis of the band structure and Fermi surface would allow the identification of Fermi surface nesting for a given \mathbf{q} vector, however the number of band crossings in this compound hinder such a simple analysis. To illustrate this point, we include the ferromagnetic band structure near the Fermi level, where 30+ band crossings are clearly visible—figure 6.

4. Conclusions

Density functional theory calculations for YFe_4Al_8 allowed the comparison of several magnetic configurations. The lowest energy structure agrees with the structure deduced from neutron diffraction results [4], however with a simpler description: the YFe_4Al_8 magnetic structure can be described by a cycloid with the moments on the a - b plane and propagation vector $\mathbf{q} = \frac{2\pi}{a}(\tau, \tau, 0)$, with $\tau = 0.85$ if we consider the calculated value, or $\tau = 0.865$, to agree with the neutron diffraction data. There is no need for a second Fe lattice with an arbitrary phase difference to explain the neutron diffraction results. The coupling between the two identical Fe sublattices is not attributed to the Y sublattice but is explained by the hybridization between the Fe atoms. Although the calculated band structure is available, there is no way of identifying a simple Fermi-surface nesting to justify the value of $\tau = 0.85$.

We show that the same reinterpretation can be applied to DyFe₄Al₈ and HoFe₄Al₈, whose magnetic structures can be described as having just one Fe lattice.

The Fe moment is found to have a value of 1.3 μ_B within the range of experimental values, and to be relatively insensitive to the value of the spin spiral propagation vector.

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