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Reconstruction of the Exchange Integrals Map of $ScFe_4Al_8$ Magnetic Structure

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The influence of the dipolar as well as crystal field interaction as the main reasons of the noncollinearity and incommensurability of the $3d$–$3d$–$3p$ alloy belonging to $ThMn_{12}$ family is investigated. Available data on the single crystal neutron scattering experiments for $ScFe_4Al_8$ compound are discussed. The values and directions of the spin ordering found by magnetic diffraction are compared to that one derived from ground state configurations obtained by a simulated annealing algorithm.

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1. Introduction

Most of the $ThMn_{12}$-type structures reveal wide variety of magnetic ordering. The incommensurate spin-canted magnetic structures are particularly complicated ones. The neutron experiment carried out on the single crystal of $ScFe_4Al_8$ [1] disclosed two incommensurate and noncollinear magnetic modulations. This result is hard to interpret because it can be expected that, in contrast to systems of lanthanides and actinides crystallizing in this structure [2–5], the spin–orbit couplings in $ScFe_4Al_8$ should be of lesser importance. The direct exchange mechanism in $3d$ metals involves the overlap of the partly localized atomic orbitals of adjacent atoms. When delocalized electrons are present in a system, the calculation of the exchange interactions is a delicate matter. The exchange energies involved are only about 1 meV, compared with bandwidths of order 1–10 eV. Competing exchange interactions may coexist with different signs of coupling. Since the orbital moments of Fe as well as Sc atoms are quenched by the crystal field, the effects of the crystal field and spin–orbit coupling on the magnetic energy levels are conveniently represented by an effective spin Hamiltonian. The Heisenberg Hamiltonian with exchange interactions coupling the atomic spins can be applied directly to the $3d$ elements. This follows from the fact that a weak crystal field ensures that spin is a good quantum number and the ions have no orbital moments.

In order to construct a map of the exchange integrals reproducing the observed spin ordering, the atomic magnetic moments, modulation vector and phase transition temperature the MCMag [6] and MCPhase [7] simulation programs were used. Both of them base on an algorithm of simulated cooling or heating, see Kirkpatrick [8], while the configuration space is examined by random sampling in accordance with the Metropolis [9] procedure. However, two crucial aspects make both methods different. While MCPhase allows finding the exchange integrals by means of a fitting (self-consistent Monte Carlo procedure) and treats the spins quantum-mechanically, much earlier developed MCMag treats the spins classically and the exchange constantans have to be guessed. Both approaches indicate for the same microscopic mechanisms of the non-collinear magnetic structure formation and estimation of the range of the exchange interactions.

2. Experimental background

This paper deals with $ScFe_4Al_8$ crystallising in a tetragonal structure with space group no. 139 [2–5]. The system has four inequivalent high-symmetry sites. The crystal structure of $ScFe_4Al_8$ with $I4/mmm$ symmetry and cell parameters $a = 860.2$ pm, $c = 499.8$ pm contains: 2(a) — occupied by Sc, (8f) — Fe site, while 8(i) and 8(j) sites are occupied by Al atoms.

The single crystal measurements were carried out on 4-circle 6T2 spectrometer (Orphée, Saclay), using the (002) reflection of pyrolytic graphite monochromator, with wavelength $\lambda = 2.35$ Å. In accordance with unpolarised neutron data $ScFe_4Al_8$ exhibits flat double modulated structure with magnetic vectors: $k_n^a = \{\tau_x, \tau_x, 0\}$ and $k_n^b = \{-\tau_x, \tau_x, 0\}$, where $\tau_x = \tau_y, n = 1, 2$ while $\tau_1 = 0.13$ and $\tau_2 = 0.18$ [1], respectively. Spin-canted system is realized in ab-plane with $\Delta\varphi^a = 0$ between iron spins along $[\bar{1}10]$ direction and with $\Delta\varphi^b \neq 0$ phase difference along [110] (see Fig. 1). The magnetic structure can be thought of as a superposition of two: non-collinear and collinear ferromagnetic lattices with the μ_{Fe} equal to $1.23(6)\mu_{\mathrm{B}}$ and $1.19(8)\mu_B$, respectively. Such a magnetic cell with experimental amplitudes and phases of Fe moments was used as the input file to search exchange integrals and phase transitions temperatures.

3. Theoretical background

The appearance of the magnetic modulated structure in $ScFe_4Al_8$ compound with double-phase transition is analysed on the basis of the Heisenberg Hamiltonian. In order to reconstruct the exchange integrals map reproducing the observed spin ordering, the atomic magnetic

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moments, modulation vectors and phase transition temperature the incommensurate magnetic structure modulation vector components $\tau_1 = 0.13$ were approximated by $\tau_1 = 2/15$ related to commensurate magnetic supercell, while $\tau_2 = 0.18$ were approximated by $9/50$.

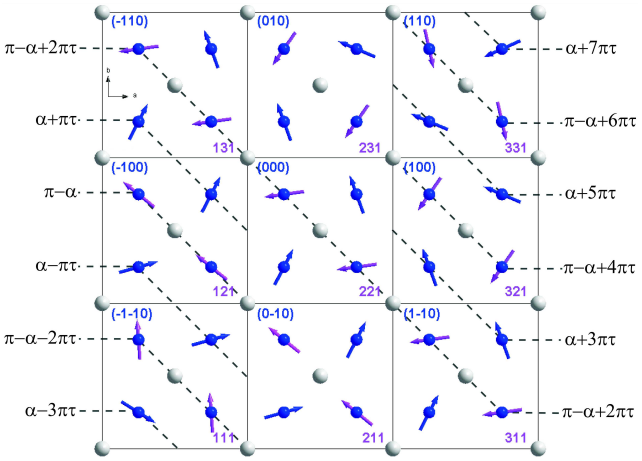

Fig. 1. Schematic representation of the experimentally obtained iron moment arrangement, observed at 8 K in the plane containing the wave vector $(\tau\tau 0)$.

The phase difference $\Delta\varphi^a$ between the nearest neighbours (nn) along a direction, attributable to antiferromagnetic couplings where α is a canting angle [4, 5], implemented during calculations was as close as possible approximation of the experimental results [1]:

$$\Delta\varphi_1^a = \pi - 2\alpha + \pi\tau = 1.13_{\text{exp}} \cong 17/15\pi_{\text{MCPhase}}.$$

In the case of the second modulation the appropriate phase shift is equal to:

$$\Delta\varphi_2^a = 1.18\pi_{\text{exp}} \cong 59/50\pi_{\text{MCPhase}}.$$

Thus, the magnetic supercell is treated as the set of $15 \times 15 \times 8 = 1800$ atoms for τ_1 modulation and 20000 magnetic atoms in the case of τ_2 mode, respectively. Indirect exchange through conduction electrons couples very distant magnetic moments. It is known as the Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction and is the dominant exchange interaction in metals where there is a little or no direct overlap between neighbouring magnetic electrons. In the case of $3d$ metals the on-site interaction between core spin and a conduction electron spin creates the oscillating spin polarization in the conduction band which falls off with the distance as r^{-3}. It leads to long-range oscillatory coupling between atomic spins. For free electrons the RKKY interactions lead to polarization density proportional to $F(\xi) = (\sin\xi - \xi\cos\xi)/\xi^4$, where $\xi = 2k_{\text{F}}r$ and k_{F} is the Fermi wave vector. Spin only magnetism as a common in the transition metal compounds justifies using Hamiltonian with the Heisenberg spins and anisotropic or biquadratic corrections. The starting point of the MCPhase calculation is the Hamiltonian:

$$H = \sum\nolimits_s D_x^2 J_x^2 + D_y^2 J_y^2 + D_z^2 J_z^2$$

$$-\frac{1}{2}\sum\nolimits_{ni\ldots nj=1\ldots 8} J^{ni}\overline{\overline{J}}(n_i n_j) J^{nj},$$

where the crystal field anisotropy is determined by parameters D_x^2, D_y^2, D_z^2 and magnetic interaction tensors between Fe atoms are presented in 1st column of Table. In the following model, anisotropy in the exchange is restricted to the nearest neighbours and the RKKY interactions were treated as the playing dominant role ones. This simple model exchange does not require to take into account the exchange constants, which couple moments parallel to c axis with those in the ab planes, therefore the components J_{ac}, J_{bc} and J_{ca}, J_{cb} as well as J_{cc} of the exchange interaction tensor $\overline{\overline{J}}(n_i n_j)$ have been omitted. The interaction tensor in the case of XY Hamiltonian reduces to square matrix $\overline{\overline{J}}(n_i n_j)$ (see Table). Moreover, the Dzyaloshinskii–Moriya interactions (DM), frequently observed in this class of materials [3–5], enforces the asymmetric off-diagonal components, i.e. $J_{ab} = -J_{ba}$.

TABLE

The solution of bilinear magnetic tensors according to magnetic mode τ_1 dedicated to 6nn of each i-th iron atom located at (8f) positions of the $I4/mmm$ space group, taking into account DM interactions where the appropriate distances with respect to $i = 1\ldots 8$ are expressed in vectors $\boldsymbol{r_i} - \boldsymbol{r_j}$. The elements of the square matrix are shown in the 3th and 4th column.

Exchange tensor $\overline{\overline{J}}(n_i n_j)$	$(r_i - r_j)$ [pm]	MCPhase J_{ij} [meV]	MCMag J_{ij} [meV]
$\begin{bmatrix} A & A & 0 \\ A & A & 0 \\ 0 & 0 & 0 \end{bmatrix}$	$\begin{pmatrix} 0 \\ 0 \\ \pm 249.9 \end{pmatrix}$	$\begin{bmatrix} 0.63 & 0.63 \\ 0.63 & 0.63 \end{bmatrix}$	$\begin{bmatrix} 0.66 & 0.66 \\ 0.66 & 0.66 \end{bmatrix}$
$\begin{bmatrix} -B & -D & 0 \\ D & C & 0 \\ 0 & 0 & 0 \end{bmatrix}$	$\begin{pmatrix} 0 \\ 430.1 \\ 0 \end{pmatrix}$	$\begin{bmatrix} -0.87 & -0.66 \\ 0.66 & 0.59 \end{bmatrix}$	$\begin{bmatrix} -0.72 & -0.84 \\ 0.84 & 0.56 \end{bmatrix}$
$\begin{bmatrix} -E & -G & 0 \\ G & F & 0 \\ 0 & 0 & 0 \end{bmatrix}$	$\begin{pmatrix} 0 \\ -430.1 \\ 0 \end{pmatrix}$	$\begin{bmatrix} -0.78 & -0.69 \\ 0.69 & 0.71 \end{bmatrix}$	$\begin{bmatrix} -0.97 & -0.99 \\ 0.99 & 0.83 \end{bmatrix}$
$\begin{bmatrix} -H & -K & 0 \\ K & I & 0 \\ 0 & 0 & 0 \end{bmatrix}$	$\begin{pmatrix} 430.1 \\ 0 \\ 0 \end{pmatrix}$	$\begin{bmatrix} -0.87 & -0.88 \\ 0.88 & 0.71 \end{bmatrix}$	$\begin{bmatrix} -0.94 & -0.77 \\ 0.77 & 0.69 \end{bmatrix}$
$\begin{bmatrix} -E & -G & 0 \\ G & F & 0 \\ 0 & 0 & 0 \end{bmatrix}$	$\begin{pmatrix} -430.1 \\ 0 \\ 0 \end{pmatrix}$	$\begin{bmatrix} -0.78 & -0.69 \\ 0.69 & 0.71 \end{bmatrix}$	$\begin{bmatrix} -0.97 & -0.99 \\ 0.99 & 0.83 \end{bmatrix}$

During MCMag calculations the DM interaction has been simulated for a sample containing the iron spins lying in the ab-plane with the almost antiferromagnetically coupled moments tilted from b axis. In order to produce this feature, the positive D_z anisotropy coefficients along c, K_{xy} along b axis and a negative K_{xy} coefficient along a axis has been used

$$H = \sum\nolimits_s D_z^2 J_z^2 - K_{xy}^2 \left[J_x^2 - J_y^2\right].$$

During further tests all of the exchange integrals have been modified up to values calculated by MCPhase in the heating scenario. Next, starting from disordered

spin structure (MCPhase and MCMag) with previously found exchange constants the stable magnetic structures of cooling scenario were searched.

4. Results and discussion

During simulations and calculations presented below the distance of the exchange interactions was restricted up to 430.1 pm (nn = 6). Were the iron spins ordered ferromagnetically, the observed transition temperatures $T_1 \approx 230$ K and $T_2 \approx 115$ K [1] would, at the first step, correspond to exchange integrals close to $J_1 \approx 20$ meV and $J_2 \approx 10$ meV, respectively. As dominant terms of Hamiltonian the exchange interactions $J^{\mathrm{RKKY}}_{\mathrm{Fe-Fe}}$ with $k_{\mathrm{F}} = 0.5$ Å^{-1} were considered. The MCPhase programs were used to find the exchange parameters reproducing the ordering temperature and the magnetic structure in the ordered state (see Table).

Taking into account 6 nn, we face the problem of minimizing energy which is described by formula: $E{=}{-}8S^2\,(J_{aa}{+}J_{bb})\cos\pi\tau\,[\cos^2\pi\tau{-}\sin^2\alpha]\,{+}6S^2J_{cc}$. Thus, $\partial E/\partial\tau{=}8\pi S^2\,(J_{aa}{+}J_{bb})\,(3\cos^2\pi\tau{-}\sin^2\alpha)\sin\pi\tau$, $\partial/E\partial\alpha{=}8S^2\,(J_{aa}{+}J_{bb})\sin2\alpha\cos\pi\tau$. Next the incommensurability and noncollinearity dependence on the position of energy minima leads to the simple relations $\partial E/\partial\tau{=}0 \implies \tau{=}0$ (experimentally rejected) or $3\cos^2\pi\tau{=}\sin^2\alpha$ and $\partial E/\partial\alpha{=}0 \implies \tau{=}1/2$ requires $2a\times2b\times c$ magnetic unit cell or $\sin2\alpha{=}0 \implies \alpha{=}0,\pi/2$. $\partial^2E/\partial\tau^2{=}8\pi^2S^2\,(J_{aa}{+}J_{bb})\cos\pi\tau\,[9\cos^2\pi\tau{-}\sin^2\alpha{-}6]$ >0, $\partial^2E/\partial\alpha^2{=}16S^2\,(J_{aa}{+}J_{bb})\cos2\alpha\cos\pi\tau>0$, finally, $\cos\pi\tau>0$ and $9\cos^2\pi\tau{-}\sin^2\alpha>6$, $\cos\pi\tau>0$ and $\cos2\alpha>0$. All these conditions are rejected by experimental results. $J_{aa}=-J_{bb}$ is the only consistent condition and undoubtedly resulted in DM-type anisotropy.

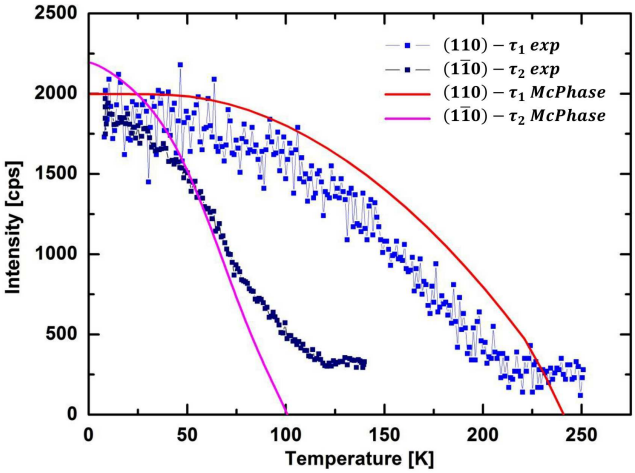

Fig. 2. Thermal variation of the integrated neutron intensities of the strongest magnetic satellites obtained experimentally and during McPhase calculations of simulated cooling for the case of 14 nn.

For a given temperature several possible magnetic structures are stabilized usually and the free energy is calculated. For stable structures all components of the magnetic moment have to be obtained. The MCPhase procedure leads to less anisotropic interactions ($B = H$, see Table) with the resultant magnetic moment 1.38 μ_{B}/atom stable up to $T = 11$ K (MCPhase) with mostly weaker exchange constants while MCMag lets imitate 1 μ_{B}/atom stable up to $T = 51$ K. The exchange constants obtained by use of McPhase program leads to slightly overestimated intensities of the magnetic sattelites observed in neutron scattering (see Fig. 2). The reasons are: (i) calculated iron magnetic moment larger than the experimental one, (ii) the RKKY and DM interactions only are taken into account. The intensities are overestimated by about 16% and 10% for τ_1 and τ_2, respectively, in the range of 2θ up to 50° and in the temperature range up to 100 K.

5. Conclusions

Irrespectively of the applied method, treating spins quantum mechanically or classically, we get similar, satisfactory description of the experimental results. The map of the exchange integrals were reconstructed. We have shown that the dominant antiferromagnetic interactions between iron atoms should be treated as of RKKY + DM origin. In spite of the complexity of the proposed description, the obtained results are encouraging.

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