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EXCHANGE INTERACTIONS IN BARIUM HEXAFERRITE[1]

V. Procházka*, H. Štěpánková[2,*], J. Štěpánek*, A. Snezhko‡, V. Chlan*, K. Kouřil*
*Faculty of Mathematics and Physics, Charles University Prague, V Holešovičkách 2, 18000 Prague 8, Czech Republic
‡Argonne National Laboratory, 9700 South Cass Avenue, Argonne, IL 60439, USA

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Temperature dependences of the hyperfine magnetic fields at ^{57}Fe nuclei in five magnetic sublattices of $BaFe_{12}O_{19}$ hexagonal ferrite were analyzed to obtain parameters characterizing the local exchange interactions. We report theoretical analysis of the temperature dependences of magnetization carried out in the framework of various cluster models based on mean field approaches including the Bethe-Peierls-Weiss approximation. Exchange integrals were extracted from the experimental data reported earlier by G. Albanese et al. (1974) and P. Novák et al. (1989).

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1 Introduction

Magnetic hexagonal ferrites are well known as important materials for permanent magnets and microwave applications. Their magnetic properties are determined by the magnetic moments of the iron ions occupying several magnetic sublattices. Temperature dependences of the individual sublattice magnetizations are detectable by means of hyperfine methods due to a reasonably good proportionality between the hyperfine field at iron nucleus and the corresponding sublattice magnetization. The use of an appropriate theoretical model for the description of the temperature behavior of magnetization in different sublattices makes possible to obtain detailed information about the exchange interactions for iron ion pairs.

Our work is devoted to the barium hexaferrite of magnetoplumbite structure $BaFe_{12}O_{19}$ (BaM). For this structure several papers dealing with fitting the experimentally determined temperature dependences of sublattice magnetizations were published [1–7]. They were based on a relatively simple model of molecular fields and differed in a type and quality of experimental data or in a number of evaluated exchange integrals. Our asset is the utilization of several theoretical approaches for the analysis of the available experimental data; these approaches are based on mean field approximations and include those not used yet for systems with more than two magnetic sublattices.

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[2] E-mail address: helena.stepankova@mff.cuni.cz

The crystal structure (space group = $P6_3/mmc$) of BaM contains Fe^{3+} ions in five crystallographic sites forming five magnetic sublattices [8]. It is a collinear ferrimagnet with the easy magnetization axis parallel to the hexagonal symmetry axis; the Curie temperature is $T_c \sim 730$ K. The five ferric sites (denoted as 2a, 2b, $4f_1$, $4f_2$ and 12k) differ in the arrangement of the neighboring ions, local site symmetry, the spin orientation as well as their number per a unit cell. As a rule, cation sites in the closest vicinity of a given ferric ion belong to several different sublattices. For this reason, one must take into account complicated interaction schemes between a given ferric ion and its Fe^{3+} neighbors.

2 Employed experimental data and theoretical approaches

The magnetization of individual sublattices can be measured by the both well-known hyperfine methods: nuclear magnetic resonance (NMR) technique and Mössbauer spectroscopy. While the former method provides more precise data at low temperatures due to its good spectral resolution, the latter is better applicable up to higher temperatures due to its sensitivity. Very precise NMR data are available from [5]. For higher temperatures only the results of Mössbauer spectroscopy are accessible. We considered the agreement of various published Mössbauer data with the NMR results in the overlapping temperature regions and chose the data published in [2] as the most reliable. We brought together these sets of data and the resulting temperature dependences were used for our analysis.

The magnetization at a given temperature for the ith sublattice is proportional to the mean value of the z-component S_{zi} of the ionic spin at the corresponding site (z-axis lies along the easy axis of magnetization). We approximated the exchange interaction energy of a pair of ions i and j by Hamiltonian of Ising model [9] $H_{ex}^{i,j} = 2J_{ij}S_{zi}S_{zj}$, which neglected correlation of transversal components of the spins in our calculations. Here J_{ij} is an exchange integral. Total spin of 5/2 was assumed for the ferric ions. The range of the exchange interactions was limited to the nearest Fe^{3+} ions, all evaluated J_{ij} were supposed to be positive without any other restrictions. We considered five different spin clusters corresponding to the five sublattices. Each cluster consisted of the central Fe^{3+} ion belonging to the given sublattice and of the shell of its Fe^{3+} neighbors. The mean value S_{zi} for the central ion spin was calculated according to

$$< S_{zi} >= Tr(S_{zi} \exp(-H^i/kT))/Tr(\exp(-H^i/kT)), \quad (1)$$

where H^i corresponds to the Hamiltonian of the ith cluster. The set of five calculated $< S_{zi} >$ temperature dependences was compared with the normalized experimental sublattice magnetizations. Minimization of the sum of squared deviations yields the values of the exchange integrals.

The models we used differed mainly in the way how H^i were constructed. The simplest models based on molecular field considered only exchange interactions between the central spin and the shell spins approximated by their mean values. In the 'full self-consistent' approach ('MF-full') the mean values of the shell spins z-components were found as a solution of a system of self-consistent equations. These equations express for particular ions an equality of their spin z-component mean value for the case when being a central ion and of that when being a shell ion (of the same or another cluster). In the 'direct' treatment of the model fitting ('MF-direct') the mean values for the shell spins were directly taken proportional to the experimental sublattice magnetizations.

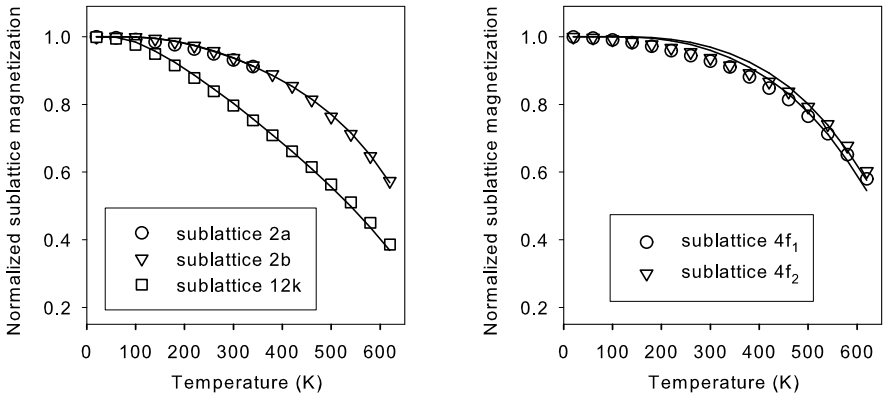

Fig. 1. Temperature dependence of normalized sublattice magnetizations in five magnetic sublattices of BaM. Experimental data (symbols) according to [2,5] and fitted lines for the model 'cluster-all pairs' (CAP).

The 'cluster - all pairs' model ('CAP') represented an expansion of the 'MF-direct' model. The cluster Hamiltonian included Ising terms describing exchange interactions between the central spin and the shell spins, and all exchange interactions of pairs in the shell. The exchange interactions of the shell spins with the neighbors not included in the cluster (outer spins) were approximated by using experimental mean values for the z-components of outer spins.

In Bethe-Peierls-Weiss method the cluster Hamiltonian includes the Ising terms for the exchange interactions between the central and the shell spins, and additional terms representing interactions of the shell ions with their neighbors except for the central ion. These interactions were approximated by means of phenomenological effective magnetic fields at the sites of the shell ions. To determine the effective fields, mean spin z-components values were calculated not only for the central ion but also for the shell ions

$$< S_{zj} >= Tr(S_{zj} \exp(-H^i/kT))/Tr(\exp(-H^i/kT))$$

for the jth site in the ith cluster. The effective fields were set to achieve agreement between the mean spin z-component value obtained when the given ion was considered as the central ion and when it was the shell ion ('BPW-full'). Besides this full consistent procedure for the determination of the effective fields, a routine when the mean z-component values of the shell spins were compared directly to the experimental data ('BPW-semidirect') was employed.

3 Results and conclusions

All five theoretical models provide acceptable convergence. In all cases, a quite good agreement between the optimized theoretical sublattice magnetizations and the experimental ones was achieved with only a slight systematic deviations for the $4f_1$ and $4f_2$ sublattices (see Fig. 1). Comparison of the obtained exchange integrals (Tab. 1) revealed that the models differ in sensitivity to variations of some exchange integrals. Particularly the MF models were found to be

Tab. 1. Exchange integrals J_{ij} determined by using five particular models described in the text (MF-full, MF-direct, CAP, BPW-full, BPW-semidirect) and J_{ij} obtained in ab initio study [10] of electronic structure based on GGA+U method for three values of the parameter U (U_1=3.47 eV, U_2=6.94 eV, U_3=10.4 eV).

J_{ij}(K)	MF-full	MF-direct	CAP	BPW-full	BPW-semid.	U_1	U_2	U_3
J_{af1}	22(4)	20.5(1)	20.96(8)	25.3(6)	20.53(3)	45.4	27.9	17.3
J_{ak}	3(3)	0.002(2)	0.01(1)	0.000(0)	0.000(0)	5.1	0.4	-1.1
J_{bf2}	20.7(1)	20.60(2)	20.57(4)	21.5(3)	21.37(1)	47.9	29.1	18.0
J_{bk}	0.004(4)	0.000(0)	0.000(0)	0.01(1)	0.000(0)	16.4	9.3	5.6
J_{f1f1}	7(7)	4(4)	2(1)	0.1(1)	0.9(1)	4.3	1.1	0.76
J_{f2f2}	67(36)	27(18)	2(1)	0.000(0)	0.03(3)	7.4	2.9	0.4
J_{f2k}	35(9)	24(4)	22.5(5)	24(1)	19.5(1)	46.5	28.6	18.3
J_{kk}	9(2)	2.0(6)	4.97(7)	1.7(6)	0.001(1)	18.1	8.1	3.9
J_{f1k}	10(6)	10(3)	13.2(9)	9.5(4)	11.22(7)	32.1	20.1	12.7

insufficient to determine J_{f2f2} or J_{f1f1}. On the other hand, no significant differences were obtained for the strong exchanges (J_{af1}, J_{bf2}, J_{f2k}, J_{f1k}).

In addition to our results, the exchange integrals calculated in ab initio study [10] based on GGA+U method are given in Tab. 1. Comparison of J_{ij} values from [10] with those we obtained shows a reasonable agreement for the parameter U between 7 and 11 eV and relatively more pronounced exchange interactions corresponding to parallel spins (especially J_{bk}).

Our results have shown that the interactions of the cluster shell ions with other than the central one must be considered (CAP, BPW-full, and BPW-semidirect models) in order to obtain reliable values of some exchange integrals. The determined exchange integrals (Tab. 1) describe exchange interactions in the BaM structure and can be employed to predict effect of diamagnetic substitution on the temperature dependence of z-component mean spin value for neighboring ferric ions (CAP and BPW-full models are applicable for this purpose).

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