

PARAMETERS OF THE DZYALOSHINSKY-MORIYA TYPE WEAK FERROMAGNETISM FOR SOME PEROVSKITE COMPOUNDS

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ПАРАМЕТРИ НА СЛАБИЯТ ФЕРОМАГНЕТИЗЪМ НА ДЗЯЛОШИНСКИ – МОРИЯ
ПРИ НЯКОИ СЪЕДИНЕНИЯ С ПЕРОВСКИТНИ СТРУКТУРА

Изследвани са магнитните свойства на някои чисти ферити с перовскитова структура, както и на смесени съединения от вида $\text{Ho}_x\text{Tb}_{1-x}\text{O}_3$, $\text{HoMn}_x\text{Fe}_{1-x}\text{O}_3$, $\text{Ho}_x\text{Cr}_{1-x}\text{O}_3$, $\text{DyFe}_x\text{Cr}_{1-x}\text{O}_3$ в широк температурен интервал. Показано е, че ъгълът на който се отклонява спонтанното намагнитване на преходния елемент от групата на желязото от оста на антиферомагнитно подреждане е постоянен при всички температури.

От стойността на този опитно определен ъгъл и от изчисления коефициент на взаимодействието между $4f$ и $3d$ решетките е определен енергетичния параметър на асиметричния обмен на Дзялошински – Мория.

Dimitar Dimitrov, Iliya Radulov, Vassil Lovchinov, Andrei Apostolov. PARAMETERS OF THE DZYALOSHINSKY-MORIYA TYPE WEAK FERROMAGNETISM FOR SOME PEROVSKITE COMPOUNDS

Magnetic properties of some pure ferrites with perovskite structure, as well as of some mixed compounds of the type $\text{Ho}_x\text{Tb}_{1-x}\text{O}_3$, $\text{HoMn}_x\text{Fe}_{1-x}\text{O}_3$, $\text{Ho}_x\text{Cr}_{1-x}\text{O}_3$, $\text{DyFe}_x\text{Cr}_{1-x}\text{O}_3$ are investigated in wide temperature range. Results manifested the constancy of angle α , at which the rare-earth element spontaneous magnetization separates from the axis of antiferromagnetic ordering. The Dzyaloshinsky – Moriya energy parameter of asymmetric exchange is defined

from the experimentally determined angle α and from the calculated coefficient of the $4f$ and $3d$ lattices exchange.

Keywords: single crystal manganites, magnetoelectric and ferroelectric properties, multiferroics

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1. INTRODUCTION

Compounds with distorted perovskite structure of the $4-f$ and $3-d$ transition metals with the common formula LnTO_3 (where Ln is rare-earth element, T is an element from the Fe group) are the most multifold binary oxides of these two groups elements. Wide range of stability for this structure allows the realization of combinations of the Lanthanides with all the transition metals except the Nickel. Quite interesting physical phenomena take place in these oxides like charge and orbital ordering; relatively independent magnetic lattices of the both metals; particular magnetic structures; high optical indicators; giant magneto-resistance; peculiar dielectric and ferroelectric properties etc.

We have investigated the magnetic properties of pure ferrites and chromium-based materials as well as of some mixed type oxides like $\text{Ho}_x\text{Tb}_{1-x}\text{O}_3$, $\text{HoMn}_x\text{Fe}_{1-x}\text{O}_3$, $\text{HoMn}_x\text{Cr}_{1-x}\text{O}_3$ and $\text{DyFe}_x\text{Cr}_{1-x}\text{O}_3$.

The samples magnetic properties are investigated between 2 K and 1000 K. Magnetic fields up to 2.5 T and the ballistic method are applied below 300 K and a magnetic relaying balance is used above this temperature.

Aim of the investigation is to measure the magnetic properties in respectively wide temperature and magnetic regions in order to determine the Dzyaloshinsky-Moria effect which appears in the perovskites.

2. RESULTS AND DISCUSSION

The investigations show that above a certain transition temperature T_{NI} all the compounds are in paramagnetic state and the measured susceptibility χ_1 follows exactly the Curie-Weiss law with an effective magnetic moment near to the calculated of magnetic ions at the applied proportions and with negative asymptotic temperature θ_1 (see fig.1). Negative value of this quantity indicates that the negative (anti-ferromagnetic) interactions predominate between the magnetic ions. Each of the measured magnetizations (see fig.2) between T_{NI}

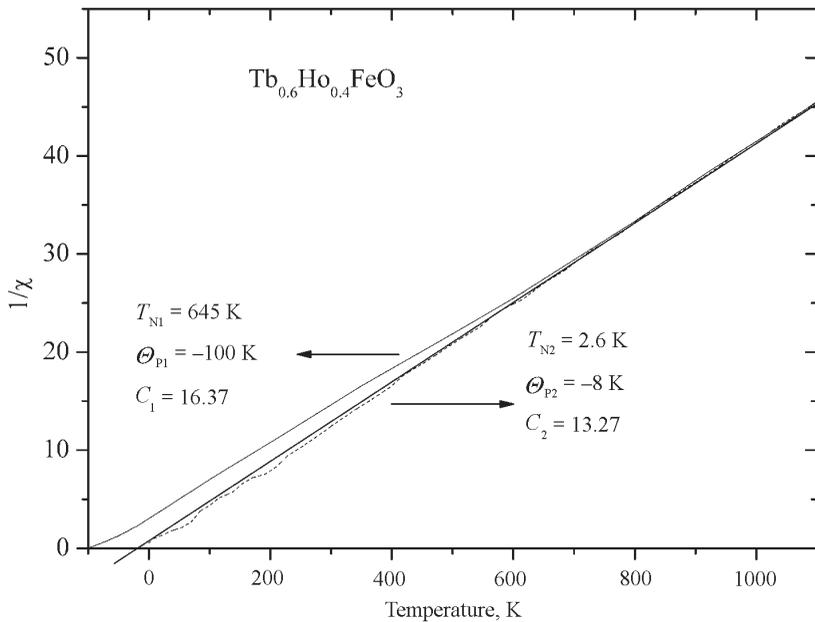


Fig. 1. (Ho-Tb)FeO₃ susceptibility

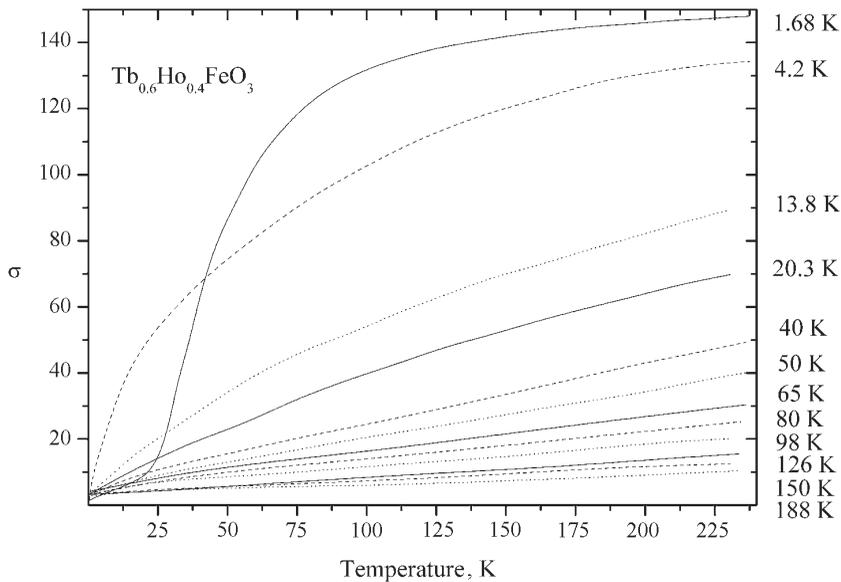


Fig. 2. Magnetic dependences $M=f(H)$ for (Ho-Tb)FeO₃ compound

and a certain lower transition temperature T_{N2} is a sum of two terms: one which is proportionally changes to the applied magnetic field and characterizes a second susceptibility χ_2 ; and second ferromagnetic term which allows measuring the spontaneous magnetization at each temperature. Temperature dependence of the reciprocal χ_2 value is a straight line with declination different from that of $1/\chi_1$ and gives the opportunity to determine second negative paramagnetic temperature θ_2 (see fig. 1). At temperatures, e.g. lower than 15 K the susceptibility experimental points become detached from the straight line $1/\chi_2$ and show phase transition which is taking place at different temperatures for the respective compounds (see table 1)

The total magnetization observed experimentally in the regarded area is:

$$M + M_{4f} + M_{3d} = M_{4f} + \chi_1.H + m_{3d} = \frac{C_{4f}}{T - \theta_{p2}} (H + n_{3d-4f}m_{3d}) + \chi_{3d}H + m_{3d}, \quad (1)$$

where χ_{3d} presents the weak antiferromagnetic susceptibility of the arranged 3d metals, c_{4f} is the Curie constant for the Lanthanide. It is much weaker than the paramagnetic susceptibility of the Lanthanides (this can be directly measured for compounds including Lu, La or Y) and, therefore, can be neglected. Thus, for the magnetization with reasonable approximation can be written:

$$M = \frac{C_{4f}}{T - \theta_{p2}} (H - n_{3d-4f} \sin\alpha M_{Fe}) + M_{3d} \sin\alpha. \quad (2)$$

Spontaneous ferromagnetic magnetization measured at $H = 0$ fulfils the equation

$$\frac{M_{\text{spon}}}{M_{3d}} (T - \theta_{p2}) = n_{3d-4f} \sin\alpha c_{4f} + \sin\alpha (T - \theta_{p2}). \quad (3)$$

Here m_{3d} is substituted by $M \sin\alpha$ which is evident. If in the left part of the equation θ_{p2} is replaced by $\theta_{\text{pR.E.}}$ ($\theta_{\text{pR.E.}}$ means the paramagnetic temperature for the rare-earth element) and it is plotted versus $(T - \theta_{\text{pR.E.}})$ the result should be a straight line. This is graphically verified. The declination of the line and the piece form the x -axis define the angle α and the coefficient n_{4f-3d} of weak interaction between the Lanthanides and the rest transient elements. These results are presented in Table 1. It is worth to notify that the angle α for the HoFeO_3 compound was directly identified by pulse neutronography [5] and the obtained value does not essentially differ from those given in the Table 1.

Table 1

Compound	α (mrad)	n	D (erg/gauss)
HoMn _{0.1} Fe _{0.9} O ₃	3	-273	1.4
HoMn _{0.2} Cr _{0.8} O ₃	12	-115	2.8
HoMn _{0.25} Cr _{0.75} O ₃	5	-104	1.0
HoMn _{0.3} Cr _{0.7} O ₃	30	-127	7.6
HoMn _{0.4} Cr _{0.6} O ₃	30	-105	6.4
HoMn _{0.7} Fe _{0.3} O ₃	14	-202	6.2
Ho _{0.6} Tb _{0.4} FeO ₃	18	-326	4.0
HoFeO ₃	4	-263	4.0
TbFeO ₃	6	-368	4.4
DyFeO ₃	7	-335	2.3
LuFeO ₃	17	-210	7.1

We have applied an entire investigation to the lutetium and yttrium ferrite systems. Lutetium ferrite, for example, orders anti-ferromagnetically below 625 K with weak ferromagnetic moment associated with the anti-ferromagnetism. The ferromagnetic moment value is low (about 0.03–0.05 μ_B) and remains unchanged up to 20 K. The ratio between experimentally determined weak ferromagnetic moment and calculated by the Brilluen's law is constant and indicates that the angle of deflection of the Fe spontaneous magnetisation from the anti-ferromagnetic axis is constant at the whole temperature region.

These results are checked out for the yttrium ferrite as well as using the available data for Lu and Y chromites in the literature [1, 2].

Explanation of the observed features [2, 3] is classic: At high temperatures the compounds are in paramagnetic state; under the first transition temperature only the $3d$ -lattice orders anti-ferromagnetically; the $4f$ -lattice (if the lanthanide is paramagnetic) retains in disordered state. The $3d$ -lattice anti-ferromagnetism is accompanied by weak ferromagnetic moment which is measured for La, Lu, and Y in pure mode. The weak ferromagnetic moment polarises the paramagnetic lanthanides and can induces a total ferromagnetic moment up to 3 μ_B (e.g. for Ho ferrite). The low angle α assessment (at which $3d$ magnetisations of different compounds decline from the anti-ferromagnetic axis) is one of the aims of the present work.

Here below we shall use the Neel model [4] concerning the molecular (self arranged) field applied to the complicated perovskite structure in the following way:

- $3d$ -lattice divides into two antiparallel lattices, each including half of the all atoms;

- Molecular field coefficients of the $3d$ -lattice are n_{3d} and n'_{3d} . n_{3d} accounts for the near neighbour interactions, i.e. between the both sublattices and n'_{3d} accounts for the interactions between the magnetic atoms which are inside of each from the sublattices;
- Noncolinearity of the both antiferromagnetically situated sublattices leads to their diversion from the antiferromagnetic axis with a small angle α and therefore, leads to weak magnetic moment m_{3d} ;
- Rare earth ions can be divided also into two antiparallel lattices with coefficients for them $n_{R.E.}$ and $n'_{R.E.}$, respectively.
- The molecular field coefficient n_{3d-4f} defines the weak interaction between the magnetic lanthanides and $3d$ ions.

Lanthanide ions are in paramagnetic state in the temperature region between T_{N1} and T_{N2} and are under the influence of the external magnetic field and the $3d$ -molecular field through the coefficient n_{3d-4f} :

$$M_{4f} = \frac{C_{4f}}{T - \theta_{p2}} (H + n_{3d-4f} m_{3d}), \quad (4)$$

here C_{4f} is the experimentally measured by the $1/\chi_2$ value of the Curie constant for the lanthanide (see table1).

The physical origin of the weak ferromagnetism, i.e. the uncolinearity of the $3d$ antiferromagnetic sublattices can result from:

- One ion anisotropy (classical example – NiFe) originates from the existence of two nonequivalent crystallographic nodes occupied by equivalent ions. Thus, each ion is affected by different crystallographic field and, therefore, nonparallel axes of anisotropy are created [6];
- Dzyaloshinsky asymmetric exchange (classical example α -Fe₂O₃) where the simultaneous action of the spin-orbital and mediate exchange junction leads to anisotropic interaction of the type $D(M_1 \times M_2)$,

where D is vector power parameter and M_1 and M_2 are the magnetisations of the both antiferromagnetic lattices [7].

Gorodetsky et al. show for the first time [8] that the probable mechanism for the weak ferromagnetism of the rare-earth perovskites is of the second type and now it is commonly accepted opinion. It is of particular interest to determine experimentally not only the angle α but the power parameter D (Dzyaloshinsky-Moriya parameter) also.

The energy level of the $3d$ -system in the examined compounds can be defined with reasonable approximation only from the isotopic and asymmetric exchange forces (the crystallography anisotropic energy is proportional to the $\sin\alpha$ and in our relatively crude model can be neglected). For the case it can be written

$$W = -\frac{1}{2} \sum_{i=1}^2 h_i M_i - D[M_1 M_2], \quad (5)$$

$$h_1 = n_{3d} M_2 + n'_{3d} M_1,$$

$$h_2 = n_{3d} M_1 + n'_{3d} M_2,$$

and thus $|M_1| = |M_2|$, $D \parallel (M_1, M_2)$, then

$$W = nM_{3d}^2 \cos 2\alpha - DM_{3d}^2 \sin 2\alpha - 2n'_{3d} M^2, \quad (6)$$

$$\frac{dW}{d\alpha} = -n \sin \alpha - D \cos \alpha = 0, \quad (7)$$

$$\operatorname{tg} 2\alpha = \frac{D}{n}, \quad (8)$$

$$|\alpha| = |\alpha| = \frac{D}{2n}. \quad (9)$$

There are few other direct measurements of this parameter which is of the order of ten miliradians as it is shown in table 1.

3. CONCLUSION

Parameters characterising the weak ferromagnetism associated to the antiferromagnetism in double oxides with perovskite structure of the $3d$ and $4f$ elements were determine applying direct magnetic measurements in wide temperature range.

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