The order of magnetic phase transitions in disordered double perovskite oxides Sm_2FeCoO_6 and Dy_2FeCoO_6

Cite as: AIP Advances **8**, 101340 (2018); https://doi.org/10.1063/1.5042757
Submitted: 04 June 2018 . Accepted: 15 October 2018 . Published Online: 29 October 2018

G. R. Haripriya, R. Pradheesh, K. Sethupathi, and V. Sankaranarayanan

AIP ADVANCES **8**, 101340 (2018)

The order of magnetic phase transitions in disordered double perovskite oxides Sm_2FeCoO_6 and Dy_2FeCoO_6

G. R. Haripriya,[a] R. Pradheesh, K. Sethupathi, and V. Sankaranarayanan
Low Temperature Physics Laboratory, Dept. of Physics, IIT Madras, Chennai 600036, India

(Received 4 June 2018; accepted 15 October 2018; published online 29 October 2018)

We present the results on the order of phase transition around the spin re-orientation region using isothermal magnetization data performed on the sol-gel synthesized disordered double perovskite oxides Sm_2FeCoO_6 and Dy_2FeCoO_6 by Arrott plot method. The temperature variation of the DC magnetization data of both the samples show that there are two magnetic phase transitions; one at high temperature regime, the conventional paramagnetic (PM) to ferro or ferrimagnetic (FM or FIM) and the other at low temperatures, ferro or ferrimagnetic (FM or FIM) to antiferromagnetic (AFM). From the Arrott plot method, it is inferred that spin re-orientation transitions of both the compounds has a first order nature corresponding to the FM (FIM) to AFM transition. https://doi.org/10.1063/1.5042757

I. INTRODUCTION

Magnetic double perovskite oxides have gained their importance in material science due to their efficiency in exhibiting different tunable physical properties such as magnetoresistance (MR), magnetocaloric effect (MCE), multiferroicity, etc.[1–3] The rare earth based transition metal perovskites of the form $R_2MM'O_6$, where R is the rare earth cation and M and M' are transition metal cations, are one of the afore mentioned category which usually exhibit interesting magnetic phenomena due to oxygen mediated cationic exchange interactions.[2,3]

The spin re-orientation is commonly observed in Fe, Cr and Mn based magnetic alloys, rare earth perovskites, etc. in which the transition ion spin changes its orientation from one crystallographic direction to the other influenced by temperature, magnetic field, internal pressure, external irradiation etc.[4–6] In general, it can be of a first order abrupt transition or a slower broad transition.[5] The order of the magnetic phase transition can be preliminarily determined by means of Arrott plots,[7,8] which include modified isotherms with M^2 and H/M on the axes.

The magnetoresistive double perovskite Sr_2FeMoO_6 geared up the material research in double perovskites due to technology related reasons.[1] The disordered double perovskites Dy_2FeCoO_6 (DFCO) and Sm_2FeCoO_6 (SFCO) are found to exhibit re-entrant magnetization, thermal hysteresis, ZFC-FC bifurcation, spin re-orientation with an antiferromagnetic ground state.[9] The transition temperatures are found to be lower than that of the corresponding orthoferrite end members.[10,11] The origin of spin re-orientation in these samples are assumed to be the R^{3+} - Fe^{3+} interactions. The occurrence of multiple magnetic transitions in the samples with change in temperature and magnetic field leads us to think about the application of Arrott plot method at the spin re-orientation region to see the behaviour of the order of transition. In this work, we present the characterization of the order of transition around the spin re-orientation region using Arrott plot method for the above referred samples.

[a] haripriyagr@gmail.com

II. EXPERIMENTAL

The sol-gel synthesized samples[9] were subjected to DC magnetization measurements in a commercial SQUID based vibrating sample magnetometer (SVSM, MPMS 3, QD USA). The temperature variation of magnetization was taken up to 320 K from 5 K in zero field cooled (ZFC) mode with a magnetic field of 100 Oe. One quadrant isothermal curves were recorded up to a maximum magnetic field of 70 kOe for temperatures close to the observed transitions for the Arrott analysis.

III. RESULTS AND DISCUSSIONS

The Fig. 1(a) and (b) show the temperature variation of magnetization for the samples Dy_2FeCoO_6 (DFCO) and Sm_2FeCoO_6 (SmFCO). The different transition temperatures observed were: Neel temperature for the samples around ~250 K (248 K for DFCO, 246 K for SmFCO), the spin re-orientation temperature around ~86 K for DFCO and around ~210 K for SmFCO. The double peaks observed at ~86 K (for DFCO) and ~210 K (for SmFCO) in comparison with the orthoferrite counter parts indicate the spin re-orientation feature in the samples.[9–11]

The derivative curves indicate that the transition around 250 K has a ferromagnetic-like behaviour, while those at spin re-orientation temperatures show a double peak behaviour. From the first observation the spin re-orientation of DFCO, which is sharp, can be of first order. While the one for SmFCO with a broadened peak appears to be that of a second order phase transition due to the broad nature.

In order to classify the order of different magnetic transitions nearer to spin re-orientation in the samples, Arrott plot method is adapted.[7,8,12,13] For this purpose, one quadrant M vs H curves were recorded around the respective transitions observed for the samples in a magnetic field range of 0 Oe – 70 kOe. The unsaturated nature of the M vs H curves (not shown here), even at a magnetic field of 70 kOe, point towards the AFM/Weak ferromagnetic nature of the samples.

For DFCO, the spin re-orientation temperature is found to be around ~86 K, so the M vs H curves were recorded from 75 K to 91 K with an interval of 2 K, where M vs T exhibited a sharp transition. For SmFCO, the spin re-orientation is around 210 K and the M vs H curves were recorded from 175 K to 277 K with an interval of 3 K, and this appeared as a broad transition.

From the equation of state, $\frac{H}{M} = a + b\,M^2$, where M is the magnetization, H is the applied magnetic field in Oe, a and b are the coefficients. If the slope of the H/M vs M^2 curve is negative then the transition is considered to be of first order and for positive slopes it is of second order.[12,13]

Figures 2 and 3 represent the Arrott plots for DFCO and SmFCO respectively. The insets are the enlarged view of low field regions. The linear fits were utilized for finding out the intercept values to distinguish the order of phase transitions. The value of intercept at 211 K, for high field region is found to be negative for SmFCO, which indicates the possibility of a first order magnetic transition.

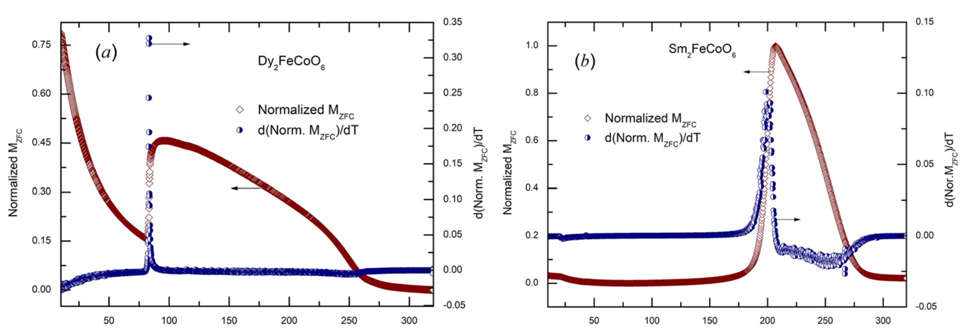

FIG. 1. The normalized ZFC magnetization (maroon diamonds) and its derivative (blue circles) of (a) DFCO and (b) SFCO.

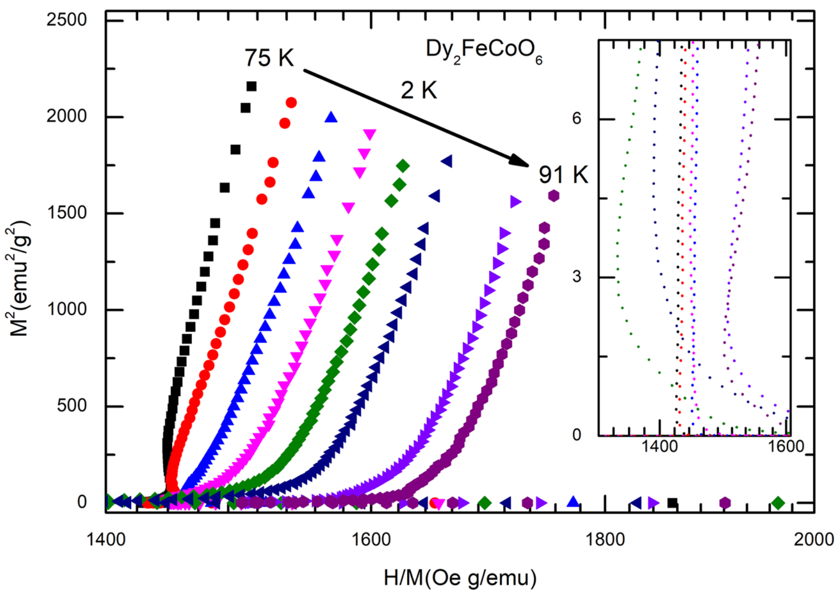

FIG. 2. Arrott plots for Dy_2FeCoO_6 for the low temperature region (75 – 91 K).

In view of the slopes, it is found that the curves for the low temperature region of Dy_2FeCoO_6 show a negative slope ('S- shape behaviour') which is considered as a first order phase transition below the spin re-orientation temperature 86 K (refer Fig. 2), while the high temperature region has positive slope inferring a second order phase transition (not shown).[12,13] For the sample Sm_2FeCoO_6, the one nearer to spin reorientation (~210 K) has a first order behaviour (refer Fig. 3). From the Arrott plot analysis it can be inferred that the first order spin re-orientation results in a FM-AFM transition in the samples. The presence of disordered B-sites with the magnetic ions of Fe and Co in mixed valence states may be the cause for the complicated magnetic structure observed in these compounds.

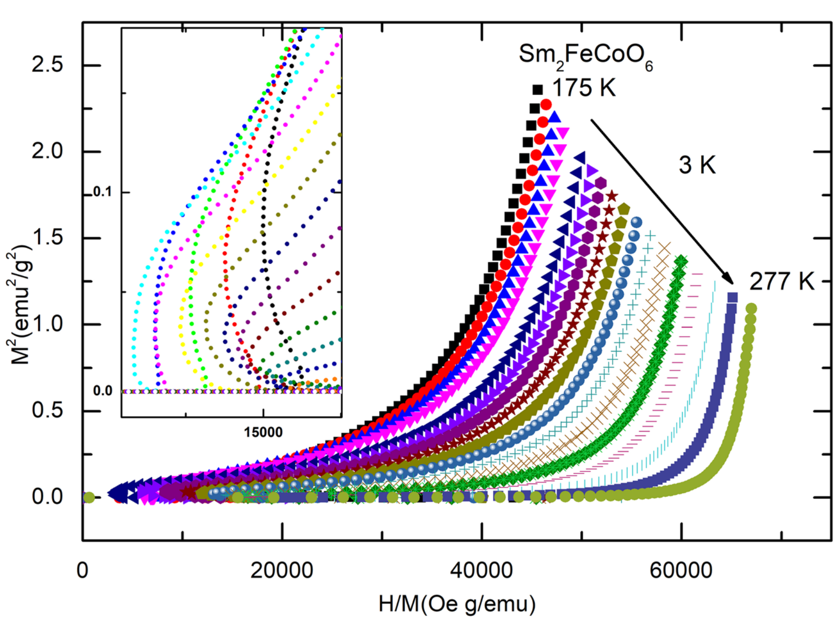

FIG. 3. Arrott plots for Sm_2FeCoO_6 for the high temperature region (175 – 277 K).

IV. CONCLUSIONS

The disordered double perovskites Dy_2FeCoO_6 and Sm_2FeCoO_6 show spin re-orientation transition around ~86 K and ~210 K respectively. The Arrott plot analysis showed that both the samples have a first order spin re-orientation. Detailed analysis and measurements are yet to be performed for getting more information on the complicated magnetism exhibited by the samples.

ACKNOWLEDGMENTS

Authors acknowledge IITM for funding SVSM. HGR thank Mr. Seenivasan (IIT Madras) for extended experimental support.

[1] J. Navarro, C. Frontera, L. I. Balcells, B. Martinez, and J. Fontcuberta, Phys. Rev. B **64**, 092411 (2001).
[2] L. H. Yin, J. Yang, R. R. Zhang, J. M. Dai, W. H. Song, and Y. P. Sun, Appl. Phys. Lett. **104**, 032904 (2014).
[3] S. K. Giri, P. Dasgupta, A. Poddar, and T. K. Nath, J. Alloys and Compounds **631**, 266–271 (2015).
[4] H. Horner and C. M. Verma, Phys. Rev. Lett. **20**(16), 845 (1968).
[5] T. Yamaguchi, J. Phys. Chem. Solids **35**(4), 479 (1974).
[6] H. Nhalil, S. Harikrishnan, S. R. Nair, M. S. André, and E. Suja, J. Appl. Phys. **117**(17) 173904 (2015).
[7] S. K. Giri and T. K. Nath, J. Mag. Mag. Mater. **324**, 2277–2285 (2012).
[8] W. Zhong, C.-T. Au, and D. You-Wei, Chin. Phys. B **22**(5), 057501 (2013).
[9] G. R. Haripriya, R. Pradheesh, M. N. Singh, A. K. Sinha, K. Sethupathi, and V. Sankaranarayanan, AIP Advances **7**, 055826 (2017).
[10] Z. Zhou, L. Guo, H. Yang, Q. Liu, and F. Ye, J. Alloys and Compounds **583**, 21 (2014).
[11] S. Cao, H. Zhao, B. Kang, J. Zhang, and W. Ren, Scientific Reports **4**, 5950 (2014).
[12] A. K. Pramanik and A. Banerjee, Phys Rev B **79**, 214426 (2009).
[13] H. S. Shin *et al.*, Solid State Commun. **118**(7), 377–380 (2001).