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1998 J. Phys.: Condens. Matter 10 11049

(http://iopscience.iop.org/0953-8984/10/48/024)

J. Phys.: Condens. Matter **10** (1998) 11049–11054. Printed in the UK PII: S0953-8984(98)95898-2

The relationship between field-cooled and zero-field-cooled susceptibilities of some ordered magnetic systems

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Received 14 July 1998, in final form 7 October 1998

Abstract. Analysis of the irreversible field-cooled (FC) and the zero-field-cooled (ZFC) magnetic susceptibilities of one ferrimagnetic and three ferromagnetic systems, measured at different applied magnetic fields, shows that the irreversibility indicated by the difference between the FC and the ZFC susceptibilities arises from magnetic anisotropy. The two susceptibilities are related to each other through the coercivity (H_c) which is a measure of the anisotropy. The ZFC susceptibility can be calculated from the FC susceptibility (or vice versa) and the coercivity.

1. Introduction

Certain magnetic systems undergoing transitions to ordered ferromagnetic [1–6], antiferromagnetic [7] and ferrimagnetic [8] states are reported to show irreversibility, indicated by the difference between their field-cooled (FC) and zero-field-cooled (ZFC) susceptibilities. The magnetic susceptibility is larger when the sample is cooled in a magnetic field (χ_{FC}) than when it is cooled in zero magnetic field (χ_{ZFC}). The irreversible FC and ZFC magnetic behaviour is one of the characteristic features of a spin glass [9, 10]. Other characteristic properties of a spin glass include the relaxation of magnetization (the aging effect) and the presence of a cusp in the ac susceptibility (χ_{ac}); the cusp temperature (T_f) increases with increasing frequency of the ac signal. However, the aging effect and a cusp in χ_{ac}, with frequency dependence of T_f, are observed for ferromagnetic systems also [1–5]. The origin of the spin-glass-like features of the magnetically ordered systems are explained in terms of magnetic frustrations, arising from the competing ferromagnetic and antiferromagnetic exchange interactions, deformed lattices, random distribution of the magnetic ions etc.

Our recent studies on the low-field magnetic behaviour of some ferromagnetic [11] and ferrimagnetic [12] oxides indicated that their spin-glass-like magnetic properties originate from the magnetocrystalline anisotropy. The temperature at which a cusp (or maximum) is observed in the zero-field-cooled susceptibility and the temperature below which irreversibility is observed, indicated by the difference between M_{FC} and M_{ZFC}, were found to be related to the magnitude and the temperature variation of the coercivity, which is a measure of the magnetic anisotropy. Though spin glasses and materials showing spin-glass-like features such as superparamagnetic [13–15] and cluster glass [16, 17] systems, superconducting oxides [18], the ordered magnetic systems etc show irreversible FC and

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0953-8984/98/4811049+06$19.50

ZFC magnetic behaviour and a cusp in their ZFC susceptibilities, the origin of such a behaviour is not yet properly understood. In this paper we report a relationship between the field-cooled and zero-field-cooled susceptibilities of some magnetically ordered systems. The results indicate that the ZFC susceptibility at each measuring field is directly related to the corresponding FC susceptibility through the coercivity.

2. Experiments

The perovskites, $SrRuO_3$ [19], $La_{0.7}Ca_{0.3}MnO_3$ [20] and $La_{0.5}Sr_{0.5}CoO_3$ [21], and the Ni–Zn ferrite, $Ni_{0.8}Zn_{0.2}Fe_2O_4$ [22], were synthesized and characterized as reported previously. FC and ZFC magnetization measurements at different applied magnetic fields were made on a PAR 4500 vibrating-sample magnetometer (VSM).

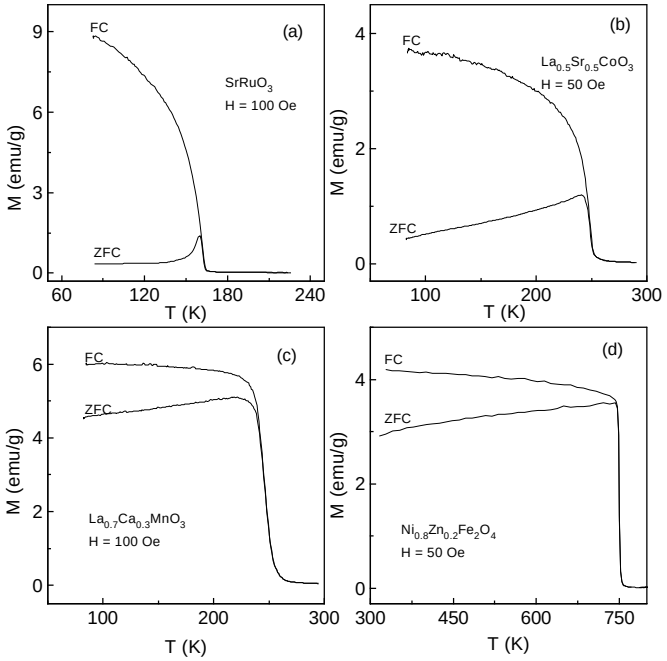

Figure 1. Curves showing M_{FC} and M_{ZFC} for $SrRuO_3$, $La_{0.5}Sr_{0.5}CoO_3$, $La_{0.7}Ca_{0.3}MnO_3$ and $Ni_{0.8}Zn_{0.2}Fe_2O_4$, measured at the low magnetic fields indicated.

3. Results and discussion

Typical FC and ZFC magnetization curves for polycrystalline samples of three ferromagnetic systems, $SrRuO_3$, $La_{0.5}Sr_{0.5}CoO_3$ and $La_{0.7}Ca_{0.3}MnO_3$, and that of a ferrimagnetic system, $Ni_{0.8}Zn_{0.2}Fe_2O_4$ (Ni–Zn ferrite), measured at low magnetic fields, are compared in figure 1. For $SrRuO_3$, H_c varies between 3600 and 100 Oe in the temperature range 82–160 K ($T_c = 161$ K); for $La_{0.5}Sr_{0.5}CoO_3$, H_c varies between 320 and 30 Oe in the temperature range 82–250 K ($T_c = 252$ K); for $La_{0.7}Ca_{0.3}MnO_3$ the variation of H_c is between 35 and 17 Oe in the temperature range 82–240 K ($T_c = 245$ K); for $Ni_{0.8}Zn_{0.2}Fe_2O_4$, H_c varies between 21 and 4 Oe in the temperature range 323–723 K ($T_c = 750$ K) [11,

12]. The differences between the magnitude and temperature variations of the coercivities of the four compounds are reflected in the extent of the irreversibility, indicated by the difference between the M_{FC}- and the M_{ZFC}-curves. For $SrRuO_3$, with very large coercivity at low temperatures, M_{FC} continuously increases below T_c as the temperature is decreased and M_{ZFC} shows a large drop below T_f giving a cusp. On the other hand, for the low-coercivity materials, M_{FC} remains almost constant below T_c and M_{ZFC} shows only a small decrease, with a broad maximum at T_f. Since coercivity is related to magnetic anisotropy, the irreversible magnetic behaviour reflects the role of anisotropy in determining the shapes of the FC and the ZFC curves below the ordering temperature.

Magnetic anisotropy aligns the spins in a preferred direction. Since no magnetic field is applied while cooling the sample through the ordering temperature during the process of zero-field cooling, the spins are locked in random directions for a polycrystalline specimen. When a small magnetic field is applied at the lowest temperature, far below the Curie temperature, the magnitude of the resultant magnetization will depend on the anisotropy of the system. If the system is highly anisotropic, the small applied field will not be sufficient to rotate the spins in the direction of the applied field and therefore the magnetization will be very small (as in figures 1(a) and 1(b)). The reverse is true if the system is less anisotropic (as in figures 1(c) and 1(d)). During the FC process, the sample is cooled through its Curie temperature, T_c, in the presence of a magnetic field. Therefore the spins will be locked in a particular direction depending on the strength of the applied field, as soon as the system is cooled below its ordering temperature. The FC magnetization will remain almost constant if the anisotropy of the sample is very low (figures 1(c) and 1(d)) or increase with decreasing temperature for highly anisotropic materials (figures 1(a) and 1(b)). Therefore, during both the FC and the ZFC processes, the anisotropy field plays a crucial role in determining the magnetization at a given field strength.

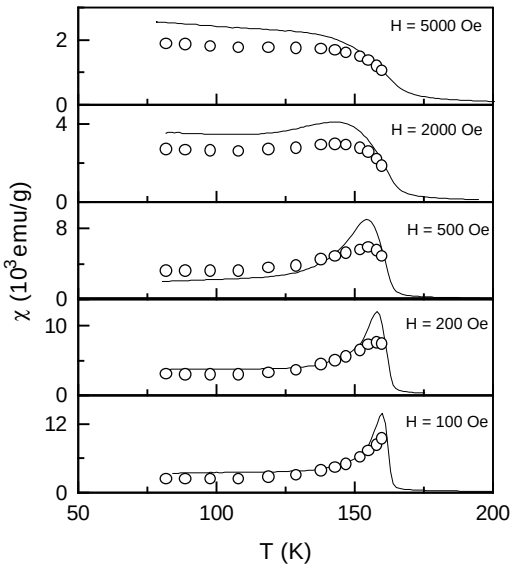

Figure 2. Curves showing χ_{ZFC} (——) and χ'_{FC} (○) for $SrRuO_3$, measured at the different magnetic fields indicated.

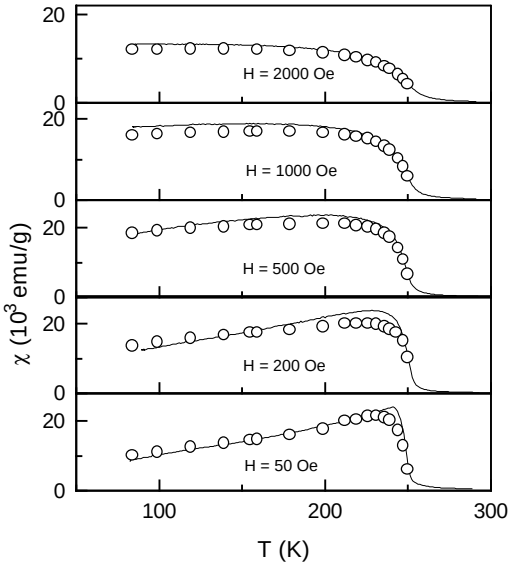

Figure 3. Curves showing χ_{ZFC} (——) and χ'_{FC} (○) for $La_{0.5}Sr_{0.5}CoO_3$, measured at the different magnetic fields indicated.

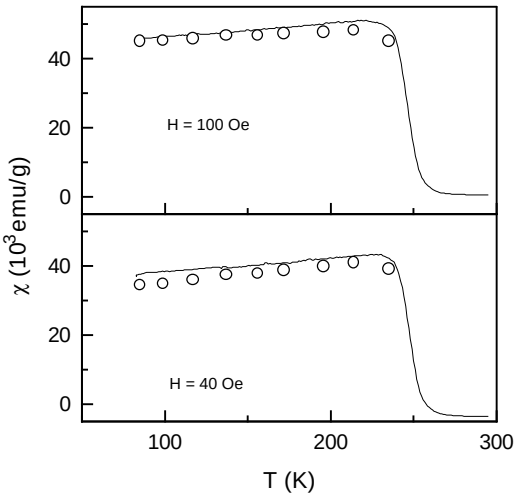

Figure 4. Curves showing χ_{ZFC} (——) and χ'_{FC} (○) for $La_{0.7}Ca_{0.3}MnO_3$, measured at the two different magnetic fields indicated.

Figures 2–5 show the magnetic susceptibilities calculated from M_{FC} using the relation

$$\chi'_{FC} = \frac{M_{FC}}{H_{app} + H_c} \tag{1}$$

for $SrRuO_3$, $La_{0.5}Sr_{0.5}CoO_3$, $La_{0.7}Ca_{0.3}MnO_3$ and $Ni_{0.8}Zn_{0.2}Fe_2O_4$, respectively, at different applied fields (H_{app}). The notation χ'_{FC} is used to distinguish this susceptibility from the normal field-cooled susceptibility $\chi_{FC} = M_{FC}/H_{app}$. The normal zero-field-cooled

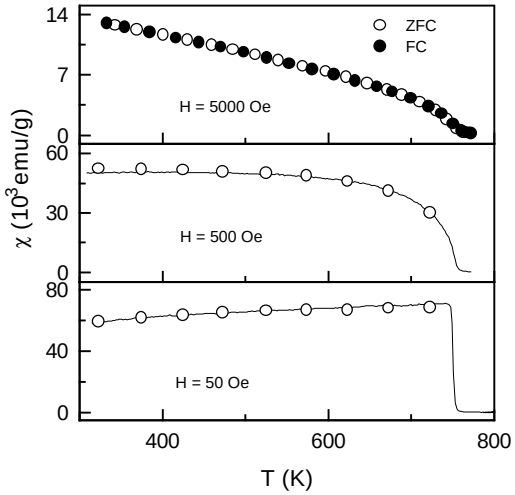

Figure 5. Curves showing χ_{ZFC} (——) and χ'_{FC} (○) for $Ni_{0.8}Zn_{0.2}Fe_2O_4$, measured at the two different magnetic fields indicated (lower and middle panels). The upper panel shows χ_{FC} and χ_{ZFC}, measured at $H_{app} = 5000$ Oe.

susceptibility, $\chi_{ZFC} = M_{ZFC}/H_{app}$, is also shown in the figures for comparison. Since the coercivity is measured only at selected temperatures, χ'_{FC} is calculated only at these temperatures from the corresponding M_{FC}-values. Interestingly, the χ'_{FC}-curves show most of the features of the χ_{ZFC}-curves. The shapes of the χ'_{FC}- and the χ_{ZFC}-curves are almost the same, and their magnitudes are comparable for all four magnetic systems at each applied field.

The total magnetic field, $H_{app} + H_c$, is a function of the temperature, since H_c varies with temperature. The similarity between χ'_{FC} and χ_{ZFC} indicates that M_{FC} and M_{ZFC} are related through the coercive field (anisotropy field). The zero-field-cooled magnetization (susceptibility) is the field-cooled magnetization (susceptibility) modified by the anisotropy field. Therefore,

$$\frac{M_{FC}}{H_{app} + H_c} \approx \frac{M_{ZFC}}{H_{app}} \tag{2}$$

$$H_{app} + H_c \approx \begin{cases} H_{app} & \text{for } H_{app} \gg H_c \\ H_c & \text{for } H_c \gg H_{app}. \end{cases} \tag{3}$$

According to equation (3), when the applied magnetic field is much greater than the coercive field, χ_{FC} and χ'_{FC} will be identical and there will not be any difference between χ_{FC} and χ_{ZFC}, as shown in figure 5 for $Ni_{0.8}Zn_{0.2}Fe_2O_4$ (at $H_{app} = 5000$ Oe compared to H_c at 300 K $\approx$ 20 Oe, no irreversibility, which would be indicated by the difference between χ_{FC} and χ_{ZFC}, is observed down to 300 K). Similarly, at very low applied magnetic fields, $\chi_{ZFC} \propto H_c^{-1}(T)$ for highly anisotropic materials ($H_c \gg H_{app}$), as observed for the ferromagnetic compound $SrRuO_3$ [11].

Though the shapes of the χ'_{FC}- and χ_{ZFC}-curves are almost identical for all four compounds at each applied field, there is a small difference in magnitude, and the difference is greater for the highly anisotropic compounds. One possible reason for this difference is that in the ZFC experiments the sample is not cooled under ideal zero-field conditions. It was observed that a small positive or negative field was always present when the instrument

was set for zero magnetic field. Neither the magnitude nor the sign of this stray field could be controlled precisely with the vibrating-sample magnetometer used in the present study. When a small net magnetic field is present while cooling the sample, the resulting ZFC magnetization value will be different if the sample is highly anisotropic.

The present results show that the ZFC and the FC susceptibilities (magnetizations) of the four magnetically ordered systems are related through the coercivity. Given the values of $M_{FC}(T)$ and $H_c(T)$, $M_{ZFC}(T)$ can be calculated (or vice versa). It is expected that the present results can be extended to spin glasses and other systems showing spin-glass-like features, namely superparamagnetic (fine-particle) systems, high-T_c oxide superconductors, cluster-glass (mictomagnetic) systems etc. However, for the magnetic systems showing relaxation effects, a time variable may have to be introduced into the expression for comparing the two susceptibilities.

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