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Research Article

Magnetothermopower in $A_{2-x}La_xFeMoO_6$ (A = Sr, Ba)

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A magnetothermopower has been observed in electronically spin-polarized polycrystalline $Sr_{2-x}La_xFeMoO_6$ and Ba_2FeMoO_6. The magnetothermopower is linear up to ~50 K for $Sr_{2-x}La_xFeMoO_6$ and linear up to ~270 K for Ba_2FeMoO_6. We suggest that the magnetothermopower may arise from a spin-tunneling magnetothermopower between the grains.

1. Introduction

The double perovskites are an interesting class of compounds where some of them are predicted to display 100% electronic spin polarization and hence they are called half-metals [1]. Most research has focused on Sr_2FeMoO_6 and Ba_2FeMoO_6 [1–11] where the Curie temperature is above room temperature and hence there are potential device applications that include magnetic field sensing [12, 13]. The appearance of electronic spin polarization in ferromagnetic metals means that spin-dependent tunneling can occur in thin films that are separated by a thin insulating layer [13–15]. Spin-dependent tunneling also occurs in Sr_2FeMoO_6 and Ba_2FeMoO_6 polycrystalline samples [1, 2, 8, 11, 12] where it has been shown that the resultant magnetoresistance can be modeled in terms of tunneling between the grains and through a thin insulating layer on the surface of the grains [8]. It has recently been shown that ferromagnetic/insulating/ferromagnetic tunneling junctions containing ferromagnetic metallic layers with some degree of electronic spin polarization can also display a spin-tunneling magnetothermopower [16, 17]. Since spin-tunneling is also known to occur between the grains in Sr_2FeMoO_6 and Ba_2FeMoO_6, then it might be expected that a spin-tunneling magnetothermopower also occurs in these compounds.

In this paper we report the results from magnetothermopower and magnetoresistance measurements on polycrystalline $Sr_{2-x}La_xFeMoO_6$ and $Ba_{2-x}La_xFeMoO_6$. We show that magnetothermopower is observed and it may possibly be due to a spin-tunneling magnetothermopower between the grains.

2. Materials and Methods

The $Sr_{2-x}La_xFeMoO_6$ and $Ba_{2-x}La_xFeMoO_6$ polycrystalline samples were made using a method described elsewhere [10]. X-ray diffraction (XRD) measurements showed that the samples were phase pure within the limit of detectability. Magnetothermopower, magnetoresistance, and magnetization measurements were made at different magnetic fields and temperatures using a Quantum Design PPMS. The magnetothermopower measurements were made using the TTO option where the applied magnetic field was parallel to the thermal gradient. The magnetoresistance measurements were done using a four-terminal configuration and with the applied magnetic field parallel to the current.

Some high field magnetization measurements were done to estimate the Fe/Mo antisite disorder (ASD). The saturation magnetic moment per formulae unit (f.u.) was 2.66, 1.74, and 1.13 μ_B/f.u. for $Sr_{2-x}La_xFeMoO_6$ with $x = 0$, 0.2, and 0.4, where μ_B is the Bohr magneton. The maximum measured value for no ASD and $x = 0$ is 4 μ_B/f.u. [18]. The resultant ASD is 15%, 25%, and 32% for $x = 0$, 0.2, and 0.4 using the correlation between the saturation magnetic moment per f.u. and the ASD [15, 18]. These estimates of the ASD are close to those obtained from the XRD data using a ratio of the (101) peak divided by the (200) peak and using the correlation between ASD and this XRD ratio [10]. The saturation moment

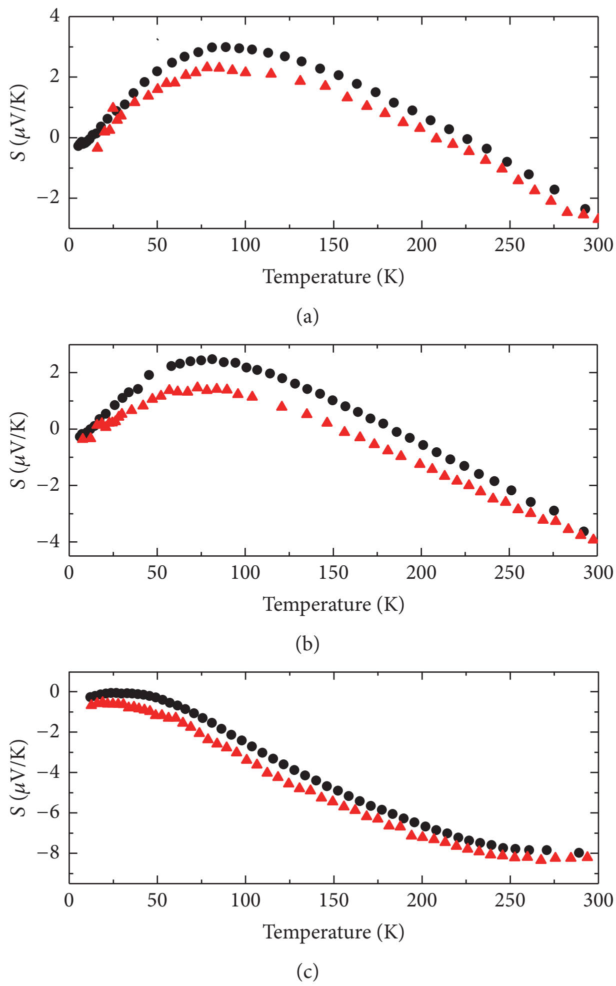


Figure 1: Plot of the thermopower from $Sr_{2-x}La_xFeMoO_6$ with (a) $x = 0.4$, (b) $x = 0.2$, and (c) $x = 0$ at 0 T (filled circles) and 6 T (filled up triangles).

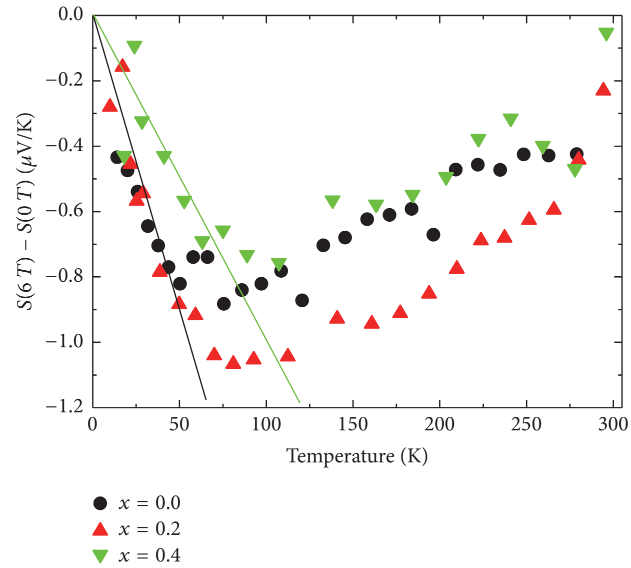


Figure 2: Plot of the $Sr_{2-x}La_xFeMoO_6$ thermopower at 6 T minus the thermopower at 0 T for $x = 0$ (filled circles), $x = 0.2$ (filled up triangles), and $x = 0.4$ (filled down triangles). The lines are guides to the eye.

for Ba_2FeMoO_6 is much higher and it is 3.78 μ_B/f.u., which corresponds to a low ASD of 3%.

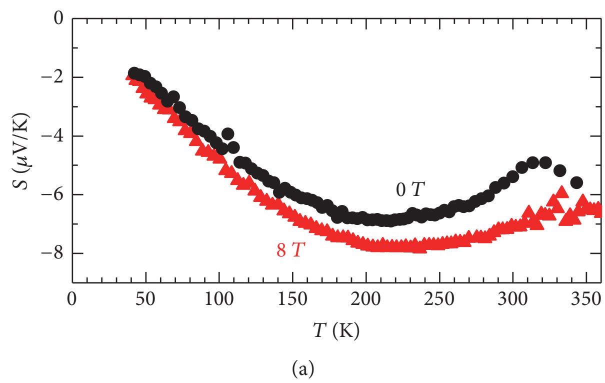
3. Results and Discussion

The $Sr_{2-x}La_xFeMoO_6$ thermopower, S, is plotted in Figure 1 for $x = 0$, 0.2, and 0.4 at 6 T and for no applied magnetic field. The thermopower in the absence of an applied magnetic field systematically increases with increasing x. This has been noted before and it has been suggested by comparison with the superconducting cuprates that it is indicative of the expected electron doping by La [10]. We have previously shown that the temperature dependence of the thermopower can be modeled with a diffusion term and a phonon-drag term where it was assumed that phonon-electron scattering is still significant at 300 K when compared with phonon-phonon scattering [10].

It is apparent in Figure 1 that there is a magnetic field induced decrease in the $Sr_{2-x}La_xFeMoO_6$ thermopower for temperatures up to 300 K, which is still below the Curie temperature of ~425 K [5]. This change can be seen more clearly in Figure 2 where the difference in thermopower, $\Delta S = S(6\,T) - S(0\,T)$, is plotted and it is apparent that ΔS is linear in temperature up to ~50 K. A linear ΔS is also observed in Ba_2FeMoO_6 up to a higher temperature of ~270 K, as can be seen in Figure 3. We note that the Ba_2FeMoO_6 thermopower at 0 T is similar to that reported in another study where there is an anomalous peak in the thermopower near the Curie temperature of 321 K [19].

There are a number of possible mechanisms that can lead to a magnetothermopower below the magnetic ordering temperature. For example, a spin-entropy thermopower can occur in compounds with strong electron-electron interactions [20, 21]. If these interactions are large enough then there will exist a very narrow band and the spin degrees of freedom can result in a spin-entropy thermopower, $S = -\sigma/e$, where σ is the entropy per electron [21]. At high temperatures the spin-entropy thermopower can be described by the Heikes function, $S = k_B \times \ln(g_s \times g_c)$, where g_s is the spin degeneracy, g_c is the configurational degeneracy, and k_B is Boltzmann's constant [21]. This produces a positive contribution to the thermopower. For high applied magnetic fields, g_s will tend towards 1 and hence there will be a reduction in the spin-entropy thermopower. A spin-entropy thermopower is unlikely to occur in $Sr_{2-x}La_xFeMoO_6$ because the conduction band is too broad [22] and the magnetothermopower strongly depends on temperature.

Magnon-drag is another mechanism that can lead to a magnetothermopower [23]. It is analogous to phonon-drag in nonmagnetic metals. It should increase at low temperatures as the magnon density increases and it will decrease at high temperatures due to magnon-magnon and magnon-phonon scattering. It has been argued that the magnon populations



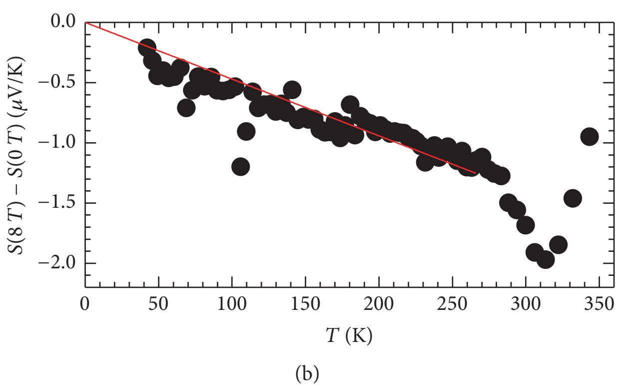


FIGURE 3: (a) Plot of the thermopower from Ba_2FeMoO_6 at 8 T (filled up triangles) and 0 T (filled circles). (b) The resultant thermopower difference $S(8\ T) - S(0\ T)$. Also shown is the best fit line with a zero intercept.

can be low when the applied magnetic field is parallel to the magnetization and high when it is antiparallel to the magnetization. The net result can be a reduction in the magnon-drag thermopower at high magnetic fields and when all magnetic domains are aligned in the direction of the applied magnetic field. At low temperatures the magnon-drag thermopower, S_{MD}, is proportional to $T^{3/2}/2/(nD^{3/2})$, where T is the temperature, n is the carrier concentration, and D is the exchange stiffness constant [23]. This predicts that S_{MD} will be proportional to $T^{3/2}$ at low temperatures and S_{MD} should decrease with increasing carrier concentration. However, it is apparent in Figure 2 that S is linear below ~50 K for $Sr_{2-x}La_xFeMoO_6$ and linear below ~270 K for Ba_2FeMoO_6 rather than being proportional to $T^{3/2}$, and S is still finite at 300 K. This suggests that magnon-drag thermopower is not the origin of the magnetothermopower in $Sr_{2-x}La_xFeMoO_6$ or Ba_2FeMoO_6.

It may be that the magnetothermopower from polycrystalline $Sr_{2-x}La_xFeMoO_6$ and Ba_2FeMoO_6 arises from a spin-dependent Seebeck effect. This type of magnetothermopower has been reported in magnetic tunneling junctions where one or more of the layers are ferromagnetically ordered and display electronic spin polarization [16, 17]. In the case of magnetic tunnel junctions with an insulating barrier between the two spin-polarized metallic layers or if there is no exchange coupling between the layers, the spin-tunneling magnetothermopower, S_T, can be written as [16]

$$S_T = \frac{-e}{k_B T G} \int t(E)(E - E_F)\left(\frac{-\partial f}{\partial E}\right) dE, \quad (1)$$

where e is the electron charge, G is the tunneling conductance, $t(E)$ is the tunneling transmission function, E is the energy, and f is the Fermi function. The tunneling conductance can be written as [16]

$$G = \frac{e^2}{h} \int t(E)\left(\frac{-\partial f}{\partial E}\right) dE, \quad (2)$$

where h is Planck's constant.

If $t(E)$ does not strongly depend on energy within a few $k_B T$ of E_F, where k_B is Boltzmann's constant and E_F is the Fermi energy, then it can be shown using (1) and (2) that S_T reduces to the Mott formulae [17]

$$S_T = -\frac{\pi^2}{3}\frac{k_B^2 T}{e}\frac{1}{G}\left.\frac{\partial G(E)}{\partial E}\right|_{E_F}, \quad (3)$$

where $G(E)$ is the energy-dependent tunneling conductance. The change in the magnetothermopower is then [17]

$$\Delta S_T = S_P - S_{AP} = -\frac{\pi^2}{3}\frac{k_B^2 T}{e}\left[\frac{1}{G_P}\left.\frac{\partial G_P(E)}{\partial E}\right|_{E_F} - \frac{1}{G_{AP}}\left.\frac{\partial G_{AP}(E)}{\partial E}\right|_{E_F}\right], \quad (4)$$

where S_P and G_P are the spin-tunneling thermopowers for the magnetization parallel to both layers and S_{AP} and G_{AP} are the spin-tunneling thermopowers when the magnetization from both layers is antiparallel. Equation (4) predicts that ΔS_T will be linearly dependent on T.

If a spin-tunneling magnetothermopower occurs in $Sr_{2-x}La_xFeMoO_6$ and Ba_2FeMoO_6 at the grain boundaries then the thermopower can be written as $S(B) = S_T(B) + S_{Bulk}$, where S_{Bulk} is the thermopower away from the grain boundaries that is assumed be independent of the applied magnetic field. Thus, $\Delta S(B) = \Delta S_T(B)$. Since $\Delta S(B)$ is linear in temperature up to ~50 K for $Sr_{2-x}La_xFeMoO_6$ and up to ~270 K in Ba_2FeMoO_6 as predicted from (4), then our data suggests that the magnetic field dependence of ΔS is due to a spin-tunneling magnetothermopower.

The slope of ΔS below ~50 K for Sr_2FeMoO_6 is nearly the same for $x = 0$ and $x = 0.2$ and it is smaller for $x = 0.4$. It is possible that this is due to a change in E_F for $x = 0.4$. The effect of changes in E_F on ΔS can be illustrated using a simple model where $t_p(E)$ can be written as $t_P(E) = (N_\uparrow N_\uparrow + N_\downarrow N_\downarrow)t$ and $t_{AP}(E) = 2N_\uparrow N_\downarrow t$, where $N_\uparrow$ is the majority carrier DOS, $N_\downarrow$ is the minority carrier DOS, and $t_P(E)$ and $t_{AP}(E)$ are the energy independent tunneling transmission functions for the parallel and antiparallel configurations, respectively [24]. It can then be shown from (2) and (3) that

$$S_P = -\frac{\pi^2}{3}\frac{k_B^2 T}{e}\frac{1}{N_\uparrow N_\uparrow + N_\downarrow N_\downarrow}\left[2N_\uparrow \left.\frac{\partial N_\uparrow(E)}{\partial E}\right|_{E_F} + 2N_\downarrow \left.\frac{\partial N_\downarrow(E)}{\partial E}\right|_{E_F}\right],$$
$$S_{AP} = -\frac{\pi^2}{3}\frac{k_B^2 T}{e}\frac{1}{N_\uparrow N_\downarrow}\left[N_\uparrow \left.\frac{\partial N_\downarrow(E)}{\partial E}\right|_{E_F} + N_\downarrow \left.\frac{\partial N_\uparrow(E)}{\partial E}\right|_{E_F}\right]. \quad (5)$$

If we take a simple DOS with $N_\uparrow = a_1(E)^{0.5}$ and $N_\downarrow = a_2(E - E_0)^{0.5}$ then from (5)

$$\Delta S = S_P - S_{AP} = -\frac{\pi^2}{6}\frac{k_B^2 T}{eE_F}\left[\frac{2}{1-zx} - \frac{2-x}{1-x}\right], \quad (6)$$

where $x = E_0/E_F$, $x < 1$, and $z = a_2^2/(a_1^2 + a_2^2)$. This is negative provided that $z > 1/(2-x)$. This simple example shows how a negative spin-tunneling magnetothermopower can occur purely from majority and minority DOS effects. ΔS will decrease if E_F increases or if the spin polarization decreases. The smaller ΔS gradient for $Sr_{2-x}La_xFeMoO_6$ with $x = 0.4$ may suggest that E_F is larger for $x = 0.4$, which would appear to be consistent with electron doping by La.

The departure from linearity for ΔS above ~50 K for $Sr_{2-x}La_xFeMoO_6$ can occur if $t(E)$ is energy-dependent. The envelope functions $(E-E_F)(-\partial f/\partial E)$ and $(-\partial f/\partial E)$ in (1) and (2) probe $t(E)$ with $\sim \pm k_B T$ of E_F. Thus, if $t(E)$ displays large energy dependence for $|E - E_F| >\sim 6$ meV then ΔS will be approximately linear only for low temperatures, which is what is observed. The departure from linearity for Ba_2FeMoO_6 only occurs above ~270 K and close to the Curie temperature. This suggests that the lower temperature departure seen in $Sr_{2-x}La_xFeMoO_6$ may be due to the ASD. It is also possible that the departure in ΔS from linearity in $Sr_{2-x}La_xFeMoO_6$ above ~50 K is due to inelastic scattering that has not been included in the simple model above.

The discussion above concerns the relatively well defined conditions where there is no applied magnetic field or when the magnetic field is high enough so that the magnetization directions are the same in the regions close to the grain boundaries. For intermediate magnetic fields, it is reasonable to assume that $S(B)$ will vary smoothly between S_{AP} and S_P as the magnetic field is increased. This is indeed observed as can be seen in Figure 4 where $\Delta S(B)$ is plotted as a function of the applied magnetic field for Sr_2FeMoO_6 at 77 K.

If there is a spin-tunneling magnetothermopower then there should also be a spin-tunneling magnetoresistance. We show in Figure 4 that a magnetoresistance is observed where the magnetoresistance from Sr_2FeMoO_6 at 77 K is plotted as a function of the applied magnetic field. Here we define the magnetoresistance as $\mathrm{MR} = [R(B) - R(0)]/R(0)$, where $R(B)$ is the resistance for an applied magnetic field and $R(0)$ is the

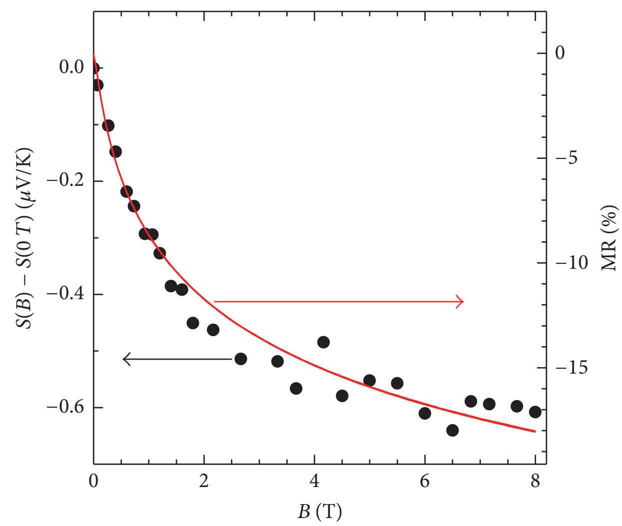


Figure 4: Plot of the thermopower difference, $S(B) - S(0\ \mathrm{T})$ (left axis, circles), and the magnetoresistance (right axis, solid curve) from Sr_2FeMoO_6 against the applied magnetic field at 77 K.

resistance with no applied magnetic field. The magnetic field dependence and the magnitude of the magnetoresistance are similar to those found in other studies on polycrystalline Sr_2FeMoO_6 and Ba_2FeMoO_6 where it has been attributed to spin tunneling between the grains [2, 8, 11, 12]. We find that the magnetoresistance and $\Delta S(B)$ have similar magnetic field dependence.

4. Conclusions

In conclusion, we observed a magnetothermopower in polycrystalline $Sr_{2-x}La_xFeMoO_6$ and $BaFeMoO_6$ that is linear up to ~50 K in $Sr_{2-x}La_xFeMoO_6$ and ~270 K in Ba_2FeMoO_6. The magnetothermopower may be due to a spin-tunneling thermopower where the magnetothermopower can be linear when the tunneling transmission function is only weakly dependent on energy within a few $k_B T$ of E_F. In this scenario, the magnetothermopower may arise from spin tunneling across the grain boundary. The departure from linearity above ~50 K in $Sr_{2-x}La_xFeMoO_6$ may be due to a tunneling transmission function that has significant energy dependence only for $(E - E_F) >\sim 6$ meV. It is also possible that there is significant inelastic scattering at higher temperatures.

Competing Interests

The authors declare that there is no conflict of interests regarding the publication of this paper.

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