Large magnetoresistance, structure and magnetic properties of the double perovskite LaKFe$_{1.2}$Mo$_{0.8}$O$_6$ compound

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Abstract

The double perovskite LaKFe$_{1.2}$Mo$_{0.8}$O$_6$ ceramics have been successfully synthesized. The structure, magnetic and transport properties of this compound have been studied. Its structure belongs to cubic system with space group $Fm\bar{3}m$. Magnetization slowly reduced with temperature and large magnetoresistance of the sample under 1.0 T magnetic field remains constant around 0.95 in the studied temperature range. Magnetic field dependence of magnetoresistance is described and magnetoresistance increases abruptly under 0.65 T. The electrical transport properties can be described by the thermally activated hopping mechanism.

1. Introduction

Recently the double perovskite compound, Sr$_2$FeMoO$_6$, has attracted a considerable scientific interest in view of its remarkable room-temperature low-field magnetoresistance properties [1–3]. The double perovskite Sr$_2$FeMoO$_6$ has an ordered structure with alternating localized up spin Fe^{3+} (3d^5; t$_{2g}^3$e$_g^2$) and itinerant down spin Mo^{5+} (4d^1; t$_{2g}^1$e$_g^0$). The parallel magnetic moments of Fe^{3+}, antiferromagnetically coupled with the spins of Mo^{5+} induce an ideal saturation magnetization (M_S) of $4\mu_B$ per formula unit (f.u.). Recently, it has been reported that the substitution of La^{3+} for Sr^{2+} in Sr$_2$FeMoO$_6$ promotes a rising of the T_C and a decreasing of M_S as doping level increases [4]. Recently, Kim et al. [5] reported that replacement of Ba^{2+} by K$^+$ in Ba$_2$FeMoO$_6$ reduces the T_C as K$^+$ content increases. With the La^{3+} or K$^+$ doping in double perovskite A$_2$FeMoO$_6$ compounds (where A is the alkaline-earth metals) carriers doping are inevitable in these materials. In order to figure out some effects of without changing ionic valence states of B and B$'$ sites by A sites doping we study the structure, magnetic properties and magnetoresistance of Sr$_2$Fe$_{1.2}$Mo$_{0.8}$O$_6$ compound substituted all Sr^{2+} by La^{3+} and K$^+$.

2. Experimental

The polycrystalline LaKFe$_{1.2}$Mo$_{0.8}$O$_6$ sample has been synthesized using standard solid state reaction technique. As the K$^+$ is easy to volatilize during calcined process at high temperature, therefore, more than nominal K content was added in order to compensate loss during sintered process at high temperature. The raw materials La$_2$O$_3$, K$_2$CO$_3$, Fe$_2$O$_3$ and (NH$_4$)$_6$Mo$_7$O$_{24}$·4H$_2$O of high purity (more than 99.99%) were mixed by hand in an agate mortar for at least 40 min. Then it was pressed into pellets under 10 MPa pressure for 1 min following preheating in air at 600 and 900 °C for 5 h, respectively. The calcined mixtures were pulverized and pressed into pellets. The pellets were sintered at 920 °C for 16 h in a stream of 5% H$_2$/Ar with intermediate grindings. Phase analysis and characterization were carried out by X-ray diffraction using Cu Kα radiation with a graphite monochromator on Rigaku model D/max-2400 X-ray diffractometer.

Temperature dependence of magnetization curve was measured by a vibrating-sample magnetometer (VSM) in a field of 0.5 T over the temperature range 80–300 K. Isothermal magnetization curve was performed on VSM apparatus. Transport properties were determined by the standard four-probe DC method in the range 80–300 K.

3. Results and discussion

Powder XRD pattern of LaKFe$_{1.2}$Mo$_{0.8}$O$_6$ sample is shown in Fig. 1 which shows that the specimen is single phase. No impurity was detected based on powder XRD pattern. The diffraction peaks could be indexed in the cubic system with space group $Fm\bar{3}m$. The lattice parameter is found to be: $a = 7.870$ (6) Å. Our sample structure is similar to that of Sr$_2$Fe$_{1.2}$Mo$_{0.8}$O$_6$

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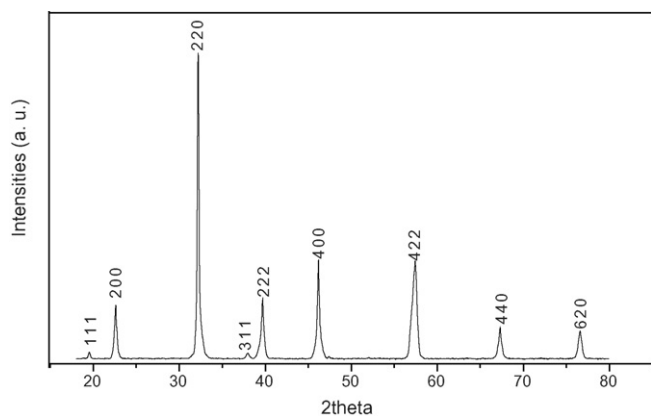
Fig. 1. X-ray diffraction pattern of $LaKFe_{1.2}Mo_{0.8}O_6$ sample.

specimen synthesized by Liu et al. [6] and differs with that of $LaKMnMoO_6$ sample prepared by Megdiche et al. [7], which the structure belongs to orthorhombic system with space group P222. The diffraction lines of (1 1 1) and (3 1 1) display that Fe and Mo ions partly order to occupy B and B′ sites, respectively.

Magnetization measurement made as a function of temperature $M(T)$ on warming the $LaKFe_{1.2}Mo_{0.8}O_6$ sample from 80 to 300 K in a magnetic field of 0.5 T is plotted in Fig. 2. It can be seen from Fig. 2 that the magnetization reduces slowly with temperature warming from 80 to 300 K and no transition temperature is observed in this temperature range. This indicates that the Curie temperature of $LaKFe_{1.2}Mo_{0.8}O_6$ sample is higher than 300 K. We calculated that reduction rate with temperatures of magnetization of the sample is 0.009 emu/g K from 80 to 300 K.

The isothermal magnetization $M(T, H)$ at 77 K with magnetic fields up to 1.5 T, after cooling in zero magnetic fields, was measured. The magnetic moment is saturated in field higher than 0.7 T, and $M(77 K, H)$ changes linearly with reciprocal of the magnetic field. A linear extrapolation at $1/H = 0$ allows us to obtain the saturation magnetization, $M(77 K)$ and, thus, the net magnetic moment per unit formula in the direction of the magnetization, $1.05\mu_B$ (77 K). We also measured the isothermal magnetization at 300 K with magnetic fields up to 1.5 T
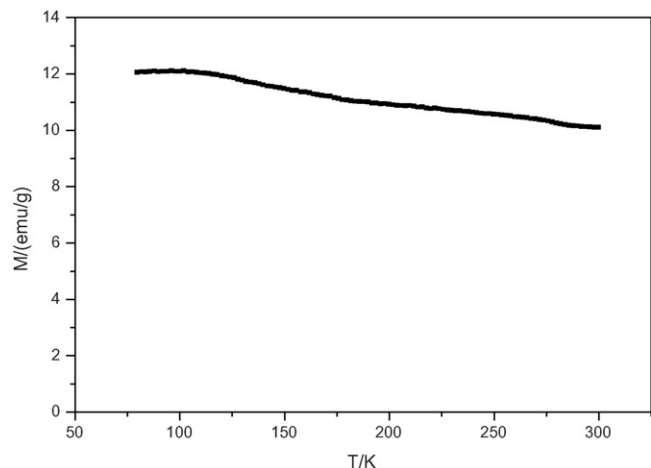

Fig. 2. Temperature dependent magnetization of $LaKFe_{1.2}Mo_{0.8}O_6$ double perovskite registered on warming in 0.5 T magnetic field.
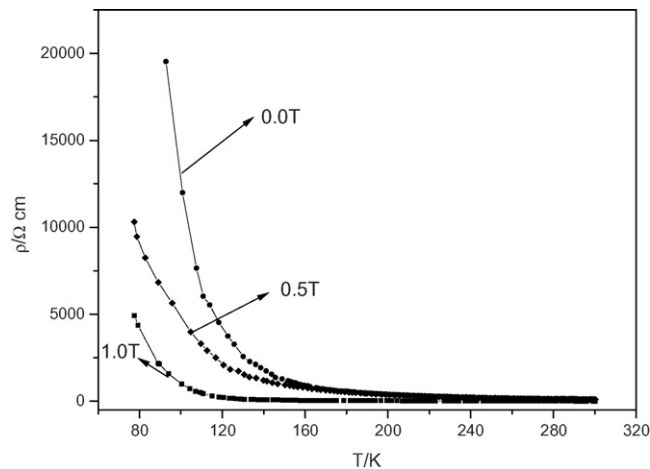

Fig. 3. Temperature dependent resistivity registered upon warming in zero, 0.5 and 1.0 T magnetic fields.

and the saturation magnetization obtained is $0.95\mu_B$ (300 K). From the saturation magnetization at 77 and 300 K it is clear to see that the saturation magnetization is influenced very weak by temperature agreed with result obtained by thermal magnetization. Taking saturation magnetization at 77 K as that at 0 K we calculated the mixing of Fe and Mo species on the B and B′ sites based on literature reported model [8]. In $LaKFe_{1.2}Mo_{0.8}O_6$ not all Fe ions and/or Mo ions retain the valence state of +3 and +5, respectively, and some Fe ions and/or Mo ions change their valence states to +4 and/or to +6.

The $\rho(T)$ curves registered upon warming in zero fields, 0.5 and 1.0 T magnetic fields are shown in Fig. 3. As the sample resistance in zero fields is too high to measure on our apparatus at low temperatures we cannot obtain the value of resistance below 92 K. It can be seen from Fig. 3 that $LaKFe_{1.2}Mo_{0.8}O_6$ sample exhibits semiconducting behavior under zero, 0.5 and 1.0 T magnetic fields over the temperature range, from 80 to 300 K, and shows a significant magnetoresistance effects in 0.5 and 1.0 T applied magnetic fields, respectively. We define $MR = [\rho(H) - \rho(0)]/\rho(0)$, where $\rho(H)$ and $\rho(0)$ are the resistivity in a magnetic field and without a magnetic field, respectively. Fig. 4 shows the temperature dependence of the magnetoresistance (MR) in 0.5 and 1.0 T magnetic fields. The −MR of this sample under 0.5 T field decreases monotonous from 0.69 at 92 K to 0.04 at 181 K and the −MR almost unchanges with temperature rising from 181 to 300 K. However, the −MR of the sample under 1.0 T field is almost constant, 0.91–0.95, in the temperature range investigated. In order to obtain influence of applied magnetic fields for the −MR of the sample we measured the resistivity of the sample under different magnetic fields at 300 K. Fig. 5 presents the magnetic field dependence of the −MR of the sample. It can be seen that the −MR increases with applied magnetic fields and that the −MR increases abruptly from 0.04 to 0.60 with the magnetic field from 0.5 to 0.65 T. Following the −MR are 0.80 and 0.95 under 0.80 and 1.0 T, respectively.

In order to understand the origin of the semiconducting behavior of sample, a detailing analysis of $\rho(T)$ was performed. The resistivity behavior without magnetic fields can be described by the thermally activated hopping of the charge carriers across
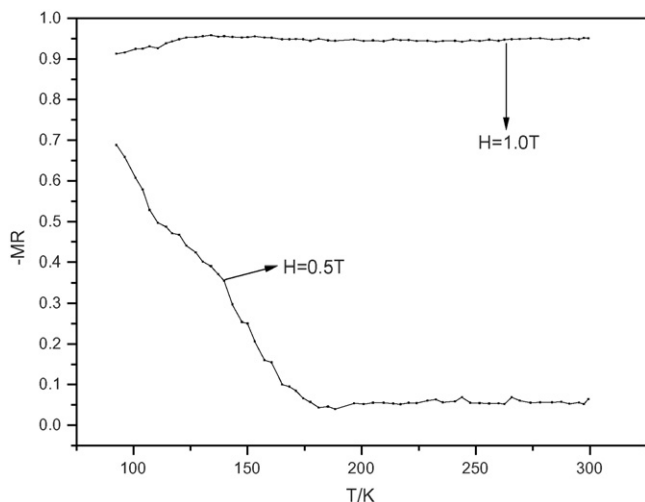
Fig. 4. Temperature dependence of magnetoresistance in 0.5 and 1.0 T magnetic fields.
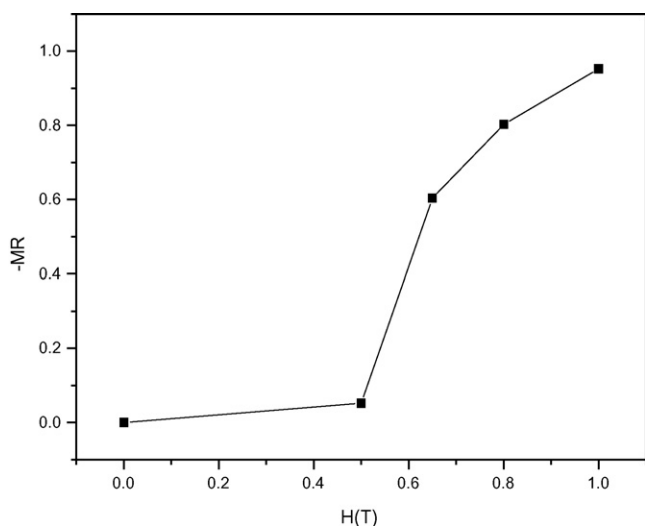

Fig. 6. Dependence of resistivity (ρ) on the temperature (T) in a log ρ against $1/T$.

The large magnetoresistance of the sample in 1.0 T field stays near constant, 0.91–0.95, with temperature from 92 to 300 K. The $-MR$ increases with magnetic fields at 300 K and abruptly increases largely under 0.65 T. In the studied temperature range, the resistivity without magnetic field appears to be best described by thermally activated hopping mechanism.

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Fig. 5. Magnetic field dependence of magnetoresistance at 300 K.

the band gap, which is the linear dependence of log ρ on $1/T$. Fig. 6 shows the dependence of resistivity (ρ) on the temperature (T) in a log ρ against $1/T$ for temperature range.

In summary, we have successfully synthesized double perovskite $LaKFe_{1.2}Mo_{0.8}O_6$ specimen which structure belongs to cubic system with space group $Fm\bar{3}m$. Magnetization of the specimen slowly decreases with temperature. Magnetoresistance of the sample in 0.5 T field decreases monotonous for low-temperature and keeps a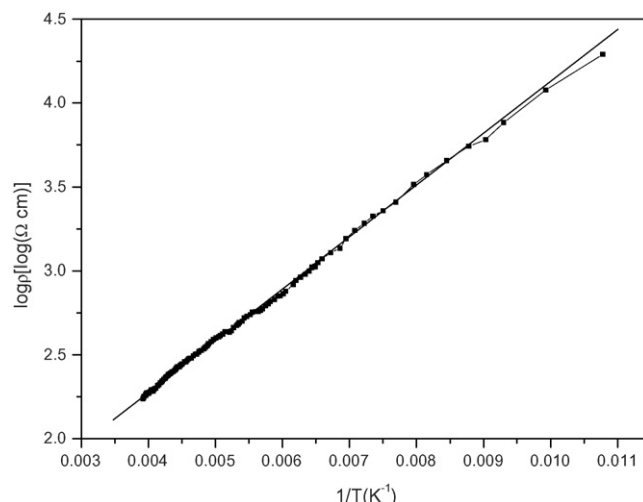 constant for high temperature.