CHIN.PHYS.LETT. Vol. 18, No. 1 (2001) 114

A New Method to Measure the Drift Mobility in Doped Manganite Films *

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(Received 3 March 2000)

The travelling wave method has been used to measure the drift mobility in the paramagnetic–insulator state of $La_{0.75}Sr_{0.11}Ca_{0.14}MnO_3$ films. The thermal activation of the drift mobility in manganites has been measured for the first time. The mobility varies from 2.5×10^{-2} cm^2/Vs at 310 K to about 9.2×10^{-2} cm^2/Vs at 400 K. The Arrhenius behaviours of the conductivity and drift mobility indicate that the transport process in manganites above the Curie temperature is dominated by the thermally assisted hopping of small polarons.

PACS: 75. 30. Un, 62. 80. +f, 71. 38. +i, 73. 50. Dn

Lanthanum-based transition-metal oxide perovskites have received much attention from theoretical and experimental researchers,[1−3] since these materials exhibit a large negative magnetoresistance, called the colossal magnetoresistance (CMR) effect, near the point of ferromagnetic–metal to paramagnetic–insulator phase transition. Due to this characteristic, it can be applied in magnetic record and switching devices.[4] And these materials in paramagnetic–insulator phase are suitable for acting as the semiconductor layer in ferroelectric field effect transistors[5] (FET) since they are closely lattice-matched to the common perovskite-type ferroelectrics. Although CMR was discovered a few decades ago, its mechanism is still not clear. At first, the mechanism of CMR was ascribed to the double exchange model.[6] But now, many theoretical[7,8] and experimental papers[9−13] have shown that magnetic interactions alone are insufficient to explain CMR and a coupling between charge and lattice is significant. Under this situation, it is necessary and important to clearly understand the transport mechanism of the materials in ferromagnetic and paramagnetic states. Recently, Hall measurements[12,14,15] have been used to investigate the transport properties in Lanthanum-based manganites. Since the anomalous part of the Hall coefficient is not well understood, it is impossible to get more information on transport mechanism in these materials, such as the drift mobility or the carrier concentration, using Hall measurements.[14] However, information on the drift mobility and its temperature dependence is indispensable for understanding the mechanism clearly.

In order to get insight into the transport process and to overcome the difficulties in Hall measurements, we have adopted the travelling wave (TW) technique to investigate the transport mechanism in manganite films. Since it was introduced by Adler *et al.*,[16] the TW method has been used to investigate the transport in amorphous semiconductors for several years.[17−19] One of the characteristics of this method is the possibility to obtain information on the transport mechanism in materials that have high resistance and low mobility.

A top-view schematic diagram of the travelling wave experiment is shown in Fig. 1. The sample is placed above the surface of an $LiNbO_3$ single crystal plate with a gap of 12 μm. The transducers on $LiNbO_3$ generate a surface acoustic wave (SAW) that propagates along the surface of the $LiNbO_3$ plate in the z direction. Due to the piezoelectric properties of $LiNbO_3$, an electric field associated with the SAW couples into the sample to bunch the carriers into charge packets and drifts these carriers through the material. Consequently, an acoustic–electronic direct current is produced and can be measured from two electrodes. Fritzsche and Chen[17−18] and Jahanson[19] have derived the relationship between the drift mobility and the acoustic–electric current or the acoustic–electric voltage as the following:[19]

$$\mu = 2I_{\rm ae}\{w\Phi_0 \sinh(kd)(k\sigma/v_{\rm s})[(|A|^2+|B|^2)\cosh(kd) - 2AB\sinh(kd)]\}^{-1} \quad (1)$$

$$\sigma = \frac{I_{\rm ae}L}{V_{\rm ae}dw}, \quad (2)$$

where $I_{\rm ae}$ is the acoustic–electronic short current, $V_{\rm ae}$ the acoustic–electronic open voltage, k the wave vector of the SAW, $v_{\rm s}$ the velocity of the SAW, d the thickness of the sample, L the distance between the electrodes, w the width in the x direction of the sample, and Φ_0 is the potential at the surface of the piezoelectric plate and is given by the Rayleigh wave power.[20] It increases with the square of the power $P_{\rm SAW}$ carried by the SAW, and depends on the properties of the piezoelectric plate and propagation axis. The proportionality constant depends on the properties of the piezoelectric plate and propagation axis of the SAW.

*Supported by the Climbing Program-National Key Project for Fundamental Research.
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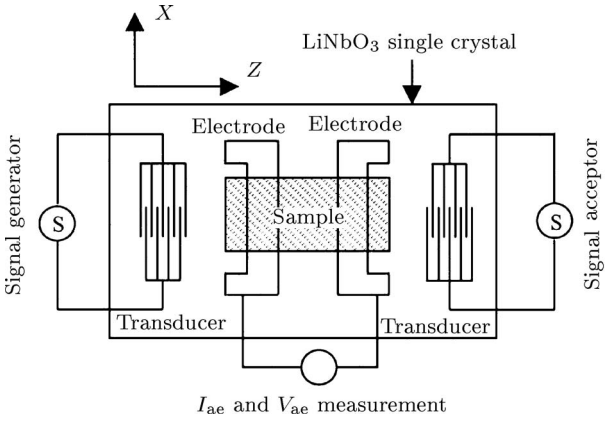


Fig. 1. A top-view schematic diagram of the travelling wave experiment.

The coefficients A and B are determined by the boundary conditions imposed on the electric field. They are generally complex, which means that the charge waves and the field have nontrivial phase relations. And they depend on the conductivity and the dielectric constants of both the sample and the substrate. They can be described as

$$A = [\cosh(kh) + \varepsilon^*(\frac{B}{A})\sinh(kh)]^{-1}, \tag{3}$$

$$\frac{B}{A} = \frac{\varepsilon_s + \varepsilon^* kd}{\varepsilon^* + \varepsilon_s kd}, \tag{4}$$

$$\varepsilon^* = \varepsilon - \mathrm{i}4\pi\sigma/\omega, \tag{5}$$

where h represents the gap between the sample and the piezoelectric plate, ε_s the dielectric constant of the substrate, ε the dielectric constant of the sample, w the frequency of the SAW.

The TW method can be used to investigate the transport process in materials with high resistivity and low mobility, for which the Hall measurement meets some difficulties. The transport process in doped La-based manganites in the paramagnetic state is semiconductor-like, and these materials usually have high resistivity and low mobility. In this case, the TW method is suitable and advantageous for investigating these materials. Therefore we can obtain conductivity and drift mobility in these samples with I_{ae} and V_{ae} from Eqs. (1) and (2), as all the other quantities are either known or measurable. Another advantage of this method is that the sign of I_{ae} or V_{ae} depends on the type of the majority carriers and the propagation direction of the SAW. Since the direction of the SAW is controlled by the experiments, the type of majority carriers can be directly determined from the sign of I_{ae} or V_{ae}.

In this letter, we focus on the application of the TW method to measure the transport properties of $La_{0.75}Sr_{0.11}Ca_{0.14}MnO_3$ thin films in the paramagnetic–insulator phase. The samples for this study were fabricated by the pulsed laser deposition method using an $La_{0.75}Sr_{0.11}Ca_{0.14}MnO_3$ ceramic target. A 248 nm KrF excimer laser source, with a 5 Hz repetition rate and 30 ns pulse width, was used. The average laser energy density was 2.5 J/cm^2. The target and the substrate holder were rotated during the deposition. The films were deposited on fused silica substrates, with a thickness of 500 nm, at 800°C in 30 Pa of oxygen. Immediately after the deposition, the films were *in situ* annealed at 850°C in pure oxygen of 0.5 atm for one hour. A more detailed description on sample preparation procedure has been published in our previous work.[21] The chemical composition of the films was analysed by an inductively coupled plasma (ICP) quantemeter. The result of the ICP measurement shows that the films are stoichiometric. The samples are single-phase, as checked by x-ray diffraction. The Curie temperature T_c of the target used in this letter is 280 K. To meet the need of the TW measurements, co-planar electrodes with a distance of 1 mm were deposited by evaporation. The measurements were carried out in vacuum.

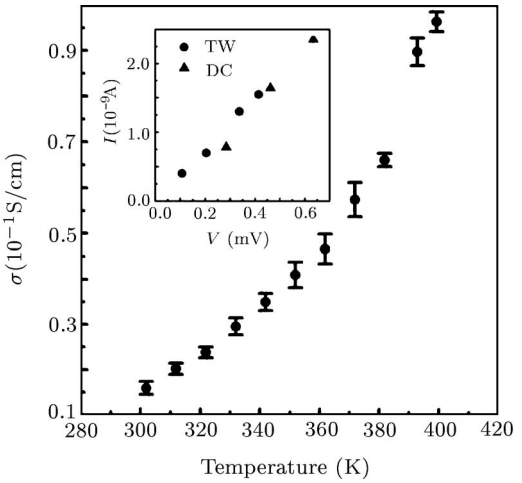


Fig. 2. Conductivity versus temperature. Inset: I–V characteristics measured by TW method (solid circle) and normal direct current and voltage method (solid triangle) at room temperature.

I_{ae} and V_{ae} measured at room temperature are given in the inset of Fig. 2. It is obvious that a good liner correlation between I_{ae} and V_{ae}, which follows the Ohm law. The conductivity is calculated to be 1.6×10^{-2} S/cm. For comparison, the results of normal I–V measurements are also shown in the inset of Fig. 2, from which the conductivity is derived to be 1.8×10^{-2} S/cm. The close agreement between the conductivity from the TW method and that from the I–V measurements indicates that the conductivity measured by the TW method is the bulk properties of the samples. Therefore, the TW method is valid to investigate the transport properties of this kind of material. The conductivity of the films was measured by the TW method over the temperature range from 300 to 400 K, as shown in Fig. 2. The conductivity varies from 1.6×10^{-2} S/cm at 300 K to 9.6×10^{-2} S/cm

at 400 K, which shows that the conduction property of the samples above T_c is universally semiconductor-like. This transport behaviour is consistent with previous reports.[11,12]

In addition, the drift mobility of this kind of material has been measured by the TW method for the first time. The drift mobility at 300 K is about $2.2 \times 10^{-2}\,\mathrm{cm^2/Vs}$, which approaches the value of mobility, $0.03\,\mathrm{cm^2/Vs}$, given in Refs. 12 and 22. The temperature dependence of the drift mobility is shown in Fig. 3. The drift mobility increases while the temperature increases and extends to $9.2 \times 10^{-2}\,\mathrm{cm^2/Vs}$ at 410 K. And the sign of I_{ae} in our work shows that the behaviour of the majority carriers in the films is electron-like.

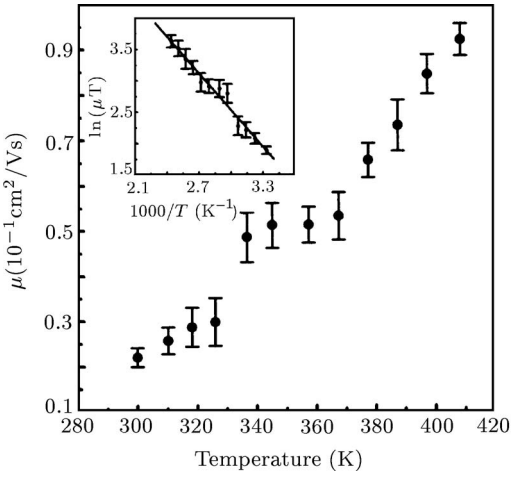


Fig. 3. Drift mobility versus temperature. Inset: the drift mobility plotted in the adiabatic limit. The fine line is a linear fit indicating an activation energy of 171 meV.

As shown in Figs. 2 and 3, the Arrhenius behaviour of the conductivity and drift mobility in our samples is clearly observed. We plot $\ln(\mu\mathrm{T})$ versus $1000/T$ in the inset of Fig. 3, and it exhibits a good liner correlation, which is a distinctive characteristic of the small polaron conduction.[23] From the $\ln(\mu\mathrm{T})$ versus $1000/T$ curve, the thermal activation energy of the mobility is calculated to be 171 meV and is much larger than the activation energy of the measured Hall coefficient (91 meV).[12] Small polaron conduction theory[12] predicts that the activation energy of the drift mobility is always more than that for the Hall mobility. And the value ($2.2 \times 10^{-2}\,\mathrm{cm^2/Vs}$) obtained for the drift mobility in our samples is much smaller than $1\,\mathrm{cm^2/Vs}$, which is also a distinct characteristic of small polaron conduction. Based on the above analyses, we consider that the transport behaviour in the films above T_c is dominated by the hopping process of small polarons.

In summary, we have successfully employed the travelling wave method to measure the drift mobility in manganite films. The drift mobility in the $La_{0.75}Sr_{0.11}Ca_{0.14}MnO_3$ films varies from $2.5 \times 10^{-2}\,\mathrm{cm^2/Vs}$ at 310 K to about $9.2 \times 10^{-2}\,\mathrm{cm^2/Vs}$ at 400 K. The thermal activation phenomenon of the drift mobility reveals that the transport process in doped manganites is dominated by the thermally assisted hopping of small polarons. Our results show that the travelling wave method has more advantages compared to the Hall measurement for which more information on the transport process can be obtained by this method. This method also can be used to investigate the transport process of the interface carriers in the perovskite heterostructure and all-perovskite FET and will attract much attention in the future.

One of the authors (L. Wang) is grateful for the financial support of the Motorola fellowship.

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