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Anomalous phonon transport enhancement at first-order ferromagnetic transition in $(Gd,Sm,Nd)_{0.55}Sr_{0.45}MnO_3$

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Abstract

The thermal conductivity $\kappa(T)$, thermal dilatation $dL(T)/L$ and the electrical resistivity $\rho(T)$ were measured for $(Gd_{1-Y}Sm_Y)_{0.55}Sr_{0.45}MnO_3$ (GSSMO) and $(Nd_{1-Z}Sm_Z)_{0.55}Sr_{0.45}MnO_3$ (NSSMO) systems. For $0.50 \leqslant Y \leqslant 1.0$ of GSSMO and $0.75 \leqslant Z \leqslant 1.0$ of NSSMO, the ferromagnetic (FM) transition is of the first order, while it is of the second order for $0 \leqslant Z \leqslant 0.50$ of NSSMO. In the samples which exhibit the first-order FM transition, $\kappa(T)$ is enhanced and $dL(T)/L$ contracts abruptly just below the FM transition temperature T_c. These behaviors can be interpreted as originating from the reduction of the local lattice distortion below T_c.

Keywords: Thermal conductivity; Ferromagnetic transition; Manganese oxides; First-order transition

Carrier-doped perovskite manganese oxides, $(RE_{1-X}AE_X)MnO_3$ (RE: rare-earth ion, AE: alkaline earth ion), undergo a variety of dramatic phenomena such as the colossal magnetoresistance (CMR), the insulator–metal (I–M) transition and the charge/orbital (CO/OO) ordering. In addition to the double-exchange mechanism, the CMR effect is believed to involve the strong electron–phonon interaction, which may originate from the Jahn–Teller (J–T) effect due to Mn^{3+} ions. At around $X \sim 0.50$, the ferromagnetic (FM) metallic state (FM–M) becomes unstable against the charge-ordered antiferromagnetic (AFM) state in these systems. In $Sm_{1-X}Sr_XMnO_3$, for example, the first-order FM transition takes place at around $X = 0.45$ [1], where the FM state competes with the CO/OO state. This first-order FM transition has been theoretically predicted as being caused by a fluctuation-induced mechanism [2], and is expected to be closely related with the average ionic radius of (RE,AE) site through its effect on the one electron bandwidth W. The effect of the difference in the ionic radius can be systematically interpreted in terms of the tolerance factor f [1], which takes the value of 1 for the cubic perovskite structure without distortion. In this paper, we investigate the thermal conductivity $\kappa(T)$ and thermal dilatation $dL(T)/L$ of $(Gd_{1-Y}Sm_Y)_{0.55}Sr_{0.45}MnO_3$ (GSSMO) and $(Nd_{1-Z}Sm_Z)_{0.55}Sr_{0.45}MnO_3$ (NSSMO) systems mainly at a fixed nominal hole concentration of $X = 0.45$ and discuss the relation between the first-order FM transition and the average ionic radius of the (RE,Sr) site (or the tolerance factor f).

$(Gd_{1-Y}Sm_Y)_{0.55}Sr_{0.45}MnO_3$ $(0 \leqslant Y \leqslant 1.0)$ and $(Nd_{1-Z}Sm_Z)_{0.55}Sr_{0.45}MnO_3$ $(0 \leqslant Z \leqslant 1.0)$ samples were prepared by a conventional solid-state reaction method. $(Nd_{1-Z}Sm_Z)_{1-X}Sr_XMnO_3$ ($Z = 0$ and 1.0; $X = 0.30-0.50$) samples were also fabricated. The mixtures of raw powders were calcined

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twice at 1000°C for 24 h in air, pressed into pellets and sintered at 1500°C for 8 h in air. $\kappa(T)$ was automatically measured by a steady-state heat-flow method [3] and $dL(T)/L$ was monitored by the change in the resistivity of the strain gauge. The electrical resistivity $\rho(T)$ was measured by a standard four-terminal method. The average ionic radius of (RE,Sr) site, r_A, and the tolerance factor $f = (r_A + r_O)/\sqrt{2}(r_B + r_O)$ (r_B: average Mn ionic radius and r_O: O ionic radius), was calculated from the tabulated radii of (RE,Sr) site cations with 12-fold coordination ($Gd^{3+} = 1.21$ Å, $Sm^{3+} = 1.24$ Å, $Nd^{3+} = 1.27$ Å, $Sr^{2+} = 1.44$ Å) [4].

Fig. 1 shows the temperature dependence of the resistivity $\rho(T)$. $\rho(T)$ shows a sharp decrease just below the FM transition temperature T_c and behaves metallic below T_c except for the $Y = 0$ and 0.25 samples. From the magnetization measurement, the $Y = 0$ and 0.25 samples did not show the FM transition. For the $0.50 \leqslant Y \leqslant 1.0$ of GSSMO and $0.75 \leqslant Z \leqslant 1.0$ of NSSMO samples with relatively small r_A (= 1.321–1.334 Å), an anomalous reduction and the hysteretic behavior of $\rho(T)$ characteristic of the first-order transition was observed below T_c. On the other hand, the FM transition in the samples for $0 \leqslant Z \leqslant 0.50$ ($r_A \geqslant 1.338$ Å) is of the typical second order. T_c increases and the r values decrease with increasing ionic radius of RE site ($r_{Gd} < r_{Sm} < r_{Nd}$) probably because of the reduction in the structural departure from the ideal cubic. In the present GSSMO and NSSMO systems, the tolerance factor f approaches 1, the ideal cubic value, with increasing r_A.

Fig. 2 shows the temperature dependence of the thermal conductivity $\kappa(T)$ of the typical samples. $\kappa(T)$ is almost entirely contributed by the phonon component in the present systems. Corresponding to the first-order FM transition for respective samples ($0.50 \leqslant Y \leqslant 1.0$ of GSSMO and $0.75 \leqslant Z \leqslant 1.0$ of NSSMO), a step-like $\kappa(T)$ enhancement can be seen just below T_c. For the second-order FM transition ($0 \leqslant Z \leqslant 0.50$), $\kappa(T)$ shows a gentle increase below T_c. In our previous investigations for $La_{1-X}Ca_XMnO_3$ and $La_{1-X}Sr_XMnO_3$ systems, the $\kappa(T)$ anomalies around T_c is closely correlated with those of the thermal dilatation $dL(T)/L$ [5,6]. Fig. 3 shows $dL(T)/L$ for the NSSMO system. The sharp and hysteretic reduction of $dL(T)/L$ was observed just below T_c only for the $Z = 1.0$ and 0.75 samples. In these samples, the decrease of the temperature derivative of $dL(T)/L$ is also noticeable above T_c, which means the gradual accumulation of the lattice distortion on approaching T_c from higher temperatures. In contrast, for the $Z \leqslant 0.50$ samples, the anomaly of $dL(T)/L$ around T_c is gentle and become weaker with decreasing Z. The lattice

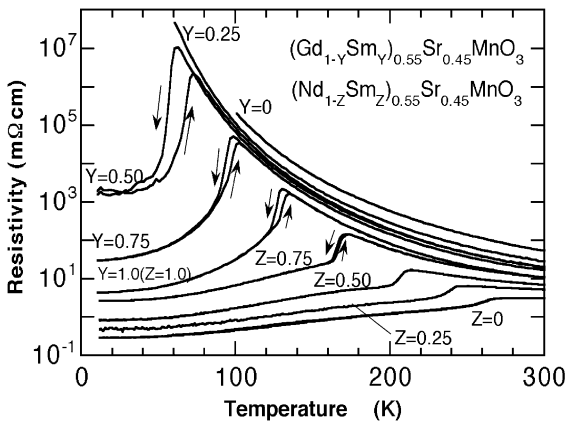

Fig. 1. The temperature dependence of the resistivity $\rho(T)$ for the $(Gd_{1-Y}Sm_Y)_{0.55}Sr_{0.45}MnO_3$ ($0 \leqslant Y \leqslant 1.0$) and $(Nd_{1-Z}Sm_Z)_{0.55}Sr_{0.45}MnO_3$ ($0 \leqslant Z \leqslant 1.0$) samples.

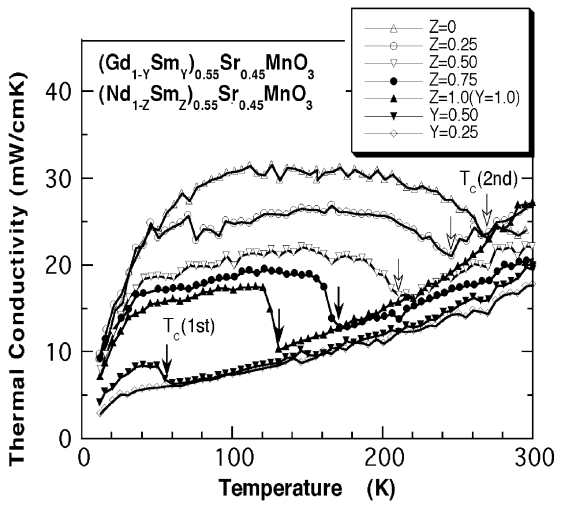

Fig. 2. The temperature dependence of the thermal conductivity $\kappa(T)$ of the typical samples. The closed and open arrows indicate the first- and second-order FM transition.

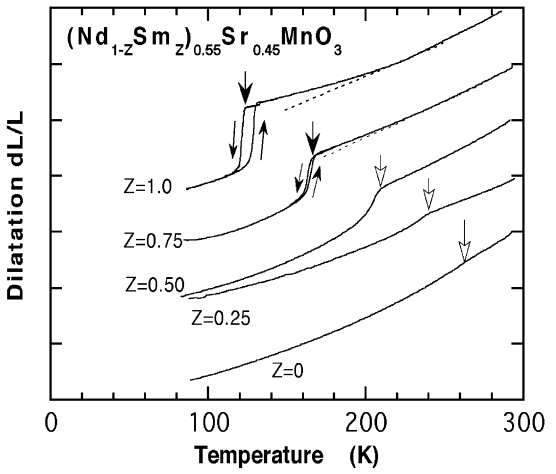

Fig. 3. The temperature dependence of the thermal dilatation $dL(T)/L$ for the NSSMO system. The closed and open arrows represent the first- and second-order FM transition.

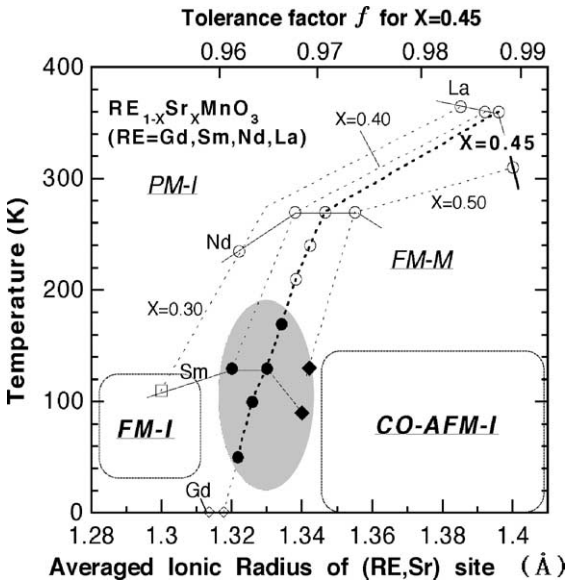

Fig. 4. The FM transition temperature T_c versus the average ionic radius r_A and the tolerance factor f (scale on the upper abscissa) for $RE_{1-X}Sr_XMnO_3$ system (RE = Gd, Sm, Nd, La). ♦ shows the first-order FM transition temperature by Kuwahara et al. [8].

contraction at and below T_c exemplified in $dL(T)/L$ may be at least partly attributable to the reduction of the local J–T distortion below T_c as a result of the increased itinerancy of the charge carriers. For $T > T_c$, the random local J–T distortions strongly scatter phonons, limiting $\kappa(T)$ to very small values, while the local J–T distortions are relaxed for $T < T_c$ and the phonon scattering is diminished in the FM–M phase.

Fig. 4 shows the FM transition temperature T_c versus the average ionic radius r_A. The data for $La_{1-X}Sr_XMnO_3$ system by us [7] and for the $(Nd_{1-y}Sm_y)_{1/2}Sr_{1/2}MnO_3$ system by Kuwahara et al. [8] are also included. The upper horizontal axis shows the tolerance factor f for the $X = 0.45$ system. The closed circles, open circles and open squares represent the FM transition temperatures T_c of the first-order accompanied with metallic conduction, the second-order transition with metallic conduction and the second-order transition with insulating behavior (FM–I), respectively. It should be noted that there is a peculiar first-order transition region into the FM-metallic phases in the $T_c - r_A$ plane. This region is situated between the charge-ordered AFM insulating and the FM insulating phases. Millis et al. theoretically predicted that the first-order transition may be possible between paramagnetic insulator and FM metallic phases for an intermediately strong electron–phonon coupling [9]. These first-order FM–M region shown in Fig. 4 is similar to the case of $(La_{1-y}Pr_y)_{1-X}(Ca_{1-Z}Sr_Z)_XMnO_3$ $(X \sim 0.25)$ [10].

In summary, the thermal conductivity $\kappa(T)$, thermal dilatation $dL(T)/L$ and the electrical resistivity $\rho(T)$ were measured for $(Gd_{1-Y}Sm_Y)_{0.55}Sr_{0.45}MnO_3$ (GSSMO) and $(Nd_{1-Z}Sm_Z)_{0.55}Sr_{0.45}MnO_3$ (NSSMO) systems. The thermal conductivity was very small above the FM transition temperature T_c probably because of strong phonon scattering by the local and spatially random J–T lattice distortions. For $T < T_c$, the local distortion was partially relaxed and $\kappa(T)$ was enhanced. The degree of the distortion relaxation seems to be more substantial for increasing charge carrier mobility, resulting in higher phonon thermal conduction. The relation between the first-order FM transition and the average ionic radius of the (RE,Sr) site, r_A was discussed. There is a peculiar region of the first-order FM-metallic phase in the $T_c - r_A$ plane,

which is situated between the charge-ordered AFM insulating and the FM insulating phases.

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