実験報告書様式（一般利用課題・成果公開利用）

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CROSS TOKAI **Experimental Report** J-PARC	承認日 Date of Approval 2014/5/26 承認者 Approver Ryoichi Kajimoto 提出日 Date of Report 2014/05/26
課題番号 Project No. 2014A0083 実験課題名 Title of experiment Study of phonons in La and Mn doped $SrTiO_3$: a potential new *n*-type thermoelectric material 実験責任者名 Name of principal investigator Ryoichi Kajimoto 所属 Affiliation J-PARC Center, JAEA	装置責任者 Name of Instrument scientist Ryoichi Kajimoto 装置名 Name of Instrument/(BL No.) 4SEASONS/(BL01) 実施日 Date of Experiment 2014/05/19 11:00 – 2014/05/21 09:00

試料、実験方法、利用の結果得られた主なデータ、考察、結論等を、記述して下さい。（適宜、図表添付のこと）

Please report your samples, experimental method and results, discussion and conclusions. Please add figures and tables for better explanation.

1. 試料 Name of sample(s) and chemical formula, or compositions including physical form.

$SrTiO_3$

$SrTi_{0.98}Mn_{0.02}TiO_3$

$Sr_{0.95}La_{0.05}TiO_3$

$Sr_{0.95}La_{0.05}Ti_{0.98}Mn_{0.02}O_3$

powder

2. 実験方法及び結果（実験がうまくいかなかった場合、その理由を記述してください。）

Experimental method and results. If you failed to conduct experiment as planned, please describe reasons.

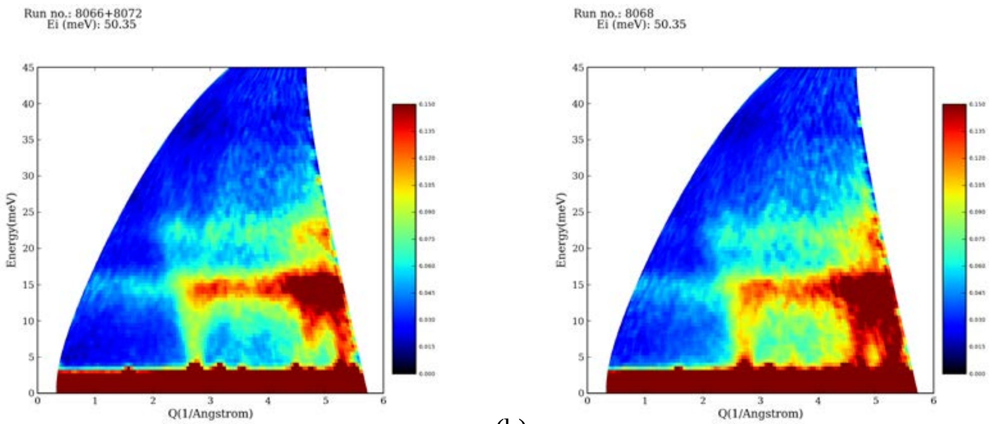
Very recently, it was found that the thermal conductivity (κ) in $SrTiO_3$ at room temperature (RT) is significantly reduced by co-doping of La and Mn in Sr and Ti sites, respectively (H. Hata *et al.*, JPS 2013 Autumn Meeting, 27aKQ-11). This finding makes this series of materials a promising candidate of an n-type oxide thermoelectric device. Interestingly, the anomaly in κ is induced by the doping of only a few percent of both La and Mn, but the La or Mn doping alone cannot induce the anomaly. It is well known that κ in $SrTiO_3$ is governed by the contribution from phonon. Accorddingly, we measured phonons in La and Mn doped compounds in addition to the mother compound by inelastic neutron scattering (INS), to investigate the microscopic mechanism of the suppression of κ.

We prepared 5 g powder samples of $SrTiO_3$, $SrTi_{0.98}Mn_{0.02}TiO_3$, $Sr_{0.95}La_{0.05}TiO_3$, and $Sr_{0.95}La_{0.05}Ti_{0.98}Mn_{0.02}O_3$. The INS measurements were performed using the chopper spectrometer 4SEASONS. Utilized incident neutron energies (E_i's) were 131, 50, and 26 meV, which were chosen by rotating the Fermi chopper at 250 Hz. The measurements were performed at RT.

2. 実験方法及び結果（つづき） Experimental method and results (continued)

Figure 1 compares the excitation spectra of the mother compound and the La-Mn co-doped sample measured with E_i = 50 meV, shown as functions of momentum (Q) and energy transfer ($\hbar\omega$). Overall features of both the spectra are apparently similar: several dispersive acoustic modes of phonons are clearly observed at Q ~ 2.8, 4.5, and 5.2 $Å^{-1}$ in addition to non-dispersive optical modes at $\hbar\omega$ ~ 15 and 22 meV. However, the low-energy region for the co-doped sample has higher intensity. To see the difference more quantitatively, we integrated the data for all the samples over Q = 2-4.5 $Å^{-1}$, and show them as functions of $\hbar\omega$ in Fig. 2. The profile for the Mn doped sample looks very similar to that for the mother compound. On the other hand, the La doped sample shows slightly higher spectral weight below $\hbar\omega$ ~10 meV. The increase of the spectral weight at this energy region is much enhanced for the co-doped sample.

One of the possible origins of the enhancement in the low-energy phonon spectrum is that the softening of the acoustic mode. This should lead decrease in the velocity of phonon around the Γ point, resulting in the decrease in κ. Another possible scenario is that the damping of acoustic phonon. The damping should shorten the mean free path of the phonon, which also suppresses κ. Although the single crystal measurement should be required to unambiguously elucidate the origin of the phonon anomaly, the observed differences in the phonon spectra show clear correspondence to the difference in κ of the compounds studied, and should give an important clue to understand the origin of the suppression of κ.



(a) (b)

Fig. 1. Q-ω maps of the excitation spectra of (a) $SrTiO_3$ and (b) $Sr_{0.95}La_{0.05}Ti_{0.98}Mn_{0.02}O_3$.

Fig. 2. Energy dependence of the excitation spectra integrated over Q = 2-4.5 $Å^{-1}$. Purple, light blue, red, green, and blue points indicate the data for $SrTiO_3$, $SrTi_{0.98}Mn_{0.02}O_3$, $Sr_{0.95}La_{0.05}TiO_3$, $Sr_{0.95}La_{0.05}Ti_{0.98}Mn_{0.02}O_3$, and empty run, respectively.