Thermoelectric Power Studies on the CuO\textsubscript{2} Plane Contributions of 
(Y\textsubscript{1-x}Ca\textsubscript{x})Ba\textsubscript{2}Cu\textsubscript{3}O\textsubscript{6.1} and (Pb\textsubscript{0.5}Cd\textsubscript{0.5})Sr\textsubscript{2}(Y\textsubscript{1-x}Ca\textsubscript{x})Cu\textsubscript{2}O\textsubscript{7}

C. W. Chang\textsuperscript{1,2}, J. G. Lin\textsuperscript{1}, C. Y. Chang\textsuperscript{3}, R. S. Liu\textsuperscript{3} and C. Y. Huang\textsuperscript{1,2}

\textsuperscript{1}Center for Condensed Matter Sciences, National Taiwan University, Taipei, Taiwan, R.O.C.
\textsuperscript{2}Department of Physics, National Taiwan University, Taipei, Taiwan 106, R.O.C.
\textsuperscript{3}Department of Chemistry, National Taiwan University, Taipei, Taiwan 106, R.O.C.

(Received August 15, 1997)

The thermoelectric power (TEP) for (Y\textsubscript{1-x}Ca\textsubscript{x})Ba\textsubscript{2}Cu\textsubscript{3}O\textsubscript{6.1} and (Pb\textsubscript{0.5}Cd\textsubscript{0.5})Sr\textsubscript{2}(Y\textsubscript{1-x}Ca\textsubscript{x})Cu\textsubscript{2}O\textsubscript{7} have been measured. The temperature dependence of TEP for these two series show very similar characteristic curves, which may be associated with the CuO\textsubscript{2} plane contributions. By comparing the data of these two different compound systems, we have shown that (Pb,Cd)O layers have negligible contribution to TEP in underdoped region. And, by comparing our results with those of Zn-doped Y-123 compounds, we suggest that the suppression of interlayer coupling may be the reason for the disappearance of a hump in the normal state TEP.

PACS. 74.25.Fb -- Transport properties.
PACS. 74.72.Bk -- Y-based cuprates.
PACS 74.72.Jt -- Other cuprates.

I. Introduction

Thermoelectric power (TEP) has become a simple and ideal probe for the studies of high \(T_c\) superconductors. Typical studies of TEP enable one to learn about the carrier concentration, flux-flux interaction, fluctuation effects and phonon drag effects, and, recently, even the normal state pseudogap can also be investigated by TEP measurements [1,2].

Recently, it has been shown that for compound YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7-\(\delta\)} both CuO\textsubscript{2} plane and CuO chain contribute to TEP, and from the underdoped to overdoped region, each sample shows a very different characteristic curve. It has been pointed out that for fully oxygenated YBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7-\(\delta\)} (\(\delta < 0.3\)) the CuO\textsubscript{2} plane contributes to the positive value of TEP with a negative slope of \(dS(T)/dT\), whereas the CuO chain to the negative TEP with a positive slope [3].

Although many measurements of TEP upon Y-123 cuprates have been performed, the lack of precise oxygen control disables one to distinguish the pure contribution of CuO\textsubscript{2} plane or CuO chain because the oxygen concentration has severe effects on TEP around the optimally doped Y-123 cuprates. In this paper, we show the pure plane contribution from the highly deoxygenated tetragonal (Y\textsubscript{1-x}Ca\textsubscript{x})Ba\textsubscript{2}Cu\textsubscript{3}O\textsubscript{6.1} samples and compare the results to that of less known series of (Pb\textsubscript{0.5}Cd\textsubscript{0.5})Sr\textsubscript{2}(Y\textsubscript{1-x}Ca\textsubscript{x})Cu\textsubscript{2}O\textsubscript{7}, for which (Pb,Cd)O layers, instead of CuO chains, act as charge reservoir. Since most experiments of TEP in
the underdoped region were done on the samples of removing the oxygen atoms out of the CuO chains, the chain contribution on the underdoped region certainly gets smaller when oxygen deficiency increases. Therefore, it is quite hard to study the chain contribution in underdoped region without disturbing CuO chains. By taking the advantage of (Pb-based) layers, we are able to study the contribution from (Pb,Cd)O layers on TEP without removing any oxygen atoms of the layers. And, by comparing the results to that of the empty-chain tetragonal \((Y_{1-x}Ca_x)Ba_2Cu_3O_{6.1}\) compounds, we are able to estimate the contribution from (Pb,Cd)O layers at various temperatures.

II. Experiments

The samples of \((Y_{1-x}Ca_x)Ba_2Cu_3O_{6.1}\) were prepared by mixing high purity powders of \(Y_2O_3\), \(BaCO_3\), \(CaCO_3\) and CuO. The mixtures were calcined at 950 °C for 20h in air. The resulting mixtures were ground and pressed into pellets. The pellets were sintered at 950 °C for 20h in air. Following, the samples were annealed at 730 °C for 20h in Ar then quenching into liquid nitrogen. Samples with nominal composition of \((Pb_{0.3}Cd_{0.3})Sr_2(Y_{1-x}Ca_x)Cu_2O_{7.4}\) were prepared by mixing the oxides of PbO, CdO, SrCO_3, \(Y_2O_3\), CaCO_3 and CuO in the appropriate molar ratio. The mixtures were calcined at 850 °C for 15h in air. The resulting mixtures were ground and then pressed into pellets. The pellets were sintered at 950 °C for 10h in air. Oxygen content was determined by the chemical titration method.

The absolute thermoelectric power of the sample was obtained by correcting the measured Seebeck coefficient with the absolute thermoelectric power of copper at various temperatures. The reference Cu leads were calibrated against the Pb standard. Chromel/Constantan thermocouple were used to detect the temperature gradient which was imposed by a heater and kept in a range of 0.8 - 1.2K.

III. Results and discussions

Figure 1 shows the TEP data for the tetragonal \((Y_{1-x}Ca_x)Ba_2Cu_3O_{6.1}\) samples with \(x = 0.2, 0.25\), respectively. It demonstrates that by doping \(Ca^{2+}\) into \(YBa_2Cu_3O_{6.1}\), the superconductivity can be induced with \(T_c \sim 30K\) and \(T_c \sim 45K\), respectively [4].

Figure 2 shows the TEP of \((Pb_{0.3}Cd_{0.3})Sr_2(Y_{1-x}Ca_x)Cu_2O_7\) with \(x = 0, 0.1, 0.2, 0.3, 0.4, 0.5\). The sample with \(x = 0.5\) is a superconductor with \(T_c \sim 35K\). Although the susceptibility measurement shows a drop around 15 K for \(x = 0.4\) [5], the drop does not show on the TEP vs. T curve which was measured down to 10 K. All the six samples show systematic characteristic curves of underdoped TEP. With increasing doping \(Ca^{2+}\), \(S(290K)\) decreases as expected since the hole concentration increases. Notably, the normal state weak temperature dependent range broadens as the hole concentration increases and the absence of humps in TEP for all these six compositions is also a general feature. The hump of TEP at the normal state was recently reported to be an another kind of universal behavior of HTSC, and some authors have correlated it to the opening of the pseudogap in the normal state of a superconductor [1]. Based on the recently established empirical hole-pseudogap relation [1], we can roughly estimate that the hump of TEP lies above 290K for all these compositions.
FIG. 1. The thermoelectric power $S$ vs. temperature $T$ for $(\text{Y}_{1-x}\text{Ca}_x)\text{Ba}_2\text{Cu}_3\text{O}_{6.5}$ samples with $x = 0.2$ and $x = 0.25$, respectively.

FIG. 2. The thermoelectric power $S$ vs. temperature $T$ for $(\text{Pb}_{0.5}\text{Cd}_{0.5})\text{Sr}_2(\text{Y}_{1-x}\text{Ca}_x)\text{Cu}_2\text{O}_7$ samples with $x = 0, 0.1, 0.2, 0.3, 0.4, 0.5$, respectively.
The \((\text{Pb}_{0.5}\text{Cd}_{0.5})\text{Sr}_2(\text{Y}_{1-x}\text{Ca}_x)\text{Cu}_2\text{O}_7\) (Pb-based 1212 type) compounds have a different kind of charge reservoir structure than \((\text{Y}-123)\) compounds, namely the (Pb,Cd)O layers replace \(\text{CuO}_2\) chains in \(\text{Y}-123\), and, oxygen atoms of (Pb,Cd)O layers locate just below the Sr atoms while the (Pb,Cd) atoms replace Cu atom in \(\text{CuO}_2\) chains of \(\text{Y}-123\) compounds. Clearly, this structure will not have the \(\text{CuO}_2\) chain contribution in the underdoped region. Some authors have proposed that the (Pb-based) layers have negligible effects on TEP like \((\text{Bi,Pb})_2\text{O}_2(\text{Tl,Pb})_2\text{O}_2(\text{Tl,Pb})\O\) layers in the Bi and Tl systems [3].

In Figure 3, we compare the TEP curves of \((\text{Y}_{0.8}\text{Ca}_{0.2})\text{Ba}_2\text{Cu}_3\text{O}_{6.1}\) and \((\text{Y}_{0.75}\text{Ca}_{0.25})\text{Ba}_2\text{Cu}_3\text{O}_{6.1}\) with \((\text{Pb}_{0.5}\text{Cd}_{0.5})\text{Sr}_2(\text{Y}_{0.8}\text{Ca}_{0.2})\text{Cu}_2\text{O}_7\) and \((\text{Pb}_{0.5}\text{Cd}_{0.5})\text{Sr}_2(\text{Y}_{0.7}\text{Ca}_{0.3})\text{Cu}_2\text{O}_7\). The similarities among these curves are easily seen. For \((\text{Y}_{0.75}\text{Ca}_{0.25})\text{Ba}_2\text{Cu}_3\text{O}_{6.1}\) and \((\text{Pb}_{0.5}\text{Cd}_{0.5})\text{Sr}_2(\text{Y}_{0.8}\text{Ca}_{0.2})\text{Cu}_2\text{O}_7\), not only the values of \(S(T)\) but also the shape of the curve, is almost the same above 100K. This indicates that nearly the same magnitude of plane contributions in both \(x = 0.2\) compounds. The small deviation of \(S(T)\) in \((\text{Pb}_{0.5}\text{Cd}_{0.5})\text{Sr}_2(\text{Y}_{0.8}\text{Ca}_{0.2})\text{Cu}_2\text{O}_7\) from that in \((\text{Y}_{0.8}\text{Ca}_{0.2})\text{Ba}_2\text{Cu}_3\text{O}_{6.1}\) may be accounted for by the presence of slight oxygen deficiency or from (Pb,Cd)O layers. If the former dominates, then \(S(T)\) value will be lower and the contribution from the (Pb,Cd)O layers is negligibly small. Even if the (Pb,Cd)O layers contribute to this deviation significantly, we can still conclude that the contribution to TEP value from (Pb,Cd)O layers is smaller than that from \(\text{CuO}_2\) plane, and their contribution to the TEP slope is much smaller.

FIG. 3. Comparison of \(S(T)\) vs. T for \((\text{Y}_{1-x}\text{Ca}_x)\text{Ba}_2\text{Cu}_3\text{O}_{6.1}\) with \(x = 0.2, x = 0.25\) (denoted as \(\text{Y}-x = 0.2, \text{Y}-x = 0.25\)) and \((\text{Pb}_{0.5}\text{Cd}_{0.5})\text{Sr}_2(\text{Y}_{1-x}\text{Ca}_x)\text{Cu}_2\text{O}_7\) with \(x = 0.2, x = 0.3\). Note that \(\text{Y}-x = 0.25\) and \(\text{Pb}-x = 0.2\) curves are nearly the same above 100K.
Comparing our results with that of Tallon's[1], it is surprising that there is a similarity on the shapes and the values of their TEP data of \( \text{YBa}_2\{(\text{Cu}_{1-x}\text{Zn}_x)\}_3\text{O}_{7-\delta} \) \((x = 0.35 \text{ and } 0.53)\) with ours. They suggested that Zn doping can suppress the pseudogap, i.e. the hump of TEP. If this is the case, the similarities of ours and their Zn-suppression TEP curve is quite surprising because there is no destructive dopant at all in our samples. Although there is some evidences of Zn-effects on the pseudogap [6], the relations between the hump of TEP and the pseudogap are still not well established [7]. According to the paper of Zhou et. al.[4], the hump of TEP may originate from the interlayer coupling. From our data, we would rather propose that Zn-dopant suppresses the interlayer coupling instead of the pseudogap. In another word, our TEP results prompt us to believe that the hump of TEP is mainly from the interlayer coupling.

IV. Conclusions

We have measured the TEP of \( (\text{Pb}_{0.5}\text{Cd}_{0.5})\text{Sr}_2\{(\text{Y}_{1-x}\text{Ca}_x)\}_2\text{Cu}_2\text{O}_7 \) and \( (\text{Y}_{1-x}\text{Ca}_x)\text{Ba}_2\text{Cu}_3\text{O}_{6.1} \). The similarity of the TEP curves in these two different systems indicates that the contributions from (Pb,Cd)O layers are negligibly small. Based on a comparison of our results with that of Tallon et. al. who have proposed a universal relation between \( S(290) \) and the hump of TEP, we propose that the suppression of interlayer coupling might be the origin of the absence of the TEP hump.

Acknowledgments

We want to thank Dr. Z. J. Huang[5] for the setup of our TEP measurements and his valuable suggestions. This work is supported in part by the National Science Council of the R.O.C. under grant No. NSC-86-2112-M-002-011.

References