PHYSICAL REVIEW B **79**, 075105 (2009)

Seebeck coefficient of Na_xCoO_2: Measurements and a narrow-band model

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(Received 17 May 2008; revised manuscript received 10 November 2008; published 9 February 2009)

We have performed Seebeck coefficient (S) measurement for a series of high-quality Na_xCoO_2 single crystals for $0.23 \leq x \leq 0.84$ from 10 to 300 K. The room-temperature S values were found to be around 30–100 $\mu V/K$ and increase with increasing Na content. The temperature dependence of Seebeck coefficient of this system was found to be very sensitive for $x=1/2$, which is most likely due to the significant modifications of band structure as a result of the magnetic and charge orderings below 88 and 51 K. The overall observed features of the $S(T)$ were analyzed in the framework of a narrow-band model. The physical parameters obtained from the fitting provide a satisfactory description of the composition and temperature dependences of Seebeck coefficient in the Na_xCoO_2 system.

DOI: 10.1103/PhysRevB.79.075105 PACS number(s): 71.28.+d, 72.15.Jf, 71.27.+a, 74.25.Fy

I. INTRODUCTION

The sodium cobalt oxide Na_xCoO_2 system, because of its peculiar properties of large thermoelectric power coexisting with low electrical resistivity, has attracted much attention in recent years.[1–3] In particular, the discovery of superconductivity in the water-intercalated Na_xCoO_2 (e.g., $Na_xCoO_2:yH_2O$, where $x \approx 0.33$ and $y \approx 1.3$) compound is a breakthrough in the search for new layered transition-metal oxides superconductors.[4,5] A lot of studies have been focused on the hydrate superconductor to clarify the mechanism of the superconductivity and to understand the underlying electronic states realized in the triangular lattice.[4–8] Likewise, a characteristic of unhydrated Na_xCoO_2 is the sensitivity of electronic states to change slightly in x.[9,10] In fact, a charge ordering state with an anomalous change in electrical, thermal, and magnetic properties occurs in the sample $Na_{0.50}CoO_2$ with average Co valence of +3.5.[9,11,12] Recent studies have given evidence of strong electron-electron correlation in Na_xCoO_2 (Refs. 13–16) and demonstrated that the physical properties of unhydrated Na_xCoO_2 compounds are very sensitive to doping.

Indeed, a rich phase diagram has been revealed for Na_xCoO_2 as a function of x.[9] A spin ordered phase is found for $x > 3/4$. With decreasing Na contents, the material becomes a "Curie-Weiss metal" for x near 2/3, then a charge-ordered insulator with $x \sim 1/2$, and finally a paramagnetic metal with $x \sim 1/3$. It shows a low metallic resistivity of 200 $\mu\Omega$ cm accompanied by a large Seebeck coefficient of about 100 $\mu V/K$ at room temperature, which is about 1 order of magnitude larger than that of typical metals.[1] Koshibae *et al.*[15,16] proposed a theory for the large Seebeck coefficient based on the generalized Heikes formula in strongly correlated electron systems. The Seebeck coefficient of Na_xCoO_2 compounds is enhanced due to their large spin entropy, and the large absolute value of the Seebeck coefficient can be realized as the existence of an unbalance of the spin and orbital degrees of freedom between Co^{3+} and Co^{4+} sites. However, the generalized Heikes formula was proposed for the calculation of Seebeck coefficient in a system of interacting localized carriers with hopping conduction in the high-temperature limit.[17]

It appears that some experimental results support the spin entropy theory. In particular, Wang *et al.*[13] found a suppression of the Seebeck coefficient in a longitudinal magnetic field at low temperature and argued that the spin entropy is the likely source for the observed large Seebeck coefficient in Na_xCoO_2. On the other hand, within the standard Boltzmann transport theory, Singh[18] calculated the Seebeck coefficient of $Na_{0.5}CoO_2$ using the local-density approximation. The author argued that the Boltzmann theory may reproduce the magnitude of the Seebeck coefficient. Further, using the t-J model, Haerter *et al.*[19] suggested that the field suppression of the Seebeck coefficient may arise from strong electron correlations, although they also pointed out that their results agree with the interpretation in terms of spin entropy as the main contribution to the field suppression. Even though these models could quantitatively describe the high-temperature features of Seebeck coefficient, understanding of the complexity of its temperature and composition dependences is not available so far. On the other hand, the Seebeck coefficient of Na_xCoO_2 is highly sensitive to the Na content, indicating that the charge carriers in the CoO_2 planes may be from a very narrow band.[20,21]

One of the established consensus on the Na_xCoO_2 is that the two bands, an a_{1g} and one of the e_g' bands, from Co t_{2g} manifold cross the Fermi level (E_F).[18,22] They form a large hole Fermi surface (FS) centered at around zone center (by a_{1g}) and six small Fermi pockets near the zone boundary (by e_g'). Angle-resolved photoemission spectroscopy (ARPES) measurements reveal a doping-dependent evolution of the holelike FS, which shows no sign of the e_g' hole pockets for $0.3 \leq x \leq 0.72$.[23–26] The observed FS is centered on the zone-center point and has mostly a_{1g} character. In fact, it has been pointed out in these studies that the extremely narrow quasiparticle bands are essential in the evolution of such a holelike FS. Therefore, the narrowness of the bands must indeed be a factor, in addition to the strong electron correlations and large spin entropy, involved in the complexity of Na_xCoO_2 system.

All these results motivate us to perform a thorough investigation of the transport properties of Na_xCoO_2 with a wide range of Na content to address some of these issues. In particular, the Seebeck coefficient is a sensitive probe of the degree of itinerary of the carriers, and a coherent interpretation of the thermoelectric response is highly desirable. In this study we have investigated the temperature dependence of Seebeck coefficient (S) of Na_xCoO_2 single crystals with $x \sim 0.23$, 0.34, 0.50, 0.55, 0.70, 0.76, and 0.84. The observed features of the $S(T)$ were analyzed in the framework of a narrow-band model, which gives a satisfactory description to the observed data. The obtained bandwidth of narrow bands and asymmetry involved in the data analyses provide a reasonable explanation to the large and anomalous temperature variation in the Seebeck coefficient of the Na_xCoO_2 system, and our results represent an alternative view in understanding the thermoelectric properties of these materials.

II. EXPERIMENT

Single crystals of Na_xCoO2 ranging from $x=0.23\pm0.02$ to $x=0.84\pm0.01$ used in this study are prepared using floating-zone method with grown $Na_{0.84}CoO_2$ crystal, and the Na content x is tuned with additional electrochemical Na deintercalation as described in Ref. 27, with error bars of ±0.02 for $x<0.5$ and ±0.01 for $x \geqq 0.5$, as verified by electron microprobe analysis (EPMA). The chosen Na content levels correspond to stable Na ordered phases, as verified by the electrochemical properties and Laue diffraction.[27] Seebeck coefficient measurements were carried out in the helium closed-cycle refrigerator using a heat-pulse technique. A detailed description of the experimental technique can be found elsewhere.[28]

III. RESULTS AND DISCUSSION

Figures 1 and 2 show the temperature variation in the Seebeck coefficient, $S(T)$, for the Na_xCoO_2 single crystals with $x \sim 0.23$, 0.34, 0.50, 0.55, 0.70, 0.76, and 0.84. The room-temperature $S(S_{300\text{ K}})$ values for studied samples were found to be around 30–100 μV/K and increase with increasing x. Such a typical range of S is also reported by others on single crystal[1,9] and bulk[29] samples. A diffusionlike behavior at low temperatures with positive Seebeck coefficients was observed in the sample with concentration $x=0.23$. Note that there is a sign change in the T-dependent Seebeck coefficient of Na_xCoO_2 system between $x=0.34$ and 0.55. The Seebeck coefficient of these samples start with negative in low-temperature regime and then crosses over zero at higher temperatures. Such a sign reversal indicates that there may be a substantial change in the band structure or the conduction mechanism of the dominant carriers at these Na concentrations. For $x=0.50$, the Seebeck coefficient shows somewhat complex temperature dependence. Peculiarity of this sample is the appearance of distinct peaks around 50 K in the negative quadrant and S becomes positive at around 85 K presumably due to the charge and antiferromagnetic (AF) spin ordering, respectively.[9,11,12] The deviation of S from linear behavior at $T=85$ K provides evidence of microscopic mechanism, i.e., the onset of a long-range ordered nature of the Na superstructure.

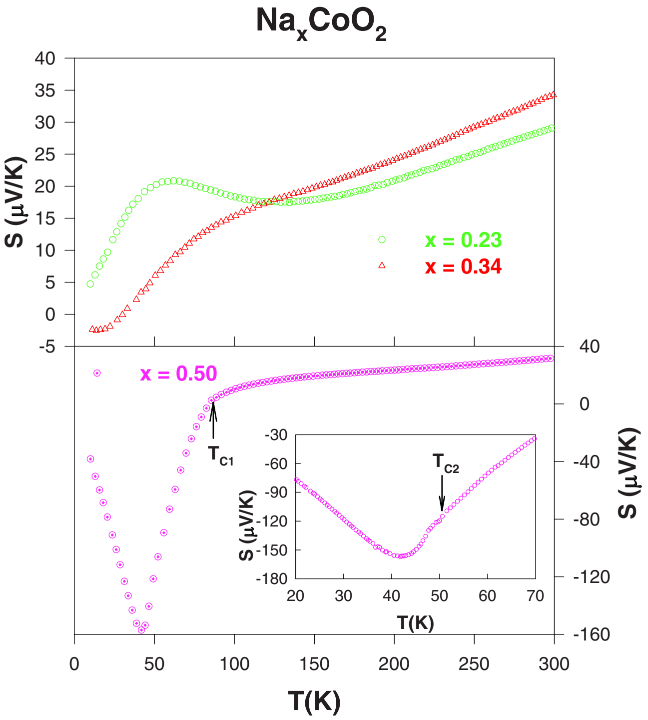

FIG. 1. (Color online) Thermopower as a function of temperature for Na_xCoO_2 with $x=0.23$, 0.34 (upper panel), and 0.50 (lower panel). Two transitions are observed in the $x=0.50$ sample as indicated by the arrows. The inset shows a close-up plot near T_{C2}.

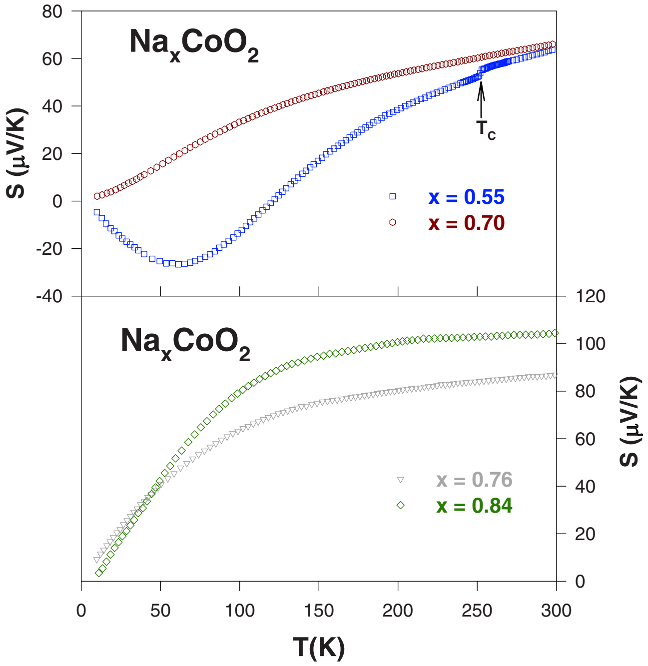

FIG. 2. (Color online) Thermopower as a function of temperature for Na_xCoO_2 with $x=0.55$, 0.70 (upper panel), and 0.76, 0.84 (lower panel).

Phonon drag can also contribute a hump to $S(T)$ when the temperature is low enough that phonon-phonon interactions are negligible but high enough for the density of phonon state being important. A likely explanation of the origin of low-temperature peak presented here in $Na_{0.5}CoO_2$ originated from the charge ordering and not by phonon drag, which we will discuss shortly. For higher x (≥ 0.70), S increases monotonically with increasing temperature and reaches above 65 $\mu V/K$ at room temperature. As far as the $x > 0.55$ samples are concerned, their $S_{300\ K}$ ($> 65\ \mu V/K$) values are good enough for the materials to be considered as a thermoelectric material[1–3] with further scope of optimization. It should be noted that the magnitude of $S_{300\ K}$ is somewhat smaller compared to that reported by Lee and co-workers[10] for high sodium samples ($x \geq 0.70$). Several factors may cause such a difference in the magnitude of S. First, they used as-grown single crystal of $Na_{0.75}CoO_2$ and then added Na content through high-temperature (100 °C) chemical intercalation,[10] which is expected to show different Na ordering (and thus spin entropy) compared with present electrochemical deintercalation route performed at room temperature. Second, our samples were prepared electrochemically with prolonged equilibrated applied potential, which guarantees perfect Na ordering. Since spin entropy is thought to be the main source for the high value of $S_{300\ K}$, the relatively ordered phases in the present set of samples may have a reduced contribution from the spin entropy.

At $x=0.5$, the Na ions are chemically ordered to form zigzag chains in an orthorhombic superstructure with twofold symmetry,[31] and the ground state is believed to be a magnetically ordered state, leading to $Co^{+3.5+\delta}/Co^{+3.5-\delta}$ charge order within each CoO_2 layer.[9,11,12] For example, Gasparovic *et al.*[11] performed neutron scattering, susceptibility, and transport study measurements to investigate the electronic ground state of the half-doped CoO_2 plane. The key observations are as follows. First, the electrical resistivity (ρ) in this sample increases gradually as T falls toward T_{C1} (=88 K) and T_{C2} (=51 K), where it exhibits anomalies. However, below T_{C2}, ρ rises rapidly and reaches its maximum. Second, a negative magnetoresistance is observed below ~51 K in the direction of the *ab* plane, while it is positive below 88 K in the direction of the *c* plane. In fact, the anisotropy in the magnetoresistance coincides with the anomalies observed in magnetic susceptibility in $Na_{0.50}CoO_2$. All these results clearly demonstrate that the sharp peak and complex behavior of Seebeck coefficient of $Na_{0.50}CoO_2$ below 88 K is most likely due to the onset of charge and magnetic orderings. The separation of Co spin states into $Co^{+3.5+\delta}$ and $Co^{+3.5-\delta}$ would allow an increase in the magnetic stabilization via local-moment formation on the Co^{3+} sites, and the cost in kinetic energy of charge carriers would lead to a large value of S as the bands are quite narrow.

Now, we will discuss an alternative possibility in the explanation of our findings through the effect of some special band structure in the Na_xCoO_2 system. The observed features needed to be considered here, including sign anisotropy, temperature dependence of S, and the magnitude increase with increasing x in $S_{300\ K}$, are all quite consistent with previous results for Na_xCoO_2.[1,9,29] However, the high quality of the Na_xCoO_2 single crystals and the wide range of the Na content are chosen to have specific Na superstructure orderings,[20] which allows us to look into the details of the observed behaviors. Let us now elaborate the electronic structure studies on Na_xCoO_2 system and deduce its possible implication on transport properties of Na_xCoO_2 in the framework of narrow-band model, which has been successfully applied to high-temperature superconductors, another system of narrow bandwidth.[32–34] Several studies on band-structure calculations revealed that there is a clean gap between the manifolds for Na_xCoO_2 system, which, in turn, is due to the narrow bandwidths (~1 eV).[18,22] Furthermore, low average Fermi velocity and a very high value of the density of states (DOS) at E_F are also considered to be the consequence of the narrow bandwidth in the present system.[35] However, ARPES studies show that the bandwidths are about few tens of meV.[23–26] The reduction in bandwidth is explained in terms of a strong mass renormalization due to the strong correlation in these materials. Therefore, the present compounds (Na_xCoO_2) provide a unique example of a narrow bandwidth system, and later we will show that such a narrow bandwidth might be one of the important factors responsible for the observed large and anomalous nature of $S(T)$.

TABLE I. Parameters (described in text) obtained from the fitting of experimental data using Eq. (2).

Samples	F	W_D (meV)	$c(W_\sigma/W_D)$
$Na_{0.23}CoO_2$	0.56	16	0.78
$Na_{0.34}CoO_2$	0.58	18	0.70
$Na_{0.50}CoO_2$	0.60	50	0.68
$Na_{0.55}CoO_2$	0.64	69	0.64
$Na_{0.70}CoO_2$	0.66	140	0.60
$Na_{0.76}CoO_2$	0.72	147	0.58
$Na_{0.84}CoO_2$	0.74	165	0.56

To begin with, we have analyzed our data using narrow-band model with a few phenomenological parameters such as the bandwidth (W_D), width of the conduction window (W_σ), and the degree of band filling (F). Here, the narrow band could be either a single band in the DOS or a narrow peak on a broad background.[18] The parameters calculated (see Table I) using this model are in agreement with the fact that the magnitude of bandwidth is determined by the intercalation of Na atoms in CoO_2 layers, i.e., band filling, and hence it is strongly affected by the overlapping of the O p bands and Co t_{2g} bands.[36] If the Fermi level is located inside this narrow band, within which the density-of-states values are much larger than the ones outside this band, then this portion of the band will play a dominant role in the transport properties of this compound. On applying the narrow-band limitation, $W \ll k_BT$, the Seebeck coefficient can be express as[32]

$$S = \frac{k_B}{e}\ln\left(\frac{F}{1-F}\right) \approx \frac{E-\mu}{eT}. \tag{1}$$

Here, μ is the chemical potential. To highlight the influence of the bandwidth, Martynova and Gasumyants[32,33] performed

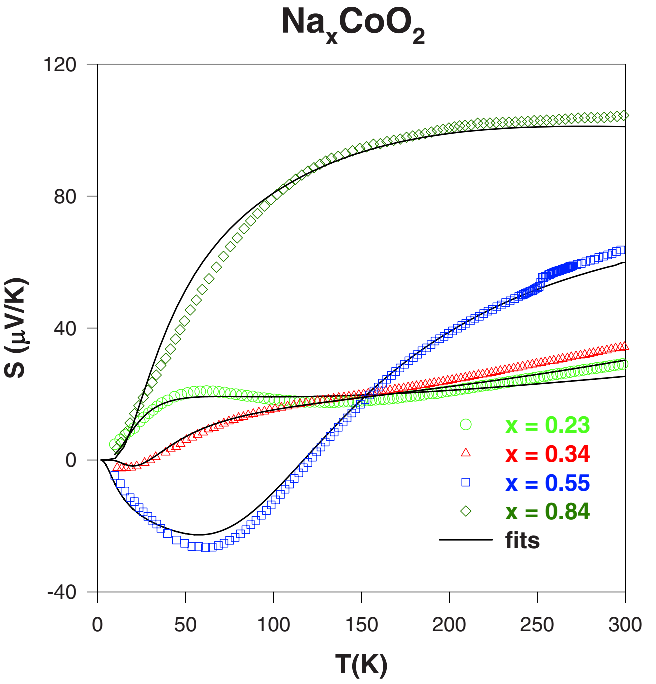

FIG. 3. (Color online) Seebeck coefficient as a function of temperature for $Na_{0.23}CoO_2$, $Na_{0.34}CoO_2$, $Na_{0.55}CoO_2$, and $Na_{0.84}CoO_2$ samples along with theoretical fit using Eq. (2). For $Na_{0.55}CoO_2$, a small asymmetry factor ($b=0.03W_D$) is added to the band near E_F in Eq. (2), as described in text.

the calculations of the transport integrals and neglected the higher-power terms in the distribution function expanded as a Taylor series. Under this approximation, $S(T)$ for $W \approx k_BT$ is given as

$$S=-\frac{k_B}{e}\left\{\frac{W_\sigma^*}{\sinh(W_\sigma^*)}\left[e^{-\mu^*}+\cosh(W_\sigma^*)-\frac{\cosh(\mu^*)+\cosh(W_\sigma^*)}{W_\sigma^*}\ln\frac{e^{\mu^*}+e^{W_\sigma^*}}{e^{\mu^*}+e^{-W_\sigma^*}}\right]-\mu^*\right\}, \tag{2}$$

where $\mu^*=\mu/k_BT=\ln\{\sinh[FW_D^*]/\sinh[(1-F)W_D^*]\}$, $W_\sigma^*=W_\sigma/2k_BT$, and $W_D^*=W_D/2k_BT$.

We have used the above equation to deduce the Seebeck coefficient profile as a function of temperature, and four representative examples of the fits of Eq. (2) to the Seebeck coefficient data for Na_xCoO_2 system are shown in Fig. 3. It is evident from Fig. 3 and Table I that the peculiar behavior of $S(T)$ in Na_xCoO_2 could be satisfactorily described in terms of the narrow bandwidth of the expected bands as follows. First, we suppose the band to be symmetric and the energy to be evaluated from the midpoint of the band. It seems that the consideration of symmetric bands alone is sufficient to explain the observed temperature dependence of $S(T)$ for lower (≤ 0.34) and higher (≥ 0.70) Na concentrations (see Fig. 3). However, it does not hold completely for intermediate Na concentrations in the Na_xCoO_2 system. Second, we therefore consider the influence of a slight band asymmetry to the temperature dependence of $S(T)$. Actually, the observed renormalization anisotropy in Fermi velocity along the principal axes confirms the existence of asymmetry involved in the bands.[25] Furthermore, it has been revealed that the peculiar shape of the band, such as the a_{1g} band of the cobaltates, which consists of a dispersive portion and a somewhat flat portion, plays an important role not only for the large Seebeck coefficient and low resistivity but also for the superconductivity and the magnetism in the Na_xCoO_2 system.[37] Moreover, the temperature dependence of the Seebeck coefficient of this system was found to be very sensitive for sodium content near 0.5, which is most likely due to the significant modifications of band structure as a result of the existence of magnetic and charge ordering transitions.[9,11,12] Therefore, the peculiar shape or anisotropy in the bands could play an important role near half doped. Thus here we have taken the asymmetry into account by introducing some distance (bW_D) away from the symmetric band in the approximating DOS. In such a case, Eq. (2) remains valid except the parameter μ^* which is displaced by an amount of $2bW_D/k_BT$. It is found that if a small asymmetry factor ($b=0.03W_D$) is added to the band near E_F in Eq. (2), as described in the literature,[32,33] a dramatic effect would occur in a nearly half-filled band. Such an effect enables us to explain the peculiarities, particularly the low-temperature hump, of the observed $S(T)$ for the samples near $x=0.05$. In Fig. 3, we clearly demonstrate that the small asymmetry involved in the bands may retrace the experimental data for $x=0.55$. It should be noted here that the $x=0.5$ sample exhibits an anomalous sharp peak at low temperatures that could not be reproduced quantitatively with the presently used model. However, the set of fitting parameters for this sample is able to give qualitative understanding of $S(T)$ and a good fit above 100 K (not shown here). Therefore, the observed peculiar behavior of $S(T)$ near $x=0.50$ is most likely due to the combination of band asymmetry and bandwidth broadening with increasing Na content.

In order to further capture the essential feature of the $S(T)$ in the Na_xCoO_2 system, we are in the position to rationalize the deduced parameters listed in Table I. It is found that the width of the narrow band, W_D, increases from 16 meV for the $x=0.23$ to 150 meV for $x=0.84$. It is noted that the deduced value of bandwidth for low concentration is very close to the Fermi temperature.[30] The band filling factor F is also found to increase with increasing Na content. These are accompanied by a simultaneous reduction in the values of the parameters $c=(W_\sigma/W_D)$, characterizing the degree of delocalization of states in the conduction band of Na_xCoO_2. In fact, an increase in the Na concentration increases the substitutional disorder, and consequently, causes a broadening of the band (W_D) due to localization effects. On the other hand, a relative reduction in the energy interval occupied by the delocalized states is expected due to an increase in the Na content. The increase in F suggested that the reduction in effective charge-carrier density, which is equal to the ratio of the total number of electrons to the number of states available in the band, results in the enhancement of the room-temperature value of the Seebeck coefficients ($S_{300\ \mathrm{K}}$) with increasing x or Na content.

It is worth mentioning that the Wang *et al.*[13] found a suppression of the Seebeck coefficient in a longitudinal magnetic field at low temperature and argued that the spin entropy dominates the enhancement of the Seebeck coefficient in Na_xCoO_2. Another possibility is that, other than the a_{1g} bands which are the main source for FS evolution, the e_g' bands could also contribute to enhance the Seebeck coefficient if these bands approach E_F and cross it due to broadening of bandwidth with higher Na content. Such a possibility has been pointed out to produce a very large power factor, which might be the combined effect of both a_{1g} and e_g' pudding mold bands.[37] However, a broad peak, which forms the e_g' bands, approaches but never reaches E_F near the Brillouin-zone boundary for the range of $0.3 \leq x \leq 0.72$.[23] There is presumably a delicate balance between narrow bandwidth and electronic spin entropy that causes an enhanced Seebeck coefficient which depends sensitively on doping. Furthermore, we note that this Na content dependence of the bandwidth is in agreement with the doping-dependent shift of the valence bands observed in ARPES studies.[23] For the Co 3*d* orbitals, the average energy difference between $x=0.48$ and 0.72 is about 0.1 eV. However, for the O 2*p* orbitals, the energy shifts are larger (~ 0.5 eV) between these two doping levels. A similar conclusion was reached on the basis of the nature of a detectable shift in Raman-active modes with Na content, which is closely correlated with the ordering of Na ions in the Na_xCoO_2 system.[38]

Further, it is noted that, for high Na content samples, the reduction in the area of holelike FS centered on zone center has also been observed, which is in accordance with the Luttinger theorem.[23] It might be possible that some electron carriers become localized at higher Na doing levels due to increase in either W_D or F, resulting in a reduction in the number of itinerant electrons that contribute to the area of the FS. It is also noteworthy that the reduction in charge-carrier density with x is consistent with previous report in the Na_xCoO_2 system.[9] The compounds with higher Na content have apparently higher electrical resistivity $\rho(T)$ due to the reduced charge-carrier concentration. Since bandwidth and carrier concentration can be controlled to a large degree in this class of materials, our results could provide a guideline for tuning the figure of merit and realize the potential of applications in this class of system. Finally, a step up in $S(T)$ around $T_C=250$ K for $x=0.55$ was observed which is presumably associated with the Na ordering transition. However, this issue is beyond the scope of this study and will be presented in a separate paper.

IV. CONCLUSIONS

In conclusion, we have reported a detailed investigation of the Seebeck coefficient of Na_xCoO_2 single crystals of $0.23 \leq x \leq 0.84$ with highly ordered Na superstructures. The temperature dependence of the Seebeck coefficient of this system was found to be anomalous, particularly, near the half doped, which is most likely due to the onset of charge ordering or the long-range nature of the Na superstructure in this composition range. The observed features of the $S(T)$ were analyzed in the framework of a narrow-band model, which provides a satisfactory description to the observed data. Our present work thus provides an alternative interpretation in understanding the anomalous thermoelectric properties of the Na_xCoO_2 system.

ACKNOWLEDGMENTS

The authors would like to thank the National Science Council of Taiwan for financially supporting this research under Contracts No. NSC-96-2112-M-259-003 (Y.K.K.) and No. NSC-95-2112-M-002 (F.C.C.).

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