Transport Properties of Strongly Correlated Material $Na_{0.75}CoO_2$

Prosenjit Sarkar *

Department of Physics, Serampore College, Serampore 712201, India.

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Abstract

The transport properties of strongly correlated material $Na_{0.75}CoO_2$ has been studied. Both resistivity and thermoelectric power increase with temperature. The change in the slope of resistivity curve at around 20 K is associated with the magnetic transition. In thermopower, there is a kink at around 290 K possibly due to a second-order structural phase transition. The value of thermopower ($\sim$ 139.4 V/K) and resistivity ($\sim$2mΩ-cm) at 300 K are good enough for the material to be considered as a good thermoelectric material.
Keywords : Strongly correlated system, thermoelectric power, resistivity.

1. Introduction

A thermoelectric device is a device that converts heat into electric energy through the thermoelectric power (S) of solids. These kinds of devices can be very much efficient, since there is no theoretical upper limit of S. Another important factor is that the lifetime of the device can be as long as the lifetime of the material from which the device is made. More importantly the device is environmentally friendly in the sense that it produces no waste matter. To become a good thermoelectric material large thermoelectric power and low resistivity (ρ) are required. In fact, materials with figure of merit ($Z = S^2/\rho\kappa$), reaching a value close to unity, are considered as good thermoelectric materials, where κ is the thermal conductivity. Sometimes power factor (S^2/ρ) is frequently used to represent the quality of a thermoelectric material[1;2]. For good thermoelectric material power factor should be large. The strongly correlated compound Na_xCoO_2 has been studied extensively due to its good thermoelectric response[3;4;5;6]. In general, the oxides are thought to be poor thermoelectric materials because of their low electrical conductivity but the compound Na_xCoO_2 proved it otherwise. The magnetic structure of the compound is reported to be complex and the exact arrangement of the Co spins is yet unclear. Na_xCoO_2 became the focus of interest due to the appearance of superconductivity with deintercalation of Na and inclusion of water in the parent structure, resulting in $Na_{0.35}CoO_2 : 3H_2O$ with a superconducting transition temperature of around 4.5 K[7;8]. Here we report the transport

*Email babu2jit@yahoo.com

properties of $Na_{0.75}CoO_2$. We have measured the resistivity and thermoelectric power of the sample as a function of temperature (T) and the behavior is consistent to those reported in the literature[3;4;5;6;7;8].

2. Experimental Details

The sample $Na_{0.75}CoO_2$ was synthesized by a solid-state reaction route by using Na_2CO_3 and Co_3O_4 in stoichiometric proportions. The heating procedure was basically *rapid heat-up* technique in which the mixed powders were put directly in a preheated furnace at 750^0C and fired for 24 h. The heated powders were reground and calcined at 850^0C for another day. In this way we have prepared polycrystalline material. But in polycrystalline materials, the properties of the grain boundaries often manifest themselves stronger than the properties of the material itself. So it is crucial to obtain good quality single crystal for transport measurement. We have tried to make single crystal of that sample by using optical floating zone furnace. Polycrystalline sample was shaped into cylindrical bars of $\sim 5 \times 100$ mm by pressing at an isostatic pressure of ~ 5 Ton and then sintered at 850^0C for 1 day in flowing oxygen to form feed and seed rods. The sintered feed and seed rods were premelted by traveling the upper and lower shafts in synchronized mode at a velocity 27 mm/h to densify the feed rod under pure oxygen flow of 200 ml/min. Principle of the Floating Zone crystallization method is shown in Fig. 1. Here a molten zone is formed and held between two solid rods by its own surface tension.

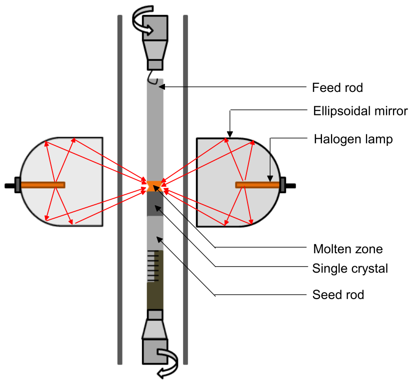

Figure 1: Schematic diagram of the floating zone technique.

Once a small section of the rod has been melted, the molten (floating) zone is translated along the sample length by moving the material with respect to the radiation focus with the help of a stepper motor. Crystal material is grown on the solidifying end of the float zone. Large crystals can be grown by this technique. In this process of premelting, considerable difficulties appear on account of the high Na_2O vapor pressure, followed by a noticeable evaporation as reported earlier. So we could not able to grow the prefect single crystal. From this melting behavior described so far it indicates that the compound melts *incongruently*. Basically incongruent melting is the temperature at which one solid phase transforms to another solid phase and a liquid phase of different chemical compositions than the original composition. In this case the compound decomposes into a sodium-rich liquid and a cobalt-rich solid phase. WE have measured the transport properties in polycrystalline sample. For that purpose we made a circular pellet. The pellet was finally fired at 850^0C for 24 h, before cooling it slowly to room temperature over a span of 10 h in flowing oxygen. Resistivity measurements were carried out by the conventional four-probe method. Thermoelectric power (TEP) measurements were carried out by dc differential technique over a temperature range 80−300 K, using a homemade setup. The temperature gradient of ∼1 K was maintained through out the TEP measurements.

3. Results and Discussions

The temperature dependence of resistivity of $Na_{0.75}CoO_2$ sample are shown in Fig. 2.

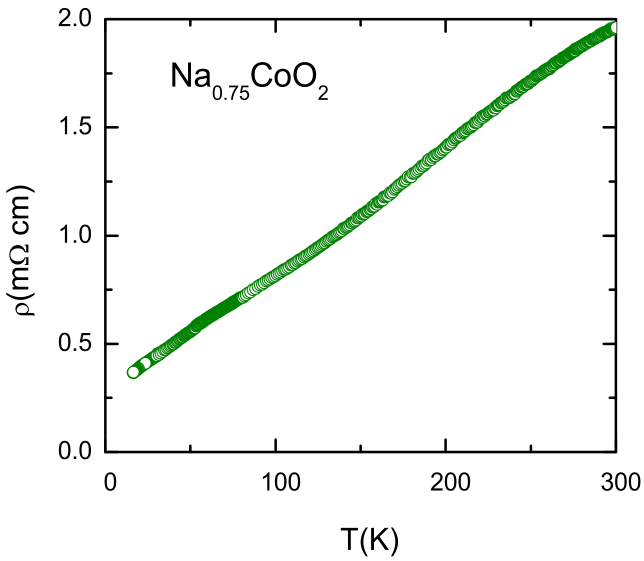

Figure 2: Temperature dependence of resistivity of $Na_{0.75}CoO_2$.

Sample exhibit a metallic behavior down to ∼15 K with room temperature (∼300 K) resistivity nearly equal to 2 mΩ-cm. Though the sample exhibits qualitatively the metallic-type of conduction, quantitatively there are a couple of finer interesting differences. For example, sample exhibits a constant $d\rho/dT$ in the temperature ranges of 50-300 K. Below 50K the $d\rho/dT$ value is increased, demonstrating changed conduction process below this temperature. Basically the change of the slope of resistivity curve represents the magnetic transitions around the temperature ∼ 20 K. If we could measure the resistivity up to 3-4 K then the exact transition temperature would be very much prominent. From magnetization measurements some groups have already reported that there is a magnetic transition around 21 K.

The thermoelectric power verses temperature plot for $Na_{0.75}CoO_2$ is shown in Fig. 3.

The magnitude of S is 139.4 V/K at 300 K. Thermopower increases with temperature but there is a kink at around 290 K possibly due to a second-order structural phase transition. For $Na_{0.75}CoO_2$, the values of thermopower (∼ 139.4 V/K) and resistivity (∼2mΩ-cm) at 300 K are good enough for the material to be considered as a good thermoelectric material, with further scope of optimization. The value of power factor S^2/ρ for the present sample at 300 K is 0.97×10^{-13} $V^2/K^2\Omega$-cm. Normally TEP is not very sensitive to grain boundaries effect in oxide materials. On the other hand resistivity is extremely sensitive to the presence grain boundary. In most of the cases, including sodium cobaltates, resistivity in single crystals is an order of magnitude less than that for polycrystalline samples. So if one can grow single crystals then the ratio S^2/ρ would be very high.

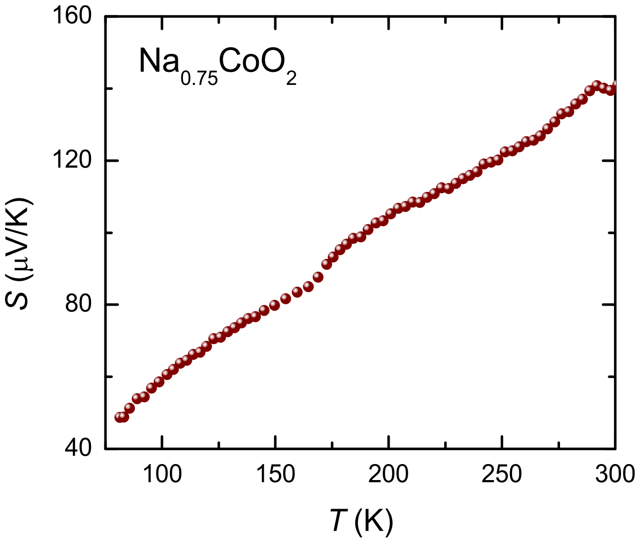

Figure 3: Thermoelectric power (S) vs. temperature (T) for $Na_{0.75}CoO_2$.

4. Conclusions

We have studied the transport properties of $Na_{0.75}CoO_2$. With increasing temperature, resistivity increases almost linearly but at around 20 K the slope of the resistivity curve changes. The change in the slope of resistivity curve is associated with the magnetic transition. Similarly, a kink in the thermopower curve at around 290 K is possibly due to a second-order structural phase transition. The values of resistivity and thermopower indicate that the material can be considered as a good thermoelectric material.

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