

# Electrical transport study of metallic delafossites by tuning thickness and dopant concentration

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## Abstract

**The metallic delafossites have a layered triangular structure and high in-plane conductivity. By comparing two delafossites PdCoO<sub>2</sub> and PdCrO<sub>2</sub>, the research aims to understand the special antiferromagnetic (AFM) ordering of PdCrO<sub>2</sub> and its correlation with Molecular Beam Epitaxy (MBE)-grown film thicknesses. Through Resistivity versus temperature measurements using the Quantum Design Physical Property Measurement System (PPMS), the phase transition resulted from AFM ordering in PdCrO<sub>2</sub> films was determined for thicknesses ranging from 3 unit cells to 20 unit cells. Our future direction is on creating potential AFM-order delafossite material by doping Ni ions to PdCoO<sub>2</sub> and assessing its AFM properties by transport measurements.**

## Introduction

Delafossites are heterostructures of layered oxides with the chemical formula ABO<sub>2</sub> (A: Pt, Pd... and B: Co, Cr...). They are renowned for their exceptional conductivity and long mean free path. A notable example is the metallic delafossite compound PdCoO<sub>2</sub>, which exhibits an ultra-low room temperature (300 K) resistivity of 2.6 μΩ·cm, surpassing the conductivity of alkali metals[1]. PdCoO<sub>2</sub> has the highest conductivity per carrier and longest mean free path among all known oxides, reaching 20 μm at 4K for the best as-grown crystal[1]. In addition to high conductivity attributed to the metallic behavior of the Pd triangular lattice, the insulating Cr-O layer of PdCrO<sub>2</sub> has antiferromagnetic (AFM) order with spins from Cr electrons ordered into a non-collinear 120° structure induced by the

spin-3/2 state of Cr<sup>3+</sup>[1]. The combination of AFM (which is commonly associated with insulators) and the metallic conducting behavior of PdCrO<sub>2</sub> makes it intriguing to explore, especially for its potential applications in memory devices and spintronics. Our interest in this AFM-metallic behavior combination has led to our ultimate project goal of creating new AFM delafossite materials with good conductivity. The first part of the project involves investigating the electronic properties of PdCrO<sub>2</sub> with thickness dependence to further enhance our understanding in AFM of this compound. By comparing PdCoO<sub>2</sub> and PdCrO<sub>2</sub>, it was found that the unpaired electrons in the Cr ions lead to AFM frustration, whereas the Co ions have paired electrons resulting in a zero spin state. Since the AFM non-collinear spin directions are not only in-plane but also

out-of-plane, the AFM properties are estimated to vary with  $\text{CrO}_2$  layer thicknesses. Therefore, the first part of the project aims to understand the correlation between  $\text{PdCrO}_2$  thicknesses and its AFM ordering. To create a new AFM metal, a dopant with unpaired electrons can be added to  $\text{PdCoO}_2$ , with Ni chosen as a suitable candidate. The second part of the project aims to study a potential AFM metal by growing samples of Ni-doped  $\text{PdNi}_x\text{Co}_{1-x}\text{O}_2$  with varying dopant levels ( $x = 5\%, 10\%, 15\%, 20\%, 33\%$ ) through MBE.

## Method

The thickness-dependence of AFM will be studied through transport measurement, comparing the resistivity-temperature measurement results of  $\text{PdCrO}_2$  samples with a thickness range from 3 unit cells to 20 unit cells. Upon the successful growth of  $\text{PdNi}_x\text{Co}_{1-x}\text{O}_2$ , the second part of my project aims to compare the resistivity-temperature dependence of this newly created metallic delafossite with different Ni dopant levels and study the AFM behavior by determining the Néel transition temperature. To perform transport measurements, the samples will be wire-bonded following the four-point geometry Van der Pauw method. The resistance-temperature measurements will be carried out using the Quantum Design Physical Property Measurement System (PPMS) over a temperature range from room temperature (300 K) down to 2.5 K.

## Results and Discussion

The measured resistance versus temperature raw data was converted into resistivity versus temperature data using the equation

$R_{sheet\ 2D} = \frac{\pi}{\ln 2} R_{measured}$  [2] to convert measured resistance to sheet resistance, then resistivity is obtained by multiplying sheet resistance with each sample film thickness. The resistivity versus temperature plot shows the trend that as  $\text{PdCrO}_2$  film thickness increases the resistivity decreases. Notably, the thinner films such as the 3-unit-cell and 4-unit-cell films have an insulating behavior whereas the thicker films have a metallic behavior and the phase transition associated with AFM-ordering.

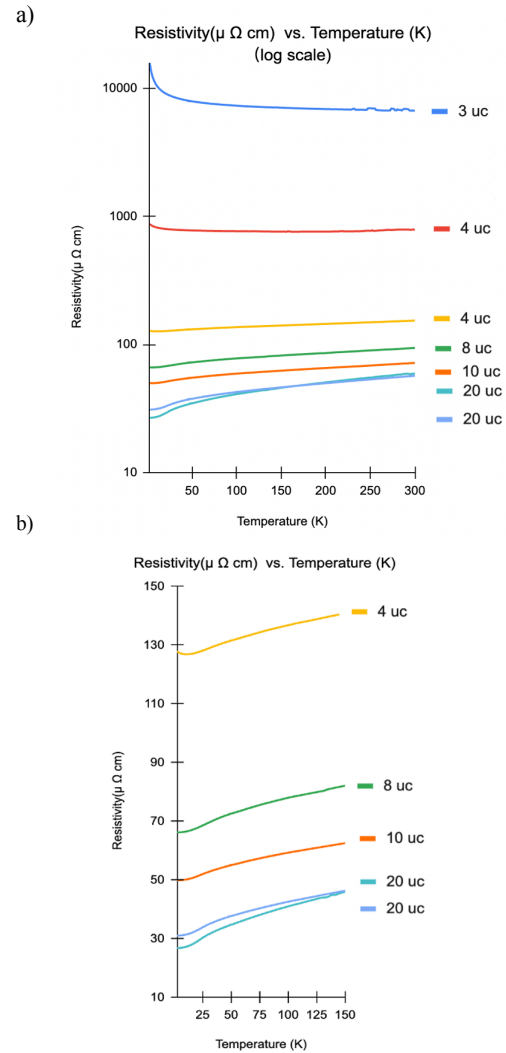
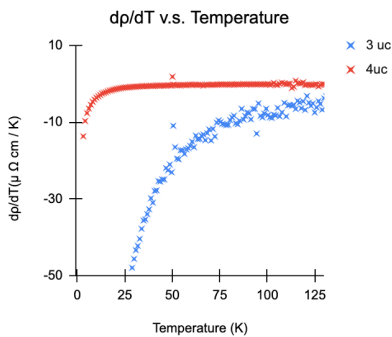


Figure 1. Resistivity versus Temperature plots. (a) For  $\text{PdCrO}_2$  thicknesses 3, 4, 8, 10, 20 unit cells and temperature range from 0

to 300K. (b) For PdCrO<sub>2</sub> films with AFM-orderings and temperature range from 0 to 100K.

By plotting the first derivative of the resistivity versus temperature plot, one can clearly see the magnetic transition as indicated by each peak for the thicker films. The insulating thin films without the magnetic ordering do not have any magnetic transition and a corresponding peak, as shown in Figure 2.

a)



b)

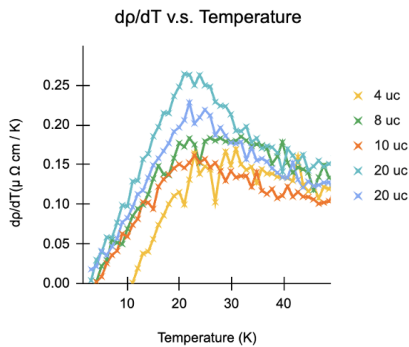


Figure 2. The first derivative of Resistivity versus Temperature plots. a) For PdCrO<sub>2</sub> thicknesses 3 and 4 unit cells and a temperature range from 0 to 125K. b) For PdCrO<sub>2</sub> thicknesses 4, 8, 10, and 20 unit cells and a temperature range from 0 to 40K.

The transition temperature is determined by Gaussian fitting to each peak in Figure 2 b). Figure 3 shows the transition temperature versus film thickness plot and an inverse relationship is observed. This decreasing trend of transition temperature as thickness increases is against the expectation that as the thickness of PdCrO<sub>2</sub> samples increases, the transition temperature would increase

and approach the bulk single crystal transition temperature, which is at 37K.

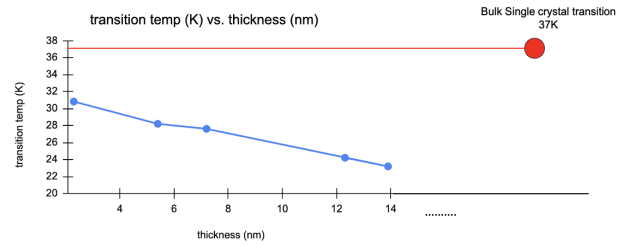


Figure 3. The transition temperature of PdCrO<sub>2</sub> films versus thickness plot.

The trend comes from only 5 data sets, which suggests the limitation in our scope of sampling. Thus, more PPMS data collection is needed in order to clearly make a conclusion on the behavior of AFM-order transition temperature versus the thickness of PdCrO<sub>2</sub> samples

## Conclusions and Future Work

The resistivity versus temperature of 7 PdCrO<sub>2</sub> samples were successfully collected, suggesting insulating behavior for ultrathin samples and AFM-metallic behavior for the thick samples. A decreasing trend for the AFM-order transition temperature versus thickness is observed and will be further verified through additional PPMS measurements on other PdCrO<sub>2</sub> samples. In the future, PPMS measurements will be conducted on the newly created Ni-doped metallic delafossite compounds to provide insights into their electrical transport properties and AFM behaviors.

## References

- [1] A P Mackenzie 2017 Rep. Prog. Phys. 80 032501
- [2] I Miccoli et al 2015 J. Phys.: Condens. Matter 27 223201