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We report the electrical, magnetotransport, and specific heat of the layered polycrystalline RECoPO (RE = La, Nd, and Sm) samples. These compounds are iso-structural to recently discovered superconductor LaFeAs(O/F). Bulk polycrystalline samples are synthesized by a solid state reaction route in an evacuated sealed quartz tube. All these compounds are crystallized in a tetragonal structure with space group *P4/nmm*. Cobalt in these compounds is in an itinerant state, with its paramagnetic moment above 1.4 μ_B and the same orders ferromagnetically (FM) with saturation moment of around 0.20 μ_B below, say, 80 K. Though, LaCoPO shows single paramagnetic (PM) to ferromagnetic (FM) transition near 35 K, the NdCoPO and SmCoPO exhibit successive PM-FM-anti-ferromagnetic (AFM) transitions. Both FM and AFM transition temperatures vary with applied fields. Although the itinerant ferromagnetism occurs with small saturation moment, typical anti-ferromagnetic (AFM) transitions (T_{N1}, T_{N2}) are observed at 60 K and 14 K for Nd and 70 K and 40 K for Sm. This FM-AFM transition of Co spins in NdCoPO and SmCoPO is both field and temperature dependent. The magneto-transport of NdCoPO and SmCoPO distinctly follows their successive PM-FM-AFM transitions. It is clear that Sm/Nd (4f) interacts with the Co (3d) in first-time-synthesized Sm/NdCoPO. © *2011 American Institute of Physics*. [doi:10.1063/1.3662151]

I. INTRODUCTION

The quaternary equiatomic ZrCuSiAs structure type RE(Fe/Co/Ru)PnO (RE = rare-earth metal and Pn = pnictogen-like P, As, Sb, and Bi) compounds were known decades before in their non-superconducting pristine form[1,2] and hardly could attract attention. However, the situation changed dramatically after the discovery of superconductivity in a doped oxy-pnictides compound, REFePO[3] and, in particular, the REFeAsO.[4] Due to the renewed interest in these compounds, their synthesis and structural properties were reviewed more recently.[5,6] Basically, superconductivity is introduced in these compounds by carrier doping in superconducting Fe-As/Fe-P layers, and, therefore, the role of 3d metal is important. Substitution of Co^{3+} at an Fe^{2+} site effectively dopes electrons in FeAs and brings about the superconductivity for around 10 to 20% of the doping level at close to 15 K.[7–9] On the other hand, full substitution of Co^{3+} at Fe^{2+} in REFeAsO results in a non-superconducting state. In fact, not only the superconducting state, but also undoped non-superconducting oxy-pnictides show interesting behavior and, hence, their study may be helpful to understand the mechanism of superconductivity in this type of compound. The iso-structural, cobalt-based compound, i.e., RECoAsO does not show superconductivity, but exhibits

itinerant ferromagnetism[10–12] and interaction of RE^{4f} and Co^{3d} moments.[13–16] Spin fluctuations play an important role in the magnetic properties of RECoAsO compounds and, hence, possibly in the iron-based superconductors having similar structure as well.[15] The iso-structural LaFePO and LaNiPO are non-magnetic, possibly due to the strongly correlated electrons being present in their un-doped form. In both of these compounds, 3d metal magnetic ordering is suppressed, due to the reduction of their magnetic moments.[17,18] Interestingly, in the case of LaCoPO, the magnetic moment does not disappear completely, because Co has an odd number of electrons in the 3d orbital. LaCoPO and CeCoPO exhibit ferromagnetic transitions near 43 K and 75 K, respectively, in magnetization measurement (M-T) under a 1 kOe magnetic field.[19,20]

Needless to say, the Co-based ground state of pnictides is intriguing and complex and, hence, need to be probed. In this direction, though the RECoAsO is widely investigated[10–16] for both non-magnetic (La) and magnetic (Nd, Sm) RE, the RECoPO, on the other hand, is studied only in case of non-magnetic La, i.e., LaCoPO.[19] Feeling the necessity to check if the RE^{4f} and Co^{3d} moments interact in RECoPO, we synthesized and studied RECoPO with both non-magnetic (La) and magnetic (Nd, Sm) rare earths. We present a comparative study of structural, electrical, magnetization, magneto-transport, and heat capacity of RECoPO (RE = La, Nd, and Sm). To our knowledge, the magnetic (Nd, Sm)-based RECoPO samples are synthesized and studied for the first

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time. Both the magnetization and magneto-transport properties change drastically when non-magnetic La is replaced by magnetic Sm and Nd.

II. EXPERIMENTAL

All the studied polycrystalline samples of RECoPO (RE = La, Nd, and Sm) are prepared through a single-step, solid-state reaction route via vacuum encapsulation technique.[8,12,14] High purity (∼99.9%) rare earths (La, Nd, Sm), P, Co_3O_4, and Co in their stoichiometric amount are weighed, mixed, and ground thoroughly using mortar and pestle under a high purity Ar atmosphere in a glovebox. The humidity and oxygen content in the glovebox is less than 1 ppm. The mixed powders were palletized and vacuum-sealed (10^{-4} Torr) in a quartz tube. These sealed quartz ampoules were placed in a box furnace and heat treated at 550 °C for 12 h, 850 °C for 12 h, and then at 1150 °C for 33 h in continuum. Finally, the furnace is allowed to cool down to room temperature at a rate of 1 °C/minute.

The x ray diffraction patterns of the studied sample were taken on a Rigaku x ray diffractometer with Cu K_α radiation. All physical property measurements, including magnetization, magneto-transport, and heat capacity, are carried out on Quantum Design physical property measurement system (PPMS) with fields up to 14 T.

III. RESULTS AND DISCUSSION

The room temperature Rietveld-fitted XRD patterns of LaCoPO, NdCoPO, and SmCoPO samples are shown in Fig. 1. The Rietveld analysis of x ray pattern confirmed that all the studied samples are crystallised in the tetragonal phase with a *P4/nmm* space group, in analogy of ZrCuSiAs-type structure. All the main peaks of the x ray pattern are well indexed, with a space group *P4/nmm*. The studied samples are phase pure and good enough for physical properties measurements, having only very minute rare earth oxide impurity, which are marked on the XRD pattern with * in Fig. 1. The Rietveld-refined parameters are in good agreement with earlier reports on similar Co-based similar as-based oxy-pnictides.[10–16] The lattice parameters *a*, *c*,
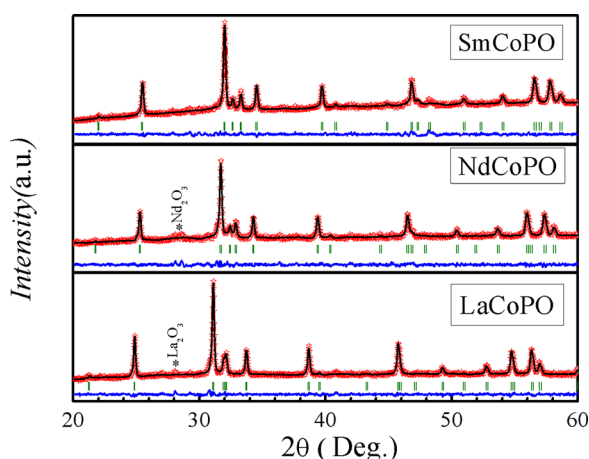

FIG. 1. (Color online) Observed and Rietveld-fitted room temperature XRD patterns of LaCoPO, NdCoPO, and SmCoPO.

TABLE I. Wyckoff position for RECoPO (space group: *P4/nmm*).

Atom	Site	x	y	z
RE	2c	1/4	1/4	z
Co	2b	3/4	1/4	1/2
P	2c	1/4	1/4	z
O	2a	3/4	1/4	0

and volume are 3.968(7)Å, 8.368(4)Å, and 131.80(5)Å³; 3.904(2)Å, 8.182(2)Å, and 124.71(9)Å³; and 3.877(4)Å, 8.072(9)Å, and 121.36(9)Å³ for RE = La, Nd, and Sm samples, respectively. The lattice parameters follow the rare earth contraction, evidencing successful replacement of rare earth metals within the same structure. The *z* positions of RE and P are 0.1503(2) and 0.6285(4), 0.1481(2) and 0.6492(4), and 0.1451(5) and 0.6439(6) for LaCoPO, NdCoPO, and SmCoPO, respectively. The Wyckoff positions of RECoPO samples are given in Table I. The details of Rietveld-refined structural and fitting parameters for all the studies' samples are given in Table II. It is clear, from Fig. 1 and Table II, that these samples are good enough for studying their physical properties.

The measured magnetic properties of RECoPO samples are shown in Figs. 2(a), 2(b), and 2(c). Though LaCoPO shows only single paramagnetic (PM) to ferromagnetic (FM) transition at around 100 K, the NdCoPO and SmCoPO both exhibit successive PM-FM-anti-ferromagnetic (AFM) transitions. The PM state moment of Co in these samples is itinerant and with effective paramagnetic moment of 1.5 μ_B, irrespective of the RE. Further, the FM saturation moment is also the same and close to 0.20 μ_B. This is similar to that as observed recently for RECoAsO.[10–16] An interesting fact to be noted in these systems is that PM-itinerant and FM-saturated moments for Co are the same, irrespective of RE in RECoPO. The non-magnetic, La-based compound undergoes only PM-FM transition, whereas (Nd, Sm) exhibit successive PM-FM-AFM transitions with a decrease in temperature.

To elucidate upon the temperature-dependent magnetic behavior of RECoPO, Fig. 2(a) shows the magnetization versus temperature plots for LaCoPO at various fields of 10 Oe, 500 Oe, and 1000 Oe. The compound orders ferromagnetically (FM) with Curie temperature (T_c) near about 35 K at the 10 Oe field. T_c is field dependent and increases significantly with increasing field, which is common for FM transition. The upper inset of Fig. 2(a) depicts the isothermal MH plots at 10, 50, 100, and 200 K, with magnetic field up to 30 kOe. At 5 and 25 K, the magnetization of the compound saturates above 2 kOe, but as the temperature increases to 40 K, it shows a hump near 10 kOe, with a gradual change, shown in the lower inset of Fig. 2(a). This

TABLE II. Reitveld-refined parameters for RECoPO.

RE	a(Å)	c(Å)	Volume (Å³)	R_P	R_{WP}	χ^2
La	3.968(7)	8.368(3)	131.80(5)	2.74	3.84	2.12
Nd	3.904(2)	8.182(2)	124.71(9)	2.45	3.16	1.16
Sm	3.877(4)	8.072(9)	121.36(9)	1.99	2.54	1.19

gradual change at 40 K is due to the competition between the FM and PM state; below this temperature, the FM state is dominant. At higher temperatures (100 and 200 K), the PM state is dominant, and small liner magnetization is seen. The saturation moment in the FM state is around 0.30 μ_B/Co.

Figure 2(b) depicts the temperature-dependent magnetization plots for NdCoPO at various magnetic fields from 10 Oe to 50 kOe. In low applied magnetic fields, NdCoPO shows a paramagnetic state from 250 K to 60 K and an abrupt increase in the magnetization at 60 K, indicating ferromagnetic (FM) ordering. The magnetization saturates below 60 K and remains nearly the same down to 20 K. Below 20 K, the magnetization is sharply decreased, sug-

gesting an anti-ferromagnetic (AFM) transition. This situation is the same for low applied fields of up to 1 kOe and change slightly at a higher field of 10 kOe. The successive FM-AFM transition is nearly disappearing at 50 kOe field; at this field, the magnetization is like a PM, but not exactly. The ordered magnetic moment under higher magnetic field is canted, and it shows the paramagnet (PM)-like behavior. It seems the FM-AFM transition temperature goes down with applied field. The isothermal MH plots of NdCoPO are shown in the inset of Fig. 2(b), which are linear at 100 K (PM state), slightly off axis in AFM (5 K and 10 K) state, and are FM-like between 20-50 K, with small opening of the loops. Unlike as in LaCoPO, the FM plots are not saturated, due to the contribution from trivalent magnetic Nd ions along with the ordered Co spin's contribution to the total moment in the NdCoPO. It is clear, from Fig. 2(b), that the magnetization of NdCoPO exhibits successive PM-FM-AFM transitions with temperature.

The MT (magnetization versus temperature) plots of SmCoPO in applied fields from 100 Oe to 30 kOe are given in Fig. 2(c). SmCoPO shows successive PM-FM-AFM transitions with lowering of the temperature, being similar to that as in the case of NdCoPO. However, there are few differences, i.e., the FM regime in SmCoPO (65 K-45 K) is much narrower than as for NdCoPO (60 K-20 K) under an applied field of 500 Oe. The saturated region of the FM state in SmCoPO disappears fast in comparison to NdCoPO with increasing magnetic field. On the other hand, the successive FM-AFM transition is not disappearing, even up to 140 kOe, which completely disappears at the 50 kOe field in NdCoPO. The transition temperature of FM-AFM is field dependent; as we increase the field, the same is shifted to lower temperature. The AFM transition temperature is about 45 K in an applied field of 100 Oe and the same comes down to below 6 K with an increase in field to 140 kOe. The MH plots of SmCoPO are shown in the inset of Fig. 2(c), which are linear in PM (200 K, 100 K) and AFM states (2.5 K, 5 K). At intermediate temperatures, i.e., 20 and 50 K, the MH are FM-like. Further, at 20 K, the magnetization under increasing field first increases slowly until 20 kOe and later saturates. At this temperature, the compound seems to be undergone in the competing AFM–FM transformation. At lower fields, magnetic moments are not fully aligned, and as the field increases above 20 kOe, the canted magnetic moments became aligned in one direction, and a FM-like saturation is seen.

The temperature-dependent reciprocal magnetic susceptibility of RECoPO is linear from 250 K to 150 K and curved below this temperature (not shown) in the FM regime, but rise in the AFM state. The Curie-Weiss fitting of the MT data between 250 and 150 K gives the effective paramagnetic moments to be 1.4 μ_B, 3.58 μ_B, and 1.7 μ_B for the LaCoPO, NdCoPO, and SmCoPO, respectively. These results are in agreement with similar RECoAsO compounds.[10] Co in the itinerant state may not be contributing to the magnetic moment, but Ohta *et al.*[11] showed that Co contributes to the total effective magnetic moment. This seems to be the situation for our RECoPO samples as well. Further, it seems that the Sm^{3+} ion effective moment is decreased in SmCoPO (1.7 μ_B), but not with Nd^{3+} in

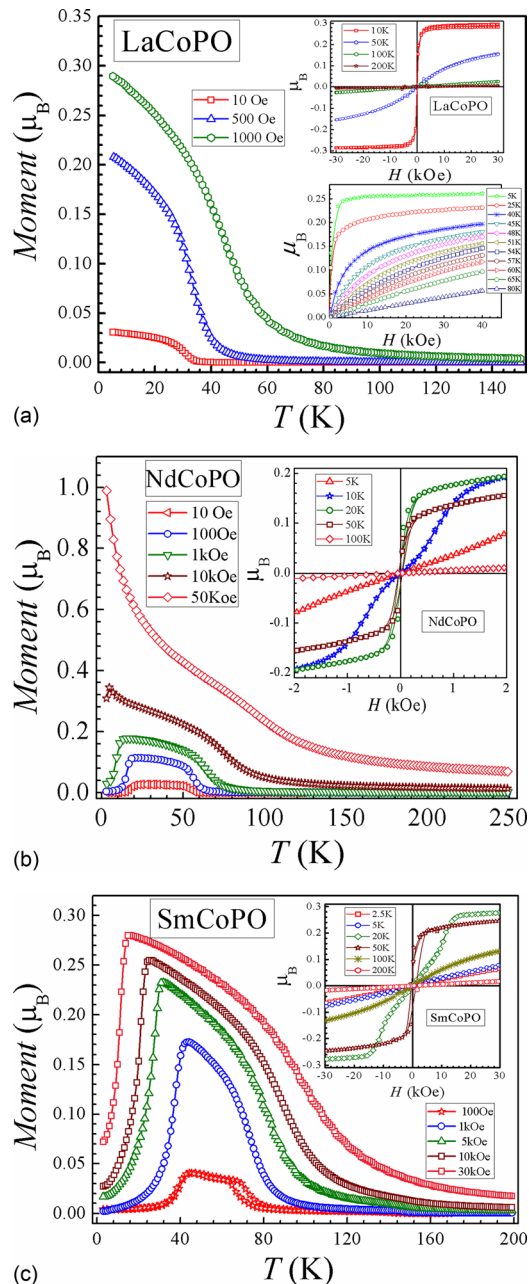

FIG. 2. (Color online) Magnetization (M) vs temperature (T) for (a) LaCoPO, (b) NdCoPO, and (c) SmCoPO at different fields. Respective insets show the isothermal magnetization $M(H)$ plots of these compounds at various temperatures.

NdCoAsO (3.58 μ_B). The total effective paramagnetic moment of the RECoPO in the PM state is related as $P_{eff}^2 = P_{RE}^2 + P_{Co}^2$, where P_{RE} is the effective movement of the rare-earth ion and P_{Co} is the effective movement of the cobalt ion. It is needless to state that the magnetic structure by neutron scattering experiments of the first-time-synthesized NdCoPO and SmCoPO is very much warranted, and that could only be able to effectively resolve the complex magnetism of RE(Nd/Sm) and Co in these pnictides oxy-phosphates.

The temperature-dependent resistivity of LaCoPO, NdCoPO, and SmCoPO at various applied magnetic fields are shown in Figs. 3(a), 3(b), and 3(c) respectively. The $\rho(T)$
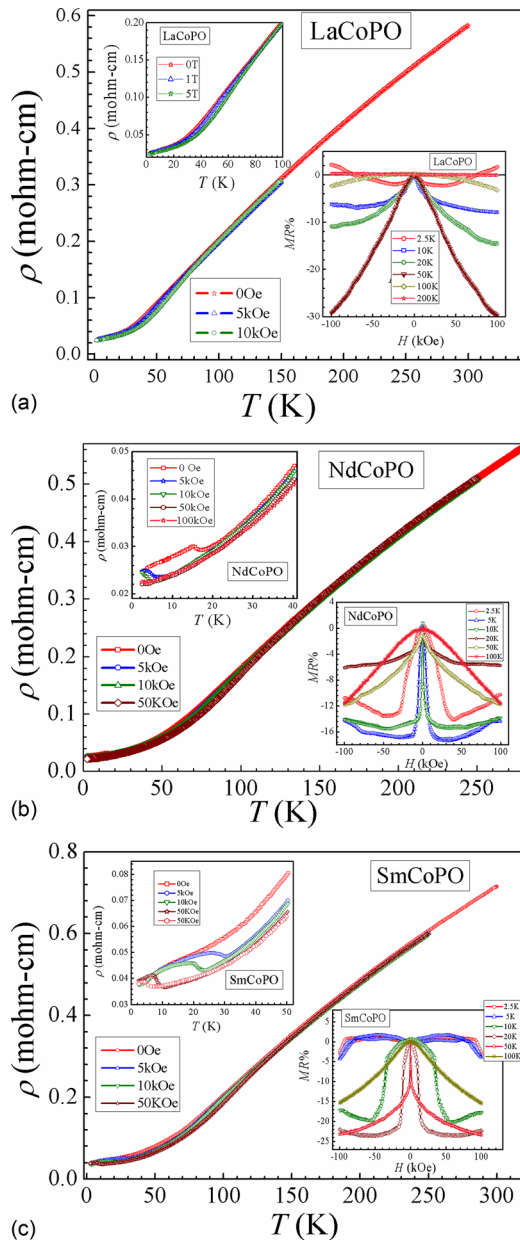

FIG. 3. (Color online) Resistivity (ρ) vs temperature (T) for (a) LaCoPO, (b) NdCoPO, and (c) SmCoPO at different fields from 300 down to 2.5 K. Upper insets show the expanded resistivity plots for respective samples. Lower insets show the isothermal magnetization plots (MR) at various temperature for respective samples.

behavior is metallic in the throughout measured (2-300 K) temperature range, with a slight increase in slope (improved metallic behavior) below, say, 50 K for all the samples. Below 50 K, the increase in conductivity could be related to the onset of the FM state in these compounds. The zero-field metallic resistivity ratios ($\rho^{300\,K}/\rho^{50\,K}$) are 6.45, 9.85, and 8.86 for La, Nd, and Sm, respectively, indicating them to be good conductors. Interestingly though, the zero field resistivity of LaCoPO and SmCoPO is metallic down to 2 K (upper inset in Figs. 3(a) and 3(c)) and the NdCoPO sample exhibits an upward (negative slope) sharp step at around 18 K (upper inset in Fig. 3(b)). The zero-field $\rho(T)$ behavior of these samples is quite similar to that as observed for RECoAsO (RE = La, Nd, and Sm).[13–15] The low temperature (18 K) step is observed nearly at the same temperature for NdCoAsO as well and is related to the complex AFM ordering of Nd being mediated by interacting Nd and Co moments.[10,14] The RE^{4f} and Co^{3d} moments' interaction[21] is certainly taking place for Sm and Nd in RECoAsO/RECoPO. A debatable question arises that, why, in the same structure either in the case of SmCoAsO[14] or in presently studied SmCoPO, the interacting Sm and Co moments do not give rise to unusual ordering of Sm at higher temperatures, the same as for Nd at around 16 K in NdCoAsO[9,13] or at 18 K in NdCoPO. One possible reason seems to be that it could be the partial ceasing of Sm moments in the paramagnetic state for SmCoAsO and SmCoPO. Also worth noting is the fact that low-temperature, free-ion Nd and Sm ordering appears at the right temperatures of 1.6 K and 4.5 K for both NdCoAsO[11,13]/NdCoPO and SmCoAsO[12]/SmCoPO, respectively. The free-ion Nd and Sm ordering for NdCoPO and SmCoPO will be further discussed in C_p results in the next paragraphs.

The resistivity versus temperature with magnetic-field, i.e., $\rho(T)H$ plots for LaCoPO at various applied fields of 0, 10, and 50 kOe, are shown in Fig. 3(a). As discussed above, LaCoPO shows metallic resistive behavior in the temperature range from 2.5 K to 300 K. Below the Curie temperature (T_c), reasonable magneto-resistance (MR) appears, reaching its maximum value at around 50 K. The isothermal magneto-resistivity, i.e., MR of LaCoPO at different temperatures, is shown in the lower inset of Fig. 3(a). The maximum MR% is $\sim -30\%$ at 50 K in the 100 kOe field, which is about 2.5 times in comparison to LaCoAsO. MR% has a cusp-like shape around 50 K, and it is decreased at higher and lower temperatures. The maximum MR% is –2% and –10% at 100 K and 20 K, respectively. In the paramagnetic state (200 K), the MR% is expectedly negligible. This is natural, as the MR% is maximum when the magnetic phase transition is taking place, due to magnetic phase separation[21] and the same is negligible in pure PM (200 K) and saturated FM (10, 5 and 2 K) states.[15]

The $\rho(T)H$ plots for NdCoPO at various applied fields are shown in Fig. 3(b). NdCoPO exhibits similar metallic behavior as LaCoPO, except the anomalous upturn step discussed in the previous paragraphs. The anomalous upturn $\rho(T)$ step temperature of NdCoPO decreases with increasing magnetic field (see the upper inset of Fig. 3(a)), similar to that observed earlier[9,13] for NdCoAsO. The step-like

transition is precisely attributed to the ordering of Nd^{4f} hybridizing with Co^{3d} moments.[9,13] The isothermal magneto-resistivity, i.e., $\rho(T)H$ with varying magnetic field at various temperatures for NdCoPO, is depicted in the lower inset of Fig. 3(b). The MR% is at maximum in the AFM-FM transformation region and least in both PM and saturated FM states. The same situation occurs at 2.5 K; at a higher field of 80 kOe, the maximum MR is nearly –12%. The unusual increase of MR at lower temperatures is possibly due to the Nd^{4f} moments ordering and their interplay with the FM-ordered Co^{3d} spins in adjacent Co-P layers. The increment of lower temperature MR% is not seen in LaCoPO, because La is non-magnetic and, hence, it does not influence the ordered Co spins.

Figure 3(c) exhibits the $\rho(T)$ plots for SmCoPO at various applied fields. No step-like transition is observed in zero-field resistivity of SmCoPO down to 2.5 K, as in NdCoPO. Interestingly, SmCoPO shows the same $\rho(T)$ step-like upward transition under magnetic field below 40 K [see the upper inset of Fig. 3(c)]. The step-like upward transition in $\rho(T)$ of NdCoPO is due to AFM ordering of Nd spins; the same transition in SmCoPO at applied fields indicates the possibility of Sm spins being ordered in these fields at high temperatures ($\sim$40 K). The step-like transition of SmCoPO under magnetic field is similar to that as for NdCoPO without field. It seems Sm^{4f} spins under field interact with Co^{3d} spins in a similar fashion as for Nd^{4f} without field. It is worth mentioning that the fact that Nd^{4f} and Co^{3d} interaction mediated higher T_N of 18 K for NdCoAsO is already reported from neutron diffraction.[9,13] The first-time-synthesized similar NdCoPO needs to be tested for its magnetic structure by neutron diffraction. Further, the SmCoAsO and SmCoPO are still to wait for cracking of their exact magnetic structure. The Nd^{4f} and Co^{3d} interaction-mediated higher T_N of 18 K for NdCoPO reminds one of the famous $PrBa_2Cu_3O_7$ high T_c cuprates situation, where the Pr spins-order AFM (T_N) are at unusual high temperature of 17 K.[22] The unusual high T_N of 17 K for Pr in $PrBa_2Cu_3O_7$ and its non-superconductivity are explained on the basis of extended Pr^{4f} hybridization with adjacent conducting Cu-O_2 planes.[23]

The isothermal MR data of SmCoPO in various applied fields up to 100 kOe at various temperatures is shown in the lower inset of Fig. 3(b). The MR% is lowest at low temperatures, i.e., in AFM (2.5 K, 5 K) states. The MR is increasing up to –22% as the temperature increases to 20 K at the applied field 10 kOe and saturates at a higher field. Below 50 K, MR% is increasing, due to the competition between FM-AFM and FM state dominates. As temperature goes below 20 K, the AFM state dominates and the MR% decreases. It is obvious from the shoulder hysteresis during increase/decrease of the field at 10 K.

The heat capacity of $C_P(T)$ of all the studied samples below 100 K are shown in Fig. 4. The expected Sm and Nd T_N (AFM) peaks are seen in $C_P(T)$ at 5.4 K and below 2 K in SmCoPO and NdCoPO, respectively. The Nd-Co mediated Nd unusual high T_N of around 18 K, observed earlier in $C_P(T)$ for NdCoAsO,[9,13] is also seen as a weak transition in $C_P(T)$ of NdCoPO, shown in Fig. 4. The C_P/T versus T plot changes first at around 80 K, which roughly coincides with
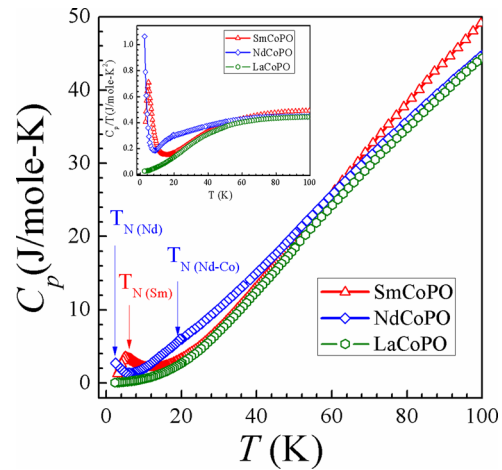

FIG. 4. (Color online) Heat capacity (C_P) vs temperature (T) for LaCoPO, NdCoPO, and SmCoPO at from 200 down to 2 K. Inset shows the expanded C_P/T vs T plots for same samples.

the FM ordering of Co spins. With further lowering of temperature, the C_P/T versus T slope change at around 20 K and 15 K for SmCoPo and NdCoPO, respectively; these temperatures roughly coincide with the complex AFM ordering of the Sm^{4f}-Co^{3d} and Nd^{4f}-Co^{3d} interplayed matrix. It seems the Sm/NdCoPO undergoes three magnetic transitions, i.e., $T_{c,Co}$ ($\sim$80 K); the Sm^{4f}-Co^{3d} and Nd^{4f}-Co^{3d} interplayed AFM below 20 K and finally Sm^{3+} and Nd^{3+} spins individual AFM at 5.4 K and below 2 K, respectively. Interestingly, the $T_{c,Co}$ ($\sim$80 K) and Sm^{4f}-Co^{3d} and Nd^{4f}-Co^{3d} interplayed AFM at low temperature are not seen as a distinct transition in $C_P(T)$ measurements. Only the distinct Sm^{3+} and Nd^{3+} spins individual AFM-ordering peaks are seen in $C_P(T)$ at 5.4 K and below 1.8 K. The $(Nd/Sm)^{4f}$ and Co^{3d}-mediated FM and AFM transition in (Nd/Sm)CoPO do not occur with a distinct change in entropy and a resulting $C_P(T)$ peak, but rather as a continuous change in entropy and, hence, no peak in heat capacity.

Summarily, we synthesized RECoPO (RE = La, Nd, and Sm) in a single phase and studied their complex magnetism and magneto-transport. Magnetic RE (Nd and Sm)-based RECoPO are synthesized and studied for the first time to our knowledge. Though LaCoPO shows a single paramagnetic (PM) to ferromagnetic (FM) transition near 35 K, the NdCoPO and SmCoPO exhibit successive PM-FM-AFM transitions. The FM-AFM transition for magnetic rare earths is mediated by $(Nd/Sm)^{4f}$ and Co^{3d} interacting moments in (Nd/Sm)CoPO. The transport measurements showed that all studied compounds are metallic in nature, and their magnetic ordering, in particular, the AFM, is clearly reflected in magneto-transport studies. NdCoPO shows an upward step-like transition in $\rho(T)$ measurement under zero field, but not for SmCoPO. The transition of Co spins from ferromagnetic (FM) to anti-ferromagnetic (AFM) in (Nd/Sm)CoPO is field dependent. The heat capacity $Cp(T)$ exhibits only the low temperature RE moments AFM ordering and, as such, the Co-mediated transitions (FM-AFM) are not seen, indicating a continuous change in entropy and, hence, no peak in heat capacity.

ACKNOWLEDGMENTS

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