



Rapid communication

Nickel-based layered superconductor, LaNiOAs

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ABSTRACT

Rietveld analysis of the powder X-ray diffraction of a new layered oxyarsenide, LaNiOAs, which was synthesized by solid-state reactions, revealed that LaNiOAs belongs to the tetragonal $ZrCuSiAs$ -type structure ($P4/nmm$) and is composed of alternating stacks of La–O and Ni–As layers. The electrical and magnetic measurements demonstrated that LaNiOAs exhibits a superconducting transition at 2.4 K, and above this, LaNiOAs shows metallic conduction and Pauli paramagnetism. The diamagnetic susceptibility measured at 1.8 K corresponded to ~20% of perfect diamagnetic susceptibility, substantiating that LaNiOAs is a bulk superconductor.

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1. Introduction

The discovery of high transition temperature (high- T_c) Cu-based superconducting oxides has led numerous efforts to explore higher T_c superconductors in a variety of materials, including Cu- and other transition metal-based oxides, because it is hypothesized that the strong electron correlation among 3d electrons plays an important role in Cu-based high- T_c superconductors. Since then, many Cu-based high- T_c superconductors have been discovered, including $HgBa_2Ca_2Cu_3O_{8+x}$ in 1993, which has T_c of 133 K [1]. On the other hand, T_c 's of other transition metal-based compounds (including oxides and pnictides) are lower than those of the Cu-based superconductors. However, the discoveries of superconductivity in new compounds such as Sr_2RuO_4 [2], $Na_xCoO_2 \cdot yH_2O$ [3], and a series of Ni-based layered borides [4–6] have provided complementary information to better understand the mechanism of superconductivity as well as clues to for exploring new material systems for higher T_c superconductors.

Recently, we have investigated a series of quaternary compounds, $LaMOPn$ (M : transition metal element such as Mn, Fe, Co, and Ni, Pn : pnictogen element such as P and As) with the expectation that this materials system will be a new correlated electron system. In this crystal structure, the La–O and M – Pn

layers are alternately stacked along the c -axis, as shown in Fig. 1. Systematic research on isostructural oxychalcogenides, $LaCuOCh$ ($Ch = S, Se, \text{ and } Te$), has indicated that the narrow energy gap Cu– Ch layer, which is sandwiched by larger energy gap La–O layers, works as a hole transport path [7]. Analogous to this, the M – Pn layer in $LaMOPn$ is also expected to be a carrier conduction path. This two-dimensional crystal structure with a transition metal element has led to the expectation that interesting electrical and/or magnetic properties, which originate from electron correlation, will be discovered.

In line with this strategy, we found new superconductors, $LaFeOPn$ ($Pn = P, As$) [8,9] and $LaNiOP$ [10], and an itinerant ferromagnet, $LaCoOPn$ ($Pn = P, As$) [11] in this materials system. Amongst them, $LaFeOAs$ exhibited extremely interesting behaviors: the temperature dependence of the resistivity of undoped $LaFeOAs$ showed a semiconductor-like behavior with a kink in resistivity at ~150 K and a superconducting transition did not occur, but its electrical property changed to a metallic behavior by F (electron) doping. Moreover, $LaFeOAs$ with a F content greater than or equal to ~4 at% showed superconducting transitions. Its transition temperature reached ~26 K around an F content of 11 at%. On the other hand, $LaFeOP$ and $LaNiOP$ appeared to be conventional superconductors with T_c 's lower than 5 K. Accordingly, the superconducting properties of $LaFeOPn$ were drastically altered by changing the Pn element from P to As. Thus, elucidating the electrical and magnetic properties of $LaNiOAs$ is important for systematic and comparative studies of the $LaFeOPn$ and $LaNiOPn$ systems. However, a study on the synthesis of $LaNiOAs$ has yet to be reported.

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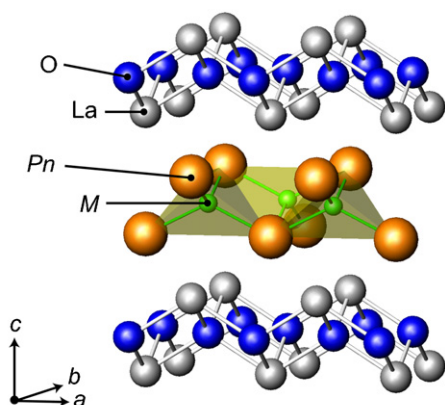


Fig. 1. Crystal structure of LaMOPn ($M = \text{Mn, Fe, Co, Ni, Zn}$, $\text{Pn} = \text{P, As}$). La–O and M–Pn layers are alternately stacked along the c -axis.

In this letter, we report the synthesis as well as the electrical and magnetic properties of LaNiOAs . LaNiOAs exhibits metallic conduction at high temperatures and a superconducting transition at 2.4 K without impurity doping, which contrasts the LaFeOPn case, suggesting that the semiconductor-like behavior in the undoped sample and the induction of the superconducting transition by F doping are characteristic of the LaFeOPn system. The present results along with those reported for the LaMOPn system would help elucidate the mechanism of superconductivity in this system.

2. Experimental

Polycrystalline samples of LaNiOAs were prepared by solid-state reactions of the starting materials, La (Shin-etsu Chemical, purity 99.5%), As (Kojundo Chemical Laboratory, 99.99%), and NiO (Kojundo Chemical Laboratory, 99.97%). Single-phase LaAs was initially synthesized by heating a stoichiometric mixture of La metal powder and As in an evacuated silica tube at 400 °C for 12 h and 700 °C for 6 h. Then a stoichiometric mixture of LaAs and NiO was pressed into a pellet and heated at 1000 °C for 1 day in an evacuated silica tube. The resulting sample was characterized by high-power X-ray diffraction (XRD, D8 ADVANCE-TXS, Bruker AXS) with $\text{CuK}\alpha$ radiation at 24 °C. Multi-phase powder X-ray Rietveld analyses were carried out using the code TOPAS3 [12] to refine the crystallographic parameters of LaNiOAs and to estimate weight fractions of LaNiOAs and the impurity phases. The initial parameters were taken from an isostructural compound, LaNiOP [10].

The electrical resistivity of the sintered pellets was measured in the temperature range of 1.8–305 K by a four-probe technique (using PPMS, Quantum Design). Ohmic contact electrodes were formed by Au sputtering and electrical connections were made using a silver paste. The magnetic properties were measured with a vibrating sample magnetometer (VSM, using PPMS, Quantum Design). The temperature dependence of the magnetic susceptibility was measured at temperatures from 1.8 to 305 K in a magnetic field of 10 Oe after zero-field cooling. The field dependence of magnetization was measured at 1.8 K.

3. Result and discussion

3.1. XRD and Rietveld analyses

The obtained samples were chemically stable in air and dark gray. Fig. 2 shows the XRD patterns measured (black circles) and

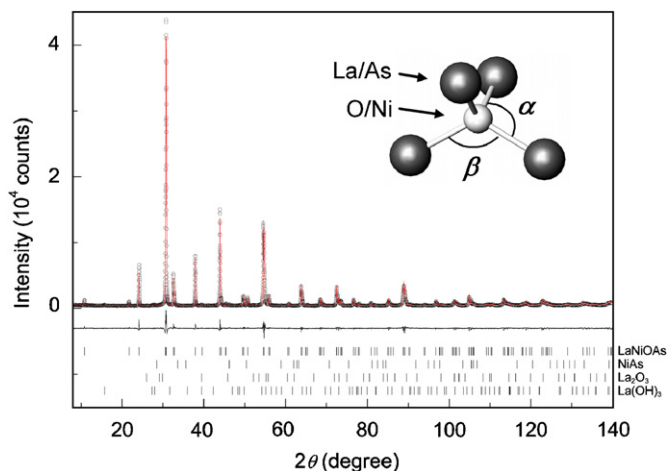


Fig. 2. XRD patterns of LaNiOAs measured (black circles) and simulated by Rietveld analysis (red lines). Line and the vertical bars below the XRD patterns show the difference profile between the observed and simulated XRD patterns, and the calculated positions of the Bragg diffractions of LaNiOAs , NiAs , La_2O_3 , and La(OH)_3 from top to bottom, respectively. The inset shows a structure of the NiAs_4 and OLa_4 tetrahedrons to identify the bond angles of α and β that are summarized in Table 1.

simulated by Rietveld analysis (red line) for the purest sample. Rietveld analyses revealed that the samples contained small amounts of impurity phases, including NiAs , La_2O_3 , and La(OH)_3 . La(OH)_3 was probably produced by hydration of La_2O_3 after the samples were taken from the silica tube to the atmosphere. Four-phase Rietveld analysis provided a weight fraction of LaNiOAs in the purest sample of 99 wt%. Table 1 summarizes the refined crystallographic parameters of LaNiOAs . Rietveld analysis also substantiated that LaNiOAs has the same crystal structure as LaMOPn ($M = \text{Mn, Fe, Co, Ni, Zn}$, $\text{Pn} = \text{P, As}$), which belongs to the tetragonal ZrCuSiAs -type structure ($P4/nmm$). The crystal structure is composed of alternating stacks of Ni–As and La–O layers, which are built from the edge-sharing networks of NiAs_4 and OLa_4 tetrahedrons, respectively. These tetrahedrons are distorted and more planar compared with regular tetrahedron, and are characterized by the two nearest-neighbor bonding angles. The As–Ni–As bond angles are 122.95° and 103.18°, and those of La–O–La are 119.45° and 104.73°. The large deviation of the bond angles from that of regular tetrahedron (109.47°) have been observed commonly in Ni- and Fe-based ZrCuSiAs -type compounds: 126.4° and 101.7° for LaNiOP [10], and 120.2° and 104.4° for LaFeOP [8]. While, those of e.g. LaCuOCh ($\text{Ch} = \text{S and Se}$), which are isostructural compounds but have not exhibited a superconducting transition [13], are much closer to that of regular tetrahedron. It implies that the distortion of the MPn_4 tetrahedron has a close relation to carrier transport including a superconducting transition, but further investigation is required to find a systematic relationship.

3.2. Electrical and magnetic properties

Fig. 3 shows the temperature (T) dependence of the electrical resistivity (ρ). ρ exhibited a metallic behavior at temperatures down to 2.4 K, and the ρ value at 305 K was very low, 3.5 mΩ cm. The inset shows a magnified view of ρ – T curves as a function of the external magnetic field (H_{ext}) in the temperature range of 1.8–2.8 K. The resistivity began to decrease at 2.4 K and became zero around 2 K. The onset temperature shifted to a lower temperature as H_{ext} increased, and the drop in ρ vanished at $H_{\text{ext}} = 3000$ Oe. These results suggest that LaNiOAs exhibits a superconducting transition at 2.4 K.

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