Electric property of the iron pnictide oxide superconductor \((\text{Fe}_2\text{As}_2)(\text{Ca}_6\text{(Al, Ti)}_4\text{O}_y)\) under hydrostatic pressure

Akihiko Hisada\(^1\), Naoki Fujiwara\(^2\), Yoshiya Uwatoko\(^1,3\), Hiraku Ogino\(^3,4\), Kohji Kishio\(^3,4\), Jun-ichi Shimoyama\(^3,4\)

\(^1\) Institute for Solid State Physics, The University of Tokyo, Kashiwanoha, Kashiwa, Chiba 277-8581, Japan
\(^2\) Graduate School of Human and Environmental Studies, Kyoto University, Yoshida Nihonmatsu-cho, Sakyo-ku, Kyoto 606-8501, Japan
\(^3\) TRiP, Japan Science and Technology Agency (JST), Sanban-cho Building 5, Sanban-cho, Chiyoda ku, Tokyo 102-0075, Japan
\(^4\) Department of Applied Chemistry, The University of Tokyo, Tokyo 113-8656, Japan
E-mail: a-hisada@issp.u-tokyo.ac.jp

Abstract. We report the electrical resistivity measurements of \((\text{Fe}_2\text{As}_2)(\text{Ca}_6\text{(Al, Ti)}_4\text{O}_y)\) polycrystalline sample under hydrostatic pressure using a cubic-anvil apparatus. The temperature dependence of the normal state resistivity exhibits metallic behavior. The \(\rho(T)\) curve is convex upward at low pressure, and it approaches \(T\)-linear dependence by applying high pressure. The superconductivity is suppressed by application of pressure. The onset temperature of superconductivity, which is 38 K at ambient pressure, decreases to 28 K at 8 GPa.

1. Introduction
Since the discovery of high-temperature superconductivity in LaFeAsO\(_{1-x}\)F\(_x\) [1], several types of iron-based superconductors have been discovered. The iron-based superconductors are commonly composed of alternate stacking of quasi-two-dimensional iron pnictide (FePn) layers and blocking layers or ions. These compounds are categorized into the so-called 1111 systems RFeAsO (R = rare earth elements), 122 systems AEFe\(_2\)As\(_2\) (AE = alkaline earth metals), 111 systems AFeAs (A = alkaline metal), and 11 systems FeSe and FeTe. The FePn interlayer distance for the 1111 systems is longer than that for other systems. For example, the typical distance for the 11, 111, 122 and 1111 systems is \(\sim 5.5\) \(\text{Å}\), \(\sim 6.4\) \(\text{Å}\), \(\sim 6.5\) \(\text{Å}\), and \(\sim 8.7\) \(\text{Å}\), respectively. Among the superconductors, the Sm-1111 system exhibits the highest superconducting transition temperature \((T_C)\). Thus the FePn interlayer distance has been pointed out to be a key factor to raise \(T_C\).

Recently, new superconducting (SC) families having perovskite-type oxide blocking layers, \((\text{Fe}_2\text{As}_2)(\text{AE}_{n+2}\text{M}_n\text{O}_y)\) (M = Sc, Cr, V, Al, (Mg, Ti), (Al, Ti), n = 2, 3, 4, and \(y \sim 3n\)) and \((\text{Fe}_2\text{As}_2)(\text{AE}_{n+1}\text{M}_n\text{O}_y)\) (M = Al, Sc, (Sc, Ti), (Mg, Ti), n = 2 \(\sim 5\), and \(y \sim 3n-1\)), were found to have relatively higher \(T_C\) than the existing systems: \(T_C\) of them exceeds 30 K at ambient pressure [2,3]. Figure 1 shows the crystal structure of \((\text{Fe}_2\text{As}_2)(\text{Ca}_6\text{(Al, Ti)}_4\text{O}_y)\). The distance
between the neighboring FeAs layers for (Fe$_2$As$_2$)(Ca$_6$(Al, Ti)$_4$O$_y$) goes beyond ~ 22 Å, which is much longer than that for the 1111 systems. In these families, the distance can be systematically controlled by changing the numbers of the blocking layers. However, so far as these families are concerned, the distance is not directly related to $T_C$, because these families exhibit high $T_C$ regardless of the numbers of the blocking layers.

Meanwhile, these families are characterized by large pnictogen height ($h_{Pn}$) from an Fe plane corresponding to a small a-axis unit length. Kuroki et. al. theoretically suggested that $h_{Pn}$ is an important factor to raise $T_C$ [4] and it is suggested that the optimal $T_C$ is realized when FePn$_4$ forms a regular tetrahedron [5]. The lattice parameters can be controlled by applying pressure, thus the effect of pressure on these families is intriguing.

In this study, we focus on (Fe$_2$As$_2$)(Ca$_6$(Al, Ti)$_4$O$_y$) having four (Al, Ti)O layers, because it contains less impurities than the families having two or three (Al, Ti)O layers. The onset and zero-resistivity temperatures are 39 and 25 K, respectively, at ambient pressure [2]. Moreover, an unusual successive phase transition from superconducting state to antiferromagnetic (AF) state was discovered from $^{75}$As and $^{27}$Al NMR studies [6]. The behavior of the AF transition under pressure is also notable.

2. Experimental procedure
Polycrystalline samples with a starting composition of (Fe$_2$As$_2$)(Ca$_6$(Al$_{0.33}$Ti$_{0.67}$)$_4$O$_{11}$) were synthesized by solid-state reaction. Details of the preparations and characterizations are given in Ref. 2. Electrical resistivity ($\rho$) measurement under pressure was performed by using a cubic-anvil high-pressure apparatus, which consists of six tungsten-carbide (WC) anvils and a pyrophylite gasket containing a teflon capsule. This apparatus has an advantage to producing highly hydrostatic pressure. As a pressure transmitting medium, we used glycerin. The pressure of the sample is calibrated by measuring resistivity anomalies of Bi, Te, and Sn, associated with their structural phase transitions at room temperature. Electrical resistivity was measured by a standard dc four-probe technique.

3. Results and Discussion
Figures 2(a) and 2(b) show the temperature dependence of the electrical resistivity under pressures up to 8 GPa. Electrical resistivity in the normal state shows metallic temperature
Figure 2. (a) Electrical resistivity ($\rho$) versus temperature ($T$) under different pressures. (b) Low temperature part of the $\rho(T)$ curve below 50 K. The arrows indicate the onset of superconducting transition temperature ($T_{\text{C onset}}$) and the temperature of the zero resistivity ($T_{\text{C zero}}$).

dependence and it monotonically decreases with increasing pressure. The $\rho(T)$ curve is convex upward at low pressure and it approaches $T$-linear dependence by applying high pressure. The onset of superconducting transition temperature ($T_{\text{C onset}}$) and the temperature of the zero resistivity ($T_{\text{C zero}}$) are shown in Figure 2(b). $T_{\text{C onset}}$ and $T_{\text{C zero}}$ are 38 and 23 K, respectively, at ambient pressure, which are in agreement with previous report [2]. They show small change below 4 GPa and they gradually decrease with increasing pressure above 6 GPa. At 8 GPa, $T_{\text{C onset}}$ and $T_{\text{C zero}}$ are 28 and 17 K, respectively. Any anomaly, which implies the magnetic ordering, was not found in the resistivity curve.

Figure 3 shows the pressure dependence of $T_{\text{C onset}}$ and $T_{\text{C zero}}$. They show the negative slope of about -2 K/GPa at high pressure. The pressure effect on $T_{\text{C}}$ is associated with $h_{\text{Pn}}$ and a-axis unit length. It is generally believed that the optimal $T_{\text{C}}$ is realized when FePn$_4$ forms a regular tetrahedron. The optimal $h_{\text{Pn}}$ is estimated to be between 1.34 and 1.38 Å. In (Fe$_2$As$_2$)(Ca$_6$(Al, Ti)$_4$O$_y$) having four (Sc, Ti)O layers, $h_{\text{Pn}}$ and a-axis unit length have been estimated to be 1.4 and 3.902 Å, respectively [3]. In (Fe$_2$As$_2$)(Ca$_6$(Al, Ti)$_4$O$_y$), a-axis unit length was estimated to be 3.815 Å, however, $h_{\text{Pn}}$ has not been estimated. Taking account of the values for (Fe$_2$As$_2$)(Ca$_6$(Sc, Ti)$_4$O$_y$), the $h_{\text{Pn}}$ of (Fe$_2$As$_2$)(Ca$_6$(Al, Ti)$_4$O$_y$) is more than 1.4 Å. For the compounds having $h_{\text{Pn}}$ larger than the optimal value, pressure application causes the decrease in $T_{\text{C}}$ because it causes reduction in a-axis unit length and thus the increase in $h_{\text{Pn}}$ [7], that is, $h_{\text{Pn}}$ deviates from the optimal value. This explanation seems valid for LiFeAs, in which $h_{\text{Pn}}$ is about 1.5 Å [8, 9].
4. Summary
We performed the electrical resistivity measurements of (Fe$_2$As$_2$)(Ca$_6$(Al, Ti)$_4$O$_y$) polycrystalline sample under hydrostatic pressure using a cubic-anvil apparatus. The temperature dependence of the normal state resistivity shows metallic behavior, and the $\rho(T)$ curve, which is convex upward at low pressure, approaches $T$-linear dependence by applying high pressure. The superconductivity is suppressed by application of pressure. At ambient pressure, $T_{C,\text{onset}}$ and $T_{C,\text{zero}}$ are 38 and 23 K, respectively. They gradually decrease with increasing pressure, and they are 28 and 17 K, respectively, at 8 GPa. The pressure effect on $T_C$ is associated with $h \rho_n$ and a-axis unit length.

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