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Pressure Dependence of Superconductivity in Filled-Skutterudite YOs_4P_{12}

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Abstract

We have studied the magnetization on YOs_4P_{12} under pressure up to ~ 4 GPa. Pure polycrystalline YOs_4P_{12} was prepared by the high-pressure high-temperature method and was evaluated by the X-ray diffraction measurement. YOs_4P_{12} shows superconducting transition at T_{SC} ~ 3 K at ambient pressure. We have investigated the pressure dependence of T_{SC} by means of DC magnetization measurements with incorporating opposed ceramic anvil pressure apparatus into MPMS. T_{SC} of YOs_4P_{12} slightly decreases with pressure at a rate of $\mathrm{d}T_{SC}/\mathrm{d}P$ = -0.11 K/GPa. This is quite contrary to the T_{SC} of related compounds YFe_4P_{12}, which rapidly increases with pressure at a rate of 1.0 K/GPa. The $\mathrm{d}T_{SC}/\mathrm{d}P$s relationship of YFe_4P_{12} and YOs_4P_{12} are similar with that of $LaFe_4P_{12}$ and $LaOs_4P_{12}$. T_{SC} of $LaFe_4P_{12}$ increases rapidly with pressure at a rate of $\mathrm{d}T_{SC}/\mathrm{d}P$ ~ 0.72 K/GPa below 2 GPa and that of $LaOs_4P_{12}$ decreases gradually with pressure at a rate of -0.095 K/GPa. Considering these similarity, we can assume that the origin of $\mathrm{d}T_{SC}/\mathrm{d}P$ of these compounds are the same and are attributed to the density of states at Fermi level and to the Hopfield parameter.

Keywords: YOs_4P_{12}, superconductivity, pressure, magnetization, ceramic anvil cell

1 Introduction

Ternary metallic pnictides with the general formula LnT_4X_{12} (Ln=Lanthanide, T=Transition metal, X=Pnictogen) crystallize in a cubic structure with a space group Im-3[1]. These compounds, called filled skutterudite, show various physical properties by the choice of combination of elements. One of the prominent physical properties is superconductivity. So far, 19 filled-skutterudite compounds are reported to exhibit superconductivity at low temperatures. The temperature of the superconducting transition (T_{SC}) has varied depending on the compounds and the highest T_{SC} among filled-skutterudite superconductor is at 17 K on $LaRh_4P_{12}$ [2]. Almost all superconducting filled skutterudite compounds obey Bardeen-Cooper-Schrieffer (BCS) theory with a negative pressure dependence of T_{SC} ($\mathrm{d}T_{SC}/\mathrm{d}P$) due to the stiffness of lattice. Among filled skutterudites which have superconducting correlations,

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only two compounds show positive dT_{SC}/dP. One is $LaFe_4P_{12}$ and the other is YFe_4P_{12} [3, 4]. T_{SC} of $LaFe_4P_{12}$ increases with pressure at a rate of 0.71 K/GPa below 2GPa. The slope decreases with increasing pressure and T_{SC} seems to reach its maximum approximately at 7 GPa [5]. When Fe in $LaFe_4P_{12}$ is replaced by Ru or Os, the respective dT_{SC}/dPs becomes negative. T_{SC} of $LaRu_4P_{12}$ monotonically decreases with increasing pressure in rates of -0.07 K/GPa in the range of 0 ~2 GPa and -0.29 K/GPa in the rage of 6~8 GPa. T_{SC} of $LaOs_4P_{12}$ monotonically decreases with increasing pressure at a rate of -0.095 K/GPa up to 1.8 GPa. At the first stage of this study, positive dT_{SC}/dPs is interpreted as the enhancement of T_{SC} due to the compression of the La metal. It is not to be denied that this conclusion is inconsistent by the appearance of positive dT_{SC}/dPs of YFe_4P_{12} [4]. Y metal does not show superconductivity above 6 mK at ambient pressure and shows superconductivity only above 11 GPa [6]. According to band-structure calculations, dT_{SC}/dP > 0 for $LaFe_4P_{12}$ can be rationalized by the enhancement of the electronic density of states at Fermi level $D(E_F)$ and the Hopfield parameter η under pressure[7]. Meanwhile, dT_{SC}/dP < 0 for $LaRu_4P_{12}$ is attributed to the reduction of $D(E_F)$. As for YFe_4P_{12}, T_{SC} monotonically increases with pressure up to 8 GPa. The mechanism of dT_{SC}/dP for YFe_4P_{12} is proposed to be the same as $LaFe_4P_{12}$ [4]. On the other hand, T_{SC} of YRu_4P_{12} and YOs_4P_{12} under pressure have not been investigated because of the difficulty of reproducibility of pure samples. YOs_4P_{12} is one of filled skutterudite compounds, which shows superconductivity at T_{SC} ~ 3 K [8]. It can only be synthesized by the high-temperature and high-pressure method. The purpose of this study is to investigate the pressure dependence of T_{SC} of YOs_4P_{12} and to reveal the origin of its dT_{SC}/dP.

	Lattice Constant (Å)	B_0 (GPa)[9]	T_{SC} (K)	dT_{SC}/dP (K/GPa)
$LaFe_4P_{12}$	7.8316	150	4.1	0.72[3]
$LaRu_4P_{12}$	8.0605	172	7.2	- 0.16[3]
$LaOs_4P_{12}$	8.0844	190	1.8	-0.095[3]
YFe_4P_{12}	7.7896	144	5.6	1[4]
YRu_4P_{12}	8.0298	183	8.5	-
YOs_4P_{12}	8.0844	189	2.8	-0.11

Table 1: The sum of structural parameters and superconducting parameters of LnT_4P_{12} (Ln=La, Y, T=Fe, Ru, Os)

2 Experimental

We have synthesized YOs_4P_{12} using the high-temperature and high-pressure method with a wedge type cubic anvil high pressure apparatus. The wedge-type cubic apparatus consists of the upper and lower stages with three anvils, respectively. The movement of the anvils is synchronized by a wedge system. The pressure from one axis gets converted to six faces. Starting material is 10% Y-rich stoichiometric rate, i.e., $Y_{1.1}Os_4P_{12}$, because Y can be easily oxidized and the amount of Y is likely to be underestimated when weighted. Each metal and red phosphorous powder were reacted at 4.5 GPa 1100℃ for 30 min. The resulting sample was evaluated by X-ray diffraction method with Co Kα radiation. We measured magnetization under pressure by incorporating an opposed anvil pressure cell into a commercial SQUID magnetometer MPMS (Quantum Design Inc.) [10]. Anvils of the pressure cell are made of non-composite ceramic (FCY20A) produced by the Fuji Die company, Japan. The outer and inner diameters, and the thickness of the NiCrAl gasket are 5.0, 0.5, and 0.5 mm, respectively. The pressure was evaluated from the superconducting transition temperature of Pb. Hydrostatic pressure is realized because the sample space was filled with glycerin as pressure medium, which will not solidify up to 6 GPa at room temperature. In order to get the accurate value of magnetization under pressure, we measured YOs_4P_{12} with pressure cell and vacant pressure cell separately. The signal of YOs_4P_{12} was obtained by subtracting the induced voltage of the vacant pressure from that of YOs_4P_{12} with pressure cell. The magnetization was calculated by fitting the subtracted data.

3 Results and Discussion

Figure 1 shows the X-ray diffraction pattern of YOs_4P_{12}. All distinct peaks can be assigned to the filled-skutterudite crystal structure. Peaks from impurity phases are within the level of noise, i.e., less than 1 %. Peaks that have a strong intensity (as indexed in Fig. 1) were picked up for calculation of the lattice parameter. The obtained lattice constant, a = 8.0619 Å, is excellent agreement with that reported by K. Kihou *et al.*, a = 8.0615(4) Å [8]. In other words, YOs_4P_{12} prepared for this study is a pure sample without impurity phases and is consistent with a previous report.

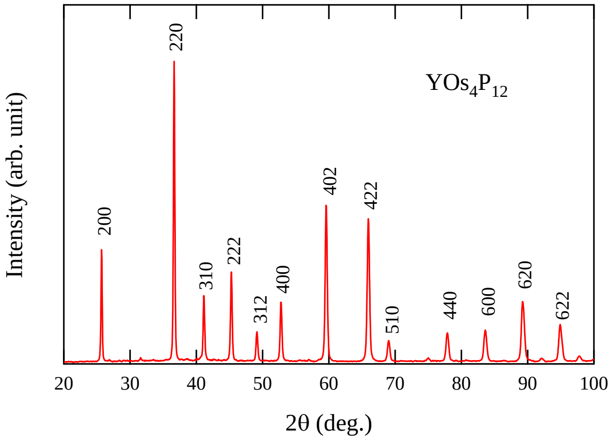

Fig. 1. X-ray diffraction pattern of YOs_4P_{12}. Only indexed peaks were used for calculation of lattice parameter

Figure 2 shows the temperature dependent magnetization divided by the field at H=10 Oe. Distinct sharp drops due to superconductivity are observed at 2.8 K in both processes of field cooled (FC) and zero field cooled (ZFC). The temperature of T_{SC} is consistent to the previous report, in which the drop of resistivity contributing T_{SC} is observed at around 3 K [8]. The diamagnetic signal can be observed below the temperature at zero resistivity as for YFe_4P_{12} [4]. The similar behavior is observed in this YOs_4P_{12}. With decreasing temperature the resistivity drops at 3 K to zero resistivity [8]. The diamagnetic signal in this study appears at 2.8 K, which is below the zero resistivity. We note that the process of ZFC is different from that of FC due to the pinning effect of a superconductor. While magnetic susceptibility (χ) at lowest temperature of FC is one-sevenths of that of ZFC on YFe_4P_{12} [4], χ at lowest temperature of FC is two-thirds of that of ZFC on YOs_4P_{12} in this study. The magnetization of YOs_4P_{12} in this study shows smaller pinning effect than that of YFe_4P_{12}. The small pinning effect implies purity of the sample. In order to confirm the purity of YOs_4P_{12}, it is important to investigate residual resistivity. However, the sample synthesized in this study is too fragile to measure electrical resistivity. In order to investigate whether the residual resistivity has the same feature found for FC and ZFC when comparing YOs_4P_{12} and YFe_4P_{12}, synthesis of a sample with high degree of sintering is in progress.

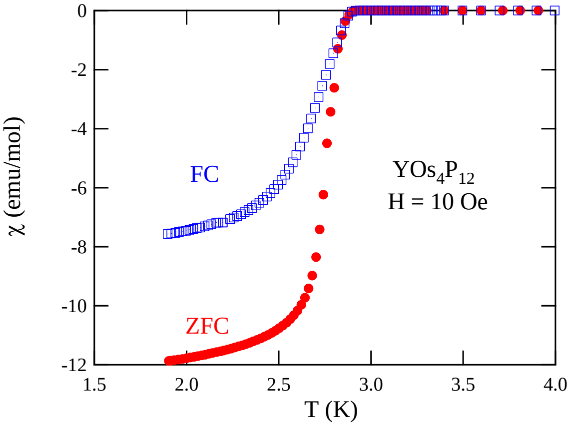

Fig. 2. Temperature dependence of χ under the field of H = 10 Oe with field cooled process (open square) and with zero field cooled process (closed circle).

Figure 3 shows the temperature dependent magnetic susceptibility up to 3.9 GPa with ZFC process. Since the weight of the sample for the measurement under pressure is too small to get the accurate value of χ, χ was calibrated by the data at ambient pressure. A distinct sharp drop due to the Meissner effect of superconductivity is observed at every pressure. We note that $\chi(T)$ at temperatures slightly above the reduction due to the Meissner effect faintly increases with decreasing temperature which cannot be seen in χ at ambient pressure. Although the pressure might cause this behavior, the signal in the normal state is too small to discuss this behavior.

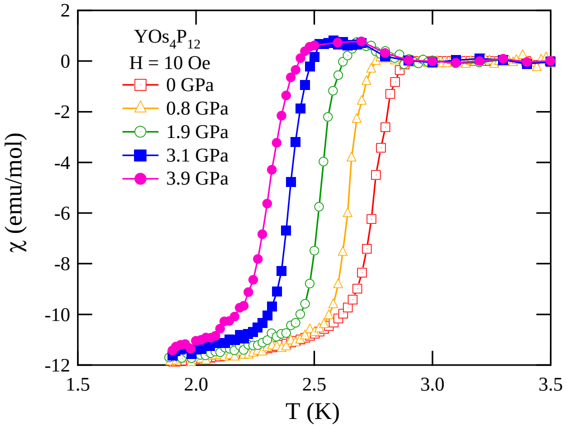

Fig. 3. Temperature dependence of magnetic susceptibility (χ-T) with zero field cooled process at 0 GPa (open square), 0.8 GPa (open triangle), 1.9 GPa (open circle), 3.1 GPa (closed square), and 3.9 GPa (closed circle).

Figure 4 shows the temperature derivative of the magnetic susceptibility. Peaks are observed, corresponding to the middle point between the onset and offset of superconductivity. We thus define the peak of $d\chi/dT(T)$ as T_{SC}. T_{SC} monotonically decreases by applying pressure.

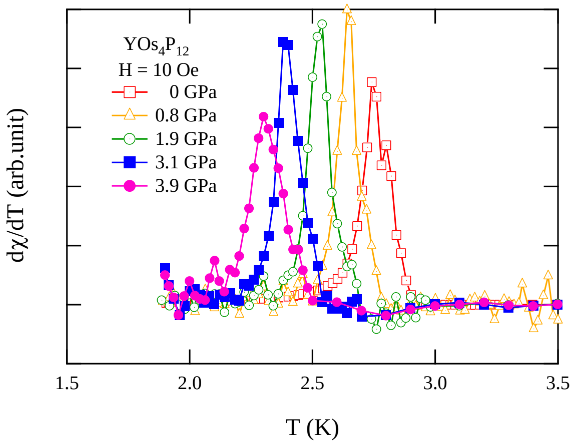

Fig. 4. Temperature derivative of magnetic susceptibility d χ /d*T* against temperature under pressure.

Figure 5 shows the shift of superconducting temperature ($\Delta T_{SC}(P) = T_{SC}(P) - T_{SC}(0)$) as a function of pressure of YOs_4P_{12}. In addition, ΔT_{SC} of related compounds LnT_4P_{12} (Ln= La, Y, T=Fe, Ru, Os) except YRu_4P_{12} are also shown. T_{SC} of YOs_4P_{12} monotonically decreases with pressure at a rate of -0.11 K/GPa. As can be seen from the graph, ΔT_{SC} of $LaRu_4P_{12}$ and $LaOs_4P_{12}$ and YOs_4P_{12} decreases with pressure at almost the same rate.

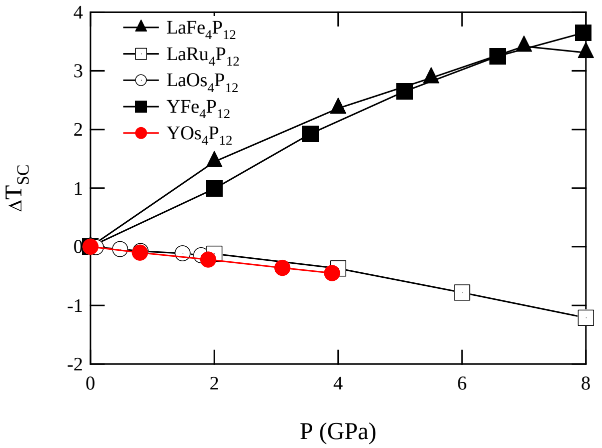

Fig. 5. Pressure dependence of ΔT_{SC} on $LaFe_4P_{12}$[5](closed triangle), $LaRu_4P_{12}$[5] (open square), $LaOs_4P_{12}$[3] (open circle) YFe_4P_{12}[4](closed square), YOs_4P_{12}(closed circle).

In the pressure region from 0 to 2 GPa, dT_{SC}/dP of YOs_4P_{12}, $LaRu_4P_{12}$, and $LaOs_4P_{12}$ are -0.11, -0.07, and -0.095 K/GPa, respectively. As it can be seen in table 1, the bulk modulus is not much different in these three compounds. Thus it would not be controversial to assume that the origin of dT_{SC}/dP for these three compounds is the same. J. G. Cheng et al. proposed that the difference of dT_{SC}/dP of YFe_4P_{12} and YRu_4P_{12} is attributed to the density of states at Fermi level and the Hopfield parameter η from the band calculation [4]. The same mechanism is proposed to the difference of dT_{SC}/dP on $LaFe_4P_{12}$ and $LaRu_4P_{12}$ [7]. From the similarity of dT_{SC}/dP of YOs_4P_{12}, $LaRu_4P_{12}$, and $LaOs_4P_{12}$, we propose that dT_{SC}/dP of YOs_4P_{12} has the same mechanism, i.e., the reduction of $D(\varepsilon_F)$ and η.

We will describe the model of the electronic density of states ($D(\varepsilon)$) of $LaOs_4P_{12}$ and YOs_4P_{12}, the band calculation of which have not been reported. The Fermi level (FL) of YRu_4P_{12} at 0 GPa is just at the peak of $D(\varepsilon)$ and that of $LaRu_4P_{12}$ is near the peak of $D(\varepsilon)$. On the other hand, FL of YFe_4P_{12} and $LaFe_4P_{12}$ is above a peak of $D(\varepsilon)$. That is why T_{SC} of YRu_4P_{12} (8.5 K) is higher than T_{SC} of YFe_4P_{12} (5.6 K) and that of $LaRu_4P_{12}$ (7.2 K) is higher than that of $LaFe_4P_{12}$ (4.1 K). In the case of $LaOs_4P_{12}$ (T_{SC}~1.8 K) and YOs_4P_{12} (T_{SC}~2.8 K), T_{SC} is lower than that of $LaFe_4P_{12}$ and YFe_4P_{12}, respectively. Thus, the FL of $LaOs_4P_{12}$ and YOs_4P_{12} cannot be at the peak of $D(\varepsilon)$. Considering the peak of $D(\varepsilon)$ moves to high energy region at 8 GPa in the band calculation of LnT_4P_{12} (Ln=La, Y, T=Ru, Os)[4,7], the FL of $LaOs_4P_{12}$ and YOs_4P_{12} should be below a maximum of $D(\varepsilon)$ and $D(\varepsilon)$ probably shifts to higher energy by applying pressure. That is why T_{SC} of YOs_4P_{12} is lower than YFe_4P_{12} and dT_{SC}/dP of YOs_4P_{12} has a negative value. The same model can be applied to the case of $LaOs_4P_{12}$ and $LaFe_4P_{12}$. To confirm this model, however, band calculations of YOs_4P_{12} and $LaOs_4P_{12}$ are desired.

4 Summary

We have investigated the pressure dependence of T_{SC} of YOs_4P_{12} up to 4 GPa. T_{SC} of YOs_4P_{12} slightly decreases with pressure by about -0.11 K/GPa. It is quite contrary to T_{SC} of $YFe_4P_{12,}$ which rapidly increases at a rate of 1.0 K/GPa. There are similar relationships between $LaFe_4P_{12}$ and $LaOs_4P_{12}$, because dT_{SC}/dP of $LaFe_4P_{12}$ is 0.72 K/GPa and that of $LaOs_4P_{12}$ is -0.095 K/GPa. Thus we propose the origin of dT_{SC}/dP of these compounds is the same and is attributed to distinct features of the density of states at Fermi level and the Hopfield parameter.

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