Ru substitution effect on the peak effect in superconducting PrOs$_4$Sb$_{12}$

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Abstract. Ac susceptibility ($\chi_{ac}$) measurements on Pr(Os$_{1-x}$Ru$_x$)$_4$Sb$_{12}$ single crystals for $x = 0.05$ and 0.1 were performed by mutual inductance method. A peak structure caused by anomalously enhanced flux pinning force, which is so-called the peak effect, appears in the $H$ dependence of $\chi_{ac}$. The peak structure shifts to lower fields as $T$ increases and disappears in $T > 1$ K in both samples, while in PrOs$_4$Sb$_{12}$ it is observable up to near $T_c$. This fact indicates that the Ru substitution suppresses the peak effect. We demonstrate that the observed $T$ dependent behavior of the peak structure can be explained roughly by the synchronization model although there remains deviation from the model curve, suggesting some modification may be needed in the model to be applied to Pr(Os$_{1-x}$Ru$_x$)$_4$Sb$_{12}$.

1. Introduction
Since the filled skutterudite compound PrOs$_4$Sb$_{12}$ was reported to be the first heavy fermion (HF) superconductor among Pr-based compounds, PrOs$_4$Sb$_{12}$ has been attracting much attention [1]. The superconducting transition temperature $T_c = 1.85$ K being higher than $T_c = 0.74$ K of no-4f-electron reference LaOs$_4$Sb$_{12}$ [1, 2] demonstrates clearly the involvement of Pr 4f electrons in the superconductivity. Actually, several measurements performed so far suggest that it is of unconventional type. The Sb NQR spin-lattice relaxation rate indicates strong-coupling isotropic superconducting gap [3]. The existence of multiple superconducting phases with point nodes is indicated by angle-resolved thermal conductivity measurements [4], although angle-resolved specific heat suggests fully opened gap [5]. The appearance of spontaneous internal fields is indicated by zero field $\mu$SR measurements [6]. Considering singlet-triplet crystalline electric field (CEF) level scheme of Pr 4f electrons[7], pairing mediated by quadrupole excitons has been discussed [8].

Another feature that characterizes the unconventional superconducting state in PrOs$_4$Sb$_{12}$ is the so-called "peak effect", i.e., anomalously enhanced flux pinning force in the mixed state below the upper critical field $H_{c2}$. An anomalous irreversible behavior is observed in the magnetization curve in the mixed state [9], a considerable peak structure in the field dependence of the surface impedance [10], and minima in the $H$ dependence of the flux-flow resistivity [11]. Similar anomalies have been observed in many of the HF superconductors [12–14] and therefore it is expected that the observed peak effect in PrOs$_4$Sb$_{12}$ reflects the involvement of the heavy quasiparticles in the HF superconductivity. In this paper, we report the Ru substitution effect on the peak effect of PrOs$_4$Sb$_{12}$ by ac susceptibility measurements. Since the unconventional HF and conventional superconducting states appear at the opposite ends in the
Pr(Os$_{1-x}$Ru$_x$)$_4$Sb$_{12}$ series with continuous $T_c$ variation in between [15–17], Pr(Os$_{1-x}$Ru$_x$)$_4$Sb$_{12}$ is an appropriate system to study how the anomalous flux pinning depends on the type of superconductivity.

2. Experimental
Ac susceptibility has been measured by mutual inductance method with ac field $H_{ac} \sim 0.3$ Oe down to 0.1 K using a dilution refrigerator equipped with a 8-T superconducting magnet. Single crystals of Pr(Os$_{1-x}$Ru$_x$)$_4$Sb$_{12}$ with $x = 0.05$ and 0.1 were grown by Sb self flux method. Obtained single crystals with the size of about $1 \times 1 \times 2$ mm$^3$ exhibit sharp superconducting transitions in $T$ dependence measurements under zero field, indicating good sample quality.

3. Results and discussion
3.1. Temperature dependence
Figure 1 shows the $T$ dependence of $\chi_{ac} (= \chi' + i\chi''$) in zero field, where $\chi'$ and $\chi''$ correspond to the real and the imaginary parts of $\chi_{ac}$, respectively. $\chi'$ represents the equilibrium differential susceptibility $\chi' = dM/dH$ while $\chi''$ represents the energy dissipation $W = \pi \chi'' H_{ac}^2$. In both samples, $\chi'$ decreases below the superconducting transition temperature $T_c \sim 1.81$ K ($x = 0.05$) and 1.76 K ($x = 0.1$), respectively. Broad peaks also appear in the $T$ dependence of $\chi''$ just below $T_c$ in both samples.

![Figure 1](image)

**Figure 1.** Temperature dependences of $\chi_{ac}$ of the single crystals of Pr(Os$_{1-x}$Ru$_x$)$_4$Sb$_{12}$ with $x = 0.05$ and 0.1 measured in zero field. $\chi'$ and $\chi''$ represent the equilibrium differential susceptibility and the energy dissipation, respectively.

3.2. Field dependence
The $H$ dependences of $\chi_{ac}$ at several temperatures are shown in fig.2. Specific peak structures indicating the peak effect are observed in $\chi'(H)$ just below $H_{c2}$ in both samples. With increasing $T$, the peak structures shift to the lower fields and becomes suppressed gradually. The $H$ dependence of $\chi''$ also shows a broad peak at the field where $\chi'$ shows the peak structure. These behaviors of $\chi_{ac}$ in the Ru doped samples are similar to those of PrOs$_4$Sb$_{12}$ [18]. However, while the peak structure remains up to $T \sim T_c$ in PrOs$_4$Sb$_{12}$, the peak structure is not visible in $\chi'(H)$ and $\chi''(H)$ for $T > 1$ K in $x = 0.05$ and 0.1. To analyze the feature of the peak structures quantitatively, $H'_{c2}$, $H_{min}$ and $H_{pk}$ representing the kink structure just below $H_{c2}$, the local minimum of $\chi'(H)$ and the local peak in $\chi'(H)$ are determined respectively by using field derivative $d\chi'/dH$. 
3.3. \(H-T\) phase diagram

Obtained \(H-T\) phase diagrams are shown in fig.3. The overall \(T\) dependence of \(H_{c2}\) is reproduced by the WHH (Werthamer-Helfand-Hohenberg) curve in the clean limit above 0.2 T [19], below which \(H_{c2}(T)\) exhibits slight deviation with a positive curvature possibly caused by the multiband effect as discussed in PrOs\(_{4}\)Sb\(_{12}\) [20].

The considerable \(T\) dependent shift of the \(H\) region of the peak structure cannot be explained by the “matching effect”, in which the pinning becomes effective in a certain field where the average pinning site spacing matches the flux line spacing. In the present case, so-called ”synchronization effect” remains as a possible origin of the peak effect [21]. With increasing \(H\), the shear modulus \(C_{66}\) of the flux-line lattice (FLL) is expected to decrease as \(\propto (1 - H/H_{c2})^2\) [22]. Therefore, in high fields, FLL can be deformed so as to be effectively pinned by randomly distributed pinning centers, leading to an increase of the pinning force density. However, when \(H\) is close to \(H_{c2}\), each flux line begins to shear due to the further decrease of \(C_{66}\) and the pinning force density starts to decrease. This is the scenario of the formation of a peak structure in the synchronization model. Considering that the pinning force density has a maximum at \(H_{\text{min}}\), we have compared the \(H-T\) data with the model.

In fig.3, shown by the dotted line is the synchronization model curve, \(H_{\text{min}} = H_{c2}(T) - \zeta H_{c2}(T)^{3/2}\), where \(\zeta\) is a fitting parameter corresponding to the density of the pinning centers and depends on neither \(T\) nor \(H\) [23]. The best fitting values of \(\zeta\) are \(2.3 \times 10^{-1} \text{ T}^{-1/2}\) and \(2.4 \times 10^{-1} \text{ T}^{-1/2}\) for \(x = 0.05\) and 0.1, respectively; these values are comparable to that of CeRu\(_2\) \((\sim 5.8 \times 10^{-2} \text{ T}^{-1/2})\) [23], whose peak effect is nicely explained by the synchronization model. The model curve roughly reproduces the \(T\) dependence of \(H_{\text{min}}\). Quantitatively, however, the \(T\) dependence of \(H_{\text{min}}\) is faster than the model curve.

As demonstrated by the disappearance of the peak effect above 1 K in Pr(Os\(_{1-x}\)Ru\(_x\))\(_4\)Sb\(_{12}\) \((x = 0.05\) and 0.1), the anomalous pinning force enhancement is suppressed by the Ru substitution. However, it is expected naively that the Ru substitution may introduce crystalline defects that could behave as pinning centers leading to the peak effect. In this sense, the suppression of the peak effect by the Ru substitution is anomalous. In CeRu\(_2\) [24], it was reported that the peak effect is more pronounced for samples with the smaller RRR values, i.e., including larger amount of conduction electron scattering centers, which may work as flux pinning centers. In Pr(Os\(_{1-x}\)Ru\(_x\))\(_4\)Sb\(_{12}\), it has been found that the singlet-triplet CEF excitation energy increases considerably by the Ru substitution; \(E/k_B \sim 8\) K for \(x = 0\) and 80 K for \(x = 1\) [17,25]. Because of this, the 4f-electron contribution to the dc magnetic susceptibility \(\chi_{\text{dc}}\) at
$T \sim T_c$ decreases with increasing the Ru substitution. The resulting smaller $\chi_{dc}$, which is less effective for the flux pinning, can be one of the possible reasons for the peak effect suppression for $x \neq 0$. However, the decrease in $\chi_{dc}$ for $x = 0.1$ is only 20% and it is questionable if such a small decrease in $\chi_{dc}$ can account for the observed suppression of the peak effect. Other changes in the electronic properties might be necessary to be taken into account; e.g., with the Ru substitution, the 5d conduction electrons from Os ions are gradually replaced by the 4d conduction electrons from Ru ions [26, 27]. We expect on-going measurements in higher Ru concentration region will be helpful to clarify the source of the anomalous flux pinning in Pr(Os$_{1-x}$Ru$_x$)$_4$Sb$_{12}$.

![Figure 3](image-url)

**Figure 3.** $H$-$T$ phase diagrams with the characteristic fields, $H^{*}_{c2}$, $H_{min}$, $H_{pk}$ and $H_{kink}$. The dashed line represents the clean-limit WHH model curve for $H_{c2}(T)$. The shaded area shows the region where the peak effect appears noticeably. The green dotted line represents the synchronization model curve. $H_{kink}$ representing the kink structure in low fields decreases almost linearly with increasing $T$ and disappears in $T > 1$K.

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