Superconductivity, magnetic order, and quadrupolar order have been investigated in the filled skutterudite system Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ as a function of composition $x$ in magnetic fields up to 9 tesla and at temperatures between 50 mK and 10 K. Electrical resistivity measurements indicate that the high field ordered phase (HFOP), which has been identified with antiferroquadrupolar order, persists to $x \sim 0.5$. The superconducting critical temperature $T_c$ of PrOs$_4$Sb$_{12}$ is depressed linearly with Nd concentration to $x \sim 0.55$, whereas the Curie temperature $T_{FM}$ of NdOs$_4$Sb$_{12}$ is depressed linearly with Pr composition to $(1-x) \sim 0.45$. In the superconducting region, the upper critical field $H_{c2}(x,0)$ is depressed quadratically with $x$ in the range $0 < x \lesssim 0.3$, exhibits a kink at $x \approx 0.3$, and then decreases linearly with $x$ in the range $0.3 \lesssim x \lesssim 0.6$. The behavior of $H_{c2}(x,0)$ appears to be due to pair breaking caused by the applied magnetic field and the exchange field associated with the polarization of the Nd magnetic moments, in the superconducting state. From magnetic susceptibility measurements, the correlations between the Nd moments in the superconducting state appear to change from ferromagnetic in the range $0.3 \lesssim x \lesssim 0.6$ to antiferromagnetic in the range $0 < x \lesssim 0.3$. Specific heat measurements on a sample with $x = 0.45$ indicate that magnetic order occurs in the superconducting state, as is also inferred from the depression of $H_{c2}(x,0)$ with $x$. 

INTRODUCTION

Since the discovery of heavy fermion (HF) superconductivity (SC) in PrOs$_4$Sb$_{12}$ in 2001, this compound has attracted intense interest. It is one of the few Pr cited states of the Pr series, and this compound exhibits some type of unconventional superconducting pairing mechanism of electrons. 

The end member compound NdOs$_4$Sb$_{12}$ is a mean-field type ferromagnet with a Curie temperature $T_{FM} \approx 1$ K. According to the analysis of magnetic susceptibility, electrical resistivity, and ultrasonic attenuation data, the CEF energy level scheme of the Nd$^{3+}$ ion in NdOs$_4$Sb$_{12}$ is consistent with a $\Gamma_8$ quartet ground state (0 K), a $\Gamma_8^{(1)}$ quartet first excited state ($\sim 220$ K), and a $\Gamma_5$ doublet highest excited state ($\sim 590$ K), all of which are magnetic in $O_h$ symmetry. Recent inelastic neutron scattering measurements support this energy level scheme with a more accurate description in $T_h$ symmetry: $\Gamma_8^{(2)}(0 K) - \Gamma_6^{(1)}(267 K) - \Gamma_5(350 K)$. Although the CEF ground state $\Gamma_8^{(2)}$ contains quadrupole moments, no features indicative of a HFOP have been detected in NdOs$_4$Sb$_{12}$. A large electronic specific heat coefficient $\gamma \sim 520$ mJ/(mol K$^2$) is inferred at low $T$. 

In addition to the typical rattling mode that is observed as an ultrasonic dispersion near $\sim 40$ K in the compounds ROs$_4$Sb$_{12}$ (R= La, Pr, Nd, Sm), an extra rattling mode is detected only in NdOs$_4$Sb$_{12}$. Because the FM transition in NdOs$_4$Sb$_{12}$ occurs at quite a low $T$ and the lattice parameter is almost the same as that of PrOs$_4$Sb$_{12}$, the substitution of Pr with Nd in the pseudoternary system Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ is well suited to investigating the effect of FM on the evolution of the unconventional SC of PrOs$_4$Sb$_{12}$.

In this paper, we report the temperature vs Nd concentration $T-x$ phase diagram of the Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ system. We find that the superconducting critical temperature $T_c$ of the Pr end member compound decreases linearly with Nd concentration $x$, while the Curie temperature of the ferromagnetic end member compound decreases linearly with the Pr concentration $(1-x)$. The superconducting region extends to $x \approx 0.55$, where it meets the ferromagnetic phase, with evidence for ferro-
magnetic ordering of the Nd moments in the superconducting phase at \( x \approx 0.45 \), from specific heat measurements in magnetic fields up to 0.429 tesla. In order to gain information about the evolution of the HFOP with Nd concentration and to probe the nature of the superconducting state, magnetoresistive measurements were performed in fields up to 9 T throughout the range 0 \( \lesssim x \lesssim 0.5 \). An analysis of the upper critical field \( H_{c2}(x,0) \), involving a comparison to measurements of \( H_{c2}(x,0) \) on the La\(_{3−x}\)Gd\(_x\)In system and the multiple pair breaking theory of Fulde and Maki, provides evidence for magnetic ordering of the Nd ions in the superconducting state for concentrations above \( x \approx 0.3 \) and suggests that the superconducting electrons of Pr\(_{1−x}\)Nd\(_x\)Os\(_4\)Sb\(_{12}\) are paired in spin singlet states. On the other hand, the evolution of \( H_{c2}(x,0) \) might be described in a two band model of superconductivity. Magnetic susceptibility measurements indicate that the magnetic correlations of the Nd ions change from ferromagnetic for 0.3 \( \lesssim x \lesssim 0.6 \) to antiferromagnetic for 0 < \( x \lesssim 0.3 \).

**EXPERIMENTAL DETAILS**

Single crystals of Pr\(_{1−x}\)Nd\(_x\)Os\(_4\)Sb\(_{12}\) were grown by the molten flux method as described in Ref 24. Pr and Nd were mixed in an arc furnace prior to the flux growth to ensure a uniform distribution of the rare earth elements. The cubic LaFe\(_3\)P\(_{12}\)-type structure [25] is observed by means of X-ray powder diffraction measurements throughout the entire doping series. The lattice parameter vs Nd concentration \( x \) is plotted in the inset of Fig. 1 where it is apparent that the lattice constant is minimally affected by Nd substitution, consistent with previous measurements by Jeitschko et al. [26].

The DC magnetic susceptibilities \( \chi_{dc}(T) \) (Fig. 1) for collections of single crystals with a total mass near 30 mg were measured using a Quantum Design SQUID magnetometer MPMS-5.5. Together with the X-ray diffraction data, the \( \chi_{dc}(T) \) data show that Nd can be substituted continuously for all values of \( x \): i.e., higher values of \( \chi_{dc}(T) \) are observed in samples with higher \( x \). The ac magnetic susceptibility \( \chi_{ac}(T) \) measurements were performed using home-built 1st-order gradiometers as pick-up coils in the temperature range from 0.05 K to 2.5 K. Each pick-up coil is coupled with a primary coil, which supplies a 17 Hz ~ 0.05 – 0.15 Oe ac magnetic field. Typically, \( \approx 6 – 18 \) mg collections of single crystals were used for the \( \chi_{ac}(T) \) measurements. Electrical resistivity measurements \( \rho(T, H) \) were performed using a standard 4-wire technique in a transverse geometry \((H \perp current)\) on individual single crystals mounted in a \(^3\)He-\(^4\)He dilution refrigerator in magnetic fields \( H \) between 0 T and 9 T. Specific heat \( C(T, H) \) measurements were performed on a collection of single crystals with a mass of 51.07 mg for \( x = 0.45 \) using a standard heat pulse technique.

**RESULTS**

Displayed in Fig. 2(a) are the ac magnetic susceptibility data \( \chi_{ac}(T) \) for various \( x \), where the paramagnetic background signal at 2.5 K has been set to zero and the data for each \( x \) have been normalized to the largest signal in either the superconducting (SC) or the ferromagnetic (FM) transitions. When a sample enters the SC state, \( \chi_{ac}(T) \) drops below the paramagnetic background in a rounded step shape, where the SC transition temperature \( T_c \) is defined at 50% of the change in \( \chi_{ac} \) and the transition width is defined as the difference in the temperatures associated with the 10% and 90% values. When FM ordering occurs, \( \chi_{ac}(T) \) exhibits a peak above the paramagnetic background at the FM transition (Curie) temperature \( T_{FM} \) and the transition width is taken to be the difference in the temperature corresponding to 90% of the peak value of \( \chi_{ac}(T) \) and \( T_{FM} \). As \( x \) increases, the SC transition is suppressed to lower \( T \) (at almost the same rate as for Pr\((\text{Os}_{1−x}\text{Ru}_x)\)Sb\(_{12}\) [27]) and, above \( x = 0.6 \), only a FM signal appears. For \( x = 0.45 \) and 0.55, features associated with both SC and FM are observed. Figure 2(b) summarizes the \( x \) dependence of \( T_c \) and \( T_{FM} \) determined from measurements of \( \chi_{ac}(T) \), \( \rho(T) \), and \( C(T) \). We also note the discrepancy between the two \( x = 0.45 \) samples in Fig. 2(a), which may be due to supercurrent surface screening in one of the samples which obscures the FM feature in the \( \chi_{ac}(T) \) measurements. On the other hand, sample dependence cannot be completely ruled out.

The specific heat of a sample with \( x = 0.45 \) was measured in order to explore the possible coexistence of SC and FM at this concentration (Fig. 3). A broad peak appears in \( C(T, H = 0) \) at 0.9 K with a maximum near 0.48
FIG. 2: (a) Temperature $T$ dependence of the ac magnetic susceptibility $\chi_{ac}$ in arbitrary units for various Nd concentrations $x$. (b) Superconducting transition temperature $T_c$ and ferromagnetic transition (Curie) temperature $T_{FM}$ vs $x$ for Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ determined from measurements of the ac magnetic susceptibility $\chi_{ac}$, electrical resistivity $\rho$, and specific heat $C$. Vertical bars indicate transition widths as defined in the text.

K. The inset to Fig. 3 shows the corresponding $\chi_{ac}(T)$ data, where the onset of $T_c$ at $\sim 0.9$ K matches the beginning of the upturn of the peak in $C(T, H = 0)$. The FM feature then appears in $\chi_{ac}(T)$ near 0.48 K, in agreement with the maximum in $C(T, H = 0)$. From these observations, we conclude that FM and SC features are present in both $\chi_{ac}(T)$ and $C(T, H = 0)$ and that the broadness of the peak in $C(T, H = 0)$ may be due to the proximity of the two transitions. Upon application of small magnetic fields, the peak gradually shifts to higher $T$, indicating that the SC phase is suppressed and the FM phase is enhanced with $H$.

From measurements of $\rho(T)$ at constant $H$ and $\rho(H)$ at fixed $T$ (Fig. 4), $H_{c2}(T)$ curves were determined for various values of $x$ (Fig. 5). The data points are defined as the temperatures and fields associated with the 50% value of $\Delta \rho$ at the SC transition, and the transition width is defined as the differences in temperatures and fields corresponding to the 10% and 90% values of $\Delta \rho$ at the SC transition. Above $x = 0.25$, the transition width becomes very large. Measurements of $\rho(H)$ isotherms also reveal the HFOP phase, which appears as a shoulder in $\rho(H)$ above 4 T are due to the HFOP.

FIG. 3: Specific heat $C$ of Pr$_{0.55}$Nd$_{0.45}$Os$_4$Sb$_{12}$ vs $T$ at $H = 0$, 0.043 T, 0.086 T, and 0.429 T. As $H$ increases, the broad peak moves to higher $T$. Inset: the corresponding $\chi_{ac}(T)$ measured on single crystals from the same batch.

FIG. 4: Electrical resistivity $\rho$ vs magnetic field $H$ at various temperatures $T$. The rapid drop in $\rho(T, H)$ to zero is due to the superconducting transition, while the shoulders in $\rho(H)$ above 4 T are due to the HFOP.

Since the FM in NdOs$_4$Sb$_{12}$ conforms to the mean field model, a Curie-Weiss analysis of the Nd contribution to the magnetic susceptibility should be a good in-
The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$.

We note that in order to apply this type of analysis, it is necessary to assume that the contribution to $\chi_{dc}(T)$ from the Pr$^{3+}$ ions must retain the same $T$ dependence for all values of $x$. This seems like a reasonable approximation because the nearest neighbors of the rare earth ions are Sb ions, which form the cages of the filled skutterudite structure. As such, the CEF that influences each Pr ion is, to first order, unchanged as Pr is replaced with Nd. Due to the curvature for 20K-50K caused by the effect of the CEF on the Nd$^{3+}$ ions, the Curie-Weiss analysis of $\chi_{Nd}(T)$ is only applied in the low $T$ regime from 2K to 10K by using the expression:

$$\chi_{Nd}(T) = \frac{C}{T - \Theta}.$$  \hspace{1cm} (2)

where $C$ is the Curie constant and $\Theta$ is the Curie-Weiss temperature. The fitting results are displayed in Fig. 7. The CW temperature is positive and decreases between $x = 1$ and $x_{cr,2} \sim 0.3$, where it crosses over to a negative value. The Curie constant undergoes a modest increase with decreasing $x$, with a possible local maximum near $x_{cr,1} \sim 0.6$. These results suggest the presence of a weak FM phase between $x \approx 0.3$ and 1 and that AFM correlations appear below $x \approx 0.3$.

**ANALYSIS AND DISCUSSION**

Taken together, these results reveal a rich phase diagram for the Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ system which includes superconductivity, magnetic order, and quadrupolar order.

Figure 6(a) shows the 0K $H - x$ phase diagram for Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$. The lower boundary of the high field ordered phase (HFOP) is determined from kinks in the $\rho(H)$ isotherms (Fig. 4). The HFOP in Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ persists to $x = 0.5$, above which resistivity measurements have not yet been performed, although we note that the features associated with the HFOP are not observed for NdOs$_4$Sb$_{12}$. The break in slope in the lower $H - x$ phase boundary of the HFOP seems to be correlated with the one observed in $H_{c2}(0) - x$. In addition, the HFOP persists throughout

**FIG. 5:** Temperature dependence of upper critical field $H_{c2}$ for Nd concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$. The horizontal and vertical bars represent the transition widths, defined from the 10% and 90% values of the drop in $\rho(T)$, $H_\text{c2}$ for Pr concentrations $x \leq 0.5$.

**FIG. 6:** (a) Magnetic field $H - x$ phase diagram of Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ at $\sim 0$K. (b) Zero-kelvin extrapolation of the experimentally determined upper critical field $H_{c2}(x,0)$. The solid line is a curve based on Eqs. (2). Inset: Normalized $H_{c2}(x,0)$ and $x$ with respect to $H_{c2}(0,0)$ and $x_{cr,1}$ for Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ and La$_{1-x}$Gd$_x$In. Note the difference between the curvature at low concentrations.
the SC phase in Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$, whereas it vanishes beyond $x \sim 0.1$ in Pr(Os$_{1-x}$Ru$_x$)$_4$Sb$_{12}$. This may be due to changes in the CEF that are larger for Ru substitution for Os than for Nd substitution for Pr.

From the $H_{c2}(x, T)$ data shown in Fig. 5, it can be seen that the magnitude of $H_{c2}$ decreases rapidly with $x$ in the range $0 < x \lesssim 0.3$, more slowly in the range $0.3 \lesssim x \lesssim 0.55$, and appears to vanish near $x \approx 0.6$. This trend is illustrated more clearly in Fig. 5(b), which shows the zero temperature value of the upper critical field $H_{c2}(x, 0)$. The $H_{c2}(x, 0)$ data can be fit with the equations (solid line in Figure 5(b)):

$$H_{c2}(x, 0) \approx \begin{cases} 2.18 - 19.78x^2 & 0 \leq x < 0.3, \\ 0.471 - 0.8x & 0.3 < x < 0.6. \end{cases}$$

(3)

where the $x$ dependence of $H_{c2}(0)$ is quadratic for $x \lesssim 0.3$ and is linear for $0.3 \lesssim x \lesssim 0.6$, resulting in an obvious break in slope near $x \sim 0.3$. Thus, there appear to be two critical concentrations, $x_{cr,2} \approx 0.3$ and $x_{cr,1} \approx 0.6$.

Since evidence for two-band SC has been observed in PrOs$_4$Sb$_{12}$, this result could indicate that Nd substitution has a different effect on the SC of the different bands. If so, then this empirical formula may suggest that one channel has a critical concentration $x_{cr,1} \sim 0.6$, for which $H_{c2}$ has a linear $x$ dependence over the entire SC region with a maximum value of $\sim 0.471$ T, while the other channel has a critical concentration $x_{cr,2} \sim 0.3$ and only exists in the region $0 \leq x \leq 0.3$ with a maximum value of $H_{c2} \sim 2.2$ T.

In order to explore an alternative route to analyzing the $H_{c2}(x, 0)$ data, we turn to the multiple pair breaking theory of Fulde and Maki. For example, this theory has previously been used to analyze the $H_{c2}(x, T)$ curves for the system La$_{3-x}$Gd$_x$In, that is formed by substituting Gd impurity ions that carry localized magnetic moments into the singlet BCS superconductor La$_3$In. 27

The $H_{c2}(x, 0)$ data in the La$_{3-x}$Gd$_x$In system reveal that $H_{c2}(x, 0)$ exhibits a rapid linear decrease with $x$ in the lower concentration region and a slower linear decrease with $x$ in the higher concentration region. In the inset of Fig. 6, the normalized upper critical field data $H_{c2}(x, 0)/H_{c2}(0, 0)$ for both systems are plotted vs the non-dimensionalized concentration $x/x_{cr,1}$. Here, $H_{c2}(0, 0)$ is the 0 K $H_{c2}$ of the parent compound without magnetic substitutions (i.e., PrOs$_4$Sb$_{12}$ and La$_3$In), and $x_{cr,1}$ is the concentration where SC disappears. $H_{c2}(0) \sim 2.25$ tesla for PrOs$_4$Sb$_{12}$ and $\sim 6.8$ tesla for La$_3$In, while $x_{cr,1} \sim 0.6$ for Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ and $0.076$ for La$_{3-x}$Gd$_x$In. The ratio of $x_{cr,1}$ of Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ to that of La$_{3-x}$Gd$_x$In is $\sim 7.6$, or $\sim 23$, if the magnetic substituent is expressed as the percentage of the over all rare-earth concentration in each compound. For $0 \leq x/x_{cr,1} \lesssim 0.5$, the suppression of $H_{c2}(x, 0)/H_{c2}(0, 0)$ in Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ is much less than that of La$_{3-x}$Gd$_x$In, but for $0.5 \lesssim x/x_{cr,1} \lesssim 1$, both systems have a similar linear monotonic suppression. Interestingly, a break in curvature occurs at $x/x_{cr,1} \sim 0.5$ for both systems.

The generalized Abrikosov-Gorkov (A-G) theory of Fulde and Maki includes three effects that can break Cooper pairs in a BCS SC in the presence of magnetic moments and magnetic field: (1) spin-polarization of conduction electrons by an applied magnetic field, (2) spin-flip scattering of conduction electrons by magnetic moments, and (3) spin-polarization of conduction electrons by the exchange field generated by the applied field or magnetic order mediated by the RKKY interaction between the magnetic moments. The generalized A-G formula for three effects into account has the following form,

$$\ln\left(\frac{T_c}{T_{c0}}\right) - \Psi\left(\frac{1}{2} - 0.14\left(\frac{T_{c0}}{T_c}\right)\left(\sum_{i=1}^{3} \frac{\alpha_i}{\alpha_{cr,i}}\right)\right) - \Psi\left(\frac{1}{2}\right) = 0,$$

(4)

where the SC depairing parameters are denoted as $\alpha_i$’s, their critical values as $\alpha_{cr,i}$’s, and $T_{c0}$ is the superconducting transition temperature of the parent compound in zero applied magnetic field. Equation 5 describes the total pair-breaking effect in the magnetically substituted SC system, which is equivalent to the pair breaking effect due only to the applied field on the parent compound:

$$\sum_{i=1}^{3} \frac{\alpha_i}{\alpha_{cr,i}} = \frac{H_{c2}(x, T)}{H_{c2}(0, 0)} + \frac{x}{x_{cr}} + \frac{P}{P_{cr}} = \frac{H_{c2}(0, T)}{H_{c2}(0, 0)},$$

(5)

where $x$ is the concentration of substituted magnetic ions, $H_{c2}(x, T)$ is the upper critical field for concentration $x$ and temperature $T$, and $P$ is the Pauli polarization term corresponding to the effect of the magnetic exchange field.
on the conduction electrons. Following this argument, the temperature dependence of the upper critical field for concentration $x$ can be expressed as,

$$H_{c2}(x, T) \approx H_{c2}(0, T) - H_{c2}(0, 0) \left( \frac{x}{x_{cr}} + \frac{\tau_{so} J^2}{P_{cr} x^2} \right). \quad (6)$$

The Pauli polarization term $P = \tau_{so}(x \Im \sigma < J_z >)^2$, where $\tau_{so}$ is the spin-orbit scattering time, $\Im$ is the s-f exchange interaction parameter, and $< J_z >$ is the average value of the total angular momentum along the direction of the exchange field, which is defined as the $z$ direction. $< J_z >$ has a Brillouin-function dependence on $T$ but approaches a constant value as $T \to 0$ K, that is proportional to the magnetic substituent’s “ground-state” magnetic moment. To simplify the analysis, we focus on the behavior at 0 K. In this case Eqs. 5 and 6 are reduced to

$$H_{c2}(x, 0) = H_{c2}(0, 0)[1 - \left( \frac{x}{x_{cr}} + \frac{\tau_{so} J^2}{P_{cr} x^2} \right)]. \quad (7)$$

In the low concentration regime, $H_{c2}(x, 0)$ of La$_{3−x}$Gd$_x$In has a linear $x$ dependence, but $H_{c2}(x, 0)$ of Pr$_{1−x}$Nd$_x$Os$_4$Sb$_{12}$ shows a more quadratic behavior. The linear dependence of $x$ in the low concentration region of La$_{3−x}$Gd$_x$In indicates that the Gd magnetic moments are in the dilute region and no magnetic ordering has developed so the Pauli polarization contribution is negligible. This suggests that if Pr$_{1−x}$Nd$_x$Os$_4$Sb$_{12}$ conforms to BCS SC, magnetic correlations may be significant in the low $x$ region. According to the Curie-Weiss analysis, $\Theta$ is negative for $x \lesssim 0.3$, indicating that AFM correlations may be dominant in this concentration range and that the internal magnetic field found in the SC state is associated with AFM order. Thus, the SC in PrOs$_4$Sb$_{12}$ may be influenced by both AFM and FM correlations that may have comparable strengths over certain parts of the phase diagram. It is possible that the slower than linear suppression of $H_{c2}(0)$ vs $x$ in the range of $x$ from 0 to $x_{cr,2} \approx 0.3$ is due to the combination of pairbreaking by the applied field and the AFM exchange field, while the suppression of $H_{c2}(x)$ vs $x$ in the range $x_{cr,2} \approx 0.3$ to $x_{cr,1} \approx 0.6$ could be primarily due to the FM exchange field, although the curvature of $H_{c2}(0)$ in this region is not in agreement with the generalized Abrikosov-Gorkov (A-G) theory of Fulde and Maki.

### SUMMARY

The effect of magnetic moments on the normal and superconducting states of PrOs$_4$Sb$_{12}$ has been investigated in the Pr$_{1−x}$Nd$_x$Os$_4$Sb$_{12}$ system. In the normal state, the feature associated with the HFOP is clearly observed up to $x = 0.5$. The kink in the lower phase boundary of the HFOP seems to correlate with a similar feature in $H_{c2}(x, 0)$. The HFOP is more robust against substituent concentrations $x$ in Pr$_{1−x}$Nd$_x$Os$_4$Sb$_{12}$ than in the Pr(Os$_{1−x}$Ru$_x$)$_4$Sb$_{12}$ system. The Curie-Weiss analysis for $T$ between 2K and 10K suggests that weak FM exists for $0.3 \lesssim x \lesssim 1$ and AFM correlations are important in the range $x \lesssim 0.3$.

In the SC state, features associated with FM were observed in the $x = 0.45$ sample. Quadratic and linear dependences of $x$ were found in $H_{c2}(0)$ for $0 \leq x \lesssim 0.3$ and $0.3 \lesssim x \lesssim 0.6$, respectively, which indicates that there are two critical concentrations in the Pr$_{1−x}$Nd$_x$Os$_4$Sb$_{12}$ system, $x_{cr,1} \sim 0.6$ and $x_{cr,2} \sim 0.3$. The break in slope for $H_{c2}(x, 0)$ may be related to the existence of two bands of SC electrons in PrOs$_4$Sb$_{12}$, which are affected by Nd substitution differently. On the other hand, the multiple-pair breaking effect could also explain the behavior in $H_{c2}(x, 0)$ where $x_{cr,1}$ and $x_{cr,2}$ are associated with the suppression of SC in the FM and AFM correlation regimes, respectively.

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