Muon spin rotation and relaxation in Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$: Magnetic and superconducting ground states

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Muon spin rotation and relaxation (µSR) experiments have been carried out to characterize magnetic and superconducting ground states in the Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ alloy series. In the ferromagnetic end compound NdOs$_4$Sb$_{12}$ the spontaneous local field at positive-muon (µ$^+$) sites below the ordering temperature $T_C$ is greater than expected from dipolar coupling to ferromagnetically aligned Nd$^{3+}$ moments, indicating an additional indirect RKKY-like transferred hyperfine mechanism. For 0.45 $\leq$ $x$ $\leq$ 0.75, µ$^+$ spin relaxation rates in zero and weak longitudinal applied fields indicate that static fields at µ$^+$ sites below $T_C$ are reduced and strongly disordered. We argue this is unlikely to be due to reduction of Nd$^{3+}$ moments, and speculate that the Nd$^{3+}$-µ$^+$ interaction is suppressed and disordered by Pr doping. In an $x = 0.25$ sample, which is superconducting below $T_C = 1.3$ K, there is no sign of “spin freezing” (static Nd$^{3+}$ magnetism), ordered or disordered, down to 25 mK. Dynamic µ$^+$ spin relaxation is strong, indicating significant Nd-moment fluctuations. The µ$^+$ diamagnetic frequency shift and spin relaxation in the superconducting vortex-lattice phase decrease slowly below $T_C$, suggesting pair breaking and/or possible modification of Fermi-liquid renormalization by Nd spin fluctuations. For 0.25 $\leq$ $x$ $\leq$ 0.75, the µSR data provide evidence against phase separation; superconductivity and Nd$^{3+}$ magnetism coexist on the atomic scale.

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I. INTRODUCTION

Rare-earth-based materials are in many ways ideal for studies of the interaction between superconductivity and magnetism in metals. An example is the filled skutterudite family of isostructural lanthanide intermetallics [1], where the unconventional heavy-fermion superconductor PrOs$_4$Sb$_{12}$ and its alloys have been the subject of considerable interest [2, 3]. The isomorph NdOs$_4$Sb$_{12}$ is a ferromagnet with Curie temperature $T_C \approx 0.8$ K [2–4]. In order to elucidate the interplay between the superconductivity of PrOs$_4$Sb$_{12}$ and the ferromagnetism of NdOs$_4$Sb$_{12}$, the alloy system Pr$_{1-x}$Nd$_{x}$Os$_4$Sb$_{12}$ has been investigated.

Figure 1 gives the phase diagram obtained from thermodynamic and transport measurements [7], together with transition temperatures from our muon spin rotation and relaxation (µSR) data as discussed in Sec. III. Superconductivity persists up to $x \approx 0.5$, and Nd$^{3+}$ “spin freezing” (static magnetism, with or without long-range order) appears above $x \approx 0.45$. This is evidence for competition between superconductivity and magnetism for the ground state of the system, as well as the possibility of a ferromagnetic quantum critical point near $x_{cr} = 0.4–0.5$. The rate of decrease of the superconducting transition temperature $T_C$ with Nd concentration is nearly the same in the (Pr$_{1-x}$Nd$_x$)Os$_4$Sb$_{12}$ and Pr(Os$_{1-y}$Ru$_y$)$_4$Sb$_{12}$ alloy systems [8], contrary to the behavior of “conventional” superconductors where magnetic impurities are much more effective than nonmagnetic ones in suppressing $T_C$. There is evidence that Nd substitution does not affect the Pr$^{4+}$ CEF splitting nearly as strongly as Ru doping [6].

A number of aspects of superconductivity and magnetism on the atomic scale are readily accessible to the µSR technique. Early µSR studies of PrOs$_4$Sb$_{12}$ revealed two important phenomena: the absence of nodes in the superconducting energy gap [9], and the onset of a spontaneous internal magnetic field below the superconducting transition temperature $T_C$ [10] that indicated broken time-reversal symmetry in the superconducting state. Later µSR experiments found no change of the muon Knight shift below $T_C$ [11], suggesting p-wave pairing, resolved a discrepancy between inductive and µSR penetration-depth measurements [12], ruled out a second phase transition in the superconducting state [13], and studied the effects of La and Ru doping [14–17].

This article reports µSR experiments in the Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ alloy series, undertaken with the goal of providing microscopic characterization of the
ordered or disordered, for the alloys, (2) the absence of any static Nd magnetism, and magnetism for 0

Pr, x = 0.55, 0.75, and 1.00, were grown using the self-flux technique [19]. Solid solubility is good across the alloy series [7, 8]. The samples were characterized by x-ray diffraction and magnetic susceptibility measurements. Each µSR sample consisted of a mosaic of crystals glued to a 11×16×0.25 mm 6N Ag plate using GE 7031 varnish, which was attached to the cold finger of the cryostat with a thin layer of Apiezon grease. The crystals were partially oriented, as their (100) faces tended to be parallel to the mounting plate, but the cubic symmetry renders most averages over µ⁺ sites (e.g., dipolar field averages) independent of orientation. To ensure isothermal conditions an Ag foil was wrapped around the sample and firmly attached to the cold finger. Magnetic fields in the range 0–200 Oe were applied.

In time-differential µSR experiments spin-polarized positive muons (µ⁺) are implanted in a sample, precess in the local fields at the interstitial µ⁺ sites, and decay with a mean lifetime τµ = 2.197 µs via the reaction µ⁺ → e⁺ + νe + τµ. The decay positrons are emitted preferentially in the direction of the µ⁺ spin, and are detected using scintillation counters. This asymmetry yields the dependence of the µ⁺ spin polarization G(t) on time t between implantation and muon decay. Typically ~10^5 events are obtained for a single measurement of G(t).

In transverse-field µSR (TF-µSR) the applied field H_T, which is usually the dominant field at µ⁺ sites, is perpendicular to the initial µ⁺ spin orientation. The µ⁺ spins precess, and the positron count rate for a given direction oscillates with time at a frequency near γµH_T [20]: here γµ = 2π × 13.553 kHz/G is the µ⁺ gyromagnetic ratio. In a disordered (inhomogeneous) static field the precession frequencies are distributed and the oscillations are damped, or do not appear at all if the disorder is sufficiently strong.

In longitudinal-field µSR (LF-µSR) the µ⁺ spins are initially oriented parallel to the applied field H_L, and hence precess only in local fields generated by the sample such that the resultant field is not parallel to H_L. Zero-field µSR (ZF-µSR) can be considered a limiting case of LF-µSR. Here again, any distribution of local-field magnitudes results in damping of the oscillation expected from a unique field, although G(t) functions for LF- and TF-µSR are quite different.

In both cases, thermal fluctuations of the µ⁺ local fields give rise to dynamic relaxation (the spin-lattice relaxation of NMR). This is the only relaxation mechanism for the µ⁺ components parallel to local static fields observed in ZF- and LF-µSR.

For a given positron counter, the count rate N(t) is related to G(t) by

\[
N(t) = N_0 e^{-t/\tau_\mu} \left[ 1 + A_0 G(t) \right]
\]

(ignoring uncorrelated background counts), where N_0 is the initial count rate, \( \tau_\mu \) is the \( \mu^+ \) lifetime, \( A_0 \) is the initial count-rate asymmetry (spectrometer-dependent but typically ~0.2), and the polarization function \( G(t) \) is the projection of the time-dependent ensemble \( \mu^+ \) spin polarization (normalized to 1 at \( t = 0 \)) on the direction to the counter. Various experimental configurations (applied field direction, counter orientations, etc.) yield specific forms for \( G(t) \), from which information on static and
tions appropriate to such cases. In zero applied field the
\( \mu \) the following sections, we consider
\( H \) component, is a monotonically increasing function of
frequencies, and
\( f \) describes the
\( \mu \) angle between \( B \) and \( S \). The projection of this longi
tudinal component back along the initial \( \mu \) spin di
rection (the axis of the spectrometer counter system) is
therefore \( S_{\mu} \cos^2 \theta \). For randomly oriented \( B \), the an
gular average \( \cos^2 \theta = 1/3 \), leading to a “1/3” longitudinal
contribution to the \( \mu \) spin polarization in zero applied
field \( \mu \). The “2/3” transverse contribution oscillates if \( B \) is reasonably constant. Any distribution of field
magnitudes (i.e., of \( \mu \) precession frequencies) leads to
damping of the oscillation. This leaves only the 1/3 com
ponent at late times, which is time independent if there are
no other relaxation mechanisms; otherwise it relaxes
dynamically. Hence dynamic and static relaxation can be
separated in ZF-\( \mu \)SR (and weak-LF-\( \mu \)SR) experiments if
the former is sufficiently slow compared to the latter.

In nonzero longitudinal field \( H_L \), the resultant \( \mu \)
field \( H_L + B \) is no longer randomly oriented. Then \( \cos^2 \theta \)
is greater than 1/3, and approaches 1 asymptotically as
\( H_L \gg B \); the \( \mu \) spins are “decoupled” \( \mu \) from their
local fields. In the absence of dynamic relaxation, one
expects a polarization function of the form
\[
G(t) = \left[ 1 - f_L(H_L) \right] g(H_L, t) + f_L(H_L),
\]
where \( g(H_L, t) \) describes the distribution of precession
frequencies, and \( f_L(H_L) \), the fraction of longitudinal
component, is a monotonically increasing function of
\( H_L \).

Since we will be dealing with disordered spin systems in
the following sections, we consider \( \mu \) polarization functions
appropriate to such cases. In zero applied field the static
Gaussian Kubo-Toyabe (KT) function \( \mu \)
\[ G_{KT}(t) = \frac{1}{3} + \frac{2}{3} \left( 1 - \Delta^2 t^2 \right) \exp \left( -\frac{1}{2} \Delta^2 t^2 \right) \] (3)
describes the \( \mu \) polarization when the distribution of
each Cartesian component of \( B \) is Gaussian with zero mean
and rms width \( \Delta \). The Gaussian KT function models \( \mu \)
relaxation in dipolar fields from a densely populated
lattice of randomly oriented moments, electronic or nuclear, when the moment magnitudes are
fixed but their orientations are random on the atomic
scale. The Gaussian distributions result from the central
limit theorem of statistics, since a given muon is
coupled to many lattice moments. For randomly located
\( \mu \) sites in a lattice with a low concentration of mo
ments, however, the wings of the distribution are dom
inated by single nearby moments, and the distribution of
field components becomes nearly Lorentzian \( \mu \). This
leads to the “Lorentzian KT” ZF-\( \mu \)SR polarization
function \( 1/3 + (2/3)(1 - at) \exp(-at) \), where \( a \) is the half
width of the field distribution \( \mu \). Clearly the form of
the polarization function changes appreciably between the
dilute and concentrated limits.

Noakes and Kalvius \( \mu \) have approached the problem of
\( \mu \) relaxation in the intermediate-dilution regime by
generalizing the KT result phenomenologically, assuming
that in a moderately diluted lattice the KT distribution
width \( \Delta \) is itself distributed. The physical meaning of
this approach is discussed further in Sec. IV. A Gaussian
distribution of \( \Delta \) with mean \( \Delta_0 \) and rms width \( w \) results in
the “Gaussian-broadened Gaussian” (GbG) KT ZF-
\( \mu \)SR polarization function \( \mu \)
\[
G_{GbG}(t) = \frac{1}{3} + \frac{2}{3} \left( \frac{1 + R^2}{1 + R^2(1 + \Delta_{\text{eff}}^2 t^2)} \right)^{3/2}
\times \left[ 1 - \frac{\Delta_{\text{eff}}^2 t^2}{1 + R^2(1 + \Delta_{\text{eff}}^2 t^2)} \right]
\times \exp \left( -\frac{\Delta_{\text{eff}}^2 t^2}{1 + R^2(1 + \Delta_{\text{eff}}^2 t^2)} \right),
\] (4)
where \( \Delta_{\text{eff}}^2 = \Delta_0^2 + w^2 \) is the effective spin relaxation rate
and \( R = w/\Delta_0 \) is the ratio of the distribution widths.

GbG KT polarization functions are shown in Fig. 2
together with the Lorentzian KT function. The ratio \( R \)
parameterizes the departure from single-Gaussian KT
behavior \( \mu \). For \( R = 0 \) \( G_{GbG}(t) = G_{KT}(t) \), and
with increasing \( R \) the minimum near \( \Delta_0 t = 1 \) decreases
in depth. For \( R \geq 1 \) \( G_{GbG}(t) \) does not approach the
Lorentzian function, so that \( G_{GbG}(t) \) is not an interpola
tion function between the Gaussian and Lorentzian
limits. Instead, \( G_{GbG}(t) \) becomes monotonic and nearly
independent of \( R \); for this reason the condition \( R \leq 1 \) was

\[
\begin{array}{c}
\text{FIG. 2. (Color online) Static zero-field Gaussian-broadened}
\text{Gaussian and Lorentzian KT polarization functions (relaxation}
\text{rates } \Delta_{\text{eff}} \text{ and } a, \text{ respectively).}
\end{array}
\]
imposed in fits of Eq. (1) to the data. For small $\Delta_{e\text{ff}}t$, $G_{G\text{pp}}(t) \approx 1 - \Delta_{e\text{ff}}^2 t^2$, independent of $R$ for all $R$.

Often the experimental $\mu$SR asymmetry signal has a contribution from muons that miss the sample and stop in the mounting plate or cryostat cold finger. This plate is usually silver, for good thermal contact and also because Ag nuclear moments are small and $\mu^+$ spin relaxation in Ag is negligible. In the following the plots of $Z\text{F}$- and $L\text{F}$-$\mu$SR data give the polarization function $G(t)$ for the sample rather than the total asymmetry $A(t)$. The relation between these quantities is

$$G(t) = \frac{A(t)}{A_0 - f_{\text{Ag}}},$$

(5)

where $A_0$ is the total initial asymmetry and $f_{\text{Ag}}$ is the fraction of muons that stop in the silver.

### III. RESULTS AND DISCUSSION

#### A. NdOs$_4$Sb$_{12}$

In the end compound NdOs$_4$Sb$_{12}$ the onset of ferromagnetism below the Curie temperature $T_C \approx 0.8$ K gives rise to a spontaneous internal field $B_\mu$ at $\mu^+$ sites, and therefore to oscillations in $Z\text{F}$-$\mu$SR and weak-$L\text{F}$-$\mu$SR. Figure 3 shows early-time data taken in a longitudinal field $H_L = 6.1$ Oe, which decouples the $\mu^+$ moment from nuclear dipolar fields above $T_C$ (cf. Sec. III, but has little effect in the ferromagnetic state; the observed internal fields are two orders of magnitude greater than $H_L$ except near $T_C$. The data are well fit by the damped Bessel function

$$G_{\text{damp}}(t) = e^{-\lambda_L t} \left[ \left(1 - f_L \right) e^{-\lambda_T t} J_0(\omega_\mu t + \varphi) + f_L \right],$$

$$\omega_\mu = \gamma_\mu B_\mu,$$

(6)

where $J_0(x)$ is the zeroth-order cylindrical Bessel function, $\lambda_L$ is the (dynamic) longitudinal spin relaxation rate, $f_L$ is the fraction of longitudinal signal component, $\lambda_T$ describes the damping of the oscillation, $\varphi$ is the initial phase, and $B_\mu$ is the dominant value of the $\mu^+$ local-field magnitude distribution. A damped cosine function does not provide as good a fit.

The choice of the Bessel function is motivated by the Fourier-transform relation between the polarization function and the distribution of precession frequencies. In an incommensurate single-$q$ sinusoidal spin-density wave, the precession frequency distribution is $g(\omega) = 2/\left(\pi \sqrt{\omega^2 - \omega_0^2}\right)$ $(0 < \omega < \omega_0)$, the Fourier transform of which is the Bessel function $J_0$. The distribution $g(\omega)$ is characterized by a singularity at $\omega_0$ and a broad distribution of lower frequencies. Thus $J_0(\omega_\mu t)$ represents the $\mu^+$ polarization when the experimental frequency distribution has this character (inset of Fig. 3). An actual incommensurate spin-density wave in NdOs$_4$Sb$_{12}$ is ruled out by the considerable evidence for a ferromagnetic ground state [32]. Broadening of the singularity is accounted for by damping the Bessel function [Eq. (6)].

$\mu^+$ spin polarizations in NdOs$_4$Sb$_{12}$ at various temperatures below $T_C$ are shown in Fig. 4 together with damped Bessel-function fits. Figure 4 shows the temperature dependencies of $\omega_\mu$, $\lambda_T$, and $\lambda_L$. The dominant frequency $\omega_\mu(T)$ exhibits an order-parameter-like increase.

![FIG. 3. (Color online) Early-time $\mu^+$ spin polarization in NdOs$_4$Sb$_{12}$, $H_L = 6.1$ Oe, $T = 25$ mK. Curve: fits of Eq. (5) to the data. Inset: Fourier transforms $g(\omega)$ of data and fit.](image-url)

![FIG. 4. (Color online) Early-time $\mu^+$ spin polarization at various temperatures $\lesssim T_C$ from weak-$L\text{F}$-$\mu$SR in NdOs$_4$Sb$_{12}$, $H_L = 6.1$ Oe. Data for $T = 25$ mK from Fig. 3. Curves: fits of Eq. (6) to the data.](image-url)
The principal axes of the external field, at least at low temperatures. Broadened unresolved resonances corresponding to the ~370-G spread in principal-axis dipolar fields (~30 µs⁻¹ spread in angular precession frequencies) might contribute to the spectral weight below the peak in g(ω) (inset of Fig. 4).

B. Pr$_{0.25}$Nd$_{0.75}$NdOs$_4$Sb$_{12}$

In Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ alloys the Pr$^{3+}$ ions are in non-magnetic crystal-field ground states, resulting in a substitutionally diluted lattice of Nd$^{3+}$ ions [33]. For $x = 0.75$ Pr doping has reduced the magnetic transition temperature $T_C$ from 0.8 K to ~0.55 K [2]. Although there has been no direct confirmation of ferromagnetic order in the diluted alloys, the Nd concentration dependence of the “Curie” temperature $T_C$ and paramagnetic-state properties in the alloys tend smoothly towards their values in NdOs$_4$Sb$_{12}$ as $x(Nd) \rightarrow 1$ [2]. In Pr$_{0.25}$Nd$_{0.75}$Os$_4$Sb$_{12}$ the paramagnetic Curie-Weiss temperature is positive and $\simeq T_C$.

Figure 6 shows early-time weak-LF-µSR spin polarization data from Pr$_{0.25}$Nd$_{0.75}$Os$_4$Sb$_{12}$ at $T = 25$ mK and $H_L = 14.8$ Oe. The weak longitudinal field was applied below $T_C$. At 0.8 K $\omega_\mu$ is small but finite (cf. Fig. 4), indicating that $T_C$ is slightly higher than this. The damping rate $\lambda_T$, being considerably larger than $\lambda_L$, is mainly due to static disorder. It is much smaller than $\mu_B$ until $T$ approaches $T_C$ from below; here it increases slightly, suggesting a spread of transition temperatures [33]. The dynamic rate $\lambda_L(T)$ increases as $T \rightarrow T_C$ from above due to critical slowing down of Nd$^{3+}$ spin fluctuations [34], followed by a decrease below $T_C$ as the Nd$^{3+}$ moments freeze. The behavior of all these quantities is that of a conventional ordered magnet.

The value of $\overline{B}_\mu = \omega_\mu/\gamma_\mu$ at low temperatures is $560 \pm 10$ G. Assuming the saturation moment $M_{sat} = 1.73\mu_B$ found from magnetization isotherms [3], the experimental $\mu^+\cdot\text{Nd}^{3+}$ coupling constant $A^{exp}$ = $\overline{B}_\mu/M_{sat}$ is $335 \pm 6$ G/$\mu_B$. For comparison we have calculated the dipolar coupling tensor $A^{calc}$ from ferromagnetically aligned Nd$^{3+}$ moments $M_{Nd}$, assuming muons stop at the probable $\frac{1}{2}$, 0.015 site as found in PrOs$_4$Sb$_{12}$ [10]. The field $B^{calc}_\mu$ at this site is given by $B^{calc}_\mu = A^{calc}_\mu \cdot M_{Nd}$. The principal axes of $A^{calc}_\mu$ are parallel to the crystal axes; the principal-axis values $A^{calc}_\mu$ and the corresponding values of $B^{calc}_\mu$ for $M_{Nd} = 1.73\mu_B$ are given in Table 1. In general three $\mu^+$ frequencies are expected from the three inequivalent $\mu^+$ sites in the cubic structure for nonzero $M_{Nd}$.

All the $B^{calc}_\mu$ are smaller in magnitude than the observed field, so that a significant RKKY-like transferred hyperfine interaction between Nd$^{3+}$ spins and $\mu^+$ spins is necessary to account for the difference. The interaction strength required to do this depends on the orientation of $M_{Nd}$, which is not known at present. The value of $B^{calc}_\mu$ and dipolar fields $B^{calc}_\mu$ at the probable $\frac{1}{2}$, 0.015 $\mu^+$ site in NdOs$_4$Sb$_{12}$.

![Diagram](image_url)

**FIG. 5.** (Color online) Temperature dependencies of weak-LF-µSR parameters in NdOs$_4$Sb$_{12}$, $H_L = 6.1$ Oe. (a) Spontaneous $\mu^+$ spin precession frequency $\omega_\mu$ and static transverse spin relaxation rate $\lambda_T$. Inset: fraction $f_L$ of longitudinal component. (b) Longitudinal spin relaxation rate $\lambda_L$. Arrow: Curie temperature $T_C$.

**TABLE I.** Calculated principal-axis values of the dipolar coupling tensor $A^{calc}_\mu$ and dipolar fields $B^{calc}_\mu$ at the probable $\frac{1}{2}$, 0.015 $\mu^+$ site in NdOs$_4$Sb$_{12}$.

| Crystal axis | $\mu^+$ site coordinate | $A^{calc}_\mu$ (G/$\mu_B$) | $B^{calc}_\mu$ (G) |
|-------------|-------------------------|----------------------------|------------------|
| a           | -1                      | 150.6                      | 280.5            |
| b           | 0                       | -62.6                      | -108.3           |
| c           | 0.15                    | -88.0                      | -152.2           |

**FIG. 6.** (Color online) Early-time $\mu^+$ spin polarization from weak-LF-µSR in Pr$_{0.25}$Nd$_{0.75}$Os$_4$Sb$_{12}$, $H_L = 14.8$ Oe. Curve: fit of Eq. (7) to the data.
Compared to data from NdOs$_4$Sb$_{12}$ at 25 mK (Fig. 1) the oscillation is almost completely damped, indicating a broad distribution of local fields. The deep minimum of the Gaussian KT function (Fig. 2) is not observed, and we have therefore fit the GbG KT polarization function $G_{GbG}(t)$ [Eq. (1)] to the data. As in Sec. IIIA we take dynamic relaxation into account via an exponential damping factor:

$$G_{GbG}^{\text{dmpd}}(t) = e^{-\lambda t}G_{GbG}(t).$$

(7)

The fit of Eq. (7) to the data at 25 mK, the early-time portion of which is shown in Fig. 3, is tolerable but not perfect; it is, however, considerably better (reduced $\chi^2 = 1.16$) than that of a number of other candidate functions as follows.

(1) The damped cosine and damped Bessel functions discussed in Sec. IIIA ($\chi^2 = 1.33$ and 1.39, respectively).

(2) The “$\delta$-function/Gaussian” function

$$\frac{1}{3} + \frac{1}{3}[\cos \omega t - (\Delta^2 t/\omega) \sin \omega t] \exp(-\frac{1}{2} \Delta^2 t^2)$$

($\chi^2 \approx 3$).

(3) The “power KT” function

$$\frac{1}{3} + \frac{1}{3}(1 - \lambda t - \Delta^2 t^2) \exp(-\frac{1}{2} \Delta^2 t^2)$$

($\chi^2 = 1.31$).

(4) The “Voigtian KT” function

$$\frac{1}{3} + \frac{1}{3}(1 - \Delta) \exp(-\frac{1}{2} \Delta^2 t^2)$$

($\chi^2 = 1.37$).

The functions in (1) and (2) above oscillate with defined nonzero frequencies, whereas the last two are phenomenological interpolations between the Gaussian and Lorentzian KT functions. The poor fits to the oscillating functions are clear evidence that for $x = 0.75$ the spread in $B_y$ is considerably greater than the average.

Figure 2 gives the temperature dependencies of $\Delta_{\text{eff}}$, $\lambda_L$ and $R$ from fits to Eq. (7). Below $T_C$, $\Delta_{\text{eff}}$ increases in an order-parameter-like fashion with decreasing temperature, to $0.2 \pm 0.3$ $\mu$S$^{-1}$ at 25 mK. This is $\sim 20\%$ of $\omega_0$ in NdOs$_4$Sb$_{12}$ at the same temperature, to be compared with the much smaller decrease in transition temperature; $T_C(x=0.75) \simeq 0.7T_C(x=1)$. The change in polarization behavior between $x = 0.75$ and 1 is drastic, and the average frequency for $x = 1$ should not be compared in detail to the spread in frequencies for $x = 0.75$. Nevertheless, the difference for this relatively light Pr doping is quite striking. In the neighborhood of $T_C$ $\Delta_{\text{eff}}(T)$ varies rather smoothly without an abrupt transition, suggesting an effect of nonzero $H_L$ and/or an inhomogeneous spread of transition temperatures. The inset of Fig. 7 shows that $R$ is nearly constant ($\sim 0.6$) at low temperatures, jumping suddenly to $\sim 1$ near $T_C$.

The longitudinal rate $\lambda_L$ behaves similarly in the $x = 1$ and $x = 0.75$ samples, exhibiting a cusp near the magnetic transition. For the $x = 0.75$ sample, however, the cusp occurs at a temperature well below $T_C$, where $\Delta_{\text{eff}}$ has increased to more than 60% of its low-temperature value. This behavior may also be due to a distribution of transition temperatures. It should be noted, though, that Eq. (7) characterizes the entire polarization function and hence the entire sample volume; the good fits to this function are thus evidence against macroscopic inhomogeneity or phase separation.

C. Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$, $x = 0.45$, 0.50, and 0.55

In the concentration range $0.45 \leq x \leq 0.55$ the transition from static magnetism to superconductivity is occurring, perhaps with coexistence of the two phases on the microscopic scale and with the possibility of one or more quantum critical points near $x = 0.5$. Figure 8 shows early-time $\mu^+$ spin polarization data for alloys with $x = 0.45$, 0.50, and 0.55. The data for $x = 0.75$ and 1.00, discussed above, are repeated for comparison. As is the case for Pr$_{0.25}$Nd$_{0.75}$Os$_4$Sb$_{12}$ (Sec. IIIIB), the data are fit best by the damped GbG KT function [Eq. (7)]. The two-component structure associated with a distribution of quasistatic local fields is present but in attenuated form, since the damping rate is large in these alloys.

Figure 9 gives the temperature dependencies of $\Delta_{\text{eff}}$, $R$, and $\lambda_L$ for $x = 0.45$, 0.50, and 0.55. Both rates increase significantly below 0.3–0.5 K. The increase of $\Delta_{\text{eff}}$ indicates the onset of a distribution of quasistatic local fields in this temperature range. The transitions occur close to temperatures determined from ac susceptibility measurements [7].

In this Nd concentration range $\Delta_{\text{eff}}$ is suppressed sig-
FIG. 8. (Color online) (a) Early-time $\mu^+$ spin polarization from LF-$\mu$SR in Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$, $x = 0.45$ ($H_L = 15.9$ Oe), $0.50$ ($H_L = 16.1$ Oe), and $0.55$ ($H_L = 16.3$ Oe), $T = 25$ mK. Curves ($x \neq 1$): fits of Eq. (7) to the data. (b) $x = 0.75$ and 1.00 (data and fits of Figs. 6 and 4, respectively) for comparison.

Significantly compared to $\omega_0$ in NdOs$_4$Sb$_{12}$, continuing the trend found in Pr$_{0.25}$Nd$_{0.75}$Os$_4$Sb$_{12}$ (Sec. III B). It can be seen in Fig. 6 that $\Delta_{\text{eff}}$ never exceeds $\sim 5.5$ $\mu$s$^{-1}$ at low temperatures. This corresponds to a spread of $\sim 65$ Oe in fields at $\mu^+$ sites, about 11% of the average field in NdOs$_4$Sb$_{12}$. A decrease in $\Delta_{\text{eff}}$ is expected due to the dilution of the Nd$^{3+}$ moment concentration [39] but not to this extent, as discussed in Sec. IV. The low-temperature values of $\Delta_{\text{eff}}$ do not vary monotonically with $x$, but exhibit a marked minimum for $x = 0.50$. The width ratios $R(T)$ [Inset of Fig. 6a] behave remarkably similarly for the $x = 0.45$, 0.50, and 0.55 alloys: at low temperatures $R \approx 0.35$, and then increases toward 1 at $\sim 0.25$ K more continuously than for $x = 0.75$ [Inset of Fig. 4]. The dynamic relaxation rates $\lambda_L(T)$ [Fig. 6b] differ considerably from the corresponding data for $x = 0.75$ and 1 in that there is no sign of a peak at or near $T_C$ and the rates remain large down to 25 mK.

Figure 10 shows LF-$\mu$SR spin polarization in Pr$_{0.55}$Nd$_{0.45}$Os$_4$Sb$_{12}$ at $T = 25$ mK for $H_L$ in the range 15–821 Oe. For intermediate fields, the field independence at early times followed by the increase in the late-time fraction is characteristic of decoupling by the applied field (Sec. III B). This is confirmation that the early-time relaxation is static and not dynamic in nature.

D. Pr$_{0.75}$Nd$_{0.25}$Os$_4$Sb$_{12}$

This alloy is superconducting below a transition temperature $T_C = 1.3 \pm 0.1$ K from ac susceptibility measurements, compared to $\sim 1.8$ K for the end compound PrOs$_4$Sb$_{12}$ [7]. The suppression of superconductivity by Nd doping has been discussed [7] in two alternative scenarios: two-band superconductivity, as found in PrOs$_4$Sb$_{12}$ [40, 41], and the Fulde-Ferrell multiple pair-breaking theory [42]. The main issues addressed by $\mu$SR experiments are therefore the magnetism associated with Nd$^{3+}$ moments, and its effect on the superconductivity of this alloy.
1. Transverse Field

TF-µSR experiments were carried out in Pr$_{0.75}$Nd$_{0.25}$Os$_4$Sb$_{12}$ in an applied field $H_T > H_{c1}$ ($T=0$). Damped oscillations were observed both above and below $T_c$. The asymmetry data $[A_0 G(t)]$ in Eq. (1) were fit to a cos function with combined Gaussian and exponential damping:

$$ G_{TF}(t) = \exp \left( -\frac{1}{2} \sigma_T^2 t^2 - \lambda_T t \right) \cos (\omega_t t + \phi), \quad \omega_T = \gamma_\mu B_\mu, $$

(8)

with an undamped oscillation from muons that stopped in the silver plate and cold finger. Combined Gaussian and exponential damping is necessary to fit TF-µSR in PrOs$_4$Sb$_{12}$.

Figure 11 shows weak-TF-µSR asymmetry data obtained from Pr$_{0.75}$Nd$_{0.25}$Os$_4$Sb$_{12}$ for $H_T = 107.5$ Oe, at 1.604 K (above $T_c$) and 25 mK (well below $T_c$). The signal from muons that did not stop in the sample has been subtracted. It can be seen that at 25 mK the precession frequency decreases and the damping rate increases markedly compared to 1.604 K.

Figure 12 gives the temperature dependencies of the parameters obtained by fitting Eq. (8) to the data. The value of $\gamma_{\mu}$ for the surrounding silver serves as a reference, and is also plotted in Fig. 12(a). The decrease in $\gamma_\mu$ and increase in $\sigma_T$ in the superconducting state of the sample are expected from the diamagnetic response and the field distribution in the vortex lattice, respectively. But normally these changes begin much more abruptly just below $T_c$, as is observed in PrOs$_4$Sb$_{12}$.

The curve in Fig. 12(b) is a fit of the relation

$$ \sigma_T(T) = \sqrt{\sigma_{ST}^2(T) + \sigma_{nT}^2}, $$

(9)

where the temperature dependence of the superconducting-state Gaussian rate $\sigma_{ST}$ is modeled by

$$ \sigma_{ST}(T) = \left\{ \begin{array}{ll} \sigma_{ST}(0) \left[ 1 - (T/T_c)^n \right], & T < T_c, \\ 0, & T > T_c. \end{array} \right. $$

(10)

and $\sigma_{nT}$ is the normal-state Gaussian rate due to nuclear dipolar fields. The two contributions are added in quadrature because the nuclear dipolar fields that give rise to $\sigma_n$ are randomly oriented and uncorrelated with the vortex-lattice field. The power law used to model $\sigma_{ST}(T)$ [Eq. (10)] is merely to indicate the form through the exponent $n$, and has little physical significance, although $n = 4$ is found in an early two-fluid phenomenology. The fits yield $n = 1.4 \pm 0.2$, much smaller than the usual values (3–4) in conventional superconductors. The exponential rate $\lambda_T$, which is quite significant, is not approximately constant, as in PrOs$_4$Sb$_{12}$, but increases below $T_c$ and exhibits an inflection point at $\sim 0.5$ K.

There is no evidence for “freezing” of the Nd$^{3+}$ moments. Static magnetism of full Nd$^{3+}$ moments would affect both the muon precession frequency and the damping much more strongly than observed. Further evidence for the lack of spin freezing is discussed below.

2. Zero Field

In most conventional superconductors the only ZF-µSR relaxation mechanism is provided by nuclear dipolar fields at $\mu^+$ sites. These are not affected by superconductivity, so that the relaxation rate is constant through the transition. The $\mu^+$ spin relaxation is then well fit by the KT function [Eq. (3)]. In PrOs$_4$Sb$_{12}$, however, it was necessary to use the exponentially damped Gaussian
FIG. 12. (Color online) Temperature dependencies of weak-TF-µSR parameters in Pr$_{0.75}$Nd$_{0.25}$Os$_4$Sb$_{12}$, $H_T = 107.5$ Oe. (a) Average µ$^+$ fields $\overline{B}_\mu$ from µ$^+$ precession in sample and Ag reference. (b) Static Gaussian transverse relaxation rate $\sigma_T$ and exponential transverse relaxation rate $\lambda_T$. Curve: fit of Eq. (11) to the data. Arrows: superconducting transition temperature $T_c$.

The KT function

$$G_{KT}^{dmpd}(t) = e^{-\lambda_\mu t} G_{KT}(t).$$

The exponential damping was attributed to dynamic fluctuations of hyperfine-enhanced $^{141}$Pr nuclear moments [43]. Equation (11) models the case where the local fields fluctuate around static averages rather than around zero [44]. Alternatively, the “dynamic KT function” [24], appropriate when the µ$^+$ local fields fluctuate as a whole around zero with a fluctuation rate $\nu$, might be considered.

Figure 13 shows the ZF µ$^+$ spin polarization in Pr$_{0.75}$Nd$_{0.25}$Os$_4$Sb$_{12}$ at a number of temperatures in the range 25 mK–2.50 K. As in PrOs$_4$Sb$_{12}$, the data can be well fit with Eq. (11) at all temperatures, with a significant contribution by the exponential damping. The dynamic KT function scenario seems unlikely, assuming that $\nu$ decreases with decreasing temperature. If $\nu \ll \Delta$ the overall relaxation of the dynamic KT function would decrease monotonically with decreasing $\nu$ [26], contrary to observation (Fig. 13). If $\nu \gg \Delta$ the relaxation is in the motionally narrowed limit and the rate increases with decreasing $\nu$, but then the relaxation would be exponential at all temperatures [26], again contrary to observation. We conclude that the damped static KT function is the better choice.

The temperature dependencies of $\Delta$ and $\lambda_L$ are shown in Fig. 14. In contrast to the transverse-field Gaussian rate $\sigma_T$ [Fig. 12(b)], the zero-field Gaussian rate $\Delta$ shows almost no temperature dependence through the superconducting transition down to 25 mK. The normal-state values of $\Delta$ and $\sigma_T$ are similar and essentially the same as in PrOs$_4$Sb$_{12}$, consistent with their contribution to Sb nuclear dipolar fields [10, 12]. The dynamic rate $\lambda_L \simeq 0.1 \mu s^{-1}$ in the normal state is also essentially the same as in PrOs$_4$Sb$_{12}$, but increases dramatically below ~0.4 K, indicating the onset of a new relaxation mechanism at this temperature.

We note that the observed constant $\Delta$ here and $\Delta_{\text{eff}}$ in Pr$_{0.75}$Nd$_{0.25}$Os$_4$Sb$_{12}$ are in contrast to the increase of $\Delta$ observed in PrOs$_4$Sb$_{12}$ below $T_c$ and attributed to broken time-reversal symmetry in the superconducting state [14, 17]. Nd doping appears to have restored time-
reversal symmetry in the superconductivity of the alloys. This seems somewhat paradoxical, since in the BCS theory spin scattering of conduction electrons breaks time-reversal Cooper pairs.

In Fig. 15 the temperature dependencies of $\lambda_L$ for $H = 0$ and $\lambda_T$ for $H_T = 107.5$ Oe are compared. Although both relaxation rates show upturns in the region 0.4–0.5 K, they are clearly different: $\lambda_T$ is larger than $\lambda_L$, and exhibits a jump at $T_c$ [15]. These features are discussed in more detail in Sec. IV.

3. $T = 25$ mK

Further characterization of the unusual $\mu^+$ spin relaxation behavior observed in ZF-$\mu$SR was obtained from LF-$\mu$SR experiments in Pr$_{0.75}$Nd$_{0.25}$Os$_4$Sb$_{12}$ at $H_L = 100$ and 200 Oe. $T = 25$ mK. These fields are an order of magnitude larger than that needed to decouple the field distribution width of $\sim$1 Oe determined from the ZF relaxation rates [20], and hence would completely suppress the relaxation if it were due solely to static fields. But Fig. 10 shows that $\mu^+$ spin relaxation in these fields is considerable, even though reduced by the field.

The LF polarization function is subexponential, i.e., exhibits more upward curvature than an exponential function, for $H_L \geq 100$ Oe. This signals an inhomogeneous distribution of dynamic relaxation rates, with the initial slope of $G(t)$ giving the average rate, and slowly relaxing regions dominating at late times after the rapidly relaxing regions have lost their spin polarization. The power exponential function

$$G_{pe}^{dmpd}(t) = G_{pe}(t) G_{KT}(t)$$

was used to fit the ZF data. The resulting value of $\beta$ for $H_L = 0$ is close to 1 (inset of Fig. 10), justifying our previous use of simple exponential damping in this case, but $\beta$ decreases with increasing field. The value of $\lambda_L$ in the stretched exponential is not the average rate but a rough characterization of the relaxation; $1/\lambda_L$ is the time at which $G_{pe}(t)$ has decreased to $1/e$ of its initial value. Different values of $\lambda_L$ should not be compared if $\beta$ is also varying, since then the shape of the polarization function is changing.

IV. CONCLUSIONS

a. Phase diagram. Magnetic transition temperatures obtained from weak-LF-$\mu$SR in Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$, $0.45 \leq x \leq 1$ and the superconducting transition from TF-$\mu$SR in the $x = 0.25$ sample are in good agreement with previous results [3] (Fig. 11). In the latter sample, however, there is no sign of a transition in other data at $\sim 0.5$ K, where a marked increase in ZF-$\mu$SR dynamic relaxation is seen (Fig. 14). Specific-heat and other measurements on a $x = 0.25$ sample in this temperature range would be desirable.

For all Nd concentrations, the $\mu$SR spectra (after subtraction of the background Ag signal) exhibit either a single component (TF-$\mu$SR) or the two-component structure that is intrinsic to LF-$\mu$SR. Furthermore, the total sample asymmetry is observed not to change within $\sim 5\%$ at the transitions, magnetic or superconducting; there is

FIG. 15. (Color online) Comparison of spin relaxation rates $\lambda_L$ ($H = 0$) and $\lambda_T$ ($H_T = 107.5$ Oe) in Pr$_{0.75}$Nd$_{0.25}$Os$_4$Sb$_{12}$. Arrow: superconducting transition temperature $T_c$.

FIG. 16. (Color online) $\mu^+$ spin polarization in Pr$_{0.75}$Nd$_{0.25}$Os$_4$Sb$_{12}$, $T = 25$ mK. Curves: fits of Eq. (13) ($H_L = 0$) and Eq. (12) ($H_L = 10$ and 200 Oe) to the data. Inset: power exponent $\beta$ vs longitudinal field.
no “lost asymmetry.” Thus there is no evidence in our data for phase separation anywhere in the phase diagram.

b. Magnetism in \(\text{NdOs}_4\text{Sb}_{12}\). The weak-LF-\(\mu\)SR data from \(\text{NdOs}_4\text{Sb}_{12}\) reveal a fairly standard ferromagnet, with evidence of disorder from the damped Bessel-function form of the \(\mu^+\) polarization function below \(T_C\). A distribution of \(B_\mu\) magnitudes is necessary for this damping: a random distribution of field directions with fixed field magnitude results in an undamped oscillation in the polarization function. The most likely origin of this distribution is disorder in \(\text{Nd}^{3+}\) orientations in the ferromagnetic state. We note, however, that as discussed below much broader spreads of \(B_\mu\) are observed in Pr-diluted alloys, the mechanism for which might play a role in \(\text{NdOs}_4\text{Sb}_{12}\).

The observation of a peak at \(T_C\) in the dynamic relaxation rate indicates critical slowing down of \(\text{Nd}^{3+}\) spin fluctuations as the transition is approached from above, and thus is evidence that the transition is second order. Critical slowing down is somewhat in disagreement with the conclusion that the transition is mean-field-like [3], however, since in that case the critical region around \(T_C\) would be expected to be quite small.

c. Magnetism in \(\text{Pr}_{0.25}\text{Nd}_{0.75}\text{Os}_4\text{Sb}_{12}\); reduced moments or reduced coupling strengths? The markedly reduced value of \(\Delta_{\text{eff}}\) (and hence \(B_\mu\)) and the striking difference in polarization function compared to NdOs4Sb12 (Sec. 11.13), are the salient results of \(\mu\)SR experiments in \(\text{Pr}_{0.25}\text{Nd}_{0.75}\text{Os}_4\text{Sb}_{12}\). Noakes [38, 41] has reported Monte Carlo calculations of \(B_\mu\) distributions under various conditions of random site dilution, moment direction, and moment magnitude. For fixed moment magnitudes and random moment orientations he found that with increased dilution the \(\mu^+\) spin polarization function retains the Gaussian KT form with its deep minimum (Fig. 2) down to moment concentration \(~0.5\). This is clearly not observed in \(\text{Pr}_{0.25}\text{Nd}_{0.75}\text{Os}_4\text{Sb}_{12}\) (Fig. 6). Noakes showed [47] that an inhomogeneous distribution of moment magnitudes can give rise to the shallower minimum exhibited by the GbG KT function [Eq. (3)], \(R \gtrsim 0.3\); cf. Fig. 2 [31].

The usual mechanism for local-moment suppression in metals is the Kondo effect, which for \(4f\) ions is normally observed only in Ce-, Yb-, and (very occasionally) Pr-based materials. This restriction to the ends of the lanthanide series is well understood [25]: the energy difference between the \(4f\) level and the Fermi energy increases with increasing atomic number. This decreases the effective \(sD-f\) exchange interaction due to \(4f\)-conduction electron hybridization at the Fermi surface, to which the Kondo temperature is exponentially sensitive, and thus quenches the Kondo effect [49]. An inhomogeneous Kondo effect may be involved in Pt- and Cu-doped CeNiSn, where the GbG polarization function also fits the ZF-\(\mu\)SR data well [50]. To the authors’ knowledge, however, Kondo screening has never been observed in Nd compounds.

As noted above, the \(\text{Pr}^{3+}\) ions are in nonmagnetic crystal-field ground states, but it should not be assumed that they are magnetically inert. Exchange interactions, mediated by a RKKY-like indirect mechanism, might admix magnetic excited CEF states into the nonmagnetic \(\text{Pr}^{3+}\) ground state so that they contribute to \(B_\mu\). It is hard to see how \(B_\mu\) would be reduced by this effect, however.

\(\Delta_{\text{eff}}\) is a sum of terms, each of which is proportional to the squared product of the static \(\text{Nd}^{3+}\) moment magnitude and the \(\text{Nd}^{3+}-\mu^+\) coupling strength [51]. An alternative mechanism for variation and reduction of \(B_\mu\) might invoke a negative contribution of admixed \(\text{Pr}^{3+}\) states to the indirect \(\text{Nd}^{3+}-\mu^+\) coupling. Variation of moment magnitudes and coupling strengths seems indistinguishable because of the product form, and the latter has the advantage of avoiding a mysterious reduction of \(\text{Nd}^{3+}\) moments. As far as we know, such a mechanism has not been addressed theoretically.

d. Magnetism in \(\text{Pr}_{1-x}\text{Nd}_x\text{Os}_4\text{Sb}_{12}\), \(0.45 \leq x \leq 0.55\). The \(\mu\)SR experiments in this concentration range are dominated by effects of \(\text{Nd}^{3+}\) magnetism; there is no sign in the data of a superconducting contribution to \(B_\mu\). The superconducting state with broken time-reversal symmetry found in \(\text{PrOs}_4\text{Sb}_{12}\) is not found in these alloys.

The pattern of good fits to the GbG polarization function with reduced values of \(\Delta_{\text{eff}}\) is continued. Figure 17 shows the dependence of \(\Delta_{\text{eff}}\) on Nd concentration for the alloys (including \(x = 0.25\), where \(\Delta_{\text{eff}}\) vanishes, and \(x = 0.75\)), together with the experimental preces-

![FIG. 17. (Color online) ZF- and weak-LF-\(\mu\)SR GbG effective spin relaxation rates \(\Delta_{\text{eff}}(x)\) (squares) and experimental \(\mu^+\) precession frequency \(\omega_{\mu}\) (circles) vs Nd concentration \(x\) in \(\text{Pr}_{1-x}\text{Nd}_x\text{Os}_4\text{Sb}_{12}\). Cross: calculated \(\mu^+\) precession frequency \(\omega_{\mu}\) from dipolar \(\mu^+\) local fields, assuming saturation \(\text{Nd}^{3+}\) moment \(\mu_{sat} = 1.73\mu_B\) (Ref. 3) and uniform (ferromagnetic) moment alignment. Curve: calculated \(\Delta_{\text{eff}}(x)\) from dipolar \(\mu^+\) local fields \([= \text{Van Vleck rate} \sigma_{VV}(x)]\), see text, assuming the saturation \(\text{Nd}^{3+}\) moment of 1.73\(\mu_B\) and random moment orientations.](image-url)
sion frequency \(\omega_\mu^{\text{eff}}\) and the maximum calculated pre-
cession frequency \(\omega_\mu^{\text{dip}}\) assuming dipolar interactions only (Sec. III A) in NdOs\(_4\)Sb\(_4\).

In general the initial curvature of \(G(t)\) is related to the \(\mu^+\) precession frequency distribution: \(G_{\mu^+}(t) = 1 - \frac{1}{2} \sigma_{\mu^+} \cdot \mathbf{t}^2 + \cdots\), where \(\sigma_{\mu^+}\) is the Van Vleck second moment of the high-field resonance line [20, 51]. This result is independent of the functional form of the static field distribution, as long as the second moment is defined [52]. For the GbG KT function \(G_{\text{GbG}}(t) = 1 - \Delta_{\text{eff}}^2 t^2 + \cdots\) (Sec. III A), so that \(\Delta_{\text{eff}} = \sigma_{\mu^+}\). Furthermore, in a di-
luted lattice of randomly oriented moments the second-
oment is proportional to the moment concentra-
tion \(\hat{n}\): \(\sigma_{\mu^+}(x)/\sigma_{\mu^+}(x=1) = \sqrt{x}\). A lattice-sum calculation of \(\sigma_{\mu^+}\) for dipolar coupling of 1.73\(\mu_B\) Nd\(^{3+}\) moments to \(\mu^+\) spins at 0.1, 0.13 sites in NdOs\(_4\)Sb\(_4\) yields \(\sigma_{\mu^+}(x=1) = 18.8\) \(\mu_s\) \(\text{s}^{-1}\). The curve in Fig. 17 gives \(\Delta_{\text{eff}}^2(x) = \sigma_{\mu^+}(x) = 18.8/\sqrt{x} \) \(\mu_s\) \(\text{s}^{-1}\) \(\text{cm}^{-1}\), a lower limit for the expected relaxation rate since an additional RKKY-based \(\mu^+\)-Nd\(^{3+}\) transferred hyperfine interaction is present (Sec. III A). The observed values of \(\Delta_{\text{eff}}\) fall significantly below this curve.

At 25 mK \(\Delta_{\text{eff}}\) depends significantly on \(x\), exhibiting a marked minimum at \(x = 0.50\) (Fig. 17). This behavior is also hard to understand: in general \(\Delta_{\text{eff}}\) would be expected to track \(T_C\), i.e., \(\Delta_{\text{eff}}\) should decrease monotonically with decreasing \(x\) in the neighborhood of \(x_C\) (Fig. III A). The minimum suggests that the moment suppress-
ion is related to an approach to quantum criticality at \(x \approx 0.5\).

The behavior of the dynamic \(\mu^+\) relaxation at low temperatures differs significantly between the \(x = 0.45, 0.50\), and 0.55 alloys and the higher-concentration ma-
erials. In the former the rate \(\lambda_L\) remains large as \(T \rightarrow 0\) (Fig. III b) rather than decreasing with decreasing temperature below \(T_C\) as in the \(x = 0.75\) alloy and NdOs\(_4\)Sb\(_4\) (Figs. 4 and 9) respectively. This behavior signals the persistence of spin fluctuations to low temperatures. Normally the amplitude of thermal fluctuations decreases at low temperatures with the decreasing popu-
lation of low-lying excitations such as spin waves. Per-
sistent spin dynamics (PSD) such as seen here are often found in geometrically frustrated spin liquids [54], but only occasionally in systems with long-range order [55]. PSD may be related to the absence of spin freezing in the \(x = 0.25\) alloy, discussed below. They might also be considered as a source of the reduced static \(\mu^+\) fields (Fig. III), except that this is also seen in the \(x = 0.75\) alloy where PSD are absent (Fig. 4).

e. \(P_{0.75}Nd_{0.25}Os_{4}Sb_{12}\): superconductivity and no spin freezing. As noted above, the observation that the TF-\(\mu\)SR asymmetry does not change in the supercon-
ducting state (Fig. III) is strong evidence that the latter occupies essentially all of the \(x = 0.25\) sample; there is no evidence for separation of superconducting and magnetic phases.

From Fig. 14 the ZF-\(\mu\)SR Gaussian KT rate \(\Delta(T)\) does not deviate from its average \(\Delta(T) = 0.125 \pm 0.008 \mu_s^{-1}\) by more than \(\sim 0.02 \mu_s^{-1}\), i.e., the \(\mu^+\) field does not change by more than \(\sim 0.8\) G over the entire temperature range. This is even more compelling evidence against static Nd magnetism than the TF-\(\mu\)SR data (Fig. 12), since frozen Nd\(^{3+}\) moments \(!1_{\mu_B}\) would produce local fields much greater than 0.8 G. An estimate of such a field can be obtained from the calculated rms dipolar relaxation rate (curve in Fig. 17), which is \(\sim 10 \mu_s^{-1} \approx 1.08 \times 120\) G for \(x = 0.25\). Since this calculation uses the measured saturation moment of 1.73\(\mu_B\)/Nd ion in NdOs\(_4\)Sb\(_4\), the data suggest an upper limit on any static moment of \(\sim 10^{-2}\mu_B\) in \(P_{0.75}Nd_{0.25}Os_{4}Sb_{12}\).

\(\mu\)SR is sensitive to the onset of static magnetism independent of the degree of order; the technique is as applicable to spin glasses as to ordered magnets. [21–25]. The absence of evidence for static Nd\(^{3+}\) magnetism, ordered or disordered, down to 25 mK in \(P_{0.75}Nd_{0.25}Os_{4}Sb_{12}\) is perhaps the most surprising result of this study. If \(T_C\) were to scale as the Nd concentration the freezing tem-
perature would be \(\sim 0.2\) K; its suppression by at least an order of magnitude is extraordinary.

In \(P_{0.75}Nd_{0.25}Os_{4}Sb_{12}\) the values at \(T = 0\) of the diamagnetic shift \(\mathcal{B}_0/\mu_B - 1 \approx -0.07\) and Gaussian relaxation rate \(\sigma_s = 0.61 \pm 0.03 \mu_s^{-1}\) are of the same order of magnitude as in the end compound PrOs\(_4\)Sb\(_4\) for comparable \(H_T\). In the London limit (penetration depth \(\lambda \gg 0\) coherence length \(\xi\)) the rms width \(\Delta B^2\) of the vortex-lattice field distribution in a conventional super-
conductor is related to the London penetration depth \(\lambda\) by \(\Delta B^2 = 0.00371\Phi_0/\lambda^4\), where \(\Phi_0\) is the flux quantum [56]. Assuming that \(\gamma_{\mu}(\Delta B^2)^{1/2}\) is approximated by the TF-\(\mu\)SR Gaussian relaxation rate \(\sigma_s\), at least near \(T = 0\) [57], the zero-temperature fit value of \(\sigma_s\) yields \(\lambda = 4190\) \(\Lambda\), compared to 3610 \(\Lambda\) in PrOs\(_4\)Sb\(_4\).

The increase below \(T_c\) of the diamagnetic shift and \(\sigma_s\) are much faster in superconducting PrOs\(_4\)Sb\(_4\) than for \(x = 0.25\) (Fig. 12). One possible mechanism for this behavior is strong pair-breaking by Nd\(^{3+}\) spins. A more speculative possibility is that the mass renormaliz-
ation characteristic of heavy-fermion superconductivity [58], which affects the temperature dependence of the penetration depth [59], is strongly modified by interaction with fluctuating Nd moments.

The increases of both \(\lambda_T(T)\) and \(\lambda_L(T)\) below \(T_c\) (Fig. 13) indicate a marked effect of superconductivity on the Nd\(^{3+}\) spin dynamics. As noted in Sec. III A an increase in relaxation rate with decreasing temperature signals slowing down of spin fluctuations. The ob-
servation that \(\lambda_T(T) > \lambda_L(T)\) is not surprising, since a contribution to relaxation from a static field distribution is possible in TF-\(\mu\)SR but not for the 1/3 component of ZF-\(\mu\)SR. The extra transverse relaxation may reflect inhomogeneity in the vortex lattice. The increases of both \(\lambda_T(T)\) and \(\lambda_L(T)\) below 0.4–0.5 K may be due to a further reduction of the Nd spin fluctuation rate at a crossover or transition, although there is no anomaly in \(H_{c2}(T)\) in \(P_{0.75}Nd_{0.25}Os_{4}Sb_{12}\) at this temperature [7].
f. Summary. μSR data from the Pr$_{1-x}$Nd$_x$Os$_4$Sb$_{12}$ alloy series reveal a disordered reduction by Pr doping of the spontaneous static $\mu^+$ local field due to static Nd$^{3+}$-ion magnetism that is is well beyond that expected from dilution (Fig. 17). Kondo-like reduction of Nd$^{3+}$ ion magnetism that is is well beyond that expected from involving the Nd$^{3+}$ moments is highly unlikely, suggesting by default an effect to elucidate their mechanisms. The origins of these phenomena remain unclear, and future work is called for to elucidate their mechanisms.

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