Successive magnetic transitions and static magnetic order in RCoAsO ($R = \text{La}, \text{Ce}, \text{Pr}, \text{Nd}, \text{Sm}, \text{Gd}$) confirmed by muon-spin rotation and relaxation

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The presence of antiferromagnetic (AF) order seems to be a common feature for the parent compounds of cuprate as well as some pnictide high-temperature superconductors. In order to search for antiferromagnetic order in the closely related rare-earth cobalt arsenic oxides (RCoAsO), we have measured muon-spin rotation and relaxation ($\mu^+\text{SR}$) spectra on a series of powder samples with $R = \text{La}, \text{Ce}, \text{Pr}, \text{Nd}, \text{Sm}, \text{Gd}$. It was found that, besides GdCoAsO, all the other five compounds enter into a static ferromagnetic ordered phase below around 70 K ($= T_C$), while additional transitions into a static AF ordered phase were found for NdCoAsO and SmCoAsO. For GdCoAsO, a static ferrimagnetic phase appeared below 60.5 K and then an incommensurate spin-density-wave ordered phase followed below 3.2 K. The AF spin structure was also found to strongly depend on $R$.

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I. INTRODUCTION

The emergence of pnictides and other Fe-based superconductors,2 such as LaFeAsO$_{1−x}$F$_x$,3 has appealed the importance of the interplay between magnetism and superconductivity (SC). This is because the phase diagram of LaFeAsO$_{1−x}$F$_x$ and related compounds5 seems to be similar to those for high-$T_c$ cuprates. For instance, the parent compound, LaFeAsO, enters into an itinerant spin-density-wave (SDW) antiferromagnetic (AF) phase below $T_N \sim 140$ K,6,7 while the parent compounds of high-$T_c$ cuprates are an AF Mott insulator. Upon F-doping for LaFeAsO, $T_N$ is steadily reduced, until finally the AF order is annihilated and superconductivity emerges. In case of F doped into the [La$_2$O$_2$] layers, the expected result is that the number of Fe-3$d$ electrons increases.

A more straightforward method is a partial substitution for Co by Ce—i.e., doping one more 3$d$ electron, and hereby doped electrons directly into the [Fe$_2$As$_2$] layers. It has been recently shown that for LaFe$_{1−x}$Co$_x$AsO the AF order is destroyed at $x = 0.05$ and maximum $T^{\text{onset}}_N = 14.3$ K is achieved for $x = 0.11$.8 For $x = 1$, namely LaCoAsO, neither AF order nor SC appears, but instead metallic resistivity and ferromagnetic (FM) order with $T_C = 60$ K.9 This is rather surprising, since the crystal structure (see Fig. 1) as well as the electronic structure around the Fermi level, consisting of Co-3$d$ and As-3$p$ hybridized orbitals, are similar to that of LaFe$_{1−x}$Co$_x$AsO. However, when La is replaced by Nd, Sm, or Gd, such rare-earth-CoAsO (RCoAsO) exhibits an additional magnetic transition at $T_m$ ($T_m < T_C$) according to susceptibility ($\chi$) measurements.10 A sudden decrease in $\chi$ at $T_m$ is likely to indicate the occurrence of an AF transition at $T_m$.10 In the context of competing AF order and SC, details of the magnetic ground state in the RCoAsO compound series is of high importance.

Indeed, recent neutron experiments on NdCoAsO$^{11,12}$ clarified the existence of long-range FM order below 70 K and long-range AF order below 15 K. Thus $T_m$ is confirmed as $T_N$ for NdCoAsO. Besides these works, the magnetic nature of RCoAsO has been investigated by macroscopic measurement techniques, such as magnetization, resistivity, and heat-capacity measurements for LaCoAsO$^{13,14}$ CeCoAsO$^{15}$ NdCoAsO$^{14}$ and SmCoAsO$^{14,16,17}$ Furthermore, the magnetism of LaCoAsO and CeCoAsO was studied by $^{75}$As- and $^{139}$La-NMR$^{15,18}$ while there are no reports on the microscopic magnetic properties of the other RCoAsO compounds.

According to neutron measurements on NdCoAsO$^{11,12}$ the ordered Co moment is estimated as about 0.2 $\mu_B$ in the FM state, but about 0.4 $\mu_B$ in the AF state. Furthermore, the Nd moment orders only in the AF state, and the ordered value reaches about 1.4 $\mu_B$ at 0.3 K. Since such FM/AF order generates a static internal magnetic field ($H_{\text{int}}$) at the interstitial sites where implanted muons are located, we can expect clear precession signals in the muon-spin rotation and relaxation ($\mu^+\text{SR}$) spectra under zero applied field (ZF) below $T_C$. If there are multiple muon sites with different $H_{\text{int}}$ in the lattice, we could confirm and/or deduce the spin structure from the intensity ratios of several $H_{\text{int}}$ signals, as in BaCo$_2$O$_3$, $^{17}$Co$_2$O$_3$, and NaV$_2$O$_4$.21 We have, therefore, measured ZF-$\mu^+\text{SR}$ spectra in the RCoAsO ($R = \text{La}, \text{Ce}, \text{Pr}, \text{Nd}, \text{Sm}, \text{Gd}$) compounds, as $\mu^+\text{SR}$ is especially sensitive to local magnetic environments.

II. EXPERIMENT

Polycrystalline samples of RCoAsO were prepared by a solid-state reaction technique using reagent grade $R$, As, and...
CoO powders. At first, RAs was prepared by the reaction \( R + As \rightarrow RAs \) in an evacuated quartz tube at 550 °C for 5 h and then at 800 °C for 12 h. Finally, the RCoAsO was prepared by the reaction \( RAs + CoO \rightarrow RCoAsO \) in an evacuated quartz tube at 1100 °C for 12 h. Powder x-ray-diffraction analyses showed that all the samples are a single phase of a tetragonal symmetry with space group \( P4/nnm \). The magnetic susceptibility \( \chi \) was measured below 400 K under a field of \( H \leq 1 \) kOe with a superconducting quantum interference device (SQUID) magnetometer. The \( \mu^+ \)SR spectra were measured on surface muon beam lines using the Dolly spectrometer of PSI in Switzerland and the LAMPF spectrometer of TRIUMF in Canada. The approximately 500-mg powder sample was placed in an envelope with \( 1 \times 1 \) cm² area made of 0.05-mm-thick Al-coated Mylar tape in order to minimize the signal from the envelope. The envelope was then attached to a low-background sample holder in a liquid-He flow-type cryostat in the \( T \) range between 1.7 and 150 K. The experimental techniques are described in detail elsewhere.

### III. RESULTS

#### A. LaCoAsO, CeCoAsO, and PrCoAsO

Figure 2(a) shows the zero-field (ZF) \( \mu^+ \)SR time spectra at 50 and 1.7 K for LaCoAsO. A clear oscillation indicates the formation of static ferromagnetic (FM) order in LaCoAsO. Also, since there is only one oscillatory signal in the whole \( T \) range below \( T_C \), all the implanted muons are found to locate at one magnetically equivalent site. In fact, the ZF spectrum was well fitted by a combination of the oscillatory signal and a nonoscillatory relaxing signal:

\[
A_0 P_{ZF}(t) = A_{FM} \cos(2\pi f_\mu t + \phi) \exp(-\lambda_{FM} t) + A_S \exp(-\lambda_S t) ,
\]

where \( A_i \) are the asymmetries (\( A_0 \) is the maximal asymmetry measured in Ag, where the muon polarization \( P = 1 \)), \( \lambda_i \) is the exponential relaxation rate, \( 2\pi f_\mu (\equiv \omega_\mu) \) is the muon Larmor frequency, and \( \phi \) is the initial phase of the oscillatory signal. Figure 2 shows the \( T \) dependences of \( f_\mu \) and \( \chi \), the normalized \( A_{FM} \) (\( A_{FM}/A_0 \)), and \( \lambda_i \) for LaCoAsO, CeCoAsO, and PrCoAsO. For LaCoAsO, the \( f_\mu(T) \) curve exhibits an order-parameter-like \( T \) dependence, suggesting that the FM phase is stable down to 1.7 K. Indeed, the \( f_\mu(T) \) curve is in very good agreement with the \( \chi(T) \) curve over the whole \( T \) range measured, except for temperatures in the vicinity of \( T_C \).

Since \( \phi \sim -17° \) at 1.7 K and is almost \( T \) independent until the vicinity of \( T_C \) (not shown), the FM order is commensurate with the lattice, as expected. In principle, \( \phi \) should be zero for a powder sample with commensurate magnetic order and an ideal Lorentzian or Gaussian field distribution at the muon site. Nevertheless, a fit using a nonzero \( \phi \) gives an overall more acceptable result for the ZF spectrum of LaCoAsO. This is most likely due to a systematic delay in the electronic setup of the present apparatus, since the \(-17°\) delay corresponds to about 1.4 ns, when \( f_\mu = 34.5 \) MHz. Unfortunately, there is no theoretical prediction for the critical \( \phi \) beyond which the magnetic order is incommensurate. However, when \( \phi \leq -45° \), such a phase shift empirically suggests possible formation of incommensurate magnetic order.

The relative amplitude of the oscillatory signal (\( A_{FM}/A_0 \)) is about 0.67, and that for the nonoscillatory signal (\( A_S/A_0 \)) is about 0.33. Both are \( T \) independent until the vicinity of \( T_C \). Therefore, the \( A_S \) signal is assigned as the \( 1/3 \) tail, which is caused by the internal magnetic-field (\( H_{int} \)) component parallel to the initial muon spin polarization. This means that the whole sample enters into the FM ordered phase below \( T_C \). This is also supported by weak transverse field (wTF) \( \mu^+ \)SR measurements, in which the normalized TF asymmetry (\( A_{TF}/A_0 \)) drops from 1 to ~0 at \( T_C \) [see Fig. 2(g)]. Here, \( A_{TF}/A_0 \) is proportional to the volume fraction of paramagnetic phases in the sample.

For CeCoAsO, the overall behavior is similar to that for LaCoAsO. However, below \sim 20 K, \( f_\mu \) decreases slightly with decreasing \( T \), whereas \( \lambda_{FM} \) increases, suggesting a shift of the field distribution at the muon site(s). Note that the \( \chi(T) \) curve also changes shape around 20 K. We wish to point out that recent specific-heat measurements showed the presence of a broad anomaly below approximately 20 K for CeCoAsO, although it was thought to be a Schottky-type anomaly.

For PrCoAsO, as \( T \) decreases from \( T_C \) (\= 7 K), \( f_\mu \) ranges below 4.2 MHz down to 50 K (\= \( T_{m1} \)), and then eventually becomes undetectable below \( T_{m1} \) due to the increase in \( \lambda_{FM} \). When \( T \) is lowered further, two oscillatory signals appear below 32 K (\= \( T_{m2} \)). Thus the ZF spectrum was fitted by a
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FIG. 2. (Color online) Top: the ZF-$\mu^+\nu$ SR time spectra (a) at 1.7 and 50 K for LaCoAsO, (b) at 1.7 and 50 K for CeCoAsO, and (c) at 1.7 and 70 K for PrCoAsO. Bottom: $T$ dependences of [f$_m$, and $\chi$, the normalized $P_{TF}(A/F, A_0)$, and $\lambda$] for [(d), (g), (j)] LaCoAsO, [(e), (b), (k)] CeCoAsO, and [(f), (i), (l)] PrCoAsO. The data (except for $\chi$ and $A_{TF}$) were obtained by fitting the ZF-spectra using Eq. (1). $A_{TF}$ represents a weak transverse field-asymmetry measured with wTF $= 30$ Oe. The susceptibility $\chi$ was measured with $H = 1$ kOe in field cooling mode. Note that the time axis of (c) is a log scale.

combination of Eq. (1) and a second exponentially relaxing cosine function, $A_{FM1}\cos(2\pi f_{m}t + \phi_2)\exp(-\lambda_{FM1}t)$. Although $f_{m1}$ ranges around 30 MHz as in the case for LaCoAsO, the larger signal shows an oscillation with $f_{m1} \sim 3$ MHz down to 1.8 K. Note that, despite the absence of anomalies in the $\chi(T)$ curve below $T_C$, $\mu^+\nu$ SR clearly detects a change in the magnetic behavior of PrCoAsO in the $T$ range between $T_{m1}$ and $T_{m2}$.

B. NdCoAsO and SmCoAsO

For NdCoAsO, SmCoAsO, and GdCoAsO, although the ZF spectra show a clear oscillation at each $T$ below $T_C$, as $T$ decreases from $T_C$, $f_m$ increases abruptly at a certain $T$. This means that each sample enters into an AF ordered phase, as speculated from $\chi$ measurements. In fact, the spectrum was fitted by a combination of Eq. (1) and a second exponentially relaxing cosine function:

$$A_0 P_{TF}(t) = A_{FM} \cos(2\pi f_{m}t + \phi) \exp(-\lambda_{FM}t) + A_{AF} \cos(2\pi f_{m}t + \phi + \lambda_{AF}t) + A_A \exp(-\lambda_A t),$$

where $A_{FM} = 0$ below $T_N$, whereas $A_{AF} = 0$ above $T_N$.

Figure 3 shows the $T$ dependences of $f_m$, $\chi$, $A_0/A_0$, and $\lambda_A$ for NdCoAsO, SmCoAsO, and GdCoAsO. In contrast to LaCoAsO, all three samples are found to enter into static AF ordered phase below $T_N = 15$ K for NdCoAsO and below 41 K for SmCoAsO. The result on GdCoAsO is explained in Sec. IV C. Since $A_{AF}/A_0 = A_{TF}/A_0 \sim 0.67$ below $T_C$, the whole volume of the samples exhibits successive magnetic transitions from a paramagnetic phase to an FM phase, and then to an AF phase, with decreasing $T$.

For NdCoAsO, as $T$ decreases from $T_N$, both $f_{m}$ and $\lambda_{AF}$ increase rapidly, particularly below $\sim 7$ K, indicating the presence of an additional AF transition, i.e., $T_{N1} \sim 15$ K and $T_{N2} \sim 7$ K. Recent neutron-scattering measurements revealed the evolution of the ordered Nd moments with decreasing $T$ below $T_{N2} \sim 3.5$ K, but the Nd moment appears to start increasing below 7 K. In addition, since $\phi \sim -10^\circ$ in the $T$ range between 1.7 K and below the vicinity of $T_C$, the FM and two AF orders should posses a commensurate spin structure. This is consistent with the neutron result.

As $T$ decreases from $T_C$, $f_m$ for SmCoAsO increases abruptly by $\sim 40$ times at $T_{N1} (= 41$ K), although $f_{m}$ for SmCoAsO in the FM phase is comparable to that for NdCoAsO. This implies that the FM spin structure for SmCoAsO is very similar to that for NdCoAsO. Below $T_{N1}$, the $f_m(T)$ curve changes slope ($df_m/dT$) around 17.5 K, where the $\lambda_{AF}(T)$ curve exhibits a broad maximum. This suggests the presence of an additional magnetic transition ($T_{N2} = 17.5$ K), which could correspond to the transition at 5 K recently reported by specific-heat measurements. Outside the vicinities of $T_C$, $T_{N1}$, and $T_{N2}$, since $|\phi| \lesssim 20^\circ$, the FM and two AF orders are thought to be commensurate with the lattice.
C. GdCoAsO

For GdCoAsO, $f_\mu$ does not exhibit a typical $T$ dependence, but rapidly increases with decreasing $T$. Then, as seen in Fig. 4, $f_\mu$ splits into $f_{\mu 1}$ and $f_{\mu 2}$ at 3.2 K, below which the relaxation rates for the two signals are rather large compared with $\lambda_{\text{FM}}$. This behavior indicates the appearance of a complex magnetic-ordered phase below $T_m = 3.2$ K. Although $|\phi| \leq 10^\circ$ above $T_m$, $\phi$ for the predominant $f_{\mu 2}$ signal ranges between $-90^\circ$ and $-54^\circ$ below $T_m$ and reaches $-54^\circ$ at 1.7 K, suggesting the formation of incommensurate (IC) SDW order below $T_N$. Thus $T_m$ should be $T_N$. In fact, the ZF spectrum below $T_N$ is fitted by a combination of a Gaussian relaxing cosine function, a Gaussian relaxing zeroth-order Bessel function of the first kind $[J_0(2\pi f_{\mu 2}t)]$, and an exponentially relaxation function:

$$A_0 P_{ZF}(t) = A_{\text{AF1}} \cos(2\pi f_{\mu 1} t + \phi) \exp\left( -\frac{\sigma_{\text{AF1}}^2 t^2}{2} \right) + A_{\text{AF2}} J_0(2\pi f_{\mu 2} t) \exp\left( -\frac{\sigma_{\text{AF2}}^2 t^2}{2} \right) + A_S \exp(-\lambda_S t),$$

where $f_{\mu 2} > f_{\mu 1}$ and $f_{\mu 2}$ corresponds to the upper limit of $H_{\text{int}}$ at the muon site, whereas $f_{\mu 1}$ corresponds to the lower limit of $H_{\text{int}}$ (see the Appendix).

The $f_{\mu 2}(T)$ curve suggests an order-parameter-like $T$ dependence [see Fig. 3(f)]. This indicates evolution of the

FIG. 3. (Color online) Top: the ZF-$\mu^+$ SR spectra (a) at 1.7 and 20 K for NdCoAsO, (b) at 1.7 and 50 K for SmCoAsO, and (c) at 1.7 and 3.5 K for GdCoAsO. Bottom: $T$ dependences of $[f_\mu, \chi, A_i/A_0,$ and $\lambda_i]$ for [(d), (g), (j)] NdCoAsO, [(e), (h), (k)] SmCoAsO, and [(f), (i), (l)] GdCoAsO. $\chi$ was measured with $H = 1$ kOe in field cooling mode. Note that the $T$ axis for GdCoAsO is a log scale. In (e') and (f'), the $f_\mu$ axis is a linear scale.

FIG. 4. (Color online) Fourier amplitude spectrum for GdCoAsO at 3.5 and 1.7 K. The magnitude of the spectrum at 1.7 K is about 1/20 of that at 3.5 K.
Concerning the variation of $M$ around $T_C = 60.5$ K, a broad peak is clearly seen at $T_C$ in the $M(T)$ curve measured with $H = 10$ and 20 Oe [Fig. 5(b)], suggesting that the transition is not a typical FM transition. Since such an anomaly shifts toward a higher $T$ side with $H$, an FM interaction plays a significant role for determining $T_C$. Furthermore, as $H$ increases further, the anomaly was reported to disappear with $H > 10$ kOe, and the $M(T)$ curve showed a typical Curie-Weiss behavior. This suggests that GdCoAsO is not a ferromagnet, but a ferrimagnet below $T_C$, i.e., $T_C = T_f$.10

IV. DISCUSSION

The parameters obtained by the present study are summarized in Table I and a magnetic phase diagram for the $R$CoAsO series is shown in Fig. 6. We also determined the field distribution width ($\Delta$) caused by nuclear moments in the paramagnetic state from ZF- and longitudinal field (LF-) $\mu^+\text{SR}$ measurements. The characteristic feature of the FM phase is that $f_\mu$ ranges between 20 and 35 MHz for the $R = \text{La}$, Ce, and Gd compounds, but $f_\mu$ is very small for PrCoAsO, NdCoAsO, and SmCoAsO. In order to explain this difference, we attempt to speculate on the FM and AF spin structures of these compounds.

For ferromagnets, the internal magnetic field at a muon site ($H_{\mu}$) is represented by25,26

$$H_{\mu} = H_{dip} + H_L + H_{hf},$$

where $H_{dip}$ is the dipole field, $H_L$ is the Lorentz field, and $H_{hf}$ is the hyperfine field. Furthermore, $H_L$ and $H_{hf}$ are connected to the saturated magnetization ($M_s$) and the local spin density at the muon sites ($\rho_{spin}$) as follows:

$$H_L = 4\pi/3 \times M_s, \quad H_{hf} = 8\pi/3 \times \rho_{spin}.$$

Since $M_s$ ranges between 0.28 and 0.16 $\mu_B$/Co for $R$CoAsO,10,17 $H_L$ is calculated as 78–48 Oe ($f_L = 1.1–0.65$ MHz). However, we cannot obtain the correct $H_{hf}$ without muon Knight shift measurements under high TF using a single crystal sample. Note that $H_L$ should be zero for antiferromagnets, and $H_{hf}$ for antiferromagnets is, in principle, very small compared with those for ferromagnets.

| TABLE I. $\mu^+$SR parameters for $R$CoAsO and the calculated $f_\mu$ ($f_\mu^{calc}$), where $f_\mu$ is converted into the local field ($H_{\mu}$) using $f_\mu = H_{\mu} \times 13.553$ kHz/Oe. The field distribution width ($\Delta$) was determined by fitting the ZF and LF spectra above $T_C$ using a dynamic Kubo-Toyabe function. For GdCoAsO, since the ZF spectrum shows a rapidly relaxing behavior even at 140 K, it was impossible to determine $\Delta$. |
|---|---|---|---|---|---|
| $R$ | $T_C$ (K) | $T_{N1}$ (K) | $T_{N2}$ (K) | $f_{\mu,FM}$ (MHz) | $f_{\mu,AF}$ (MHz) | $\Delta$ (10$^6$ s$^{-1}$) |
| La | 59 | – | – | 34.5(1) [at 1.7 K] | – | 0.22(2) [at 140 K] |
| Ce | 76 | – | – | 19.6(2) [at 1.8 K] | – | 0.300(4) [at 80 K]$^a$ |
| Pr | 71 | – | – | 3.07(2) & 34.2(4) [at 1.8 K] | – | 0.31(1) [at 120 K] |
| Nd | 70 | 15 | 7 | 3.05(1) [at 15.3 K] | 5.97(1) [at 1.7 K] | 0.192(4) [at 140 K] |
| Sm | 69 | 41.5 | 17.5 | 1.07(2) [at 42.5 K] | 45.3(1) [at 1.7 K] | 0.164(1) [at 140 K] |
| Gd | 60.5$^b$ | 60.8$^b$ | 3.2 | 25.9(4) [at 3.5 K] | 22.2(1.6) & 66.2(2) [at 1.7 K]$^c$ | – |

$^a$Only by ZF spectrum.

$^b$Not ferromagnetic, but ferrimagnetic.

$^c$The AF order is incommensurate.
A. NdCoAsO

We now begin the discussion of the AF state of NdCoAsO. First we attempt to determine the muon sites in the lattice by evaluating $H_{\text{dip}}(f_{\text{dip}})$ using dipole field calculations. First-principles calculations for LaFeAsO$^{27}$ and CeFeAsO$^{28}$ predicted that muons locate either at $(0.25, 0.25, 0.41)$ in the FeAs layer (A site) or at the vicinity of the $O^{2-}$ ion (B site). Assuming the same muon sites for NdCoAsO, the nuclear dipole field width ($\Delta$) in the paramagnetic state, which is caused by $^{143}\text{Nd}$, $^{145}\text{Nd}$, $^{59}\text{Co}$, and $^{75}\text{As}$ moments, is calculated to be $\Delta_{\text{calc}} = 0.399 \times 10^6$ s$^{-1}$ for the A site and $0.092 \times 10^6$ s$^{-1}$ for the B site (see also Table II). Here, the B site is assigned at $(0.25, 0.96, 0.93)$ by electrostatic potential calculations using a point-charge model. On the other hand, based on ZF- and LF-$\mu^+$SR measurements$^{29}$ for NdCoAsO far above $T_C$, i.e., at 140 K (see Fig. 7), $\Delta$ was estimated to be $\Delta_{\text{calc}} = 0.192 \times 10^6$ s$^{-1}$. Since $\Delta_{\text{calc}}$ corresponds to the highest limit of $\Delta_{\text{exp}}$ ($\Delta_{\text{calc}} > \Delta_{\text{exp}}$), muons are unlikely to be located at the B site.

Recent neutron experiments led to the proposal that, for the spin structures for NdCoAsO$^{1,12}$ in the FM state, only the Co moments align along the [100] direction, with $\mu^{\text{ord}}_{\text{Co,FM}} \sim 0.3 \mu_B$ at 15 K. In the AF state between $T_{N1}$ and $T_{N2} = 7$ K—i.e., in the AF1 phase—both the Co and Nd moments align antiferromagnetically with propagation vector $(0 0 1/2)$. The Co moments still align along the [100] direction, while the Nd moments align along the [100] direction. The ordered moments $\mu^{\text{ord}}_{\text{Co,AF}} = \mu^{\text{ord}}_{\text{Nd,AF}} \sim 0.4 \mu_B$ are almost $T$ independent. Below $T_{N2}$—i.e., in the AF2 phase—$\mu^{\text{ord}}_{\text{Nd}}$ increases with decreasing $T$, reaching 1.39$\mu_B$ at 300 mK. Using the proposed AF model, $H_{\text{dip}}(f_{\text{dip}})$ at the A site for NdCoAsO is predicted as a function of $\mu^{\text{ord}}_{\text{Co,AF}}$ and $\mu^{\text{ord}}_{\text{Nd,AF}}$ (see Fig. 8). It is clearly found that $f_{\mu^{\text{ord}}_{\text{Co,AF}}} = 12.2$ MHz for $\mu^{\text{ord}}_{\text{Co,AF}} = 0.4 \mu_B$ and $\mu^{\text{ord}}_{\text{Nd,AF}} = 0$. Since $f_{\mu}$ = 4 MHz at 8 K (in the AF1 phase) and 6 MHz at 1.7 K (in the AF2 phase), the ordered moments satisfy the following relations:

\begin{equation}
\mu^{\text{ord}}_{\text{Co,AF}} = 0.13 - 0.22 \times \mu^{\text{ord}}_{\text{Nd,AF}} \quad \text{at} \quad 8 \text{ K,}
\end{equation}

\begin{equation}
\mu^{\text{ord}}_{\text{Co,AF}} = 0.20 - 0.22 \times \mu^{\text{ord}}_{\text{Nd,AF}} \quad \text{at} \quad 1.7 \text{ K,}
\end{equation}

where $0 \leq \mu^{\text{ord}}_{\text{Co,AF}} \leq 0.2 \mu_B$ and $0 \leq \mu^{\text{ord}}_{\text{Nd,AF}} \leq 0.91 \mu_B$. If we assume that $\mu^{\text{ord}}_{\text{Co,AF}}$ is $T$ independent below $T_{N1}$, $\mu^{\text{ord}}_{\text{Nd,AF}}$ increases by $0.3 \mu_B$ with decreasing $T$ from 8 to 1.7 K, while the increase in $\mu^{\text{ord}}_{\text{Nd,AF}}$ is about $1 \mu_B$ from neutron data. Also, $\mu^{\text{ord}}_{\text{Nd,AF}}$ should be below $0.2 \mu_B$ in the whole $T$ range. The discrepancy between $\mu^{\text{ord}}_{\text{Nd}}$ estimated by $\mu\text{SR}$ and neutrons is probably due to the different time windows of the two techniques.

![FIG. 7. (Color online) ZF- and longitudinal (LF-) $\mu^+$SR spectra for NdCoAsO obtained at 140 K. The magnitudes of the LF were 10 and 30 Oe. Solid lines represent the fit result using a dynamic Kubo-Toyabe function.](image3)

### TABLE II. Prediction of the nuclear dipole field width ($\Delta$) at the A and B site for NdCoAsO

<table>
<thead>
<tr>
<th>$R$</th>
<th>$\Delta^{\text{calc}}_A$ (10$^6$ s$^{-1}$)</th>
<th>$\Delta^{\text{calc}}_B$ (10$^6$ s$^{-1}$)</th>
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<tr>
<td>La</td>
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<td>Ce</td>
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<td>Sm</td>
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</tbody>
</table>

![FIG. 8. (Color online) Prediction of the muon precession frequency ($f_{\mu}$) for NdCoAsO due to the dipole field [$H_{\text{dip}} = f_{\mu}/13.55342$ (kHz/Oe)] at the A site in the AF state. $f_{\mu}$ was calculated using the AF model, in which the ordered Co and Nd moments ($\mu^{\text{ord}}_{\text{Co}}$ and $\mu^{\text{ord}}_{\text{Nd}}$) are parallel/antiparallel to the [100] direction. The planes with $f_{\mu} = 4$ and 6 MHz represent the experimental results at 8 and 1.7 K. The line at which the two planes intersect corresponds to the combination of $\mu^{\text{ord}}_{\text{Co}}$ and $\mu^{\text{ord}}_{\text{Nd}}$ to satisfy the experimental result.](image4)
Returning to the FM state of NdCoAsO, we note that \( M_s \) (≈0.18 \( \mu_B \)) determined by dc-magnetization measurements is less than \( \mu_{\text{Co,AF}}^{\text{ord}} \approx 3 \mu_B \) estimated by neutrons. Since the time window of \( \mu^+ \) SR is between those of dc-magnetization and neutrons, \( \mu^+ \) SR should at least sense an FM ordered moment with a magnitude of 0.18 \( \mu_B \). Thus the dipole field calculations for the A site show that \( f_{\mu,\text{FM}} \approx 4.0 \) MHz, which corresponds to \( H_{\text{dip}} = 292 \) Oe. Also, \( H_L = 53 \) Oe from Eq. (5) and \( M_s \approx 0.18 \mu_B \). Therefore, \( H_{\text{dip}} = -120 \) Oe for NdCoAsO, since \( H_{\text{int}} = 225 \) Oe (≈3.05 MHz/13.55 kHz/Oe) at 3.05 K.

### B. SmCoAsO

For SmCoAsO, \( M_s \) was reported as 0.16 \( \mu_B \) by magnetization measurements,\(^{16,17} \) but there is no information regarding the FM and AF spin structures. Since \( f_{\mu,\text{FM}} \approx 1.07 \) MHz for SmCoAsO is comparable to \( f_{\mu,\text{FM}} \) for NdCoAsO, the FM structure is likely the same as that for NdCoAsO. Indeed, \( f_{\mu,\text{FM}} \) is predicted to be 3.4 MHz (\( H_{\text{dip}} = 249 \) Oe), resulting in \( H_L = 48 \) Oe and \( H_{\text{dip}} = -240 \) Oe at 42.5 K.

It is, however, difficult to explain \( H_{\text{int}} \) in the AF phase of SmCoAsO by the AF model for NdCoAsO, because \( f_{\mu,\text{AF}} \) for SmCoAsO is about eight times larger than that in NdCoAsO at 1.7 K. In fact, if we use such an AF model and fix \( \mu_{\text{Co,AF}}^{\text{ord}} \) at 0.4 \( \mu_B \), which is the value proposed for NdCoAsO from neutron experiments, \( \mu_{\text{Sm,AF}}^{\text{ord}} \) should be in the range \(-5 \mu_B \), inconsistent with the fact that the theoretical value of \( \mu_{\text{eff}} \) of Sm\(^{3+} \) is 0.845 \( \mu_B \). Therefore, we need to find another AF model for SmCoAsO, although \( \mu^+ \) SR is not an ideal tool for determining unknown magnetic structures—particularly for cases where there is only one muon spin precession frequency in the magnetic state. Nevertheless, assuming that an AF structure with the same propagation vector but \( \mu_{\text{Co,AF}}^{\text{ord}} \parallel [001] \) and \( \mu_{\text{Sm,AF}}^{\text{ord}} \parallel [001] \) instead, \( f_{\mu,\text{AF}} \) reaches 45.3 MHz (see Fig. 9) when \( \mu_{\text{Co,AF}}^{\text{ord}} = (0.73-0.20) \times \mu_{\text{Sm,AF}}^{\text{ord}} \), in which

\[
0.56 < \mu_{\text{Co,AF}}^{\text{ord}} < 0.73 \mu_B \quad \text{and} \quad 0 < \mu_{\text{Sm,AF}}^{\text{ord}} < 0.845 \mu_B .
\]

Such a simple structure could be a good candidate for the magnetic ground state for SmCoAsO. Here the ordered moments along the [100] direction are most likely rapidly fluctuating compared with those along the [001] direction, because of the tetragonal symmetry of RCoAsO. Therefore, it would be acceptable that both \( \mu_{\text{Co,AF}}^{\text{ord}} \) and \( \mu_{\text{R,AF}}^{\text{ord}} \) for SmCoAsO are larger than those for NdCoAsO, within the time window of \( \mu^+ \) SR.

### C. GdCoAsO

For GdCoAsO, the IC-SDW field ranges between \( \sim 22 \) and 66 MHz at 1.7 K. Since \( \mu_{\text{eff}} \) of Gd\(^{3+} \) is about 8 \( \mu_B \), either of the two AF models mentioned above could be used for the basis of the IC modulated spin structure. However, due to the large neutron absorption cross section of Gd, it would be difficult to determine the AF spin structure of GdCoAsO by a neutron-scattering technique.

The variation of the \( \mu^+ \) SR parameters of RCoAsO with \( R \) demonstrates the importance of an interaction between 3d and 4f electrons to the electronic properties. For such cases, the electronic properties of 4f ions in ZF could be described by the following Hamiltonian:\(^{31} \)

\[
\mathcal{H}_R = \mathcal{H}_{\text{so}} + \mathcal{H}_{\text{CF}} + \mathcal{H}_{\text{ex}} = \lambda \mathbf{L} \cdot \mathbf{S} + \mathcal{H}_{\text{CF}} + g_1 \mu_B \mathbf{S} \cdot \mathbf{H}_{\text{ex}},
\]

where \( \mathcal{H}_{\text{so}} \) stands for the spin-orbit coupling energy, \( \mathcal{H}_{\text{CF}} \) is the crystal electric-field energy, \( \mathcal{H}_{\text{ex}} \) the exchange energy, \( \lambda \) the spin-orbit coupling parameter, \( L \) and \( S \) the orbital and spin momenta, \( g_1 \) the spin gyromagnetic factor (≈2), and \( \mathbf{H}_{\text{ex}} \) the exchange field at \( R \) due to neighboring 3d electrons. Note that \( \lambda > 0 \) for R\(^{3+} \) with \( n < 7 \), \( \lambda = 0 \) for Gd\(^{3+} \) with \( n = 7 \), and \( \lambda < 0 \) for R\(^{3+} \) with \( n > 7 \), where \( n \) denotes the number of 4f electrons.

For most of the \( R \) ions, the spin-orbit coupling dominates the other terms in Eq. (7)—that is, \( L \) and \( S \) are coupled. As a result, the total angular momentum \( (J) \) becomes a good quantum number. As usual, Eq. (7) is further simplified as:

\[
\mathcal{H}_R = \mathcal{H}_{\text{CF}} + 2(g_J - 1) \mu_B J \cdot \mathbf{H}_{\text{ex}}.
\]

where \( g_J \) is the Landé factor given by

\[
g_J = 1 + \frac{J(J + 1) + S(S + 1) - L(L + 1)}{2J(J + 1)}.
\]

Here, \( g_J < 1 \) for R\(^{3+} \) with \( 1 \leq n < 6 \), \( g_J > 1 \) for R\(^{3+} \) with \( 7 \leq n \leq 13 \), and \( g_J = 1 \) for R\(^{3+} \) with \( n = 0, 6 \), and 14 (La\(^{3+} \), Eu\(^{3+} \), and Lu\(^{3+} \)). This leads to a parallel configuration between 3d and 4f moments for light \( R \) ions, and an antiparallel configuration for heavy \( R \) ions, but no preference for La\(^{3+} \), Eu\(^{3+} \), and Lu\(^{3+} \).

Although the overall magnetic properties of RCoAsO are determined not only by the 3d-4f interaction but also by 3d-3d and 4f-4f interactions, it is reasonable that such a change of sign in the second term of Eq. (8) induces a complex magnetic order for GdCoAsO compared with those for the other five RCoAsO compounds. In fact, despite systematic magnetization measurements for the compounds with \( R = \text{La}, \text{Ce}, \text{Pr}, \text{Nd}, \text{Sm}, \) and \( \text{Gd} \), a reliable value of \( M_s \) for

![FIG. 9. (Color online) Prediction of \( f_{\mu} \) for SmCoAsO due to dipolar fields at the A site in the AF state. \( f_{\mu} \) was calculated using the AF model, in which the ordered Co and Sm moments (\( \mu_{\text{Co}} \) and \( \mu_{\text{Sm}} \)) are parallel/antiparallel to the [001] direction. The plane with \( f_{\mu} = 45 \) MHz represents the experimental result at 1.7 K. The line at which the two planes intersect corresponds to the combinations of \( \mu_{\text{Co}} \) and \( \mu_{\text{Sm}} \) that satisfy the experimental result.](image_url)
GdCoAsO is still not available, suggesting that GdCoAsO is not a simple ferromagnet, but a ferrimagnet, as already mentioned.

Assuming that GdCoAsO is a ferrimagnet with $T_f = 60.5\,K$, $T_{N1}$ ($T_f$) is most likely to correlate with the de Gennes factor ($\xi$), \cite{10} where $\xi$ is defined as

$$\xi = (g_f - 1)^2 J(J + 1). \quad (10)$$

This could indicate that the Ruderman-Kittel-Kasuya-Yoshida (RKKY) interaction plays a certain role to determine the AF nature of $RCoAsO$, because, if crystal electric-field effects are not taken into account, $T_N$ for a series of isostructural and isoelctronic metals are expected to scale with $\xi$. However, as seen in Fig. 10, the slope of the $T_N(\xi)$ curve decreases with $\xi$, indicating that $T_{N1}$ is not simply determined by the RKKY interaction.

In order to further understand the role of the $3d$-$4f$ and/or RKKY interaction on the magnetic nature of $RCoAsO$, it is important to study the compounds with $R = Eu$ and Tl-Lu by both muons and neutrons.

### D. LaCoAsO and FM nature

Although $M_s$ is reported as $0.28\mu_B$/Co by magnetization measurements, a reliable FM ordered moment ($\mu^{\text{ord}}_{\text{Co,FM}}$) and FM spin structure are still unavailable for LaCoAsO. We therefore calculated a proportional coefficient ($f^{\text{calc}}_{\mu,\text{FM}}$) between $f^{\text{ord}}_{\mu,\text{FM}}$ and $\mu^{\text{ord}}_{\text{Co,FM}}$, i.e., $f^{\text{calc}}_{\mu,\text{FM}} = F^{\text{calc}}_{\text{Co,FM}} \times \mu^{\text{ord}}_{\text{Co,FM}}$. For the $A$ site, $F^{\text{calc}}_{\text{Co,FM}} = 41.8\,\text{MHz}/\mu_B$ when $\mu^{\text{ord}}_{\text{Co,FM}} \parallel [001]$, but $20.9\,\text{MHz}/\mu_B$ when $\mu^{\text{ord}}_{\text{Co,FM}} \parallel [100]$. If we assume that

$$M_s = \mu^{\text{ord}}_{\text{Co,FM}} \cdot f^{\text{calc}}_{\mu,\text{FM}}[100] = 11.7\,\text{MHz}, \text{ whereas } f^{\text{calc}}_{\mu,\text{FM}}[100] = 5.8\,\text{MHz}; \text{ that is, } H_{\text{dep}}[100] = 863\,\text{Oe} \text{ and } H_{\text{dep}}[100] = 428\,\text{Oe}.$$ 

Therefore, even for the case where $\mu^{\text{ord}}_{\text{Co,FM}} \parallel [001]$, $f^{\text{calc}}_{\mu,\text{FM}}$ is found to be about $1/3$ of the experimental value [34.5 MHz (=2546 Oe) at 1.7 K]. As a result, we obtain $H_{\text{hf}} = 1604\,\text{Oe}$, since $H_L = 78\,\text{Oe}$.

By means of the same procedure for the other compounds, $H_L$, $H_{\text{dep}}$, and $H_{\text{hf}}$ were estimated (Table III), based on the present $H_{\text{hf}}$ and $M_s$ in the literature. Figure 11 shows the relationships between $H_{\text{hf}}$ and $M_s$ of Co ions, and between $H_{\text{hf}}$ and theoretical $\mu_{\text{eff}}$ of $R^{3+}$. This is because $H_{\text{hf}}$ is proportional to the spin density at the muon site [see Eq. (5)]. Considering the fact that muons locate in the CoAs layer ($A$ site) and the $4f$ electrons well localize at the $R$ site, $H_{\text{hf}}$ is expected to increase with $M_s$. Indeed, there is a good correlation between $H_{\text{hf}}$ and $M_s$ of Co ions, and between $H_{\text{hf}}$ and theoretical $\mu_{\text{eff}}$ of $R^{3+}$. On the contrary, if we note the $3d$-$4f$ interaction, $H_{\text{hf}}$ could be proportional to $\mu_{\text{eff}}$ of $R^{3+}$, as in the case for an itinerant ferromagnetic system, $R_2Fe_14B$.\textsuperscript{34} In fact, except for SmCoAsO, $H_{\text{hf}}$ decreases monotonically with $\mu_{\text{eff}}$ of $R^{3+}$ [Fig. 11(b)].

Here, the slope ($dH_{\text{hf}}/d\mu_{\text{eff}}$) is steeper for $R_2Fe_14B$ than for

#### Table III. Internal magnetic field ($H_{\text{hf}}$) of $RCoAsO$ in their FM state obtained by the present $\mu$+SR work, the saturated magnetization ($M_s$), and the estimation of the Lorentz field ($H_L$), the dipole field ($H_{\text{dep}}$), and the hyperfine field ($H_{\text{hf}}$).

<table>
<thead>
<tr>
<th>$R$</th>
<th>$H_{\text{hf}}$ (Oe)</th>
<th>$M_s$ ($\mu_B$/Co)</th>
<th>$H_L$ (Oe)</th>
<th>$H_{\text{hf}} - H_L$ (Oe)</th>
<th>$H_{\text{dep}}$ (Oe)</th>
<th>$\mu_{\text{Co}} \parallel [001]$</th>
<th>$\mu_{\text{Co}} \parallel [100]$</th>
<th>$\mu_{\text{Co}} \parallel [100]$</th>
</tr>
</thead>
<tbody>
<tr>
<td>La</td>
<td>2546 [at 1.7 K]</td>
<td>0.28\textsuperscript{b}</td>
<td>78</td>
<td>2467</td>
<td>863</td>
<td>$\mu_{\text{Co}} \parallel [001]$</td>
<td>$\mu_{\text{Co}} \parallel [100]$</td>
<td>$\mu_{\text{Co}} \parallel [100]$</td>
</tr>
<tr>
<td>Ce</td>
<td>1446 [at 1.8 K]</td>
<td>0.24\textsuperscript{b}</td>
<td>69</td>
<td>1377</td>
<td>764</td>
<td>$\mu_{\text{Co}} \parallel [001]$</td>
<td>$\mu_{\text{Co}} \parallel [100]$</td>
<td>$\mu_{\text{Co}} \parallel [100]$</td>
</tr>
<tr>
<td>Pr</td>
<td>227 [at 1.8 K]\textsuperscript{a}</td>
<td>0.24\textsuperscript{b}</td>
<td>70</td>
<td>157</td>
<td>386</td>
<td>$\mu_{\text{Co}} \parallel [100]$</td>
<td>$\mu_{\text{Co}} \parallel [100]$</td>
<td>$\mu_{\text{Co}} \parallel [100]$</td>
</tr>
<tr>
<td>Nd</td>
<td>225 [at 15.3 K]</td>
<td>0.18\textsuperscript{b}</td>
<td>53</td>
<td>172</td>
<td>292</td>
<td>$\mu_{\text{Co}} \parallel [100]$</td>
<td>$\mu_{\text{Co}} \parallel [100]$</td>
<td>$\mu_{\text{Co}} \parallel [100]$</td>
</tr>
<tr>
<td>Sm</td>
<td>79 [at 42.5 K]</td>
<td>0.16\textsuperscript{c}</td>
<td>48</td>
<td>31</td>
<td>271</td>
<td>$\mu_{\text{Co}} \parallel [100]$</td>
<td>$\mu_{\text{Co}} \parallel [100]$</td>
<td>$\mu_{\text{Co}} \parallel [100]$</td>
</tr>
<tr>
<td>Gd</td>
<td>1911 [at 3.5 K]</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
</tr>
</tbody>
</table>

\textsuperscript{a}Only for the main signal with $f_\mu = 3.07\,\text{MHz}$.

\textsuperscript{b}Reference 10.

\textsuperscript{c}Reference 17.
RCOAsO. However, such difference is reasonable, because the muons locate at the center of the tetrahedron consisting of two R and two Fe in the $R_2$Fe$_{14}$B lattice. Although we cannot judge which is the predominant factor for $H_{hf}$ at present, 4f electrons might strongly affect $H_{int}$ through $H_{hf}$. However, 4f electrons are most unlikely to contribute to the FM order in RCoAsO.

For PrCoAsO, $f_\mu$ and $f_{\mu1}$ are comparable to $f_\mu$ of NdCoAsO in the FM phase, while $f_{\mu2}$ is comparable to that of LaCoAsO. This suggests a crossover of the FM spin structure from the high-$T$ NdCoAsO-type order to a complex low-$T$ order in the $T$ range between $T_{m1}$ and $T_{m2}$.

Finally, we wish to point out the possibility that $H_{hf}$ has a finite value even in the AF phase of RCoAsO, since the neighboring Co spins are reported to align parallel in the AF phase of NdCoAsO. This means that $H_{hf}$ listed in Table III should be subtracted from $H_{int}$ in the AF phase for NdCoAsO and SmCoAsO, and confirmed the absence of AF transitions for GdCoAsO, and PrCoAsO down to 1.8 K. Also, GdCoAsO and SmCoAsO. For NdCoAsO, the two planes in Fig. 8 are shifted upward by 120 Oe ($\approx$ 1.6 MHz), resulting in an increase in the ordered moments in the AF phase by about 0.05$\mu_B$. For SmCoAsO, due to its large $H_{int}$ in the AF phase, the effect of $H_{hf}$ on the total magnetic nature is considered to be negligible small.

V. SUMMARY

In summary, our systematic $\mu^+$SR study of RCoAsO has clarified the successive transitions from a high-$T$ FM phase to a low-$T$ AF phase for NdCoAsO, SmCoAsO, and GdCoAsO, and confirmed the absence of AF transitions for LaCoAsO, CeCoAsO, and PrCoAsO down to 1.8 K. Also, the FM and AF spin structures were found to vary with $T$ due to coupling between 3$d$ and 4$f$ electrons. Finally, if an SDW order between 3$d$ electrons is an essential component for the parent materials of superconductors, it is very important to clarify the AF spin structure of GdCoAsO. Such information in combination with further studies of similar compounds with $R$ = Eu and Tb-Lu could provide a valuable insight into the interplay between magnetism and superconductivity.

ACKNOWLEDGMENTS

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APPENDIX: FIT FOR IC FIELD

Although $J_0(2\pi f_\mu t)$ is widely used for fitting the ZF-$\mu^+$SR spectrum in an IC-SDW state, it should be noted that $J_0$ only provides an approximation of the generic IC magnetic-field distribution. This is because the lattice sum calculation of the dipolar field at the muon site ($H_{IC}$) due to an IC magnetic structure lies in a plane and traces out an ellipse. The half-length of the major axis of the ellipse corresponds to $H_{max}$, whereas half of the minor axis corresponds to $H_{min}$. As a result, the IC magnetic-field distribution $P_{IC}$ is generally given by

$$P_{IC} = P(H_{IC}) = \frac{2}{\pi} \frac{H}{\sqrt{(H^2 - H_{min}^2)(H_{max}^2 - H^2)}}. \quad (A1)$$

This distribution diverges as $H$ approaches either $H_{min}$ or $H_{max}$. Thus $J_0$ describes the field distribution very well except in the vicinity of $H_{min}$, and the value of $f_\mu$ should be interpreted as an accurate measure of $H_{max}$. However, $J_0$ provides no information on $H_{min}$. Hence the first term $\Delta_A \cos(2\pi f_\mu t + \phi)$ is added in Eq. (3) to account for the van Hove singularity around $H_{min}$ and to determine the value of $H_{min} = 2\pi f_\mu/\gamma_\mu$. (24) ($\gamma_\mu$ is the muon gyromagnetic ratio and $\gamma_\mu/2\pi = 13.553$ 42 kHz/Oe). In other words, only when $H_{min} = 0$ is Eq. (A1) well approximated by $J_0$. Field distributions well described by Eq. (A1) were observed for Ag$_2$NiO$_2$, Co$_3$O$_4$, and Na$_2$V$_2$O$_4$.

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