Co-existence of short- and long-range magnetic order in LaCo$_2$P$_2$

Ola Kenji Forslund$^{1,4}$, Daniel Andreica$^2$, Hiroto Ohta$^4$, Masaki Imai$^1$, Chishiro Michioka$^4$, Kazuyoshi Yoshimura$^1$, Martin Månsson$^{5,6}$ and Jun Sugiyama$^{4,7}$

$^1$ Department of Applied Physics, KTH Royal Institute of Technology, SE-106 91 Stockholm, Sweden
$^2$ Ioan Ursu Institute, Faculty of Physics, Babes-Bolyai University, 400084 Cluj-Napoca, Romania
$^3$ Faculty of Science and Engineering, Doshisha University, Kyotanabe, Kyoto 610-0321, Japan
$^4$ Department of Chemistry, Graduate School of Science, Kyoto University, Kyoto 606-8502 Japan
$^5$ Neutron Science and Technology Center, Comprehensive Research Organization for Science and Society (CROSS), Tokai, Ibaraki 319-1106, Japan
$^6$ Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan
$^7$ Authors to whom any correspondence should be addressed.

E-mail: okfo@kth.se and condmat@kth.se

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Abstract
The ferromagnetic (FM) nature of the metallic LaCo$_2$P$_2$ was investigated with the positive muon spin rotation, relaxation and resonance ($\mu^+\cdot\text{SR}$) technique. Transverse and zero field $\mu^+\cdot\text{SR}$ measurements revealed that the compound enters a long range FM ground state at $T_{C}^{ZF} = 135.00(1) \text{ K}$, consistent with previous studies. Based on the reported FM structure, the internal magnetic field was computed at the muon sites, which were predicted with first principles calculations. The computed result agree well with the experimental data. Moreover, although LaCo$_2$P$_2$ is a paramagnet at higher temperatures $T > 160 \text{ K}$, it enters a short range ordered (SRO) magnetic phase for $T_{C}^{ZF} < T \leq 160 \text{ K}$. Measurements below the vicinity of $T_{C}^{ZF}$ revealed that the SRO phase co-exists with the long range FM order at temperatures $124 \text{ K} \leq T \leq T_{C}^{ZF}$. Such co-existence is an intrinsic property and may be explained by an interplay between spin and lattice degree of freedoms.

1. Introduction

The interplay between magnetism and superconductivity is a long-term popular problem, particularly since the discovery of high-$T_c$ cuprates, because the ground states are usually considered incompatible with each other [1]. While the coexistence of both phases have been reported [2, 3], a competition between the two phases is more common and has been observed in many systems such as rare earth (R) RNi$_2$B$_2$C [4], K doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ [5] or BaFe$_{1.89}$Co$_{0.11}$As$_2$ [3]. The latter two compounds in particular crystallize in a ThCr$_2$Si$_2$-type structure, for which the general structure is described by AT$_2$X$_2$ with a metal A, transition metal T and metalloid X atoms. In these systems, the interactions are presumably dominated by low dimensional fluctuations within the edge-sharing TX$_4$ tetrahedra layers. The interlayer interaction across the A layer is heavily dependent on the X – X bonding distance, which naturally changes depending on the specific elements that occupy each site of AT$_2$X$_2$. As a result, many ground states, such as, paramagnetism (PM), ferromagnetism (FM), antiferromagnetism (AF), short range order (SRO) and superconductivity have been reported for AT$_2$X$_2$ [6–9], since this family can accommodate many elements and combinations. The flexibility in the combination of constituent elements results in ground states that greatly varies with chemical doping [8–10] and by application of hydrostatic pressure [7].

It has been observed in several AT$_2$P$_2$ (A=Ca, Sr, and Ba, and T=Fe, Co, and Ni) compounds that the magnetic phase transitions are related to subtle structural changes in the crystals [6, 11, 12]. These compounds have shown to transform from an uncollapsed tetragonal (ucT) phase to a collapsed tetragonal (cT) phase as a function of chemical doping [6, 9, 12], which naturally affects the ground state. While the majority of the AT$_2$P$_2$ compounds exhibit AF or PM ground states, LaCo$_2$P$_2$ is an itinerant FM with $T_{C}^{ZF} = 135 \text{ K}$. In fact, magnetisation measurements [13, 14] satisfy Rhodes-Wohlfarth relation, and the spin fluctuations resembles...
that of a weak itinerant ferromagnet \cite{14, 15}. Early magnetization measurements showed a Curie-Weiss behavior at higher temperatures above $T_{\text{C}^{2D}}$ \cite{6, 16}. Neutron diffraction measurements on a polycrystalline sample revealed a collinear FM structure along the $a$-axis with an ordered moment of 0.44(3)\mu_B \cite{13}. Indeed, recent magnetization measurements performed on a single crystal at low temperature suggest a highly anisotropic FM ground state \cite{14}, at least in a sense that the magnetisation show large differences depending on if the applied field is along a or $c$-axis \cite{13, 14}. Meanwhile, the spin-lattice relaxation rate reported in a $^{31}$P-NMR study \cite{14} indicated a 3D character of the spin fluctuations above $T_{\text{C}^{2D}}$.

In general, magnetic compounds exhibit signatures of low dimensional fluctuations at higher temperatures. At lower temperatures, weaker interchain/interplane interactions becomes more prominent and a 3D long range order (LRO) may be established. In fact, the relationship between such interplay and dimensionality was reported for quasi-one-dimensional Co based compounds with muon spin rotation and relaxation ($\mu^+\text{SR}$) measurements \cite{17–19}, because $\mu^+\text{SR}$ is very sensitive to local magnetic environments. Although, since LaCo$_2$P$_2$ exhibits a 3D character above $T_{\text{C}^{2D}}$ \cite{14}, but a highly anisotropic 2D character below $T_{\text{C}^{2D}}$, it is of high interest to study the crossover between these two regimes across $T_{\text{C}^{2D}}$. Therefore, we have initiated a detailed of LaCo$_2$P$_2$ using muon spin rotation, relaxation and resonance ($\mu^+\text{SR}$), taking advantage of the fact that $\mu^+\text{SR}$ can offer information about the internal magnetic field distributions of both static and dynamic characters \cite{20–22}. In this study, several magnetic regimes are observed using $\mu^+\text{SR}$. In particular, both short and long range magnetic ordered phases are likely to coexist in LaCo$_2$P$_2$ in the vicinity of $T_{\text{C}^{2D}}$. Such behavior is attributed to an intrinsic property and may originate from an interplay between spin and lattice degree of freedom.

2. Experimental setup

Polycrystalline LaCo$_2$P$_2$ was prepared from pure La, and Co and P starting materials. LaP and Co$_2$P were first synthesized by a solid state reaction between La/Co and P in evacuated quartz tubes at $800 \degree C/700 \degree C$. LaCo$_2$P$_2$ could then be synthesized from a solid state reaction between LaP and Co$_2$P, kept at 1000 \degree C for 20 hours in an Ar atmosphere. Details about the sample synthesis can be found elsewhere \cite{23, 24}.

The $\mu^+\text{SR}$ measurements were performed at the Dolly instrument at the S $\mu$S muon source at Paul Scherrer Institute, Switzerland. A top loaded $^4$He cryostat was used in order to reach temperatures down to $\sim$2 K. About 500 mg of powder sample was inserted into an Al-coated Mylar tape envelope. The envelope was attached to a low-background fork type sample holder made of Cu. The $\mu^+\text{SR}$ data was analyzed using MUSRFIT \cite{25}.

The electrostatic potential and the local spin density in LaCo$_2$P$_2$ were calculated by density functional theory (DFT) using a full-potential linearized augmented plane-wave method within generalized gradient approximation (GGA) as implemented in WIEN2k program package \cite{26}. The GGA by Perdew, Burke, and Ernzerhof \cite{27} was adopted as the exchange-correlation functional since it is suitable for 3d itinerant systems. In the calculations, the lattice parameters and atomic positions of LaCo$_2$P$_2$ were taken from \cite{6}. The magnetic moments were aligned parallel to the [100] direction through spin–orbit coupling, to be consistent to experimental results described later. The muffin tin potential radii ($R_{\text{MT}}$) for La, Co, and P were taken to be 2.50, 2.36, and 1.84 \AA, respectively. The energy cutoff was chosen to be ($R_{\text{MT}} \times K_{\text{max}} = 7.0$, and $20 \times 20 \times 20$ $k$-points meshes were used in the Brillouin zone. Here, $K_{\text{max}}$ is the maximum modulus for the reciprocal vectors. The muon sites were predicted from the DFT calculations as the positions of the local minima of the electrostatic potential. This has been shown to be a good approximation for muon site determinations, especially for metallic systems \cite{28}.
3. Results

The presentation of the $\mu^+\text{SR}$ results of LaCo$_2$P$_2$ is divided into sections based on the type of field configuration chosen for the $\mu^+\text{SR}$ experiments: transverse field (TF) or zero field (ZF). Transverse refers to the applied field direction with respect to the initial muon spin polarization. The FM transition temperature is estimated in both field configurations and are distinguished by the conventions $T_{C}\text{ZF}$ for ZF and $T_{C}\text{TF}$ for TF. Additionally, the ZF time spectra collected at base temperature is reproduced based on the published magnetic structure and muon site determined from DFT calculations.

3.1. Transverse field

While measurements in ZF contains more detailed information regarding the magnetic properties of the compound, measurements in TF configuration require shorter collection time and are usually more easily analysed compared to the measurements in performed in ZF. Therefore, the analyses and conclusion drawn from the ZF data can be confirmed by comparing the ZF fit results to the overall behaviour observed in TF.

Figure 2 shows the collected TF ($\sim$50 Oe) spectra of LaCo$_2$P$_2$ for selected temperatures. At high temperatures, a distinct oscillation is observed with a frequency of about 0.7 MHz, corresponding to the applied field TF. As the temperature is lowered, the amplitude of the 0.7 MHz oscillation, $A_{TF}$, decreases. Although, the amplitude is not completely diminished at lower temperature. Moreover, a faster oscillation can be observed in the time spectra at lower temperatures, accompanied by a positive offset. Therefore, the TF spectra were fitted using a combination of three exponentially relaxing cosine oscillations together with an non-oscillating exponential relaxation:

$$A_0 P_{TF}(t) = A_{TF}^{PM}\cos(f_{TF}^{PM}2\pi t + \phi_{TF}^{PM})e^{-\lambda_{TF}^{PM}t} + A_{TF}^{IMP}\cos(f_{TF}^{IMP}2\pi t + \phi_{TF}^{IMP})e^{-\lambda_{TF}^{IMP}t} + A_{TF}^{FM}\cos(f_{TF}^{FM}2\pi t + \phi_{TF}^{FM})e^{-\lambda_{TF}^{FM}t} + A_S e^{-\lambda_S t},$$

where $A_0$ is the initial asymmetry determined by the detector geometry of the instrument and $P_{TF}$ is the muon spin polarization function in TF configuration. $A_{TF}^{PM}, f_{TF}^{PM}, \phi_{TF}^{PM}$ and $\lambda_{TF}^{PM}$ are the asymmetry, frequency, initial phase and depolarization rate resulting from the applied TF, where the superscripts PM and imp represent the contributions from the paramagnetic (PM) and impurity (imp) phases, respectively. Furthermore, $A_{TF}^{FM}, f_{TF}^{FM}, \phi_{TF}^{FM}$ and $\lambda_{TF}^{FM}$ represent contributions from the internal FM field, together with $A_S, \lambda_S$. In particular, $A_{TF}^{FM}$ represents the internal magnetic field contributions that are perpendicular to the initial muon spin polarization, while $A_S$ are contributions from the internal field that are parallel to the initial muon spin polarization. Since the applied field is about 10 % of the internal field (about 6 MHz) at the muon site, it is natural to observe a non-oscillating fraction even below $T_{C}\text{TF}$.

![Figure 2. Transverse field $\mu^+\text{SR}$ spectra for selected temperatures ($T = 110, 130, 140$ and $200$ K) for the LaCo$_2$P$_2$ compound. Solid lines represent the best fit using equation (1).](image-url)
The oscillating fraction still present well below $T_{TF}^C$ is originating from a PM impurity phase, most likely Co$_2$P. In order to properly separate the PM and the impurity phases under the applied TF, the constraint $f = \frac{T_{PM}}{T_{imp}}$ was set. Therefore, this fraction was fixed through the whole temperature range since it should be temperature independent. An accurate estimate of the size of this impurity fraction is given below in ZF section.

Finally, the total asymmetry was fixed to 0.2351 for the measurements above 125 K, a value obtained from a high temperature measurement.

The obtained fit parameters using equation (1) with the procedure as described above are displayed in figure 3. Each asymmetry component has a temperature dependence that is expected for a magnetically ordered sample. For $T < 130$ K, $A_{PM}^{TF} = 0$ and exhibits a sharp change at a certain critical temperature. Since $A_{PM}^{TF} / (A_0 - A_{imp}^{TF})$ roughly corresponds to the PM volume fraction, the abrupt change observed at $T \approx 130$ K corresponds to the transition from a magnetically ordered state at low temperatures to magnetically disordered state at high temperatures. An accurate value of the transition temperature is obtained by fitting the $A_{PM}^{TF}(T)$ curve using a sigmoidal function, for which $T_{TF}^C = 130.17(1.18)$ K is obtained. Similarly, $A_{FM} = 0$ above $T_{TF}^C$ while $A_S$ poses none zero values even above $T_{TF}^C$, which steadily decreases with increasing temperature. Such behavior is naturally expected as these fractions stem from internal magnetic fields as described above. It should be noted that the $A_S$ term is observed well above $T_{TF}^C$. This is consistent with the results presented in ZF section and is due to the formation SRO. The presence of a SRO distorts the temperature dependence of $A_{TF}^{PM}(T)$ for what is expected for a simple FM-PM transition. This is the reason to why the accuracy of the sigmoid fit is low just above $T_{TF}^C$. Overall from TF point of view, disordered moments seems to appear below 160 K, which develops with decreasing temperature into a long range FM order below $T_{TF}^C$.

The temperature dependencies of $\lambda_{PM}^{TF}$, $\lambda_{imp}^{TF}$ and $\lambda_S$ are displayed in figure 3(b). $\lambda_{imp}^{TF}$ shows a steady decrease from lowest measured temperature up to the highest, and shows no anomaly at the magnetic transition. Such behaviour originate most likely from fluctuating Co $d$-moments and static Co and/or P nuclear moments, as also underlined in the ZF section. This temperature dependence is similar to the one obtained in ZF configuration, underlying the quality of the fits in both field configurations. On the other hand, $\lambda_{TF}^{PM}$ is none zero only above $T_{TF}^C$, as expected. It has a maximum just above $T_{TF}^C$ and starts to decrease with increasing...
temperature, reflecting an increase in the internal field dynamics. This temperature dependence will most likely follow the temperature dependence of magnetic susceptibility of a Curie-Weiss paramagnet. The small value of $\lambda_{TF}$ (below 0.1 $\mu s^{-1}$) at high temperatures suggests that the PM fluctuations eventually become motionally narrowed for the $\mu$+SR time window and only relaxation from nuclear moments are visible.

The fact that $\lambda_S = 0$ $\mu s^{-1}$ across the whole measured temperature range suggests a static behavior of the internal FM field. Both $f_{FM}$ and $\lambda_{FM}$ represent the nature of the internal FM field but is not presented here. Instead, measurements in ZF configuration provide more accurate information regarding the internal fields. Finally, it should be noted that since $f_{EM} \approx 4$ MHz at 110 K, which is not very different from the TF precession frequency with 50 Oe (about 0.7 MHz), the estimated volume fraction of the nonmagnetic impurity phase includes an ambiguity. A more accurate value of the size of this fraction will be estimated using the ZF-$\mu$+SR data.

3.2. Zero field
ZF-$\mu$+SR time spectra for selected temperatures are shown in figure 4. At 200 K, the ZF-spectrum in an early time domain exhibits a convex shape time dependence, which indicates a Gaussian-type relaxation. A notable

![Figure 4: Zero field $\mu$+SR spectra for the LaCo$_2$P$_2$ compound up to (a) 7 $\mu$s, (b) 1 $\mu$s and (c) 5 $\mu$s. (a) and (b) are plotted in their absolute asymmetries for selected temperatures ($T = 5, 135, 140, 160$ and 200 K). The time spectra in (c) have been shifted by 0.015 units for selected temperatures ($T = 124, 127, 128.5, 130, 135, 140$ and 160 K) and by 0.05 units for the $T = 105$ K time spectrum, for clarity of display. Solid lines represent the best fit using equation (2).](image-url)
A small dip around 3 μs would suggest the presence of two independent Gaussian relaxations, consistent with the presence of an impurity phase. As temperature is lowered, the Gaussian relaxation is gradually changing into a more exponential like form, until an oscillation appears at lower temperatures below \( T_C^{ZF} \). In order to take into account all the phases present over the whole measured temperature range, the time spectra were fitted using a combination of two static Gaussian Kubo-Toyabe (SGKT) functions, two exponential relaxations, and one exponentially relaxing cosine oscillating terms:

\[
A_0 P_{ZF}(t) = A_{FM} \cos(f_{FM} 2\pi t + \phi_{FM}) e^{-\lambda_{FM} t} + A_{tail} e^{-\lambda_{tail} t} + A_f e^{-\lambda_f t} + A_{KT} G(t, \Delta_{KT}) e^{-\lambda_{KT} t} + A_{imp} G(t, \Delta_{imp}) e^{-\lambda_{imp} t}
\]

where \( A_0 \) is the initial asymmetry determined by the instrument’s detector geometry and \( P_{ZF} \) is the muon spin polarization function in ZF configuration. The first row of equation (2), i.e., \( A_{FM} \) and \( A_{tail} \) represents the response of the sample at low temperatures, when it enters a magnetically long range ordered state below \( T_C^{ZF} \). The second row, i.e., \( A_f \) and \( A_{KT} \), represents the sample response above or close to \( T_C^{ZF} \). The last row, \( A_{imp} \) corresponds to the impurity phase present in the sample, a constant term throughout the whole temperature range. In detail, \( A_{FM}, f_{FM}, \phi_{FM} \) and \( \lambda_{FM} \) are the asymmetry, frequency, phase, and relaxation rate resulting from perpendicular (with respect to the initial muon spin polarisation) internal field components, while \( A_{tail} \) and \( \lambda_{tail} \) are the asymmetry and relaxation rate of the tail component that inevitably exist in powder measurement of a magnetically ordered sample. This contribution stems from the fact that on average, 1/3 of the internal magnetic fields are parallel with respect to the initial muon spin polarisation for a perfect powder. \( A_f \) and \( \lambda_f \) are the asymmetry and relaxation rate of a fast component that manifests the ZF-spectra around \( T_C^{ZF} \).

\( A_{KT} \) and \( \lambda_{KT} \) correspond to the asymmetry and relaxation rate related to the static Gaussian KT, represented by \( G(t, \Delta) \) where \( \Delta \) is the internal field distribution width. This function is given by

\[
G(t, \Delta) = \frac{1}{3} + \frac{2}{3} (1 - \Delta^2 t^2) \exp(-\frac{\Delta^2 t^2}{2}),
\]

and is derived assuming isotropically distributed random magnetic fields [29]. Similar to how the internal magnetic field in the FM state was modeled by a 1/3 tail term (parallel magnetic field component: \( A_{tail} \)) and a 2/3 oscillating term (perpendicular magnetic field component: \( A_{FM} \)), the static Gaussian KT is composed of 1/3 none relaxing term together with a 2/3 relaxing term, resulting into a characteristic dip in the time spectra. In this compound however, the value of \( \Delta_{KT} = 0.1264(12) \mu s^{-1} \) is low such that the characteristic dip is present outside the considered time window (i.e. above 7 μs). The same description holds also for the subscript \( A_{imp} \) signal, but corresponds to the impurity phase instead of the main phase.

In order to separate the various contributions, some constraints were set for the fits using equation (2). In particular, \( A_{imp} = 0.0485(9) \) was fixed across the whole temperature range, whereas \( \Delta_{imp} = 0.45(2) \) was fixed at higher temperatures, as these parameters can be expected to be temperature independent. These values were estimated at the base temperature given that the magnetic contrast between the main and impurity phase is the highest. Indeed, the value of \( \Delta_{imp} \) suggests the presence of Co, La and/or P elements in the impurity phase and the value of \( A_{imp} \) corresponds to a volume fraction of 22%. It is noted that measurements (not shown here) with externally applied longitudinal field at 200 K (5 and 10 G), i.e. field parallel with the initial muon spin polarization, was able to decouple the time spectra using the fixed values of \( A_{imp} = 0.0485 \Delta_{imp} = 0.45 \). This behavior suggest that the the origin behind the Gaussian KT is indeed nuclear (mostly \(^{59}\)Co and \(^{139}\)La nuclear moments) and are static within the lifetime of the muon, as expected. In other words, the fitting assessments described above are coherent and it also noted that the impurity phase maintains a Gaussian KT relaxation from 2 K up to 200 K. For reference, \( \Delta_{KT} = 0.1264(12) \mu s^{-1} \), which corresponds to \( 1.484 \) 237(2) Oe, was obtained for the main phase.

The temperature dependencies of the obtained fit parameters between 100 and 170 K are shown in figure 5, using the fit procedure described above. Same parameters but for the whole considered temperature range is instead shown in figure A1. Below \( T_C^{ZF} \), only two asymmetry components have none zero values (except for the \( A_{imp} \) that is not shown). This is consistent with the whole main sample phase entering a long range magnetically ordered state, for which \( A_{tail} \approx \frac{1}{2} A_{FM} \). Just below \( T_C^{ZF} \), a small upturn is observed in \( A_{tail} \) while a downturn is seen in \( A_{FM} \). This is natural given that parallel fluctuations usually increases as the static perpendicular component loses its structure close to \( T_C^{ZF} \). Intriguingly, the decrease of \( A_{FM} \) is followed by an additional fast relaxing component, \( A_f \). Such component exhibits a maximum just below \( T_C^{ZF} \) for which \( A_{FM} \) is still none zero, and slowly decreases with further increasing temperature. This behavior is directly visible in the measured time spectra (figure 4(c)). As seen, the amplitude of the oscillation slowly decreases with increasing temperature. This decrease is followed by an increase in the amplitude of an additional exponential, most visible in the time range 0-3 μs. Above \( T_C^{ZF} \), defined at the point where \( f_{FM} = 0 \) MHz and thus \( A_{FM} = 0 \), the spectra consist of \( A_{KT} \) and \( A_f \) components, where only \( A_{KT} \) persist above 160 K. It is noted that the fast exponential relaxation component (\( A_f \)
is present between the temperature range 124 and 160 K, i.e. across $T_C$. This implies that long range and short range order coexist for a narrow temperature range below $T_C$, which will be further discussed in section 4.

The muon spin precession frequency ($f_{FM}$), on the other hand, exhibits an order parameter like temperature dependence, and it can be fitted according to mean field theory: $f_{FM}(T) = f_{FM}(0 \text{ K}) \left(1 - \frac{T}{T_C}\right)^\beta$ with $f_{FM}(0 \text{ K}) = 6.48(14)$, $T_C^{ZF} = 135.00(1)$ and $\beta = 0.169(11)$. Intriguingly, the fit follows the data down to lowest measured temperature (figure A1(b)), which is surprising especially for an FM [30, 31]. Although, restricting the fitting...
Table 1. The calculated local field values at the predicted muon sites are tabulated together with the obtained local spin density \( \rho(\mathbf{r}_i) \) for the given magnetic structure. \( f_{\text{loc}} \) was evaluated using equation (5), whereas the dipole field, Lorenz field and the hyperfine contact field are given by equation (7), equation (8) and equation (6), respectively. The experimentally obtained precession frequency \( f_{\text{M}}(0 \text{ K}) \) and the expected internal field distribution width are listed as well.

| Muon site | \( \rho(\mathbf{r}_i)[\mu_\text{B} \text{Å}^{-1}] \) | \( B_{\text{dip}} \) [T] | \( B_{\text{lo}} \) [T] | \( B_{\text{li}} \) [MHz] | \( f_{\text{M}}(0 \text{ K}) \) [MHz] | \( \Delta f_{\text{M}} \) [MHz] |
|-----------|---------------------------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| \( \mu_1 \) (0,0,0.198) | -0.00237581 | [0.0872, 0, 0] | [-0.0185, 0, 0] | [0.0429, 0, 0] | 15.13 | 6.48(14) | 0.379 |
| \( \mu_2 \) (0,0.5,0.102) | -0.0015801 | [-0.0947, 0, 0] | [-0.0117, 0, 0] | [0.0428, 0, 0] | 8.61 | 6.48(14) | 0.461 |
| \( \mu_3 \) (0,0.5) | -0.00176282 | [-0.0379, 0, 0] | [-0.0137, 0, 0] | [0.0428, 0, 0] | 1.86 | 6.48(14) | 0.297 |

3.3. Magnetic structure

For the sake of completeness, we have evaluated the internal field based on the FM structure proposed in [13]. The magnetic structure was inferred from a combination of single crystal magnetization and polycrystal neutron diffraction (ND) measurements. In detail, ND measurements determined the \( \mu_\text{ord} = 0.44(3) \mu_\text{B} \) to be aligned perpendicular to [001], which naturally cannot be resolved for tetragonal structures for a powder sample. Complementary magnetisation measurements determined then that the easy axis of magnetization to be along [100].

The expected muon sites for the given electrostatic potential and the corresponding local spin density (based on the FM structure as described above) was predicted within the Wien2K framework [32, 33]. These calculations yield three possible muon sites; \( \mu_1 = (0, 0, 0.198) \), \( \mu_2 = (0, 0.5, 0.102) \) and \( \mu_3 = (0, 0, 0.5) \) and \( \mu_\text{ord} = 0.5 \mu_\text{B} \). The complete magnetic structure together with the predicted muon sites are shown in figure 1 and the calculated local spin density at these sites are listed in table 1.

With this in mind, we attempt to calculate the local field at the predicted muon sites, based on the magnetic structure presented above. For an non-magnetized FM in ZF, the internal field at the muon site \( (\mathbf{B}_{\text{loc}}) \) is given by the following three components,

\[
\mathbf{B}_{\text{loc}} = \mathbf{B}_{\text{dip}} + \mathbf{B}_{\text{li}} + \mathbf{H}_{\text{hf}}
\]

(4)

where \( \mathbf{B}_{\text{dip}} \) is the resulting dipole field within the considered Lorentz sphere, \( \mathbf{B}_{\text{li}} \) is the Lorentz field, and \( \mathbf{H}_{\text{hf}} \) is the hyperfine contact field. Such internal fields can be translated into the corresponding precession frequency via \( f = (\gamma_\mu / 2\pi) |\mathbf{B}| \), so that temperature range does not significantly alter the fit results. The value of \( \beta = 0.169(11) \) supports the scenario of the compound being a 2D magnet [30]. The obtained value of \( T_C^{ZF} \) is slightly higher than \( T_C^{C} \), obtained in TF configuration. This is most likely due to the difference in definition of transition temperature. In ZF, the transition temperature is defined at the point in which when \( f_{\text{FM}}(T) = 0 \) MHz while in TF configuration, the transition temperature is defined in which \( A_{\text{PM}}^{ZF} / (A_0 - A_{\text{PM}}) = 0.5 \). The value of the frequency corresponds to the total magnitude of the local field at the muon site. A frequency of 6.33(2) MHz is obtained at 5 K and corresponds to 0.046 70(15) T, which can be considered relatively low. Detailed calculations referencing these values are presented in section 3.3.

As for the relaxation rates, both \( \lambda_{\text{FM}} \) and \( \lambda_{\text{HF}} \) are temperature independent at low temperatures. Here, \( \lambda_{\text{HF}} \) corresponds to the spin-lattice relaxation rate. The value of \( \lambda_{\text{HF}}(2 \text{ K}) = 0 \mu_\text{s}^{-1} \) suggests that the magnetic order at low temperatures is static. \( \lambda_{\text{HF}} \) increases as \( T_C^{ZF} \) is approached, indicating that the internal field is dynamic close to \( T_C^{ZF} \). \( \lambda_{\text{FM}} \) on the other hand corresponds roughly to the spin-spin relaxation rate and its value can be interpreted as the field distribution width at the muon site. Similar to \( \lambda_{\text{HF}} \), an increase of \( \lambda_{\text{FM}} \) is observed close to \( T_C^{ZF} \). Part of it can be ascribed to increase in dynamics (in principle, \( \lambda_{\text{FM}} \) is composed of both spin-spin and spin-lattice relaxation rates).

The temperature dependencies of \( \lambda_{\text{CT}} \) and \( \lambda_{\text{imp}} \) are consistent with the findings in TF configuration. In equation (2), each static Gaussian KT is multiplied by an exponential relaxation function. This is because the internal field at the point has two independent contributions (nuclear and electronic) and the time dependence of the muon polarization, \( P(t) \), is given by the product of the expected polarization function originating from each contribution. Since the KT is the depolarization due to isotropically distributed nuclear moments, the exponential is accounting additional PM fluctuations present in the compound and it should follow the temperature dependence of \( \lambda_{\text{FM}} \). Details of \( \lambda_{\text{imp}} \) are highlighted in appendix B. Finally, \( \lambda_\text{loc} \) is the additional fast relaxation rate that manifests in the time spectra close to \( T_C^{ZF} \). Its temperature dependence shows a minimum close to \( T_C^{ZF} \), and increases again both below and above \( T_C^{ZF} \). The origin of this additional fast relaxation is discussed in section 4.
where  is the muon spin Larmor precession frequency around the internal field at the considered muon site. The modules of equation (5) corresponds to the fact that  is isotropic, i.e. assuming a spherical electron wave functions. In this case, the  is simplified to

\[
B_{\text{L}} = \frac{\mu_0}{3} \sum_{j} \rho(r_{\mu}) \overline{r_{\mu} r_{j}} \cdot m_j
\]

(6)

where  is the vacuum permeability (= \(4\pi \times 10^{-7}\) H/m) and the probability density for a spherical cloud at the muon site is given by \(|\psi(r_{\mu})|^2\), which in turn is related to the local spin density \(\rho(r_{\mu})\). In other words,  is given as a scalar coupling between \(\rho(r_{\mu})\) and the magnetic moment of the electron \(m_e = g \mu_B J\) where  is the total angular momentum.

The dipole field at the muon site on the other hand is originating from the dipolar interactions between localized electrons and the muon spin. A good approximation is to simply consider classical dipoles originating from spin polarized electron orbitals at the center of the magnetic atoms within a large sphere (the Lorentz sphere) with  N atoms:

\[
B_{\text{dip}} = \frac{\mu_0}{4\pi} \sum_{j} 3 r_{\mu j} (m_{\mu j} r_{\mu j}) - \frac{m_{\mu j}}{r_{\mu j}^3}
\]

(7)

where if  \(N \rightarrow \infty\) then  is the distance between the muon and the  j-th ion. Since the summation in equation (7) is not infinite but is instead limited up to within the so called Lorentz sphere, an additional contribution is added to the local field, known as the Lorentz field

\[
B_{\text{L}} = \frac{\mu_0}{3} M_L = \frac{\mu_0}{3V} \sum_{j} m_{\mu j}
\]

(8)

where  is the vector sum of the magnetic moments inside the Lorentz sphere divided by its volume.

Based on presented theoretical models and the determined magnetic structure, the local field components were computed for the muon sites; (0,0,0.198), (0,0.5,0.102) and (0,0,0.5). The calculations were performed using Python package MUESR [34], and the obtained local field values are presented in table 1. Among the considered muon sites, \(\mu_2\) agrees well with the experimentally obtained data with  \(f_{\text{loc}} = 8.61 \pm 6.48(14) = f_{\text{SR}}(0\ K)\), although the calculated value slightly overestimates the local field in comparison to the data.

The Lorentz field is independent on muon site, since the considered Lorentz sphere was kept constant for the all muon sites. It is however noted that given the symmetry of the positions of \(\mu_2\) and \(\mu_3\), \(B_{\text{dip}}\) and \(B_{\text{L}}\) are effectively canceling each other resulting into a low precession frequencies, especially for \(\mu_3\). The hyperfine contact fields are fairly constant across the considered muon sites and constitute a fairly large portion of the resultant local field. Such behavior is different from the A-type AF NaNiO\(_2\) [35], where the local field was found to be solely formed by dipolar fields. This is to some degree expected since the local spin density should be more considerable in a FM, compared to an AF.

4. Discussion

4.1. Main phase

The value of the local field at the \(\mu_2\) site at low temperatures is reasonably explained by the magnetic structure determined by neutron diffraction. Therefore, we will to focus our discussion on the behavior of the compound at higher temperatures. The ZF scan presented above reveals different magnetic regimes for different temperatures (figure 6). In detail, a PM order is established at higher temperatures \(T > 160\ K\), as evidenced by the exponentially relaxing KT \((\Delta > 160\ K)\). A short range order is stabilized in the temperature range 124 K \(\leq T \leq 160\ K\), for which the muon spin depolarization is made up of two separate exponential relaxations \((A_F\) and \(A_{\text{KT}}\) with  \(\Delta = 0\) \(\mu_2\)\) and a FM-LRO is stabilised below \(T_{\text{C}}^{ZF} = 135.00(1)\).

For compounds with a magnetic transition at finite temperatures, SRO are in general present above the  \(T_{\text{C}}^{ZF}\). At  \(T_{\text{C}}^{ZF}\), the SRO order is collectively developed into a LRO, and the compounds exhibit only LRO characteristics below \(T_{\text{C}}^{ZF}\). This has for example been the situation in Pu\(_{1-x}\)Ni\(_x\) alloy [36], in the triangular lattice compound NiGa\(_2\)S\(_4\) [37] or in \((\text{MnFe}_2)_2(\text{P,Si})\) [38]. The behaviour is particular in low dimensional systems, as observed in the quasi 1D \(A_{11+2} \text{Co}_{n+1} \text{O}_{3n+3}\) \((A=\text{Ca}, \text{Sr})\) [17, 18] and \(A_{11+2} \text{CoRh}_2 \text{O}_{3n+3}\) \((A=\text{Ca}, \text{Sr})\) [19].
BaCo$_2$V$_2$O$_8$ [39] or in layered compounds like NaNiO$_2$ [40] or CrCl$_3$ [41]. Being a low dimensional magnet, the situation for LaCo$_2$P$_2$ seems to be a little bit different compared to the previously mentioned cases. In LaCo$_2$P$_2$, the SRO seems to coexist with the FM-LRO for an extended temperature range 124 K < T < 160 K, for which FM-LRO is fully formed below T < 124 K (figure 6). This observation is supported by the fact that $A_F$ is split into two components at $T_C^{PF}$. While $A_{KT}$ sets a value $\sim A_F/3$, consistent with the tail component, $A_T$ shows a maximum at $T_C^{PF}$ which is separated into $A_{FM}$ and $A_F$ just below $T_C^{PF}$. Since $A_{FM}$ is the oscillating fraction, this is the long range ordered fraction of the sample. $A_F$ on the other hand is still an exponential relaxation even below $T_C^{PF}$, implying that there is a small temperature range in which FM-LRO and SRO co-exist.

While it was not clearly commented on, a similar behavior was observed in a single crystal $^{31}$P-NMR study in the raw data figure of [14]. The spin-spin relaxation rate ($1/T_2$) seems to decrease between 130 and 120 K and increases again around 110 K. Such behavior is reflected by the behavior of $\lambda_2$, presented in figure 5(c). Most likely, the smaller upturn of $\lambda_2$ is related to stray fields, induced by the ferromagnetic LaCo$_2$P$_2$ phase [21]. Although NMR does not provide the information on the volume fraction, our study clearly show that a small fraction of SRO is present while the majority of the sample forms LRO.

From the best of our understanding, compounds exhibiting both SRO and LRO characteristics for an extended temperature range is an uncommon phenomena. While the situation is different from the title compound, a coexistence of LRO and SRO has been reported for SrEr$_2$O$_4$ [42] or La$_{0.7}$Sr$_{0.3}$Mn$_{1-x}$Co$_x$O$_3$ [43]. SrEr$_2$O$_4$ is a frustrated magnet where the coexistence was attributed to complex interactions arising from geometrical frustration. For La$_{0.7}$Sr$_{0.3}$Mn$_{1-x}$Co$_x$O$_3$, on the other hand, the coexistence looks to stem from the competitions between AF and FM interactions. A similar separation of ordered and disordered phases has also been reported for Li$_3$RhO$_2$ [44]. Theoretical treatment of such phenomena revealed that the ground state is stabilized from competition between quantum fluctuations and frustration [45, 46]. This would suggest that competitions among complex multiple interactions could results in LRO + SRO phases, and may thus be present in LaCo$_2$P$_2$. As well. If this is the case, it is tempting to ascribe the mechanism, for which SRO and LRO forms, to be due to the changes in LaCo$_2$P$_2$ dimensional character at $T_C^{PF}$. Despite the layered strcuture with a highly anisotropic FM order in LaCo$_2$P$_2$ [14], because of the stronger 2D interactions in the CoP plane compared to the interplane interactions, a $^{31}$P-NMR study [14] clarified the spin fluctuations above $T_C^{PF}$ to be three dimensional. In other words, there exists a small temperature range for which the interplane [out of plane] interactions [fluctuation] is diminished with lower temperatures: the compound changes its characteristics from 3D to a low temperature 2D like states. Admittedly, this explanation is speculative and single crystal neutron scattering and complementary DFT calculations are needed in order to confirm whether a ‘3D-2D crossover’ could lead to manifestation of both SRO and LRO in LaCo$_2$P$_2$. Another scenario includes by not only considering the spin degree of freedom, but also the lattice degree of freedom. The related compound, Sr$_{1-x}$Ca$_x$Co$_2$P$_2$, could be tuned from a SRO ground state (0.5 < x < 0.7) to a LRO ground state (x > 0.7) [9]. The ground state was found to be correlated to the interplane distance [9], as the substitution of Sr with Ca gradually compresses the unit cell [24]. It may be that the interplane distance, or more precisely the interplane interaction, in LaCo$_2$P$_2$ around $T_C^{PF}$ is such that the compound is on the borderline between SRO and LRO, for which one order is favored over the other at lower temperatures. A detailed temperature dependent neutron diffraction study may clarify how the lattice affects the magnetic ordering in this compound.
4.2. Impurity phase
Finally, we wish to note the fact that a relatively large impurity phase is present in the compound, which could be the origin behind the anomaly around $T_{CF}^{ZF}$. Since $\mu^+$ SR is a local probe, for which the obtained information is based on implanted muons, each powder sample particle is large enough to observe bulk magnetic properties of the sample. This is the reason why $\mu^+$ SR is sensitive to detecting magnetic volume fractions, meaning each contribution is in principle separated. In fact, the signal from the impurity phase is nicely fitted with an exponentially relaxing Gaussian KT at 2 K, and it is highly unlikely that such behaviour should evolve into an exponential like relaxation at higher temperatures. The value of the exponential relaxation rate itself is also one order of magnitude lower than the one of $\lambda_F$ (appendix B). While the ferromagnetic LaCo$_2$P$_2$ phase may induce stray fields and affect the temperature dependence of $\lambda_F$, the effect of a paramagnetic impurity phase is negligibly small from a $\mu^+$ SR viewpoint, due to the absence of a stray field in non-ferromagnetic materials [21]. X-ray diffraction (XRD) measurements yielded, apart from the main phase, presence of minor impurity peaks. Although, the pattern of the main phase could be nicely refined using the expected crystal structure of LaCo$_2$P$_2$: tetragonal structure I4/mmm (# 139). The consistency between this and a single crystal study [14] supports the fact that the magnetic properties of this compound is not affected by the non magnetic impurities.

5. Conclusions
Transverse and zero field (ZF) $\mu^+$ SR measurements reveal that the sample exhibits a long range ferromagnetic (FM) ground state below $T_{CF}^{ZF} = 135.00(1)$ K, consistent with previous reports. The muon sites and the corresponding local spin density, based on the already reported magnetic structure, were predicted by density functional theory. The estimated local field calculations agrees well with the presented ZF-$\mu^+$ SR data. Intriguingly, this study reveals presence of multiple magnetic regimes, not observed in previous studies. In detail, a paramagnetic (PM) phase is found at higher temperatures $T > 160$ K, below which a short range order (SRO) is stabilised. In a narrow temperature range below $T_{CF}^{ZF}$, 124 K $\leq T \leq T_{CF}^{ZF}$, the SRO and a long range FM seem to coexist. The coexistence may originate from an interplay of spin and lattice degrees of freedom. Additional studies using both theoretical and experimental means would benefit the discussion of this interesting situation.

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Data availability statement
The data that support the findings of this study are available upon reasonable request from the authors.

Appendix A
For reference, the fit parameters shown in figure 5 is presented again but down to the lowest measured temperature (figure A1).
Appendix B. Temperature dependence of the impurity phase

The temperature dependencies of the obtained KT field distribution widths (ΔKT and Δimp) and relaxation rate for the impurity phase (λimp) are shown in figure B1. The fit favors ΔKT = 0 μs⁻¹ below 160 K, which is a common phenomena in samples exhibiting SRO. Δimp on the other hand show a non temperature dependent behaviour at low temperatures, as expected. In order to stabilise the fit at higher temperatures, the value obtained at base temperature, Δimp = 0.45(2), was fixed. The value of λimp saturates at lower temperatures and steadily
decreases with increasing temperature. As mentioned in the main text, the temperature dependence follows the one obtained in TF configuration, underlying the high quality of our fits in both field configuration. Moreover, the value itself is one order of magnitude lower than that of $\lambda_F$, suggesting that the origin of the additional exponential close to $T_C^{ZF}$ is not from the impurity but is truly an intrinsic behavior of LaCo$_2$P$_2$.

Appendix C. Basic characterisations

The current sample batch was measured and characterised with x-ray diffraction and magnetisation measurements. Figure C1 show the x-ray diffraction pattern collected at room temperature for the LaCo$_2$P$_2$ sample used for the $\mu^+$ SR experiments. The crystal structure, given by the space group $I4/mmm$ (#139), was used for the Rietveld refinement, which in turn was performed using the Fullprof suite [48]. A notable difference is observed between the model and data around $Q = 2.2$ Å$^{-1}$. Although, refining the occupancy of the sample does not alter nor improve the overall refinement. Therefore, the difference is mostly due to imperfect peak shapes. The peak shape around this $Q$ is harder to refine given the presence of minor peaks in between the main sample peaks. Most of these minor peaks were identified and indexed with CoP (Pnma), Co$_2$P (Pnma) and LaP (Fm-3m) phases. Detailed analysis of room temperature structure is found elsewhere [6, 13].

Figure C1 show the temperature dependence of magnetisation ($M$) and inverse magnetic susceptibility ($1/\chi$) measured at $H = 0.05$ T in zero field cooled protocol. $1/\chi$ show a linear behaviour at higher temperature and follows Curie-Weiss law, as expected. The data is coherent with the one obtained for a single crystal [14] and underlines that the magnetic properties presented in this work is not really affected by the presence of smaller none-magnetic impurities. Detailed analysis of the magnetisation data is found elsewhere [6, 13, 14, 16].
Figure C1. (a) Collected room temperature x-ray diffraction pattern together with the corresponding Rietveld refinement with $\chi^2 = 4.1$, R-factor = 19.01 and RF-factor = 16.1. The green ticks highlight the allowed Bragg reflections while the blue curve is the difference between the measured and refined pattern. (b) Temperature dependence of magnetisation (M) and the inverse susceptibility $(1/\chi)$, measured at $H = 0.05$ T in zero field cooled protocol.

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