Quantum fluctuations in the quasi-one-dimensional non-Fermi liquid system $CeCo_2Ga_8$ investigated using μSR

A. Bhattacharyya,[1,*] D. T. Adroja,[2,3,†] J. S. Lord,[2] L. Wang,[4,5] Y. Shi,[4,5,6] K. Panda,[1] H. Luo,[4,5,6] and A. M. Strydom[3]

[1]*Department of Physics, Ramakrishna Mission Vivekananda Educational and Research Institute, Belur Math, Howrah 711202, West Bengal, India*
[2]*ISIS Facility, Rutherford Appleton Laboratory, Chilton, Didcot Oxon OX11 0QX, United Kingdom*
[3]*Highly Correlated Matter Research Group, Physics Department, University of Johannesburg, P.O. Box 524, Auckland Park 2006, South Africa*
[4]*Beijing National Laboratory for Condensed Matter Physics, Institute of Physics, Chinese Academy of Sciences, Beijing 100190, China*
[5]*School of Physical Sciences, University of Chinese Academy of Sciences, Beijing 100190, China*
[6]*Songshan Lake Materials Laboratory, Dongguan, Guangdong 523808, China*

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Reduced dimensionality offers a piece of crucial information in deciding the type of the quantum ground state in heavy fermion materials. Here we have examined the stoichiometric $CeCo_2Ga_8$ compound, which crystallizes in a quasi-one-dimensional crystal structure with Ga-Ce-Co chains along the c axis. The low-temperature behavior of the magnetic susceptibility ($\chi \sim T^{-0.2}$), heat capacity ($C_p/T \sim -\ln T$), and resistivity ($\rho \sim T^n$ with $n \sim 1$) strongly confirms the non-Fermi liquid ground state of $CeCo_2Ga_8$. We study the low-energy spin dynamics of the $CeCo_2Ga_8$ compound utilizing zero-field (ZF) and longitudinal-field muon spin relaxation (μSR) measurements. ZF-μSR measurement reveals the absence of long-range magnetic ordering down to 70 mK. Interestingly, below 1 K the electronic relaxation rate rises sharply, suggesting the appearance of low-energy quantum spin fluctuations in $CeCo_2Ga_8$.

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I. INTRODUCTION

Searching for the quantum critical point (QCP) is a great challenge in strongly correlated materials since it only emerges at zero temperature by varying a control parameter such as the magnetic field, pressure, or chemical doping or alloying [1–3]. Heavy fermion (HF) materials exhibit many exotic states in the vicinity of a magnetic QCP, including non-Fermi liquid (NFL) and unconventional superconductivity [1,4–8]. At the QCP, the quantum fluctuations dominate over the thermal fluctuations that break the predictions of well-known Landau–Fermi liquid behavior [9,10], and hence the system exhibits NFL behavior. The nature of quantum fluctuations and development of magnetic correlations will depend on the dimensionality of the systems, and hence it is very important to investigate the effect of dimensionality on the QCP/NFL. So far, most Ce-based QCP/NFL systems investigated are two-dimensional (2D) or 3D, and there are no reports on 1D Ce-based NFL systems [8,11,12]. Furthermore, the physical properties of Ce-based compounds at low temperatures exhibit Fermi liquid behavior predicted by Landau theory [13]. For example, at low temperatures the electrical resistivity $\rho \sim T^2$, the heat capacity $C \sim T$, and the dc magnetic susceptibility is independent of the temperature [14–18]. Interestingly some of the Ce- and Yb-based materials deviate from conventional Fermi liquid behavior to so-called NFL behavior, which can be tuned from an antiferromagnetic ground state to a zero-temperature QCP, where quantum fluctuations are responsible for the NFL behavior. In the case of NFL compounds $\rho \sim T^n$ ($1 \leqslant n < 2$), $C/T \sim -\ln T$ or $C/T \sim a - bT^{1/2}$, and $\chi \sim T^{-p}$ ($p < 1$) [14–18].

To accommodate deeper insight into the specific nature of the QCP, both theoretical and experimental efforts have been made frequently in recent years. Still, most of them focus on the quasi-2D or 3D HF [8,11,12]. Considering that dimensionality is a fundamental component in defining the unique NFL attributes in these materials, a lower dimension predicts a substantial magnetic frustration parameter, and it is imperative to search the QCPs in quasi-1D HF compounds, whose science can be easily approximated by the density matrix renormalization-group method as well as the mean-field approximation [19]. The QCP has been observed in $CeCu_{6-x}T_x$ (T = Au, Ag) [20,21], in which the NFL at $x = 0.1$ and an ambient pressure is driven to a magnetically ordered state via further doping in antiferromagnetic ordered HF compounds such as $CeIn_3$ [22,23] and $CePd_2Si_2$ [24]. To date, there exist only a few undoped or stoichiometric materials which exhibit NFL states at ambient pressure, such as UBe_{13} [25,26], $CeNi_2Ge_2$ [27], $CeCu_2Si_2$ [27], and CeRhBi [28,29].

As chemically disordered NFL states remain challenging to understand by theoretical models, it is highly desirable to

*amitava.bhattacharyya@rkmvu.ac.in
†devashibhai.adroja@stfc.ac.uk

examine stoichiometric and homogeneous systems, which are prototypical materials for theoretical modeling. The recently discovered quasi-1D Kondo lattice system $CeCo_2Ga_8$, which crystallizes in the $YbCo_2Ga_8$-type orthorhombic structure, provides us a rare opportunity to scrutinize a QCP at ambient pressure [30]. The onset of coherence is at about $T^* \sim 20$ K, and no sign of superconductivity is found down to 0.1 K. Furthermore, 1D spin-chain behavior is also clear from susceptibility data [30], and density functional computations predict flat Fermi surfaces originating from the 1D f-electron bands along the c axis. An NFL state develops in a wide temperature range, as is apparent from the pressure-dependence resistivity data [30]. All these facts firmly suggest that $CeCo_2Ga_8$ is naturally positioned in the proximity of a magnetic QCP [30]. Nevertheless, T-linear resistivity and a logarithmically divergent specific heat are expected in a 2D antiferromagnetic QCP from the conventional Hertz-Millis theory [31,32], but not in a quasi-1D Kondo lattice system. Interestingly in the case of $CeCo_2Ga_8$, anisotropic magnetic susceptibility behavior can be explained utilizing crystal-field theory, and the ratio of the exchange interaction is $|J_{\rm ex}^{c}/J_{\rm ex}^{a,b}| \sim 4$–5 [33]. Our investigation of $CeCo_2Ga_8$ includes electrical resistivity $\rho(T)$, dc susceptibility $\chi(T)$, and heat capacity $C_{\rm P}(T)$ data used for characterization of $CeCo_2Ga_8$ and muon spin relaxation (μSR) measurement to study the low-energy spin dynamics. Our microscopic examination confirms that the stoichiometric $CeCo_2Ga_8$ compound exhibits an NFL ground state without any doping. It is interesting to note that an NFL ground state without doping is rare in Ce-based compounds, with only a few, CeRhBi [28,29], CeRhSn [34], and $CeInPt_4$ [35,36], belonging to this group.

II. EXPERIMENTAL DETAILS

For the present investigation, high-quality single crystals of $CeCo_2Ga_8$ were grown employing the Ga-flux method. The complete growth method is reported in Ref. [30]. The temperature-dependent magnetic susceptibility [$\chi(T)$] was measured using a Quantum Design Magnetic Property Measurement System (MPMS-SQUID) with the applied field parallel to the c axis. Electrical resistivity [$\rho(T)$] and heat capacity [$C_p(T)$] measurements were done using a Physical Property Measurement System (PPMS) with a dilution inset and He^3 cryostat, respectively.

μSR experiments were carried out the MUSR spectrometer in the zero-field (ZF) and longitudinal-field (LF; $H \parallel$ muon spin direction) geometries at the muon beamline of the ISIS Facility at the Rutherford Appleton Laboratory, Oxfordshire, U.K. [37]. Unaligned single crystals [as they were very small: less than 1.5 mm (length) × 1 mm (diameter)] of $CeCo_2Ga_8$ were mounted on a 99.995% silver plate, applying thinned GE varnish covered with a high-purity silver foil. The sample was cooled down to 70 mK using a dilution refrigerator. The spin-polarized incident muon thermalized on the sample, and the resultant asymmetry was determined employing $G_z(t) = [N_{\rm F}(t) - \alpha N_{\rm B}(t)]/[N_{\rm F}(t) + \alpha N_{\rm B}(t)]$, where $N_{\rm B}(t)$ and $N_{\rm F}(t)$ are the number of positrons counted in the backward and forward detectors, respectively, and α is an instrumental calibration constant determined with a small (2-mT) transverse magnetic field at a high temperature. $G_z(t)$ gives information about the spin-lattice relaxation rate and internal field distribution. We have collected ZF μSR data between 0.07 and 4 K, and LF μSR data in an applied field up to 0.3 T at 0.25 K. ZF/LF-μSR data were analyzed utilizing WiMDA software [38].

III. RESULTS AND DISCUSSION

Figure 1(a) represents the orthorhombic structure (space group *Pbam*, No. 55) of $CeCo_2Ga_8$, showing the quasi-1D chains of Ce atoms along the c axis. The individual unit cell holds four Ce atoms. The NFL state in $CeCo_2Ga_8$ is confirmed by electrical resistivity, magnetization, and heat capacity measurements, and the results are presented in Figs. 1(b)–1(d). The temperature variation of susceptibility measured under a field-cooled condition with a 10 mT applied field parallel to the c axis is illustrated in Fig. 1(b). $\chi(T)$ manifests an $\sim T^{-0.2}$ response, suggesting $CeCo_2Ga_8$ located near the quantum phase transition [30]. This type of power-law behavior of $\chi(T)$ is also observed for other NFL systems such as CeRhBi [8,29]. On the other hand, the susceptibility of the NFL system $CeCu_{5.9}Au_{0.1}$ exhibits $\chi(T) \sim \chi_0(1 - a * T^{1/2})$ behavior [1]. The high-temperature Curie-Weiss fit yields an effective magnetic moment $\mu_{\rm eff} = 2.74\mu_B$, which is larger compared to the free Ce^{3+}-ion value ($2.54\mu_B$); this may indicate a very weak magnetic contribution from the Co ion in $CeCo_2Ga_8$. This peculiarity is well known in Ce-based HF systems, for instance, CeCoAsO [39] and $CeCo_2As_2$ [40]. In the low-temperature limit as shown in Fig. 1(b), 0.1 K $\leqslant T \leqslant$ 2 K, $\rho(T)$ varies linearly with the temperature ($\rho \sim T$), characteristic of an NFL state. The heat capacity varies logarithmically, $C_p/T \sim -\ln(T)$, in the low-T limit as shown in Fig. 1(d). It is clear from the inset in Fig. 1(d) that the heat capacity exhibits a broad maximum near 90 K, which can the attributed to the Schottky anomaly arising from the crystal-field effect in the presence of the Kondo effect. All of these attributes of $CeCo_2Ga_8$ are rather similar to the quasi-1D NFL ground state and lead to further examination using a microscopic technique such as muon spin relaxation measurement.

To probe the NFL state as seen from electrical, thermal, and magnetic measurements at low T, we carried out ZF/LF-μSR measurements [41]. The ZF depolarization reveals the sum of the local responses of muons embedded at different stopping sites in $CeCo_2Ga_8$. ZF-μSR muon asymmetry spectra of $CeCo_2Ga_8$ at T = 70 mK (black symbols) and T = 4 K (blue symbols), which are representative of the data collected, are shown in Fig. 2(a). Both the 4 K and the 70 mK data in Fig. 2(a) reveal the same value of the initial asymmetry at t = 0 along with the lack of oscillations confirming the absence of long-range magnetic ordering in $CeCo_2Ga_8$ down to 70 mK. This precludes any argument for static magnetism in the sample [42]. Hence the moderate increase in the relaxation upon cooling from high temperatures reflects only a slowing-down of the electronic spin dynamics. Fits to the ZF-μSR data at different temperatures were done employing a Gaussian Kubo-Toyabe function multiplied by an exponential decaying function [43–45],

$$P_z(t) = A_1\left[\frac{1}{3}+\frac{2}{3}\left(1-\sigma_{\rm KT}^2t^2\right)\exp^{\left(\frac{-\sigma_{\rm KT}^2t^2}{2}\right)}\right]\exp\left(-\lambda_{\rm ZF}t\right)+A_{\rm bg}, \tag{1}$$

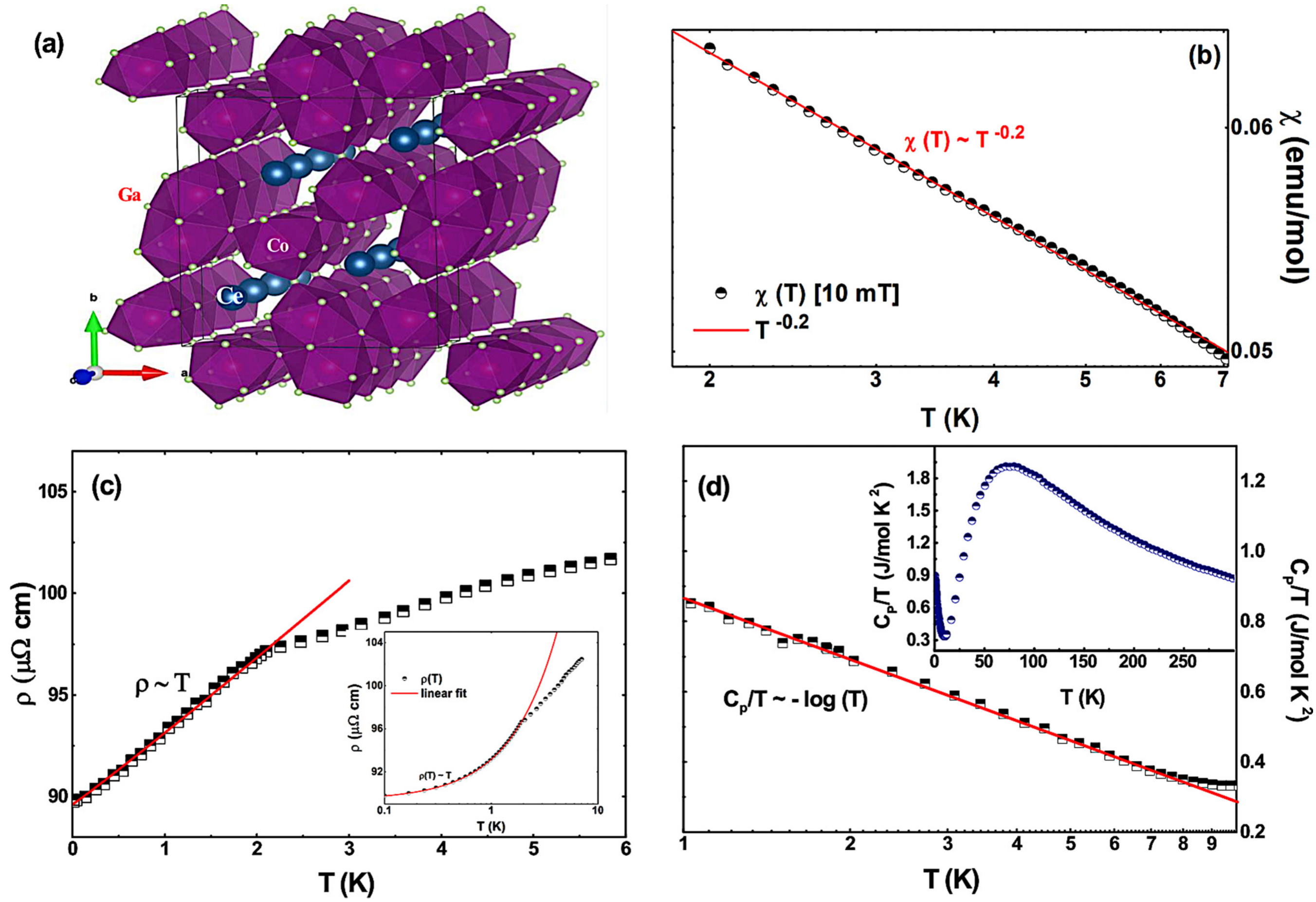

FIG. 1. (a) Crystal structure of $CeCo_2Ga_8$, showing the quasi-1D chains of Ce atoms along the c axis. The individual unit cell holds four Ce atoms. (b) Magnetic susceptibility as a function of temperature (log-log scale) parallel to the c axis ($\chi \sim T^{-0.2}$). (c) Low-temperature region of resistivity ($\rho \sim T$). Inset: $\rho(T)$ vs T plotted on the x axis as logarithmic scale down to 0.1 K. (d) Low-temperature dependence of heat capacity divided by temperature C_p/T plotted at a semilogarithmic scale in zero applied field ($C_p/T \sim -\ln T$). Inset: C_p/T over a large temperature range.

where λ_{ZF} is the ZF relaxation rate arising due to the local moment, and A_1 and A_{bg} are the asymmetries originating from the sample and background, respectively. A_{bg} was determined from the high-temperature ZF, which was kept fixed for the analysis. σ_{KT} is the nuclear contribution that emerges from the Gaussian distribution of the magnetic field at the muon site. The relaxation term $\exp(-\lambda_{ZF}t)$ is the magnetic contribution that comes from the fluctuating electronic spins, which provides information on the low-energy spin dynamics of $CeCo_2Ga_8$. Considering that we have observed only a single-exponential function in ZF-μSR data, this indicates that magnetic disorder in $CeCo_2Ga_8$ is negligible and hence the observed NFL behavior is intrinsic and has its origin in stoichiometric crystallographic-ordered $CeCo_2Ga_8$.

As shown in the left panel in Fig. 2(b), the T variation of λ_{ZF} increases sharply below 1 K, indicating the development of a NFL state as evidenced by the bulk properties. Above 1 K, λ_{ZF} decreases with increasing temperature. The right panel in Fig. 2(b) represents the Arrhenius-like behavior of $\lambda_{ZF}(T)$, i.e., follows the form $\lambda_{ZF} = \lambda_0 \exp\left(-\frac{E_a}{k_B T}\right)$, where E_a and k_B are the activation energy and Boltzmann constant, respectively. This confirms that the low-T spin dynamics of $CeCo_2Ga_8$ is thermally activated with $E_a = 2.3$ mK, which is similar to observations of $CeInPt_4$ [36] and CeRhBi [29], with E_a values of 2.9 and 140 mK, respectively. It is noteworthy that the amplitude of the thermal fluctuations decreases with decreasing temperature, and at 70 mK thermal fluctuations are not important. We therefore attribute the observed behavior of the temperature dependence of $\lambda_{ZF}(T)$ to the quantum fluctuations, which is in agreement with the observed behavior of the heat capacity, resistivity, and magnetic susceptibility of $CeCo_2Ga_8$. It is an open question why the temperature dependent relaxation of stoichiometric CeRhBi [29], $CeInPt_4$ [36] and $CeCo_2Ga_8$ compounds exhibits Arrhenius behavior in the NFL state (as $T \to 0$), while that of chemically disordered NFL systems such as $CeRh_{0.85}Pd_{0.15}$ exhibits power-law behavior [46]. We also plotted $\lambda_{ZF}(T)$ data for $CeCo_2Ga_8$ in a log-log plot [inset in Fig. 2(b)] to see the power-law behavior, but the data did not follow power-law behavior.

A longitudinal field of just 40 mT removes any relaxation due to a spontaneous field and is adequate to decouple muons from the relaxation channel, as presented in Fig. 2(c) at 250 mK. Once a muon is decoupled from the nuclear moments, the spectra can be best fitted using [47] $G_{z(t)} =$

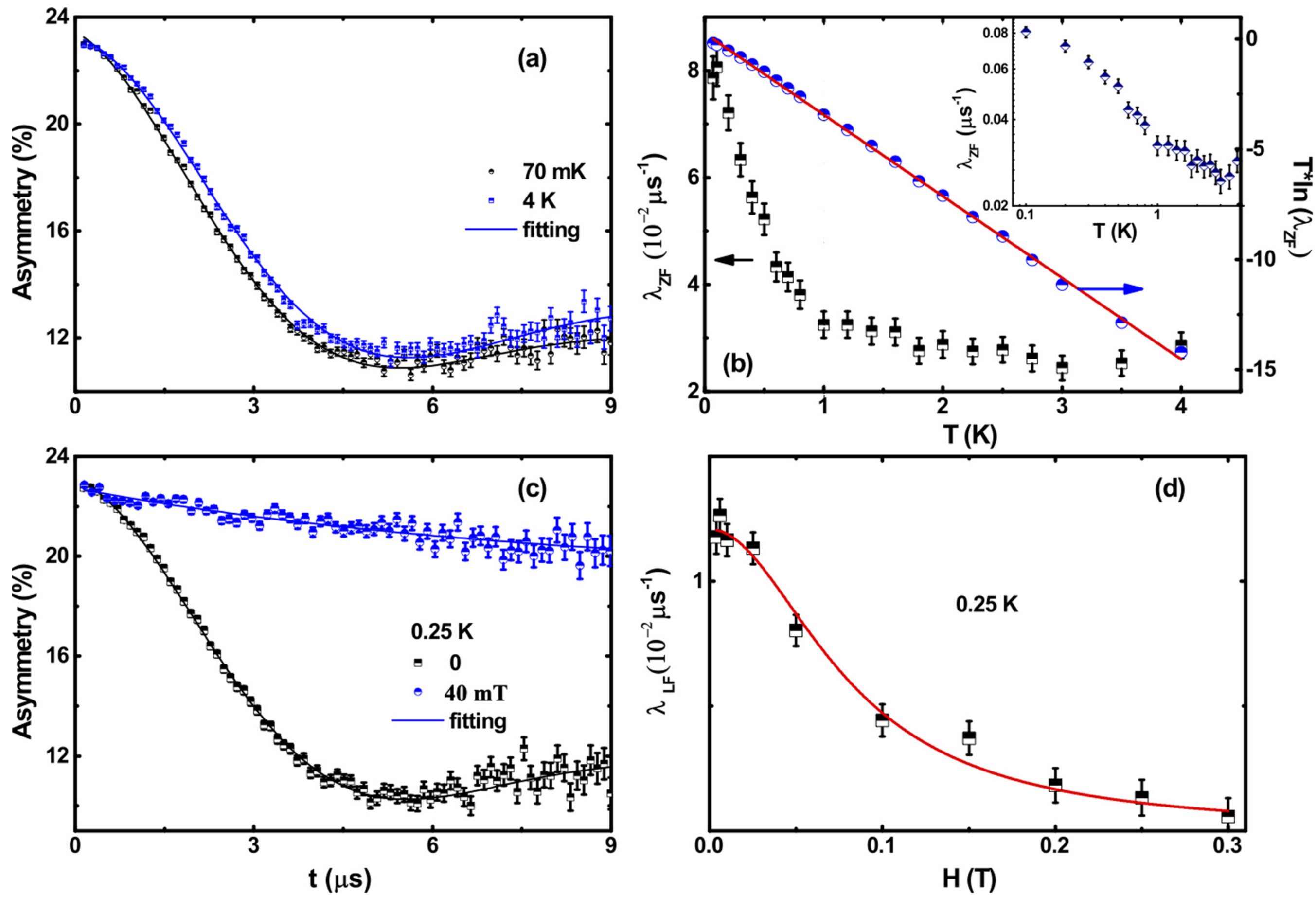

FIG. 2. (a) Time-dependent zero-field μSR spectra of $CeCo_2Ga_8$ collected at 70 mK (black symbols) and 4 K (blue symbols). Solid lines are the least-squares fit applying Eq. (1). (b) The left y axis plots the electronic contribution of the muon relaxation rate λ_{ZF}. A clear signature of quantum fluctuations is seen below 1 K, confirming the NFL state as shown by the bulk properties. The right axis demonstrates the Arrhenius behavior of λ_{ZF}. The line is the least-squares fit of the data, as presented in the text. Inset: A log-log plot of λ_{ZF} vs temperature. (c) Time-dependent longitudinal-field muon asymmetry spectra of $CeCo_2Ga_8$ measured at $T = 0.25$ K in zero field and at 40 mT. Solid lines are the least-squares fit to the raw data using Eq. (1). (d) Longitudinal-field dependence of the muon relaxation rate λ for $CeCo_2Ga_8$ at 0.25 K. The solid red line is the least-squares fit using Eq. (2).

$A_1 \exp(-\lambda_{LF}t) + A_{bg}$. λ_{LF} decreases rapidly at a low H and saturates at a high H. $\lambda_{LF}(H)$ can be adequately expressed by the standard description given by the Redfield formula [48,49],

$$\lambda = \lambda_0 + \frac{2\gamma_\mu^2 \langle H_l^2 \rangle \tau_C}{1 + \gamma_\mu^2 H^2 \tau_C^2}, \tag{2}$$

where λ_0 is the field-independent depolarization rate, and $\langle H_l^2 \rangle$ is the time-varying local field at muon sites due to the fluctuations of Ce $4f$ moments. H_l is the applied longitudinal field and the correlation time τ_C is related to the imaginary component of the q-independent dynamical susceptibility, $\chi''(w)$, through the fluctuation-dissipation theorem [50]. The red line in Fig. 2(d) represents the fit to the $\lambda_{LF}(H)$ data. The calculated parameters are $\lambda_0 = 0.19(1)$ μs^{-1}, $\langle H_l^2 \rangle = 1.3(1)$ mT, and $\tau_c = 3.1(6) \times 10^{-8}$ s. The value of the time constant of $CeCo_2Ga_8$ unveils a slow spin dynamics, which originates from the quantum critical fluctuations at low T. Similar values are observed for CeRhBi [29]: $\lambda_0 = 0.17(1)$ μs^{-1}, $\langle H_l^2 \rangle = 1.5(1)$ mT, and $\tau_c = 4.2(6) \times 10^{-8}$ s.

IV. CONCLUSION

In conclusion, we have presented magnetization, resistivity, heat capacity, and ZF/LF muon spin relaxation measurements on the quasi-one-dimensional $CeCo_2Ga_8$ compound. The linear behavior of $\rho(T)$, power-law diversion of $\chi(T) \sim T^{-0.2}$, and logarithmic divergence of $C_p(T)/T$ suggest a NFL ground state of $CeCo_2Ga_8$. Moreover, the increase in the ZF relaxation rate λ_{ZF} below 1 K and the Arrhenius-like behavior of $CeCo_2Ga_8$ suggest a NFL ground state, which is quite similar to that seen in other NFL systems, for example, CeRhBi [29] and $CeInPt_4$ [36]. Our ZF μSR measurements confirm the absence of long-range magnetic ordering down to 70 mK. Furthermore, the longitudinal-field dependence μSR study provides information on the spin fluctuation rate and the width of the field distribution at the muon sites. The observed quantum fluctuations below 1 K in the undoped $CeCo_2Ga_8$ compound make it a prototype material for investigation of the low-T quantum fluctuations in low-dimensional NFL systems and other Ce-128 counterparts with a $YbCo_2Ga_8$-type structure. This work will pave the way to our understanding of the NFL in 1D systems, both theoretically and experimentally.

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