Charge-stripe order, antiferromagnetism, and spin dynamics in the cuprate-analog nickelate La$_4$Ni$_3$O$_8$

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We report a muon spin rotation ($\mu$SR) study of the cuprate-analog nickelate La$_4$Ni$_3$O$_8$, which undergoes a transition at 105 K to a low-temperature phase with charge-stripe and antiferromagnetic (AFM) order on square planar NiO$_2$ layers. Zero-field $\mu$SR shows that this transition is abrupt and that the AFM configuration is commensurate. Comparison of observed muon precession frequencies with Ni dipolar field calculations yields Ni moments \(\lesssim 0.5\mu_B\). Dynamic muon spin relaxation above 105 K suggests critical slowing of Ni spin fluctuations, but is inconsistent with corresponding $^{139}$La NMR results. Critical slowing and an abrupt transition are also observed in the planar cuprate AFM La$_2$CuO$_{4+\delta}$, where they are evidence for weakly interplanar-coupled two-dimensional AFM spin fluctuations, but our $\mu$SR data do not agree quantitatively with theoretical predictions for this scenario.

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I. INTRODUCTION

Bednorz and Müller searched for superconductivity in nickel systems without success before they discovered superconducting La$_2$BaCuO$_4$ [1]. For many years after, high-$T_c$ superconductivity was found only in cuprate compounds with CuO$_2$-layered structures. It is now well known that besides the cuprates other layered systems (e.g., Fe-pnictide/chalcogenide and Bi$_2$S$_2$-based materials) also become superconductors, suggesting a potential universality in layered superconductivity. This in turn revives interest in the quest for Cu-analog, Ni-based, superconducting materials. The motivation for studying these materials lies in their potential to help answer current experimental and theoretical questions regarding superconductivity in layered compounds, to assess the universality of layer superconductivity, and to provide clues for where to search for new layered superconductors.

The nickelate La$_4$Ni$_3$O$_8$ is one such material [2, 3]. The crystal structure of this compound involves square planar NiO$_2$ layers, isostructural to the Cu$_2$O$_2$ layers of the cuprates if the Ni valence were 1+. A phase transition at 105 K involves both magnetic and charge stripe order [4–6].

This article reports a muon spin rotation and relaxation ($\mu$SR) study of La$_4$Ni$_3$O$_8$. $\mu$SR is a magnetic resonance technique [7], conceptually similar to NMR, that uses muon spins implanted into the sample as microscopic probes of their static and dynamic magnetic environment. We find that the transition is abrupt (discontinuous to within experimental resolution), into a commensurate antiferromagnetic (AFM) configuration. Experimental spectra of muon spin precession frequencies resemble histograms of calculated Ni dipolar fields for AFM stripe configurations, with Ni moments 0.4–0.5$\mu_B$, but the data do not pinpoint a specific pattern.

Dynamic muon spin relaxation above 105 K suggests critical slowing of Ni spin fluctuations. Critical slowing and an abrupt transition are expected for nearly two-dimensional (2D) systems, where critical behavior involves 2D AFM spin fluctuations that do not order at finite $T$ [8]. In this scenario the three-dimensional (3D) transition is due to a weak interplanar coupling, and does not dominate the critical spin dynamics. Evidence for 2D criticality is found from NMR studies of the planar cuprate AFM La$_2$CuO$_{4+\delta}$ [9], and in La$_4$Ni$_3$O$_8$ [10]. Our $\mu$SR data do not agree quantitatively with the corresponding theory [8, 11], however, and instead suggest a connection between critical fluctuations and the 105-K transition in La$_4$Ni$_3$O$_8$.

The article is organized as follows: The current understanding of La$_4$Ni$_3$O$_8$ and related systems is reviewed in Sec. II, with emphasis on the charge/stripe nature of the 105-K transition. Section III gives experimental details and a brief description of the $\mu$SR technique. Candidate muon stopping sites in the crystal are discussed in Sec. IV, and Sec. V reports $\mu$SR results in the ordered phase and their interpretation. Section VI discusses muon spin relaxation in the paramagnetic phase.

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above 105 K. Our results are summarized in Sec. VII, and the Appendix gives supplementary information on AFM configurations.

II. BACKGROUND

In 1999 Anisimov, Buhkvalov, and Rice [12] concluded “Only if the Ni ions are forced into a planar coordination with the O ions can a $S = 1/2$ magnetic insulator be realized with the difficult Ni$^{1+}$ oxidation state and possibly doped with low spin ($S = 0$) Ni$^{2+}$ holes directly analogous to the superconducting cuprates.” i.e., a system with electronic configuration Ni$^{1+}$/Ni$^{2+}$ would have the same configuration as that of Cu$^{2+}$/Cu$^{3+}$ in the cuprates, but to be a true analog it would also need to be in a square planar coordination with O ions. The Ni$^{1+}$/Ni$^{2+}$ configuration is rare in oxides but it does exist, and conforms with the condition of Anisimov et al. in the $T'$-type nickelates Ln$_{n+1}$Ni$_n$O$_{2n+2}$, Ln = La, Nd, n = 2, 3 and $\infty$ [13]. In this formula n is the number of NiO$_2$ layers in a multilayer of square-coordinated Ni ions.

Poltavets et al. [4] reported a magnetic transition at 105 K for La$_4$Ni$_3$O$_8$ (n = 3), and attributed it to Ni-spin inter-trilayer interactions in analogy with undoped La$_2$CuO$_4$. No transition was seen in the magnetization for applied fields lower than 0.1 T, but the observed change in slope of the resistivity $\rho(T)$ and the lambda anomaly in the specific heat at 105 K were taken as evidence for a transition in zero field. At low temperatures $\rho(T)$ exhibits semiconducting behavior [4]. $^{139}$La NMR measurements indicated that the transition is to an AFM phase (i.e., 105 K is a Néel temperature $T_N$), further suggesting the possibility of a cuprate analog.

The results of Ref. 4 triggered a good deal of theoretical and experimental work [10, 14–19]. Pardo and Pickett [14] predicted an insulating ground state of the Mott type, and related the transition to binding of $d^2$ orbitals of Ni ions in outer and middle layers of a trilayer (designated Ni1 and Ni2 respectively). Checkerboard AFM ordering in NiO$_2$ planes was assumed, although powder neutron diffraction has not yielded specific indications for magnetic order [3, 4]. A predicted metal-insulator transition under pressure [14] was not observed [16].

Liu et al. [17] and Wu [18] predicted La$_4$Ni$_3$O$_8$ to be a C-type AFM using different theoretical methods. A correlation of the metal/insulator character of the system with dimensionality was found from first-principle calculations in La$_{n+1}$Ni$_n$O$_{2n+2}$ by Liu et al. [20]. The quasi-2D $n = 2$ and 3 varieties were predicted to be molecular insulators in contrast with 3D ($n = \infty$) LaNiO$_2$, which according to their calculated electronic structure is metallic by virtue of Ni-La hybridization. The inclusion of electron correlation effects in first-principles calculations by Patra et al. [19] suggested that the two inequivalent Ni1 and Ni2 atoms in the La$_4$-$x$Sr$_x$Ni$_3$O$_8$ crystal structure are both in a high-spin state, with an average valence of $+1.33$ independent of $x$ doping.

There were indications, however, that the physics of the nickelates is more complicated than expected. A fit to the temperature dependence of the $^{139}$La NMR relaxation rate $1/T_1$ in La$_4$Ni$_3$O$_8$ [10] required a Korringa term $1/T_1T = \text{const.}$, indicative of a Fermi liquid phase. This is in contrast to the AFM cuprate compounds, which are Mott insulators. In addition, $1/T_1$ above $T_N$ could be fit as diverging at $T = 0$ [10], rather than at $T_N$ as expected for a second-order (continuous) transition in 3D. A $T = 0$ divergence is characteristic of critical fluctuations of a 2D Heisenberg antiferromagnet [8, 11], and is observed in the quasi-2D cuprate antiferromagnet La$_2$CuO$_4$ [9]. Zeeman-split $^{139}$La NQR experiments in zero applied field [21] determined that the transition is abrupt but continuous. By analogy the magnetic transition in La$_4$Ni$_3$O$_8$ might be due to weak interplanar exchange between large 2D AFM fluctuations (correlation length $\gg$ lattice constant) as in La$_2$CuO$_4$. But the situation was clearly not well understood.

More recently, NMR measurements by ApRoberts-Warren et al. [22] in the paramagnetic phase of the bilayer ($n = 2$) $T'$-type compound La$_2$Ni$_2$O$_6$ revealed very similar behavior of the $^{139}$La relaxation rate to that found previously in trilayer La$_4$Ni$_3$O$_8$. Although they observed no magnetic transition in the bilayer down to 5 K, their results were ascribed to similarity in the electronic structure of the two materials, presumed to be of the 2D variety.

The discussion of the 105-K transition in La$_4$Ni$_3$O$_8$ shifted in 2016, when Zhang, Chen et al. [5] reported results from synchrotron x-ray diffraction measurements on single crystals. They found a quasi-2D stripe-ordered charge configuration below 105 K, with orientation at $45^\circ$ to the Ni-O-Ni bonds in NiO$_2$ squares and a 2D supercell $3\sqrt{2}a \times \sqrt{2}a$, where $a$ is the Ni-Ni nearest-neighbor distance. This is similar to the charge order found in La$_{1.5}$Sr$_{1.5}$NiO$_4$, a related 1/3-hole doped single-layer Ruddlesden-Popper nickelate, where the average Ni valence is 2.33+. The stripe pattern in La$_4$Ni$_3$O$_8$ is two Ni$^{2+}$ ($S = 1/2$) rows followed by one Ni$^{2+}$ ($S = 0$) row, oriented at $45^\circ$ to the planar Ni-O bonds, with staggered stacking along the c-axis. The 2:1 ratio preserves the formal Ni valence 1.33+. All three layers in a trilayer possess stripes. The magnetic structure was not obtained in this study, which sampled only charge degrees of freedom.

$Ab$ initio calculations by Botana et al. [23] yielded stripe charge order from a combination of structural distortion and AFM order, in which Ni$^{2+}$ ions have spin $S = 0$, and Ni$^{1+}$ $S = 1/2$ ions order antiferromagnetically, similar to the spin-1/2 insulating antiferromagnetism of the cuprate parent materials. In this result the valences of corresponding Ni1 and Ni2 ions in a trilayer are the same. Two 2D supercells of the observed stripe pattern and the proposed 2D AFM order in a single Ni layer [23] are shown in Fig. 1.

Very recently Zhang, Pajerowski et al. [6] reported results of neutron Bragg diffraction experiments on
La$_4$Ni$_3$O$_8$. Their measurements confirm the magnetic ground state, with stripe AFM order commensurate with the charge stripes in individual trilayers but little or no correlation between trilayers. In the following we discuss in detail relations between these findings and our $\mu$SR results.

III. EXPERIMENT

a. Sample. Polycrystalline La$_4$Ni$_3$O$_8$ was prepared as described previously [3] and characterized by powder x-ray diffraction, x-ray absorption spectroscopy, magnetization, specific heat, resistivity, and NMR measurements [3, 4, 10]. For our $\mu$SR experiments we used enough fine powder to fully cover a circular area of about 1 cm in diameter and 1 mm thickness.

The two-trilayer tetragonal unit cell of La$_4$Ni$_3$O$_8$ (space group $I4/mmm$, no. 139) is shown in Fig. 2. There are two inequivalent Ni sites (Ni1 and Ni2) and three inequivalent oxygen sites, two of which coordinate the Ni ions in the planar NiO$_2$ layers. The third oxygen sites form separate square-coordinated layers, with La ions above and below the square centers forming flourite La-O blocks that separate and isolate one NiO$_2$ trilayer from the next. NiO$_2$ layers within a trilayer are separated by La-only planes. The candidate muon sites shown in Fig. 2 are discussed in Sec. IV.

b. $\mu$SR experiments. In the $\mu$SR technique [7], 100% spin-polarized muons are implanted into a sample, where they decay $[\mu^+ \rightarrow e^+ + \nu_e (\sqrt{s}) + \nu_\mu (\sqrt{s})]$ with a mean lifetime 2.197 $\mu$s. Usually positive muons ($\mu^+$) are used, which stop in interstitial sites. They are more sensitive to their magnetic environment than negative muons, which go into tight Bohr orbits around host nuclei. The decay positron is preferentially emitted in the direction of the $\mu^+$ spin at the time of decay. Thus the time-dependent positron count-rate asymmetry $A(t)$ (the asymmetry spectrum), where $t$ is the time after implantation, directly yields the evolution of the ensemble $\mu^+$ spin polarization $G(t)$ [$G(0) \equiv 1$]: $A(t) = A_0 G(t)$, where the initial asymmetry $A_0 = 0.2$–0.3 is instrument-dependent. Information on local magnetic behavior is contained in $G(t)$. Measurement of a single asymmetry spectrum typically requires $\gtrsim 10^7$ events.

$\mu$SR experiments on La$_4$Ni$_3$O$_8$ were carried out with the LAMPF spectrometer at the M20D muon beam line, TRIUMF, Vancouver, Canada, over the temperature range 1.2–300 K, in zero magnetic field (ZF) and in longitudinal applied fields (LF) (field parallel to the initial $\mu^+$ spin polarization) up to 4 kOe. Data were analyzed using the Paul Scherrer Institute musrfit fitting program [24] and the TRIUMF PHYSICA programming environment [25].

IV. $\mu^+$ STOPPING SITES

$\mu^+$ precession frequencies in ordered magnets yield static magnetic field values at the muon stopping sites, which can help to characterize the magnetic order if these sites are known. Implanted positive muons in oxides are likely to stop near O$_2^-$ ions, and in La$_4$Ni$_3$O$_8$ with stripe order the large unit cell yields a considerable number of such candidate sites. To date there are no reported
calculations of site locations or relative occupations in La$_4$Ni$_3$O$_8$ using, for example, DFT theory [26]. In lieu of such information, we consider candidate muon sites that are symmetric with respect to oxygen near neighbors. Eight such sites that are crystallographically inequivalent are shown in Fig. 2 and listed in Table I. It is customary [5, 15, 23] to describe magnetic and stripe structure in La$_4$Ni$_3$O$_8$ using the setting $F4/mmm$, in which basal-plane unit vectors are rotated 45° to define a face-centered square lattice of Ni ions. The lattice constants in these directions are then $\sqrt{2}a$ times the Ni-Ni nearest neighbor distance $a$ in the basal plane. The $c$ lattice constant remains the same as in the $I4/mmm$ setting, and the unit cell volume is doubled. The observed charge-stripe supercell [5] is $3\sqrt{2}a \times \sqrt{2}a \times c$, where the stripe direction is the $b$ axis (cf. Fig. 1). Muon sites are given in Table I for both settings.

In the $I4/mmm$ tetragonal unit cell there are a total of 58 such sites (the sum of the Wyckoff multiplicities in Table I). With two trilayers per $I4/mmm$ unit cell, there are 58 sites per trilayer in the $F4/mmm$ setting and $3 \times 58 = 174$ sites per trilayer in a stripe supercell. The two trilayers are crystallographically equivalent, so that it is sufficient to consider $\mu^+$ sites for a single trilayer provided this equivalence extends to the ordered magnetic structure. Figure 3 shows two $F4/mmm$ supercells of La$_4$Ni$_3$O$_8$ viewed along the $c$ axis, with the stripe AFM configuration of Fig. 1 [23] and $\mu^+$ sites from Table I. The number of magnetically inequivalent sites in a full 3D AFM configuration is clearly greater in stripe models than for AFM order without stripes.

The $\mu^+$ sites of Table I may not be realistic: they would be unstable if the O$^{2-}$ ions were classical point charges, and our calculations do not take account of off-site ionic positions, intrinsic to the La$_4$Ni$_3$O$_8$ crystal structure [23] or due to the $\mu^+$ charge. The candidate sites sample a large fraction of the unit cell, however, (Fig. 2) so that calculated dipolar fields at these sites in the magnetically ordered phase can be taken as rough guides to the local field distribution.

V. ANTIFERROMAGNETIC CONFIGURATIONS

A. Experimental results

A representative series of ZF-$\mu$SR asymmetry spectra for temperatures between 1.85 K and 105 K (the AFM phase) is shown in Fig. 4. Of particular interest is the unambiguous appearance of oscillations due to $\mu^+$ spin precession in well-defined magnetic fields (oscillation frequency $\omega = \gamma_\mu H$, where $\gamma_\mu$ is the muon gyromagnetic ratio). Figure 5 shows Fourier transforms of ZF-$\mu$SR asymmetry data from La$_4$Ni$_3$O$_8$ from 2.2 K to 110 K. There are six discrete peaks with nonzero frequency, indicating the presence of at least this many magnetically inequivalent $\mu^+$ stopping sites. Some of the peaks are broad, likely containing unresolved frequencies from multiple sites, but others are quite narrow except at low temperatures. In the temperature range 99–105 K the

![FIG. 3. 2D AFM structure of Fig. 1 [23] and projections of candidate $\mu^+$ stopping sites in La$_4$Ni$_3$O$_8$ (Fig. 2 and Table I) onto the $ab$ plane in the $F4/mmm$ setting of the $3\sqrt{2}a \times \sqrt{2}a \times 2d$ 2D stripe supercell. Large circles: Blue: Ni$^{1+}$ ($S = 1/2$) (♦ = up-spins, ♣ = down-spins). Yellow: Ni$^{2+}$ ($S = 0$). Red: O$^{2-}$. Small circles: $\mu^+$ sites from Table I. Rectangle: supercell boundary.

![FIG. 4. Representative ZF-$\mu$SR asymmetry spectra in La$_4$Ni$_3$O$_8$, $T \leq T_N = 105$ K. Curves: fits of Eq. (1) to the data.](image-url)
TABLE I. Candidate symmetric $\mu^+$ stopping sites in La$_4$Ni$_3$O$_8$. Coordinates are fractions of lattice parameters in the $I4/mmm$ and $F4/mmm$ settings (see text). Coordinates and O$^{2-}$ distances from lattice data of Ref. 3.

| $\mu^+$ site | $I4/mmm$ | Wycoff designation | $F4/mmm$ | O$^{2-}$ distance (Å) | Location       |
|--------------|----------|--------------------|----------|----------------------|----------------|
| $\mu1$       | $\frac{1}{4}$ | $\frac{1}{4}$ | 0        | $8h$                 | 1.4039         | nn O in $ab$ plane |
| $\mu2$       | $\frac{1}{4}$ | $\frac{1}{4}$ | 0.1256   | 16m                  | "   "          | "   "            |
| $\mu3$       | $\frac{1}{4}$ | $\frac{1}{4}$ | $\frac{1}{4}$ | 8f                  | "   "          | "   "            |
| $\mu4$       | 0         | $\frac{1}{4}$   | 0.1878   | 8g                   | 1.6238         | nn O along c axis |
| $\mu5$       | 0         | $\frac{1}{4}$   | 0.0628   | 8g                   | 1.6394         | "   "            |
| $\mu6$       | $\frac{1}{4}$ | $\frac{1}{4}$ | 0        | 2b                   | 1.9854         | centered in O square |
| $\mu7$       | $\frac{1}{4}$ | $\frac{1}{4}$ | 0.1256   | 4e                   | "   "          | "   "            |
| $\mu8$       | $\frac{1}{4}$ | $\frac{1}{4}$ | $\frac{1}{4}$ | 4e                  | "   "          | "   "            |

FIG. 5. Fourier-transform spectra from ZF-$\mu$SR asymmetry data (cf. Fig. 4) in La$_4$Ni$_3$O$_8$, $2.2\;K \leq T \leq 110\;K$. The spectra are offset for clarity.

The frequency of the high-frequency ($\sim 19\;MHz$) peak hardly changes, but its amplitude decreases smoothly to nearly zero, suggesting a spread of Néel temperatures of a few kelvin.

The curves in Fig. 4 are fits to the data using the exponentially-damped multi-frequency function

$$G(t) = f_0 \exp(-\lambda_0 t) + \sum_{i=1}^{6} f_i \exp(-\lambda_i t) \cos(\omega_i t + \phi_i)$$

for the $\mu^+$ spin polarization [27]. The frequencies $\omega_i$ give the local magnetic fields, and the line amplitudes $f_i$ are related to $\mu^+$ site occupation probabilities. Frequencies obtained from fits of Eq. (1) to asymmetry data are plotted vs temperature in Fig. 6. The peaks exhibit differences in temperature dependence, suggesting Ni moment reorientation within the overall AFM order. The damping rates $\lambda_i$ (not shown) increase with decreasing temperature, suggesting a partial disordering of the magnetic structure.

From Figs. 4–6 we conclude that the transition at $T_N = 105\;K$ has a clear magnetic component with an abrupt onset. The well-defined frequencies indicate commensurate AFM order, since in an incommensurate structure crystallographically equivalent muon stopping sites would yield a continuous distribution of frequencies. This result is consistent with recent neutron diffraction experiments [6]. An AFM transition was also observed in

FIG. 6. Temperature dependence of muon precession frequencies in La$_4$Ni$_3$O$_8$ from fits of Eq. (1) to ZF-$\mu$SR asymmetry data (cf. Figs. 4 and 5).
Neutron diffraction and NMR data both exhibit a smooth transition, however, in contrast with our results. The NMR study used a partially-aligned powder sample, so that the resolution of the $^{139}$La resonance in a 42-kOe field was limited by partial powder broadening; the field may have affected the behavior of the transition. The discrepancy with the (zero-field) neutron data is discussed below in Sec. VII.

The polycrystalline nature of our sample is not involved in the ZF results shown in Fig. 4, except that the oscillation amplitudes are averaged over local-field orientations relative to the initial $\mu^+$ polarization direction.

**B. Models of AFM order: dipolar-field calculations**

This section describes lattice-sum calculations of Ni-moment dipolar magnetic fields at the candidate $\mu^+$ stopping sites of Table I for a number of candidate AFM configurations, with and without charge stripes. The calculations assume the same moment on all Ni$^{1+}$ ions in a trilayer, consistent with the neutron diffraction results [6]. Dipolar fields are summed over moment-bearing Ni ions within a sphere of radius 200 Å centered on the $\mu^+$ site, after determining that increasing this radius does not affect the results appreciably. The Ni-ion moment is scaled for rough agreement of the dipolar-field frequencies with a typical experimental spectrum ($T = 50$ K, cf. Fig. 5).

1. **AFM configurations with charge stripes**

   a. **Stripe models.** For completeness, we consider the four charge stripe models discussed by Zhang, Chen et al. [5], which differ in the positions of the stripes on the individual layers. These models are shown in Fig. 7, where the view is along the stripe direction (the $b$ direction in the $F4/mmm$ setting), and a supercell boundary is shown. The model numbering (1–4) of Ref. 5 is used in the following. The stripes in a trilayer are stacked along the $c$ axis in Models 1 and 4 and staggered in Models 2 and 3. The Coulomb repulsion energy would seem to be lower for the latter, but x-ray diffraction data favor the stripe structure of Model 1 [5]. The latter is also used for AFM configurations in the neutron diffraction study of Ref. 6. Model 4 is assumed in the $ab$ initio treatment of charge and AFM order in La$_4$Ni$_3$O$_8$ of Botana et al. [23], where it is argued that the weak coupling between trilayers does not play a significant role in the electronic structure. We discuss Model 1 below, and Models 2–4 are described in the Appendix.

   The calculations and neutron diffraction results cited above [6, 23] both yield the 2D AFM structure shown in Fig. 1, with Ni moments aligned along the $c$ axis. We have calculated dipolar fields at $\mu^+$ sites for 16 3D AFM structures, obtained by placing 2D AFM layers with this structure on each of the four stripe models in four configurations, designated A–D in the following. In Configuration A the moment orientations are those of Fig. 1 on all layers. In Configuration B all moments on alternate trilayers of Configuration A are inverted. In Configuration C the Ni2 (middle layer) moments of Configuration A are inverted for all layers, yielding intra-trilayer AFM ordering. Configuration D can be obtained in either of two ways: by inverting the Ni2 moments of Configuration B, or by inverting alternate trilayers of Configuration C.
b. Dipolar fields in stripe Model 1. The four AFM moment configurations are shown in Fig. 8 for charge stripe Model 1, and histograms of frequencies from $\mu^+-$

![Image of AFM moment Configurations A–D in La$_4$Ni$_3$O$_8$ on charge stripe Model 1 [5]. Blue spheres: Ni$^{1+}$ ions ($S = 1/2$). Yellow spheres: Ni$^{2+}$ ions ($S = 0$) Rectangles: $3\sqrt{2}a \times \sqrt{2}a \times c$ supercell outlines. Arrows: AFM ordered moments. The view is along the stripe (b) direction.

site dipolar-field calculations for these configurations are compared with the observed 50-K spectrum in Fig. 9. The calculated histograms assume equal stopping probabilities for all $\mu^+$ sites, which may not be the case, and do not take into account possible transferred muon hyperfine fields. A rough correspondence between the calculated histograms and observed spectra is obtained with Ni$^{1+}$ moments 0.4–0.5$\mu_B$, somewhat smaller than the predicted values 0.6–0.7$\mu_B$ [23]. A high-frequency group of peaks is separated from a low-frequency group by a region with less spectral weight. The comparison suggests that the high-frequency peak is formed from a group of discrete frequencies. These are mainly from $\mu$1 sites, which is not surprising as they are the closest to Ni ions (Fig. 3).

There are a number of general features of the calculated histograms (Fig. 9 and the Appendix). Histograms for stripe Models 1 and 4 are similar, as are those for stripe Models 2 and 3, due to stacked stripes in the former compared to offset stripes in the latter. For a given model, only small differences and no change of Ni moment are found between A and B histograms, and also between C and D histograms. Inverting alternate trilayers changes the $\mu^+$-site fields considerably less than inverting Ni$^2$ moments within each trilayer.

Reference 6 concludes that trilayers are “essentially uncorrelated” along the c axis, corresponding to a random mix of Configurations 1A and 1B (or 1C and 1D). From Fig. 9 it is clear that the differences in dipolar field distributions between these configurations, or a hypothetical random mix of them, are too small to have been resolved in the experimental spectrum.

For all stripe models a calculated peak or group is
found near the observed small peak at \( \sim 12 \) MHz for AFM Configurations A and B but not for Configurations C and D; agreement with the observed spectrum is better for the former configurations. This is not strong evidence for them, however, due to the approximate nature of the treatment and the uncertainty in the muon sites. Nevertheless, we note the discrepancy between this result and the neutron scattering data \([6]\), which favor the AFM intra-trilayer order of Configurations 1C and 1D (trilayer model 5 of Ref. 6, Supplementary Information) over the FM intra-trilayer order of Configurations 1A and 1B (their model 6).

2. AFM order without stripes?

For completeness we also consider AFM configurations without stripes (although these are ruled out by the neutron diffraction results \([6]\)). Using the \( F4/mmm \) setting, Fig. 10 shows four such configurations in \( \text{La}_4\text{Ni}_3\text{O}_8 \): the ferromagnetic (FM) configuration (for completeness), and three AFM configurations \([15, 17–19]\). Figure 11 compares frequency histograms from dipolar field calculations for the configurations of Fig. 10 with the observed 50-K spectrum. The subscripts 1 and 2 for the FM and A-AFM configurations designate alternate trilayer moments as pictured in Fig. 10 or inverted, respectively. Alternate-trilayer inversion followed by 90° rotation about the \( c \) axis is a symmetry operation for the C-AFM and G-AFM configurations, and therefore does not affect the dipolar-field histogram.

These histograms have a rough correspondence with those for the stripe-model AFM configurations of the previous section and, similarly, have no clear agreement with details of the experimental spectrum. This is not surprising, since commensurate stripes are long-range order (i.e., points in reciprocal space) best probed by a diffraction technique, whereas \( \mu\text{SR} \), like NMR, is a point probe in real space. The evidence for a striped AFM configuration from by neutron diffraction \([6]\) is much more compelling.

![FIG. 10. AFM Ni-ion moment configurations without stripes in \( \text{La}_4\text{Ni}_3\text{O}_8 \), \( F4/mmm \) setting. A-AFM: FM intra-trilayer, AFM inter-trilayer. C-AFM: AFM intra-trilayer, FM inter-trilayer. G-AFM: AFM intra-trilayer, AFM inter-trilayer.](image)
VI. PARAMAGNETIC PHASE

Two mechanisms for $\mu^+$ spin relaxation are expected in the high-temperature paramagnetic phase of a local- 
moment magnet: dynamic relaxation due to local- 
moment fluctuations, and (quasi)static relaxation from 
nuclear dipolar fields. In zero or low applied fields the 
resulting Kubo-Toyabe (KT) relaxation [28] is due to the 
combined effects of these mechanisms.

LF-µSR is a standard method for separation of the 
static and dynamic contributions [7], since a longitudi- 
unal field $H_L$ “decouples” the $\mu^+$ spin polarization from 
the static local field distribution [29]. Static $\mu^+$ re- 
 laxation functions $G_s(H_L, t)$ for arbitrary $H_L$ (including 
zero field) have been determined for several local field 
distributions [7, 28, 30, 31]. The combined effect of static 
and dynamic relaxation is often modeled by exponential 
damping of $G_s(H_L, t)$ with rate $\lambda$:

$$G(H_L, t) = e^{-\lambda t}G_s(H_L, t). \quad (2)$$

$$G_s^{\text{Lor}}(H_L, a, t) = 1 - \frac{a}{\omega_L}j_1(\omega_L)\exp(-at) - \left(\frac{a}{\omega_L}\right)^2\left[j_0(\omega_L t)\exp(-at) - 1\right]$$

$$- a\left[1 + \left(\frac{a}{\omega_L}\right)^2\right]\int_0^t j_0(\omega_L \tau)\exp(-a\tau)\,d\tau, \quad (4)$$

where $a/\gamma_\mu$ is the half-width of the Lorentzian field 
distribution, $\omega_L = \gamma_\mu H_L$, and $j_0$ and $j_1$ are spherical Bessel functions. In the ZF limit Eq. (4) simplifies to

$$G_s^{\text{Lor}}(a, t) = \frac{1}{3} + \frac{2}{3}(1 - at)\exp(-at). \quad (5)$$

The stretched-exponential form of the damping in Eq. (3) 
represents an inhomogeneous distribution of local dy- 
namic relaxation rates $\lambda_{loc}$ [32, 33], where $\lambda^*$ is a 
characteristic rate (but not the average [33]) and the 
stretching power $\beta < 1$ characterizes the distribution 
function $P(\lambda_{loc})$ of the local rates.

The stretched-exponential dynamic and Lorentzian 
static forms are crude approximations (both are strictly 
valid only for a $r^{-3}$ interaction with local moments in the 
dilute limit [30, 34]). Their use is justified by their rela- 
tive simplicity, and a posteriori by the fact that they give 
good fits to the data (see below). An alternative model 
for static relaxation in disordered systems, the “Gaus- 
sian broadened Gaussian” KT function [31], does not fit 
well; for early times it varies as $t^2$ (i.e., as a Gaussian), 
whereas the data exhibit the linear early-time behavior 
of an exponential [30].

In La$_4$Ni$_3$O$_8$ $G(H_L, t)$ is complicated by the large num- 
ber of inequivalent $\mu^+$ sites. This results in a broad in- 
homogeneous distribution of $\mu^+$ dipolar fields from Ni$^{1+}$ 
 moments, as can be seen from the AFM-phase histograms 
discussed above. Similarly, the dipolar-field distribution 
from randomly-oriented$^{139}$La nuclei is further broadened 
because of the multiple $\mu^+$ sites. Obtaining $G(H_L, t)$ is 
then a difficult numerical problem even if these distribu- 
tions were known, which is not the case for La$_4$Ni$_3$O$_8$ 
because of uncertainty in the $\mu^+$ sites.

We have carried out ZF- and LF-µSR experiments in 
the paramagnetic phase of La$_4$Ni$_3$O$_8$ between $T_N$ 
and 275 K. For analysis of the data we have chosen a rough 
approximate form of Eq. (2), using the static Lorentzian 
KT function $G_s^{\text{Lor}}(H_L, a, t)$ [30] with “stretched exponent- 
ial” damping:

$$G(H_L, a, t) = \exp\left[-(\lambda^* t)^\beta\right] G_s^{\text{Lor}}(H_L, a, t). \quad (3)$$

We have taken a Lorentzian distribution of $^{139}$La dipol- 
ar fields as an approximation beyond the Gaussian that 
is appropriate for a single $\mu^+$ site [28]. The LF static 
Lorentzian KT function is [30]

A. Longitudinal field, $T = 200$ K

Figure 12 shows representative asymmetry spectra at 
200 K for $0 \leq H_L \leq 4$ kOe. The curves are fits of
laxation slows with increasing field but the early-time relaxation is not affected, which is a characteristic of static local-field decoupling. For higher fields the initial slope decreases, indicating a slight field dependence of $\lambda^*$ and/or $\beta$.

The observed spectra are quite smooth, and the parameters $a$, $\lambda^*$, and $\beta$ in Eqs. (3)–(4) are highly correlated statistically, making it hard to determine them separately. The following procedure was used: at high enough fields $G_{\text{Lor}}(H_L, t) = 1$ independent of $a$, so that stretched-exponential fits determine $\lambda^*$ and $\beta$. Assuming these are roughly field-independent, they were fixed and the lowest-field data were fit to determine $a$. Then all three parameters were iteratively fixed and freed at low fields: $a$ was fixed and $\lambda^*$ and $\beta$ freed, and vice versa, until convergence was obtained. Finally $a$ was fixed, and $\lambda^*$ and $\beta$ were freed for fits at all fields.

The field dependences of $\lambda^*$ and $\beta$ at 200 K are shown in Fig. 13. The scatter in the parameters is obviously strongly correlated. The field dependences at low fields are probably artifacts of the assumed field independence of $a$; a field dependence (as discussed in Ref. 28 for the Gaussian relaxation rate at a single $\mu^+$ site) might be expected in La$_4$Ni$_3$O$_8$ because the $\mu^+$ frequency $\gamma_\mu H_L$ is of the order of the lower of the two observed $^{139}\text{La}$ quadrupolar splittings [4] for $H_L \sim 100$ Oe. Nevertheless, for $H_L \gtrsim 50$ Oe the fits no longer depend on $a$, and the parameters are only weakly field-dependent. The stretching power $\beta$ is close to the value $1/2$ expected from a Lorentzian distribution of fluctuating fields in the motional-narrowing limit (rapid fluctuations) [30, 34], which is perhaps further justification for the use of Lorentzian distributions to describe $\mu^+$ relaxation in La$_4$Ni$_3$O$_8$.

B. Longitudinal field, $T = 110$ K

A similar analysis was applied to LF data taken for $T = 110$ K, a temperature just above $T_N$. Representative asymmetry spectra are shown in Fig. 14, and the field dependences of $\lambda^*$ and $\beta$, determined as described above for 200 K, are shown in Fig. 15. The values of $\beta$ at the two temperatures are comparable, but at 110 K a clear increase of $\lambda^*$ at low fields is observed. This increase and the temperature independence of $\lambda^*$ at high fields are discussed further in Sec. VII.
C. Zero field

Representative ZF asymmetry spectra above the transition temperature $T_N$ are shown in Fig. 16, and Fig. 17 shows the temperature dependence of $\lambda^*$ and $\beta$ in zero field from fits of Eqs. (3)–(5) to the data. The 200-K value of $a$ is used, but the results do not change qualitatively for $\sim 10\%$ variations of $a$. The stretching power $\beta(T)$ is essentially constant, whereas $\lambda^*(T)$ increases markedly with decreasing temperature as $T_N$ is approached.

The curves in Fig. 17(a) give fits of theoretical expectations for 2D AFM spin fluctuations [11, 35, 36], as discussed in Ref. 10 with respect to $^{139}$La NMR relaxation data. The solid curve is a phenomenological Curie-Weiss-like fit: $\lambda^*(T) \propto T/(T - T_0)$ [35, 36]. This yields $T_0 = 90(1)$ K, in disagreement with the NMR result $T_0 = 0$ [10]. The dashed curve in Fig. 17(a) is the function
\[ \lambda^*(T) \propto x^{3/2} e^{1/x}/(1 + x)^3, \quad x = T/1.13J, \]
derived by Chakravarty and Orbach [11] from the treatment of 2D spin fluctuations in the renormalized classical regime by Chakravarty, Halperin, and Nelson [8] in which the 2D critical point is at $T = 0$. The fit value of the AFM exchange coupling $J$ is $86(22)$ K; the corresponding NMR value is $129(5)$ K [10]. Equation (6) is, however, only valid for $T < 0.57J = 49–74$ K. Temperatures in the paramagnetic phase of La$_4$Ni$_3$O$_8$ violate this condition, so that the fits are inconsistent.

Values of $\lambda^*$ for $H_L = 0.8$ Oe at $T = 200$ K and 110 K (Figs. 13 and 15) are larger than the ZF values at those temperatures (Fig. 17). This field dependence at low fields is not presently understood.

VII. DISCUSSION AND CONCLUSIONS

A. AFM transition and configuration

Zero-field $\mu$SR asymmetry spectra in La$_4$Ni$_3$O$_8$ exhibit an abrupt and possibly discontinuous transition at $T_N$ into a commensurate AFM structure (Sec. V A). Commensurate AFM is also found by neutron Bragg diffraction [6], but it and NMR [10] both observe a smooth transition. This is similar to the situation in the layered AFM cuprate La$_2$CuO$_{4+\delta}$, where an abrupt transition is observed in NQR and other hyperfine experiments but not in neutron diffraction (See, e.g., Ref. 21 and references therein). The discrepancy can be explained by an inhomogeneous distribution of Néel temperatures, since then “... The AFM Bragg intensity increases smoothly as the temperature is decreased through the transition not because the transition is smooth, but because the AFM fraction of the sample is increasing smoothly” [21].

In La$_4$Ni$_3$O$_8$ this scenario is strongly suggested by the behavior of the high-frequency line near $T_N$ (Fig. 5): its amplitude increases smoothly through the transition, but its frequency hardly changes. $^{139}$La NQR spectra in La$_2$CuO$_{4+\delta}$ exhibited a transition that was abrupt but continuous only in a particularly homogeneous single crystal [21] but otherwise discontinuous, so that the discontinuity in La$_4$Ni$_3$O$_8$ reported here might not be intrinsic.

Histograms of calculated Ni-ion dipolar fields at candidate $\mu^+$ sites for a number of charge stripe models and AFM configurations capture general features of frequency spectra and suggest reduced Ni moments (0.4–0.5$\mu_B$), but do not uniquely determine the specific AFM configuration. Dipolar field histograms for AFM configurations without stripes have somewhat less structure than the observed spectra.
The abrupt onset of nonzero spontaneous $\mu^+$ frequencies (Fig. 5) is suggestive of a first-order transition. In contrast, the peak at $T_N$ in the temperature dependence of the specific heat [4, 5] has been taken as evidence for a second-order (continuous) transition [10], but we note that a specific-heat peak can also be due to first-order latent heat in an inhomogeneous sample with a spread of transition temperatures. In $\text{La}_4\text{Ni}_3\text{O}_8$ charge ordering may also contribute to the peak. Be this as it may, critical slowing is not normally expected for a first-order transition.

B. Paramagnetic-phase relaxation; critical behavior

The observed behavior of the ZF $\mu^+$ relaxation rate $\chi(T)$ [Fig. 17(a)] is qualitatively similar to that of the $^{139}\text{La}$ nuclear spin-lattice relaxation rate in $\text{La}_4\text{Ni}_3\text{O}_8$ above $T_N$ [10], which was attributed to critical slowing of 2D spin fluctuations. The field dependence of $\chi(T)$ at 110 K [Fig. 15(a)] suggests that the critical point is at zero field.

Critical slowing with a $T = 0$ critical point is observed in NQR experiments on $\text{La}_2\text{CuO}_4+\delta$ [9]. The NQR relaxation data [9] agree with Eq. (6) based on the theory of Ref. 8, in which the weak inter-trilayer interaction that precipitates the 3D AFM transition is not expected to affect the 2D fluctuations appreciably. This nearly-2D picture has also been suggested for $\text{La}_4\text{Ni}_3\text{O}_8$ [4, 10]. There are a number of difficulties with the results of NMR and $\mu$SR relaxation studies in $\text{La}_4\text{Ni}_3\text{O}_8$, however. As noted above, the data are not consistent with the theory of the renormalized classical regime [8, 11]. Temperatures $T_0$ from Curie-Weiss fits disagree strongly between the two techniques. The NMR relaxation rates were measured in a field of 42 kOe, whereas $\mu^+$ spin relaxation rates show no sign of critical slowing between 200 K and 110 K for nonzero but much lower fields (0.1–4 kOe, Figs. 13 and 15). For $\text{La}_2\text{CuO}_4+\delta$ $T_N \ll J$ [9], whereas for $\text{La}_4\text{Ni}_3\text{O}_8$ both fits of Fig. 17(a) yield characteristic temperatures $T_0$ and $J$ of the order of $T_N$ (which, as noted, invalidates the Chakravarty-Orbach fit). This seems more compatible with a connection between critical slowing and the 3D phase transition than a $\text{La}_2\text{CuO}_4$-like scenario.

$\text{La}_4\text{Ni}_3\text{O}_8$ is intrinsically overdoped, and a more appropriate comparison might be with results from doped $\text{La}_2\text{CuO}_4$ with charge ordering. Previous $^{63,65}\text{Cu}$ and $^{139}\text{La}$ NQR studies of underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, $0 \leq x \leq 0.15$, [9] found that critical slowing for $x = 0$ at high temperatures is rapidly suppressed by Sr doping. Recent NMR experiments [37, 38] using a single crystal of $\text{La}_{1.885}\text{Sr}_{0.115}\text{CuO}_4$ revealed inhomogeneity and considerable structure associated with staging and oxygen diffusion at low temperatures, but did not revise the earlier conclusions. A NMR study of single-crystal $\text{La}_2\text{CuO}_{4+y}$, $y \sim 0.11$ [39], showed Curie-Weiss behavior of the $^{63}\text{Cu}$ relaxation rate for temperatures $\gtrsim 60$ K. This result is similar to both NMR and ZF-$\mu$SR data from $\text{La}_4\text{Ni}_3\text{O}_8$, but the doping level is higher in the nickelate and of the opposite sign. To our knowledge no muon relaxation studies of critical slowing in the paramagnetic phases of $\text{La}_2\text{CuO}_4$-based systems, doped or undoped, have been reported.

We conclude that low-frequency spin dynamics in the two compounds are not comparable, in spite of some similarities, although the question of first-order vs second-order nature of the transition [21] is raised in $\text{La}_4\text{Ni}_3\text{O}_8$ as in $\text{La}_2\text{CuO}_4$. More work is clearly necessary to understand the transition and both phases of this enigmatic material.

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Appendix: Other AFM stripe models

AFM configurations and corresponding dipolar-field histograms for stripe Models 2–4 are shown in Figs. 18–23. The trends seen in Model -1 histograms and discussed in Sec. V B 1 (low Ni moments, low- and high-frequency groups, only small changes for inverted alternate trilayers) continue to be found.
FIG. 18. AFM moment Configurations A–D in La$_4$Ni$_3$O$_8$ on stripe Model 2 [5]. See Fig. 8 for designations.

FIG. 19. Histograms of dipolar-field $\mu^+$ frequencies from AFM stripe Models 2A–2D compared to 50-K $\mu$SR FT spectrum in La$_4$Ni$_3$O$_8$. Colors as in Fig. 9.
FIG. 20. AFM moment Configurations A–D in La$_4$Ni$_3$O$_8$ on stripe Model 3 [5]. See Fig. 8 for designations.

FIG. 21. Histograms of dipolar-field $\mu^+$ frequencies from AFM stripe Models 3A–3D compared to 50-K $\mu$SR FT spectrum in La$_4$Ni$_3$O$_8$. Colors as in Fig. 9.
FIG. 22. AFM moment Configurations A–D in La$_4$Ni$_3$O$_8$ on stripe Model 4 [5]. See Fig. 8 for designations.

FIG. 23. Histograms of dipolar-field $\mu^+$ frequencies from AFM stripe Models 4A–4D compared to 50-K $\mu$SR FT spectrum in La$_4$Ni$_3$O$_8$. Colors as in Fig. 9.
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