Muon-spin-relaxation investigation of the spin dynamics of geometrically frustrated chromium spinels

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dc magnetic susceptibility (χ) and muon-spin-relaxation measurements (μSR) on the geometrically frustrated systems $MgCr_2O_4$ and $CdCr_2O_4$ indicate a transition from a paramagnetic state to a quasistatic spin state at 12.5 K and 7.8 K, respectively. Comparison of low-temperature field strength measured by μSR to that predicted by a dipole model suggests that only about 15% of the spin is static on the muon time scale. We observe slow spin fluctuations persisting to millikelvin temperatures, indicating that extremely low-energy magnetic excitations are present in these materials to the lowest temperatures. Our results demonstrate that substantial magnetic frustration remains, in spite of the tetragonal distortion occurring at T_N.

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I. INTRODUCTION

The geometric arrangement of the magnetic atoms in a compound can frustrate the transition to an ordered state at low temperatures. Instead, the system can enter an unconventional ground state displaying unique magnetic properties. Nearest-neighbor Heisenberg Hamiltonians predict a degenerate ground state for antiferromagnetic triangular-based lattices due to intercompeting magnetic interactions.[1,2] The lowest-energy configuration only requires that $\Sigma_{i=1}^{4}\mathbf{S}_i=0$ for each triangle or tetrahedron. In systems that obey this Hamiltonian, the spins will remain unordered and in a *cooperative paramagnetic* state termed a *spin liquid* for all $T>0$.[3]

A wide variety of behavior has been observed in real systems. Small perturbations, such as next-nearest-neighbor interaction, can induce low-temperature phase transitions,[4–6] leading to novel magnetic ground states. In two dimensions the corner-sharing triangular (*kagomé*) lattice $SrCr_pGa_{12-p}O_{19}$ has been studied and found to display a constant low-temperature dynamic spin component with a relaxation rate proportional to p^3 for $p>p_c$.[7,8] This is evidence of low-energy magnetic excitations that do not freeze out even at temperatures much less than T_N. The presence of such strong low-energy excitations indicates that the ground state has many nearly degenerate excited states which can be easily excited even at low temperatures and is a characteristic feature of geometrically frustrated systems.

Oxide pyrochlores have the general formula $A_2B_2O_7$. The ions on the A and B sites each form a sublattice of corner-sharing tetrahedra. Inelastic neutron scattering on $Tb_2Mo_2O_7$ shows evidence for the development of short-range magnetic ordering between the Tb^{3+} magnetic moments as the temperature is decreased towards the freezing temperature (T_F).[9] Muon-spin-relaxation (μSR) measurements indicate a dynamic component to the internal field that remains present down to 100 mK.[10] Perhaps the most definite example of a spin liquid yet discovered is the pyrochlore $Tb_2Ti_2O_7$ where μSR studies show that the relaxation rate climbs from 0.06 μs^{-1} at 200 K to 2 μs^{-1} at 2 K, below which it is approximately constant.[6,11] Neutron scattering reveals no long-range order[6,11] and specific-heat data shows only broad peaks centered at 1.5 K and 6 K.[11]

In this paper we report an investigation of the low-temperature magnetic properties of the chromium oxide spinels $MgCr_2O_4$ and $CdCr_2O_4$ using dc susceptibility and muon spin relaxation. $MgCr_2O_4$ and $CdCr_2O_4$ (and $ZnCr_2O_4$) are normal spinels where the Cr ions are all in the 3+ oxidation state and occupy equivalent octahedral sites with no evidence of any disorder (there is no site interchange between the Cr and metal ions). In these materials only the Cr ions are magnetic and therefore the magnetic lattice consists of Cr^{3+} ($J=3/2$) atoms arranged in corner-sharing tetrahedra. An advantage to studying materials with one magnetic sublattice arises from the possibility that apparent spin dynamics or glassiness in other frustrated systems may be due to the interchange of ions in the magnetic sublattices. For example, in $SrCr_pGa_{12-p}O_{19}$, although both Cr and Ga lie on the *kagomé* lattice, only Cr is magnetic, therefore $(9-p)/9$ of the sites are nonmagnetic. Additionally, the *kagomé* planes are interspaced by triangular Cr/Ga planes. It is possible that many of the magnetic properties of $SrCr_pGa_{12-p}O_{19}$ are strongly influenced by these structural disorders.[12,13]

$MgCr_2O_4$ and $CdCr_2O_4$ are cubic and paramagnetic at high temperatures and undergo first-order phase transitions at 12.5 K and 7.8 K, respectively, characterized by the development of tetragonal lattice symmetry. Specific-heat data on $ZnCr_2O_4$, which has the same T_N and Curie-Weiss temperatures (θ_{CW}) as $MgCr_2O_4$, exhibit a sharp peak at 12.5 K.[14] The transition is also seen in susceptibility, where hysteresis of roughly 80 mK between field-cooled and zero-field-cooled measurements reflects the first-order nature of the transition. The magnetic transition is further confirmed by neutron-scattering measurements[15] where weak antiferromagnetic Bragg peaks at low temperature indicate long-range spin ordering. Comparison of the integrated weight of the elastic-

scattering data to the total magnetic scattering cross section, however, gives an ordered magnetic moment of only approximately $1.0\mu_B$/Cr atom. Assuming a total magnetic moment of $3.7\mu_B$/Cr, this indicates a fluctuating moment of $2.7\mu_B$/Cr. Additionally, inelastic scattering shows a diffraction peak at an energy corresponding to nearest-neighbor exchange energy (estimated from the Curie-Weiss temperature) and a reciprocal-lattice vector corresponding to nearest-neighbor separation. The spectral weight of this excitation is $0.59(1)\mu_B$/Cr, corresponding to 22% of the fluctuating moment and 16% of the total moment.

II. EXPERIMENTAL PROCEDURE AND RESULTS

A. Sample preparation

Crystals were prepared using a flux-growth method as described elsewhere.[16] X-ray measurements show no flux inclusions in single crystals and confirm the spinel structure (space group $Fd\bar{3}m$, $a=8.334$ Å for $MgCr_2O_4$ and 8.567 Å for $CdCr_2O_4$). Some crystals had multiple domains which prevented their orientation along any specific axis during μSR measurements. Approximately 10 crystals were used in each μSR experiment with dimensions varying from 2 to 8 mm with single crystals averaging approximately 10 mm^3. Larger ceramic samples were prepared by means of a solid-state reaction between stoichiometric amounts of MgO or CdO (99.95%) and Cr_2O_3 (99.99%) in air. The powders were annealed twice and were mechanically ground and pressed between annealings. $MgCr_2O_4$ was annealed at 1950 K but since cadmium oxide evaporates at approximately 1500 K, $CdCr_2O_4$ was annealed at 1200 K. X-ray powder-diffraction measurements gave no evidence of impurity phases.

B. dc magnetic susceptibility

The dc magnetic susceptibility was measured using a commercial superconducting quantum interference device magnetometer (Quantum Design, San Diego). The susceptibility and inverse susceptibility of $MgCr_2O_4$ and $CdCr_2O_4$ crystals in an applied field of 1000 G is shown in Figs. 1 and 2. Fits of data above 200 K to the Curie-Weiss law give an effective moment of $3.82(5)\mu_B$/Cr for $MgCr_2O_4$ and $3.98(5)\mu_B$/Cr for $CdCr_2O_4$. Curie-Weiss temperatures (θ_{CW}) of 400(5) and 330(5) K respectively lead to high frustration parameters (defined as θ_{cw}/T_N) of approximately 32 and 44 (see Ref. 17 for the frustration parameter in various systems). The inverse susceptibility remains linear well below θ_{CW} as is characteristic of frustrated systems.[14]

Transitions are evident at 12.5 K in $MgCr_2O_4$ and 7.8 K in $CdCr_2O_4$. In larger, multiple-domain crystals, the transition was broader, occurring over approximately 1 K, and was centered 3 K below the standard transition temperature possibly reflecting the effects of strains associated with the multiple-domain structure. Transitions in ceramic samples occurred at the same temperature as the best single crystals. Fits of the low-temperature tail to Curie's Law give an effective moment of $0.038\mu_B$/Cr in $MgCr_2O_4$ crystals and ceramics. Assuming the paramagnetic tail is due to unordered chromium atoms with moments of $3.82\mu_B b$ and since the effective moment is proportional to S^2, this would correspond to $0.01\% = (0.038/3.8)^2$ of all chromium atoms being unconstrained.

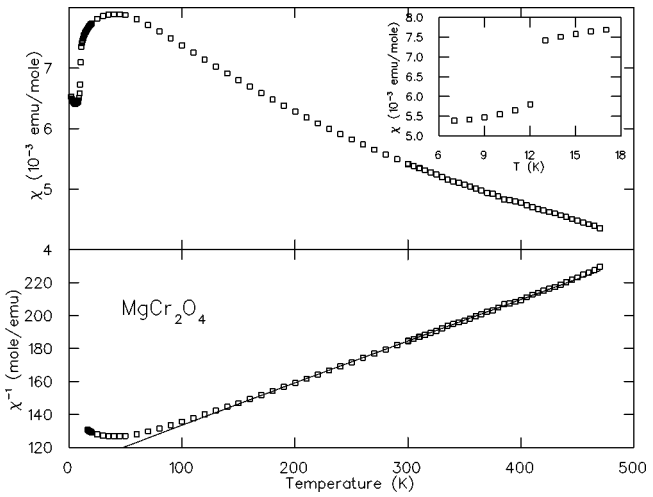

FIG. 1. dc susceptibility of $MgCr_2O_4$ crystals in 1000 G. The solid line is the fit to the Curie-Weiss equation. The inset shows the low-temperature susceptibility.

C. μSR measurements

Muon spin relaxation (μSR) is an extremely sensitive real-space probe of magnetic order and spin freezing.[18] In this technique, positive muons are implanted one at a time into a material of interest where they precess in the local magnetic field until they decay, emitting a positron preferentially in the instantaneous muon spin direction at the time of decay. One then reconstructs the spin-polarization function from the time spectra of detected positrons. In a usual commensurate antiferromagnet one observes coherent precession of the muon polarization since the local field has the same magnitude in each unit cell. With increasing inhomogeneity in the distribution of fields at the muon site(s), one sees

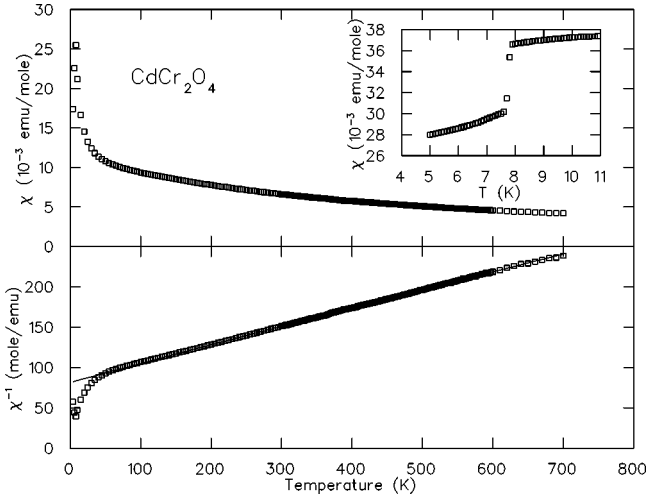

FIG. 2. dc susceptibility of $CdCr_2O_4$ crystals in 1000 G. The solid line is the fit to the Curie-Weiss equation. The inset shows the low-temperature susceptibility.

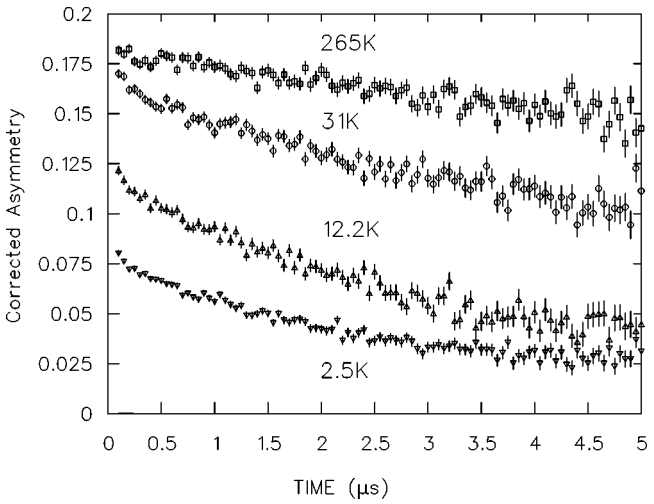

FIG. 3. Muon spin polarization in $MgCr_2O_4$ crystals at various temperatures in zero magnetic field (ZF).

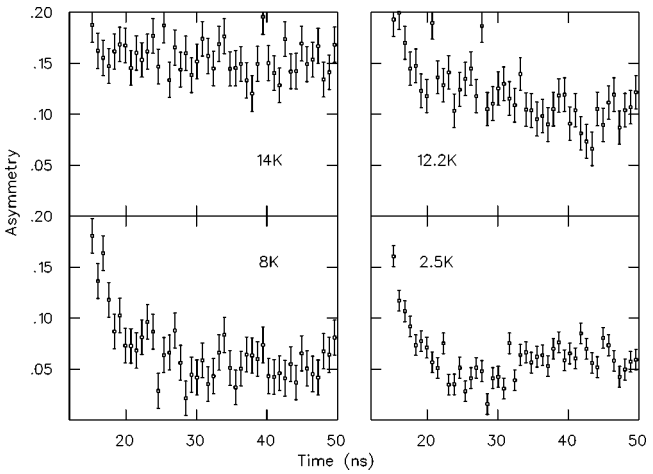

FIG. 4. Early time muon spin polarization of $MgCr_2O_4$ crystals in ZF.

increasing damping of the spin precession; in the limit of complete randomness (as in a spin glass) the polarization exhibits well-known forms depending on the details of the field distribution.

Our measurements were carried out using the M15 and M20 surface muon channels at TRIUMF in Vancouver, Canada. All regular cryostat measurements were performed on M20 using a low background holder while dilution refrigerator (DR) measurements were performed on M15 using regular (silver) sample holders. Silver has very weak nuclear magnetic moments and no electronic magnetic moment so muons landing in silver experience essentially no depolarization. Both zero-field (ZF) and longitudinal-field (LF) experiments were performed at temperatures between 20 mK and 265 K.

ZF μSR time spectra measured for $MgCr_2O_4$ crystals at various temperatures are shown in Fig. 3. The muon spin polarization is proportional to the corrected asymmetry of the positron decay, $G_z(t) \propto A(t)$. At high temperatures there is only slow relaxation characteristic of weak nuclear dipole fields. The relaxation increases with decreasing temperature; its magnitude indicates that its origin is slowly fluctuating (or static) electronic moments. We did not observe coherent precession in any of the samples studied, indicating that the distribution of local fields at the muon site(s) in these materials is extremely broad, as one would see in a spin glass. The development of two-component relaxation below T_N indicates the transition of the chromium spins to a quasistatic state. Early time data is shown in Fig. 4. The recovery of asymmetry that is visible following $t=30$ ns at 2.5 K is also characteristic of strong quasistatic internal fields. The behavior of all other samples was qualitatively similar.

With a Gaussian distribution of static fields centered about a frequency f the muon-spin-polarization function for a single magnetic site is given by

$$G_z(t) = A_1 \exp(-\lambda t)^{\beta} + A_2 \cos(2\pi f t) e^{-\sigma^2 t^2}, \qquad (1)$$

where the damping σ reflects the width of the field distribution and $A_1 = 1/3$ and $A_2 = 2/3$ in isotropic samples. When data below T_N was fit to Eq. (1), f and σ quickly reached saturation values of 10 MHz and 50 μs^{-1}, respectively, indicating a broad field distribution centered at about 700 G.

The magnetic-field distribution at possible muon sites in $MgCr_2O_4$ was calculated using a dipole model. The field was represented by static, randomly oriented $3.7\mu_B$ magnetic dipoles at each chromium site and the muon was assumed to come to rest at 1 Å from an oxygen atom, as is generally known for oxides.[19,20] We initially chose a random spin arrangement in view of the fact that we do not observe precession, indicating an inhomogeneous field distribution. This led to a field distribution with no values below 4500 G for any possible muon site. When the muon orbital radius was varied between 0.8 and 1.2 Å there was no appreciable difference in the results. Using the Ewald sum of point-charge method, the lowest electrical potential site was found to correspond to the site with a 4500-G characteristic local field. If the chromium spins are assumed to be ordered along an axis then the field at the muon site, calculated as above, is 3500 G. In all cases, the local field calculated assuming $3.7\mu_B$ Cr moments is much greater than we actually observe (4500 G or 3500 G vs 700 G). This indicates that either many of the Cr moments do not freeze or that only a portion of each individual moment freezes [i.e., $\langle S(0) \cdot S(t \rightarrow \infty) \rangle < S^2$] while the remainder of the spectral density remains at finite frequency. The early time data exhibits a Gaussian time dependence, a feature characteristic of a dense distribution of moments; this argues for the second interpretation where a portion of each moment freezes. A similar conclusion was reached in the neutron-diffraction study of Lee *et al.*[15] In this picture, we estimate that only 16% (700/4500 calculated for the random spin arrangement) or 20% (for the ordered spin arrangement) of each chromium spin becomes static on the μSR time scale. If the muon occupies any other site, then the fraction of each Cr moment which freezes is correspondingly lower.

The temperature dependence of the chromium spin-fluctuation rate can be determined by examining the relaxation rate of the muon spin polarization. Below the transition temperature, a static spin component leads to a fast reduction of asymmetry. Therefore, to compare relaxation rates above

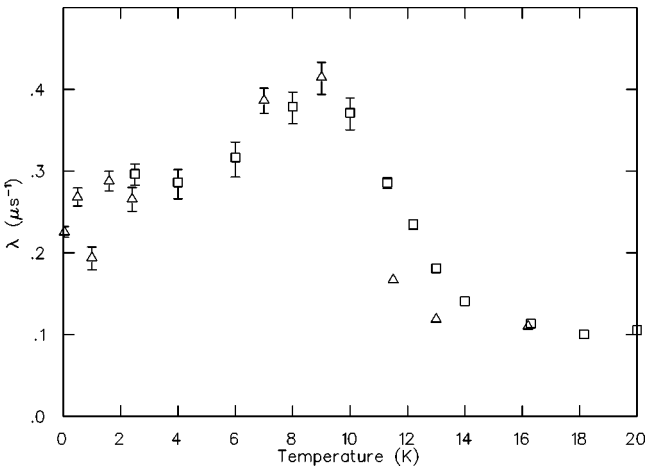

FIG. 5. Temperature dependence of λ in two different mosaics of $MgCr_2O_4$ crystals (triangles and squares).

and below the transition temperature long-time data (>100 ns) was fit to a phenomenological stretched exponential function

$$G_z(t) = A\exp(-\lambda t)^{\beta}. \tag{2}$$

The temperature dependence of λ for two mosaics of crystals (one in the regular cryostat and one in the DR) is shown in Fig. 5. Again, data for other samples (ceramic $MgCr_2O_4$, and crystal and ceramic $CdCr_2O_4$) are qualitatively similar. At low temperatures, λ becomes approximately constant at $\lambda \approx 0.25\ \mu s^{-1}$. This behavior is in contrast to the situation in usual ordered systems where $\lambda \to 0$ as $T \to 0$, reflecting the freezing out of magnetic excitations.

The peak in λ occurs significantly below 12.5 K. This can be attributed to the range of transition temperatures in the crystals used. (λ will peak when the last crystal orders since, with some spins still unordered, the muon spin is able to depolarize faster at a lower temperature. The fact that some crystals are ordered is accounted for by the decrease in asymmetry.) This explanation is supported by the fact that although asymmetry begins to decrease at 12.5 K, it does not reach a minimum until λ peaks. Additionally the decrease in the asymmetry of a $MgCr_2O_4$ ceramic specimen, which is expected to be more homogeneous, occurs over a smaller temperature range while the peak in λ occurs closer to 12.5 K.

The μSR time spectra were well described by a single exponential ($\beta = 1$) at high temperatures. β decreased to approximately 0.6 by 50 K (corresponding to the temperature at which susceptibility peaks). When β was held constant the temperature dependence of the relaxation rate was qualitatively unchanged although the quality of the fits decreased. The fact that the data must be fit to a stretched exponential indicates that the muon spin relaxation cannot be described by a single correlation time. The deviation of β from 1 at $T \gg T_N$ suggests that there is variation in the size and fluctuation rate of correlated regions as ordering develops. Similar behavior has been observed in the frustrated pyrochlores $Y_2Mo_2O_7$ and $Tb_2Mo_2O_7$,[10] as well as the dense spin glasses AgMn and AuFe.[21]

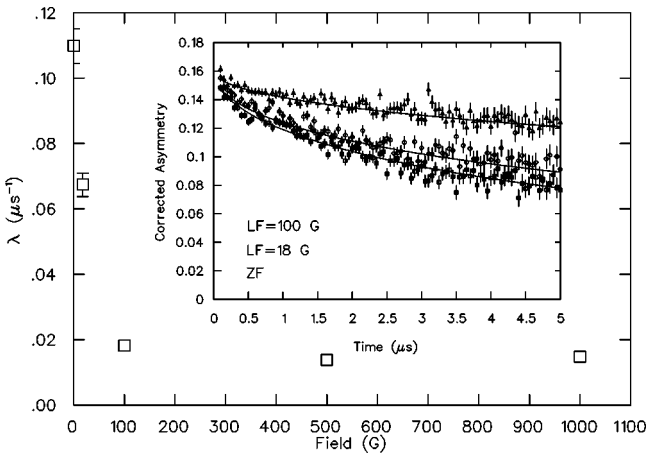

FIG. 6. LF dependence of λ in $MgCr_2O_4$ crystals at 24.9 K. Asymmetry and β are fixed to ZF values. Inset: ZF and LF muon spin polarization.

Above T_N the muon spin depolarization results from rapid fluctuations of the internal field due to Cr^{3+} atoms in the paramagnetic phase. The field dependence of the relaxation rate depends on the ratio of ν, the spin-fluctuation rate, and Δ, where $\Delta = \gamma_\mu B$ ($\gamma_\mu = 2\pi \times 135.54$ MHz/T and B is the rms internal magnetic field). When $\nu \gg \Delta$ the muon depolarization is due to spin flipping as opposed to Larmor precession and $G_z(t)$ will be a single exponential with a relaxation rate[22]

$$\lambda = 2\Delta^2 \nu/(\nu^2 + \omega_L^2), \tag{3}$$

where $\omega_L = \gamma_\mu B_L$ describes the effect of an applied longitudinal field. In conventional systems λ is only weakly dependent on the applied field when $\nu \gg \omega_L$, as observed in $Y_2Mo_2O_7$ and $Tb_2Mo_2O_7$.[10] The field dependence of muon spin polarization and λ is shown in Fig. 6. The quick decrease in λ with only 20 G is attributed to the decoupling of weak static fields at the muon site due to nuclear dipole moments. The presence of weak static fields is confirmed by the nonzero relaxation at 265 K as evident in Fig. 3. After the initial decoupling, the relaxation rate is field independent as expected.

Below T_N, we observe effects of an applied longitudinal field which are more complicated than in the paramagnetic state. Essentially, the application of a longitudinal field much weaker than the characteristic quasistatic internal field causes an appreciable change to the muon spin relaxation which implies that some part of the muon spin ensemble must actually experience a much smaller characteristic local field than is implied by the fast relaxation seen in zero field.

The field dependence of the muon spin polarization in a Gaussian distribution of static fields is well known.[23,24] The internal field is completely decoupled, and hence muon spin polarization is constant, with a LF $\sim 5\Delta/\gamma_\mu$. With quasistatic fields the expected field dependence can be modeled by assuming a strong collision model where the local field at the muon site changes discontinuously to a new field drawn from the Gaussian distribution with a certain time probability (inversely proportional to the spin-fluctuation rate). The result-

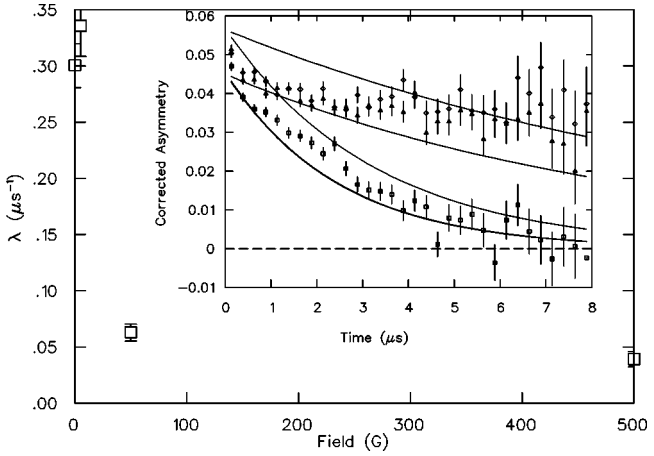

FIG. 7. LF dependence of λ in $CdCr_2O_4$ ceramics at 20 mK. Asymmetry and power β are fixed to ZF values. Inset: Muon spin polarization in $CdCr_2O_4$ ceramics for ZF (squares), 50 G (triangles), and 500 G (diamonds) at 20 mK. Overlapping lines along the ZF data are fits to ZF and 50-G data with the hopping rate set by ZF data. The 500-G fit is shifted up from the ZF and 50-G fits. Lines running through LF data are fits to 50-G and 500-G data with the fluctuation rate determined globally between the two runs. The fact that the ZF data does not accurately predict the field dependence of the relaxation rate suggests the presence of muon sites with only weak static fields (see text).

ing polarization function cannot be solved explicitly but can be analyzed numerically. Comparison of fits to actual data are shown in the inset of Fig. 7. Fits for LF data are shown with the fluctuation rate set by ZF data and with the fluctuation rate set globally between the two LF runs. While the two LF runs seem mutually consistent, the ZF run does not accurately predict the field dependence; this discrepancy implies that a weak static field (on the order of a few gauss) is the only field present in regions that account for approximately 10% of the asymmetry.

It is unlikely that the applied field is changing the dynamic behavior of the spins in these materials. The exchange energy is approximately 4.5 meV (estimated from θ_{CW}) while an applied field of 10 G is on an energy scale of 10^{-4} meV ($E=g_0\mu_B HS$ where $g_0\mu_B=2\times0.579\times10^{-8}$ eV/G, $H=10$ G, $S=3/2$).

The weak static field does not arise from the ordered component of the chromium spins, since, as we have already discussed, the early time relaxation indicates that this field is on the order of 700 G. The 10% signal could originate either in nonmagnetic regions of the sample if they exist or in sites where the field is zero by symmetry (although such sites are not predicted by the dipole model, they could perhaps correspond to spin arrangements not considered). In these situations, these low-field sites would only experience a weak static field from nuclear dipole moments that would be, as predicted by high-temperature data, decoupled with LF=10 G. A final possibility is that this abnormal field behavior may be due to muons landing outside the sample and experiencing an easily decoupled weak static field from nuclear dipole moments. This effect, however, should be reduced with larger samples (the ceramics) and in measurements performed on M20 in the low background apparatus. As evident in Table I this was not observed and hence stray muons do not likely account for the observed LF behavior. In any case, this easily decoupled field corresponds to only 10% of the muons and therefore does not affect our main findings.

TABLE I. Relaxation rates in $MgCr_2O_4$ and $CdCr_2O_4$ above and below T_N in ZF and 100-G LF.

Material	Sample	Setup	T (K)	λ_{ZF} (μs^{-1})	λ_{LF} (μs^{-1})
	crystal	M20	25	.085	0.03
$MgCr_2O_4$	crystal	M20	2.5	0.3	0.02
$T_N=12.5$ K	crystal	M15	0.035	0.3	0.05
	ceramic	M20	16		0.05
	ceramic	M20	2		0.15
	ceramic	M15	0.020	0.2	0.04
	crystal	M20	2.5	0.3	0.015
$CdCr_2O_4$	ceramic	M20	11	0.12	0.05
$T_N=7.8$ K	ceramic	M20	5.1	0.4	0.09
	ceramic	M15	10	0.2	
	ceramic	M15	0.020	0.3	0.05

As a further means of analyzing the low-temperature field dependence of the relaxation rate, the 20-mK $CdCr_2O_4$ ceramic data was fit to Eq. (2) with β and asymmetry fixed to ZF values. The field dependence of λ is shown in Fig. 7. In support of the presence of a weak static field in a small volume fraction, λ is substantially reduced with 50 G and then remains approximately constant to 500 G.

To confirm the basic spin behavior with any weak static field that is decoupled, μSR measurements were performed on $MgCr_2O_4$ and $CdCr_2O_4$ ceramics with a 100-G LF applied. Figure 8 shows the temperature dependence of λ, β, and asymmetry in $MgCr_2O_4$ ceramics. At low temperatures the relaxation rate became too small to allow determination of the temperature dependence of β, from where it was fixed

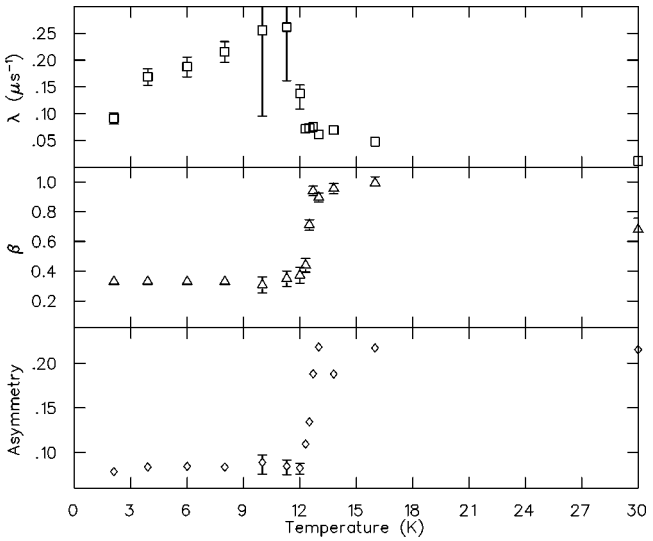

FIG. 8. Temperature dependence of λ, β, and asymmetry in $MgCr_2O_4$ ceramics in a 100-G LF. β was fixed to 0.35 below 10 K.

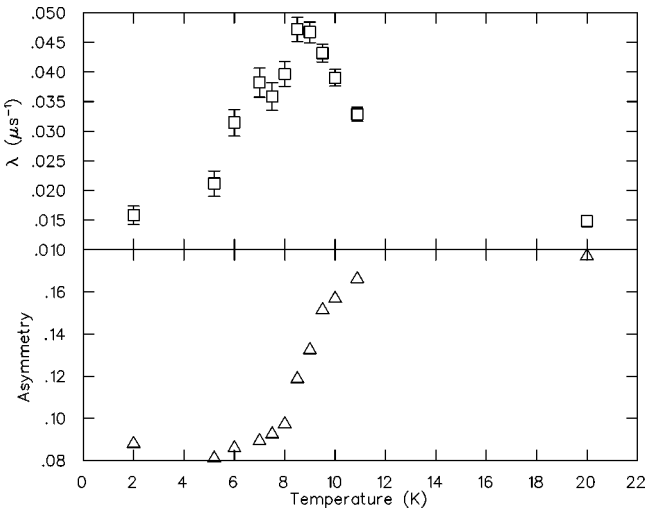

FIG. 9. Temperature dependence of λ and asymmetry in $CdCr_2O_4$ ceramics with LF=100 G. β is fixed to 0.6 for all temperatures.

to 0.33 below 10 K. Similarly, the relaxation rate in $CdCr_2O_4$ was too low to allow any temperature dependence of β to be determined. The temperature dependence of λ and asymmetry in $CdCr_2O_4$ ceramics is shown in Fig. 9 with β fixed to 0.6 (which gave the best overall fit quality). The temperature dependence of all parameters in Eq. (2) is identical between ZF and LF scans, confirming the presence of a dynamic component of the spin below T_N.

The muon-spin-depolarization rate is generally dependent upon the strength, distribution, and fluctuation rate of the internal magnetic field. In the limit of slow fluctuations, however, the tail will decay simply as[25]

$$G_z^G(t) = A\exp[-(2/3)\nu t], \tag{4}$$

where ν is the field fluctuation rate. Although this equation does not account for the observed stretched exponential behavior (as it assumes a single correlation time $\tau = 1/\nu$), it provides a characteristic estimation of the spin-correlation time. For example, $\lambda = 0.1\ \mu s^{-1}$ corresponds to a time scale of approximately 10^{-5} s. Since spins with a correlation time greater than 10^{-9} s appear static to neutron scattering the low-temperature spin fluctuations observed by μSR do not correspond to the inelastic peak observed in neutron scattering but instead reflect a more slowly fluctuating part of the Cr moments.

III. CONCLUSION

Geometric frustration prevents $MgCr_2O_4$ and $CdCr_2O_4$ from entering a typical antiferromagnetic state. At low temperatures a transition from cubic to tetragonal symmetry allows the spins to enter a quasistatic state with approximately 15% of the spin spectral density (almost) frozen on the μSR time scale. We have found that slow fluctuations ($\tau \sim 10^{-5}$ s) are present down to millikelvin temperatures, suggesting that low-energy excitations persist to $T \ll T_N$, indicating a highly nearly degenerate ground state (where the excited states are energetically accessible even at low temperatures) which is a feature characteristic of geometrical frustration. Apparently, the structural distortion at T_N only partially removes the degeneracy of the ground state, substantial frustration remains in the tetragonal phase at low temperature, and much of the Cr moment remains rapidly fluctuating to low temperatures.

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