Magnetism in the $S=1$ frustrated antiferromagnet $GeNi_2O_4$ studied using implanted muons

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We present the results of a muon-spin relaxation study of $GeNi_2O_4$. We provide further clarification of the two transitions to the antiferromagnetic state and measure the magnetic field dependence of the heat capacity up to 14 T. Both oscillatory and relaxing signals are observed below the lower transition (at temperature T_{N2}) in the muon-decay positron asymmetry spectra, arising from two distinct types of magnetic environment. A possible explanation is suggested in terms of the separate ordering of two magnetic subsystems, one of which does not order fully down to the lowest measured temperature (1.8 K).

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I. INTRODUCTION

Materials experiencing geometric frustration have been the topic of much recent interest. Their rich behavior is due to a large ground-state degeneracy, which renders them highly unstable to perturbations.[1] In the case of a frustrated model system comprising Heisenberg-type antiferromagnetic nearest-neighbor interactions, cooperative paramagnetic behavior (i.e., fluctuations at all temperatures down to absolute zero) is expected. This state of affairs is altered, however, by perturbations due to other interactions (including next-nearest neighbor, dipole, crystal field, etc.) which cause a variety of low temperature states to be realized, including spin liquids[2] and spin ices[3] in some rare-earth pyrochlores and structural phase transitions for some spinels with transition metal ions on the frustrated B sublattice.[4,5]

Many geometrically frustrated systems with half-integer spins relieve the frustration by undergoing a structural phase transition at low temperatures to a magnetically ordered state; this is the case for the spinels $ZnCr_2O_4$ ($S=3/2$),[4] $GeCo_2O_4$ ($S=3/2$), and powder samples of $ZnFe_2O_4$ ($S=5/2$)[5,6] (although we note that a magnetic transition is not observed in single crystal samples[7]). The integer spin ($S=1$) frustrated antiferromagnet $GeNi_2O_4$ has therefore been of recent interest, since it undergoes a transition to an antiferromagnetically ordered state below $\sim$12 K (observed in both powder and single crystal samples[8–10]) with no accompanying structural transition. $GeNi_2O_4$ has the normal spinel structure, having Ni^{2+} ions ($3d^8$) at the vertices of corner sharing tetrahedra (the spinel B site), coordinated by a nearly regular octahedron of oxygen ions, resulting in a $^3A_{2g}$ triplet ground state. The crystal field lowers the degeneracy of this triplet, further splitting it into a close lying spin singlet and doublet expected to be separated by only a few cm^{-1}. Recent experimental studies of this system by Crawford *et al.*[9] show that the ordered state is reached by two separate transitions at $T_{N1}=12.13$ K and $T_{N2}=11.46$ K. It was found from heat capacity measurements that the magnetic entropy S_{mag} of $GeNi_2O_4$ is only half the expected $2R\ln 3$ per mole,[9] with the same measurements suggesting the existence of both gapped and gapless excitations within the Néel state.

In recent years, muon-spin relaxation (μ^+SR) measurements have been highly successful in observing frustration-related behavior in several frustrated systems, including the observation of spin-ice behavior and cooperative paramagnetism (see, e.g., Refs. 2 and 11). In this paper we report on the first muon-spin relaxation measurements on $GeNi_2O_4$. We have probed the magnetically ordered region from a local viewpoint and provide evidence for muons stopping in separate magnetic environments in the ordered state. It is suggested that the double transition may be due to the separate ordering of these two subsystems.

II. EXPERIMENTAL

Polycrystalline $GeNi_2O_4$ was prepared in a solid-state reaction using high purity (>99.99%) GeO_2 and NiO. The stoichiometric mixed powder was calcined in an O_2 flow atmosphere at 1200 °C for 48 h. The sample was characterized using x-ray diffraction analysis (which revealed no contribution from impurity phases) and heat capacity measurements. The latter were carried out on a sintered pellet sample on warming using a 14 T Quantum Design PPMS system. Heat capacity data taken in zero field (ZF) and in an applied field of 7 and 14 T are shown in Figs. 1(a)–1(c). The ZF results show two sharp maxima[12] at T_{N1} and T_{N2}, in agreement with previous studies.[9,10] In addition, we find a small shoulder below T_{N1}, also observed by Hara *et al.*,[10] who speculated about a possible third transition. The temperature of the two main peaks decreases markedly with an applied field (as expected for an antiferromagnet; see, e.g., Ref. 13), as shown in the phase diagram in Fig. 1(d). Both main peaks broaden and decrease in intensity with increasing field, the effect being more pronounced for the lower temperature peak. Also included in Fig. 1(d) are the two spin-flop transitions measured at 4.2 K in a recent high field magnetization study,[14] and our phase diagram associates these with our heat capacity peaks.

Zero-field muon-spin relaxation measurements have been made on polycrystalline $GeNi_2O_4$ using the DOLLY instrument at the Swiss Muon Source, Paul Scherrer Institute (PSI), Villigen, Switzerland.[15] The sample was wrapped in

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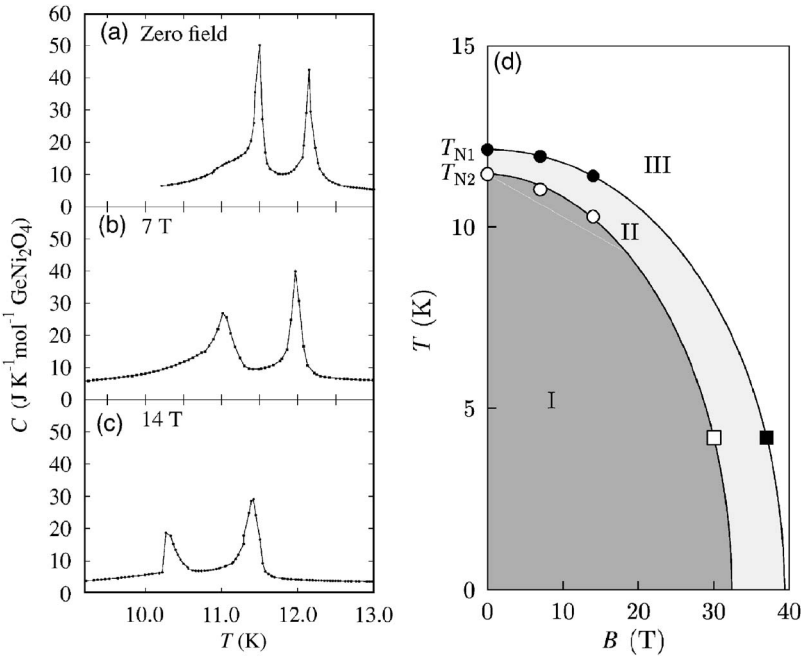

FIG. 1. Heat capacity measured in (a) zero field and in an applied magnetic field of (b) 7 T and (c) 14 T. (d) Magnetic phase diagram deduced from (a)–(c) (circles) and from spin flops observed in high field magnetization measurements (Ref. 14) (squares) (phases I and II are antiferromagnetic phases, phase III is paramagnetic).

25 μm Ag foil and mounted on a Ag backing plate. In a μ^+SR experiment, spin-polarized positive muons are stopped in a target sample, where the muon usually occupies an interstitial position in the crystal. The observed property in the experiment is the time evolution of the muon spin polarization, the behavior of which depends on the local magnetic field B at the muon site, and which is proportional to the positron asymmetry function[16] $A(t)$.

III. RESULTS

Our μ^+SR data allow us to identify three distinct regions of temperature as follows: (a) phase I ($T<T_{N2}$), (b) phase II ($T_{N2}\leqslant T\leqslant T_{N1}$), and (c) phase III ($T>T_{N1}$). We now discuss each of these regimes in turn.

A. Phase I ($T<T_{N2}$)

For phase I [Fig. 2(a)], oscillations in the asymmetry are clearly discernible, though with a rather small amplitude, and a fast initial relaxation of the muon polarization is visible at early times. The oscillations are characteristic of a quasistatic local magnetic field at the muon site, which causes a coherent precession of the spins of those muons with a component of their spin polarization perpendicular to this local field; their presence provides strong evidence for the existence of long range magnetic order (LRO) in phase I, in agreement with previous neutron diffraction measurements.[9] The frequency of the oscillations is given by $\nu_i=\gamma_\mu B_i/2\pi$, where γ_μ is the muon gyromagnetic ratio ($\equiv 2\pi\times 135.5$ MHz T^{-1}), and B_i is the local field at the ith muon site. The existence of oscillations in the asymmetry in a polycrystalline sample is a signature of a narrow distribution of the magnitudes of the static local magnetic fields, at symmetry-related muon sites in the crystal, associated with LRO. A broader distribution of these fields leads to the occurrence of the Kubo-Toyabe (KT) function,[17] which is well approximated by a Gaussian function at early times with relaxation rate $\sigma^2=\gamma_\mu^2\,\Delta B^2/2$, where $\Delta B^2=\langle(B-\langle B\rangle)^2\rangle$ is the second moment of the local field distribution. The recovery of asymmetry at late times, characteristic of the KT function, is often lost due to the presence of slow dynamics in the local field distribution. Because of the additional fast relaxation signal in the data, a full description also requires an additional component. This is well described over the measured temperature regime by a Gaussian functional form $A_2\exp(-\sigma^2t^2)$ with a large relaxation rate (see later). The large magnitude of the relaxation at very low temperatures makes it impossible to unambiguously assign it a Gaussian lineshape over the entire temperature range. However, the continuous temperature evolution of σ and the

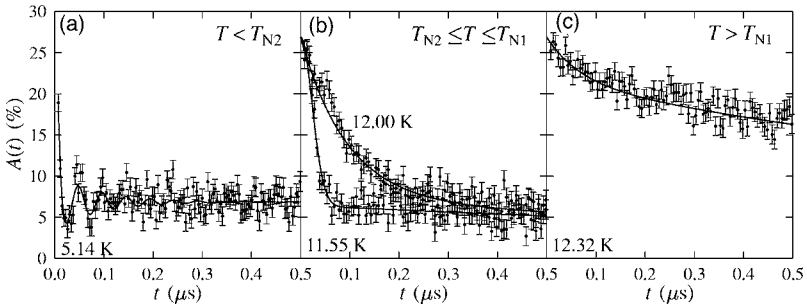

FIG. 2. ZF spectra measured (a) at 5.14 K, in phase I ($T<T_{N2}$), (b) at 11.55 K and 12.00 K, in phase II ($T_{N2}\leqslant T\leqslant T_{N1}$), and (c) at 12.32 K, in phase III ($T>T_{N1}$). The oscillations seen in (a) are observed to vanish above T_{N2} (b), with the Gaussian component vanishing above T_{N1} (c).

lineshape seen at temperatures $T>10$ K makes this a reasonable assumption.

The spectra are found to be best fitted in the range $T<T_{N1}$, with the resulting functional form

$$A(t) = A_1\{a_1 + a_2 \exp(-\lambda_1 t)\cos(2\pi\nu_1 t)\} + A_2 \exp(-\sigma^2 t^2) + A_{bg}, \quad (1)$$

where A_{bg} represents a constant background contribution from those muons that stop in the sample holder or cryostat tail.[18] The factor multiplied by A_1 accounts for the component of the spectra associated with LRO (normalized to 1), expected to be made up from contributions from those muons with spin components parallel to the local magnetic field a_1 and those with perpendicular components a_2. The value of a_1 consistent with the measured spectra is much higher than expected ($a_1=0.59$ compared to the expected value of 1/3). If allowed to vary in the fitting routine, amplitude a_1 follows the temperature evolution of the oscillating component with amplitude a_2, leading us to believe that a_1 and a_2 have the same physical origin. It could be that a portion of a_1 arises from muons stopping inside magnetically ordered regions in sites at which the local field is very small, so that an oscillating signal is not produced by those sites. However, an unambiguous assignment of the amplitudes for the lowest measured temperatures is hindered by the large value of the relaxation rate σ, so the situation may be more complex than considered here.

The amplitude A_1 is found to be approximately constant up to $T\approx 9$ K [Fig. 3(a)], where it is seen to decrease as the transition at T_{N2} is approached from below. This is accompanied by a corresponding rise in the amplitude of the Gaussian component A_2. The existence of two distinct components in the measured spectra (i.e., Gaussian and oscillatory) provides evidence for two sets of muon sites (or *subsystem*) in the material, such that a localized muon experiences one of two distinct magnetic environments. The first of these subsystems, with occupancy proportional to the amplitude A_1, has a sufficiently narrow distribution of quasistatic magnetic fields that oscillations are observable. The other subsystem, with occupancy proportional to A_2, is associated with a wider distribution of fields (preventing oscillatons being observable) and slow dynamics preventing any recovery of the asymmetry at later times. The μ^+SR data show that the fraction of the sample associated with the Gaussian component gives rise to a very different local magnetic environment to that causing the fraction associated with amplitude A_1. Since the muon is a local probe, it is not possible to conclude whether the two magnetic environments suggested by the data are spatially separated or spatially coexisting. If these two subsystems were spatially separated, then the exchange of amplitudes seen as T_{N2} is approached from below would probably be due to those regions associated with Gaussian relaxation growing at the expense of those sites related to the oscillations. If, in contrast, the two environments are coexisting, then the changes in amplitude may be due to a reordering of the magnetic moments as the temperature is increased.

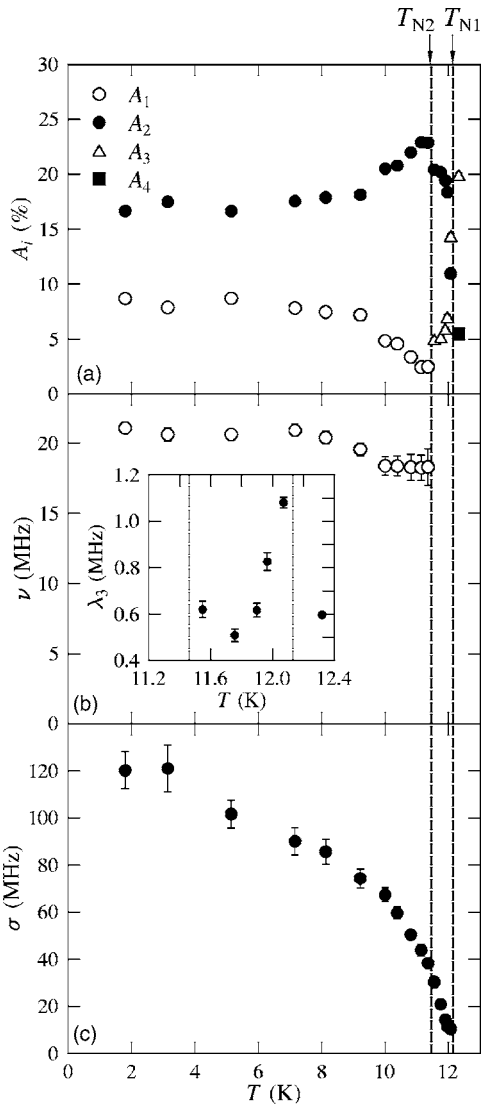

FIG. 3. Results of fits as described in the main text. (a) Amplitudes of the magnetic component A_1 (open circles) and Gaussian component A_2 (closed circles) as a function of temperature. A_2 is seen to rise as T_{N2} is approached from below. Between the transitions, A_2 decreases suddenly, while A_3 (open triangles) increases. Amplitude A_4 (closed square) in the paramagnetic regime is also shown for comparison. The error bars are smaller than the data points in this panel. (b) The precession frequency remains approximately constant for $1.8\text{K}\leqslant T\leqslant T_{N2}$. *Inset:* The relaxation rate λ_3 in the region $T>T_{N2}$ increases sharply as T_{N1} is approached from below. The transition temperatures are shown by dashed lines. (c) The relaxation rate σ is seen to fall smoothly across the transition at T_{N2}.

The frequency of the oscillations [Fig. 3(b)] varies little below the transition temperature T_{N2}, at which point $A_1\rightarrow 0$, indicating first-order behavior, in agreement with the heat capacity measurements.[9] This could suggest that the subsystem associated with the oscillating component undergoes an ordering transition at T_{N2}. The stability of the oscillation frequency across the temperature range means that we see no effect due to depopulation of the upper level of the split $^3A_{2g}$ triplet state at low temperatures.

B. Phase II ($T_{N2} \leqslant T \leqslant T_{N1}$)

In phase II Fig. 2(b) the oscillations vanish and the spectra are described by relaxing components only. The Gaussian component persists into this regime, with no discernible discontinuity in σ at T_{N2}. This suggests that this component arises from the same mechanism responsible for the corresponding component in Eq. (1). Also evident in the spectra measured in phase II is an exponential component $A_3 \exp(-\lambda_3 t)$. An exponential function is often the result of fast, dynamic fluctuations in the local field, in which case the relaxation rate $\lambda_3 \propto \gamma_\mu^2 \Delta B^2 / \delta$, where δ is the fluctuation rate.[17] This behavior points to dynamic fluctuations of a paramagnetic subsystem, coexisting with the subsystem associated with the Gaussian component seen in phase I. The data are therefore described by the resulting relaxation function,

$$A(t) = A_2 \exp(-\sigma^2 t^2) + A_3 \exp(-\lambda_3 t) + A_{bg}. \quad (2)$$

With increasing temperature, we see [Fig. 3(a)] the increase in the amplitude of the fluctuating component (A_3 increases) along with a decreasing fraction of the Gaussian component (A_2 decreases). The exponential relaxation rate λ_3 is seen to increase sharply as T_{N1} is approached from below [*inset*, Fig. 3(b)] corresponding to a slowing of fluctuations. The relaxation rate σ decreases smoothly across the entire temperature range [Fig. 3(c)], showing no features corresponding to the transition at T_{N2} but vanishing at T_{N1}. The smooth decrease of σ with increasing temperature suggests that this parameter is dominated by the magnitude of the local field distribution, as seen in other muon studies of magnetically ordered systems.[19] This may suggest that the subsystem associated with this component undergoes an ordering transition at T_{N1}.

C. Phase III ($T > T_{N1}$)

In Phase III [Fig. 2(c)] the Gaussian component is absent and the data are best described by a two exponential form $A(t) = A_3 \exp(-\lambda_3 t) + A_4 \exp(-\lambda_4 t) + A_{bg}$, typical of relaxation due to dynamic fluctuations, as would be expected in a purely paramagnetic material. The relaxation rates differ by an order of magnitude, with $\lambda_3 = 0.59(1)$ MHz and $\lambda_4 = 5.5(2)$ MHz at $T = 12.32$ K.

IV. DISCUSSION

The fact that only half of the expected magnetic entropy $S_{mag} = 2R \ln 3$ is accounted for in heat capacity measurements made below 75 K points to unconventional magnetic behavior in $GeNi_2O_4$. The double transition is seen in neutron diffraction measurements[9] through discontinuities in the intensity of the $(\frac{1}{2}, \frac{1}{2}, \frac{1}{2})$ magnetic Bragg peak, the intensity of which is proportional to the ordered volume-fraction multiplied by the square of the magnetic moment.[20] These results suggest that LRO exists throughout the material with the same wave vector at all temperatures below T_{N1}.

The muon provides us with a local perspective. Of the two magnetic subsystems identified in the data, the one giving rise to oscillations may unambiguously be attributed to the presence of LRO. In phase I the magnetic moment (proportional to the frequency ν_1) shows little temperature dependence, in contrast to the volume fraction, which decreases as T tends to T_{N2}. This component cannot be resolved in phase II, and the disappearance of magnetic order in this subsystem may cause the discontinuity in the Bragg peak intensity seen in the neutron data.

Since the neutron diffraction results strongly suggest that LRO exists throughout the sample in phase II, it may be that the Gaussian component associated with the other magnetic subsystem seen in phases I and II could also be an artifact of LRO. An incomplete ordering of the magnetic moments, for example, may provide a sufficient spin disorder in this subsystem to wash out an oscillatory signal. The magnitude of the local magnetic field in this subsystem may still be probed from the behavior of the relaxation rate σ (provided that any dynamics are slow and only weakly temperature dependent), which decreases smoothly. The amplitude of this component decreases with increasing temperature, before vanishing at T_{N1}, perhaps accounting for the other discontinuity in the Bragg peak intensity.

The observed behavior in $GeNi_2O_4$ may, therefore, be analogous to that observed in the geometrically frustrated pyrochlore $Gd_2Ti_2O_7$. In that system, there are two magnetic subsystems of $S = 7/2$ Gd^{3+} spins, with different ordering temperatures.[21] Ordering takes place at the higher transition ($T_N = 1.1$ K) in one subsystem (accounting for 3/4 of the Gd^{3+} spins). At the lower transition temperature $T' = 0.7$ K the other subsystem (comprising the remaining 1/4 of the Gd^{3+} spins) weakly orders, such that the material adopts a 4-k magnetic structure, and results in a disordered spin component. If a similar mechanism is the cause of the observed behavior in $GeNi_2O_4$, we note that the higher transition at T_{N1} would probably be associated with a weakly ordering subsystem (with sufficient spin disorder to prevent the observation of coherent oscillations) followed by the ordering of the second subsystem below T_{N2}.

Partial disorder has also been suggested recently in the case of geometrically frustrated clinoatacamite, $Cu_2Cl(OH)_3$, on the basis of μ^+SR measurements.[22] Asymmetry spectra typical of LRO throughout the material are obtained in the temperature region $6.5 \leqslant T \leqslant 18.1$ K. At the lowest measured temperatures ($\sim$20 mK), however, additional relaxation is observed, coexisting with a signal corresponding to LRO. This is explained in terms of the coexistence of static order and of a subsystem of disordered, fluctating spins.[22]

Such behavior in $GeNi_2O_4$ could also explain the missing contribution to the magnetic entropy. In their heat capacity study Crawford *et al.* assume that $\lim_{T \to 0} S_{mag} = 0$ and detect only 56.5% of the expected $S_{mag} = 2R \ln 3$ up to $T = 75$ K. If a subsystem does indeed exist that does not undergo full magnetic ordering, then the magnetic entropy would take a nonzero value below the transitions. Assuming that all of S_{mag} is accounted for by $T = 75$ K, then the amount of magnetic entropy associated with the partially ordered subsystem would be 43.5% of $2R \ln 3$. Such a value is possible given that the amplitude A_2 associated with the Gaussian component is approximately twice A_1 at low temperatures [Fig. 3(a)], suggesting that around two thirds of the spins are associated with the partially ordered subsystem. We note, however, that

the situation in $GeNi_2O_4$ may be even more complicated than considered here. In $Gd_2Ti_2O_7$, for example, where a partially ordered subsystem exists at low temperatures associated with 1/4 of the spins, 92% of the expected $S_{mag}=R\ln 8$ is accounted for in specific heat measurements.[23] It has been suggested[9] that the unusual ordering behavior and smaller than expected magnetic entropy in $GeNi_2O_4$ may stem from the integer spin, which may also be the case for the recently reported pyrochlore $Y_2Ru_2O_7$.[24]

The existence of two relaxation rates in phase III points to the coexistence of two sets of muon sites in this region also. It is not clear whether these correspond to the suggested separate magnetic subsystems persisting into the paramagnetic regime. The coexistence of two relaxation rates of differing magnitudes is usually associated with spatially separated magnetic components.[25]

V. CONCLUSION

In conclusion, we have investigated magnetic transitions in $GeNi_2O_4$ using implanted muons. We observe two separate contributions to the measured spectra below the upper magnetic transition at T_{N1} suggestive of two different magnetic environments coexisting in the material. Our data may be interpreted in terms of two distinct magnetic subsystems that undergo separate ordering transitions. The first subsystem, which accounts for the larger portion of the sample, partially orders at the higher transition temperature T_{N1} leaving a significant spin disorder that persists to the lowest measured temperature. This subsystem coexists with a second subsystem that is fluctuating dynamically down to the lower transition temperature T_{N2}, where this second subsystem undergoes full magnetic ordering. The exchange in amplitudes of the signals corresponding to each subsystem suggests that the number of Ni^{2+} spins associated with each subsystem is not constant but varies above $T\approx 9$ K. Further magnetic neutron diffraction measurements would be required to elucidate the precise nature of the proposed subsystems.

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