Rapid Communications

Quantum spin liquid ground state in the disorder free triangular lattice $NaYbS_2$

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Rare-earth delafossites were recently proposed as promising candidates for the realization of an effective $S = 1/2$ quantum spin liquid (QSL) on the triangular lattice. In contrast to the most actively studied triangular-lattice antiferromagnet $YbMgGaO_4$, which is known for considerable structural disorder due to site intermixing, $NaYbS_2$ delafossite realizes structurally ideal triangular layers. We present detailed muon spin rotation (μSR) studies on this regular (undistorted) triangular Yb sublattice based system with an effective spin $J_{\mathrm{eff}} = 1/2$ in the temperature range 0.05–40 K. Zero-field and longitudinal-field μSR studies confirm the absence of any long-range magnetic order state down to 0.05 K ($\sim J/80$). Current μSR results, together with the so far available bulk characterization data, suggest that $NaYbS_2$ is an ideal candidate to identify the QSL ground state.

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Introduction. A quantum spin liquid (QSL) is an exotic state of matter, in which electrons spins are strongly entangled, but do not exhibit any long-range magnetic ordering down to $T = 0$. Despite considerable effort in the past, so far experimental realizations of a clean QSL remain scarce. At the outset, Anderson proposed that the QSL state can be stabilized in materials where $S = 1/2$ forms a perfect triangular lattice. Such a scenario has proved exceptionally hard to realize [1,2]. The organic compounds κ-(BEDT-TTF)$_2$Cu$_2$(CN)$_3$ and EtMe$_3$Sb[Pd(dmit)$_2$]$_2$ appear to be two promising examples of triangular lattices with $S = 1/2$ moments and fluctuating disordered spin ground states [3,4]. However, $S = 1/2$ inorganic analogs such as $Ba_3CoSb_2O_9$ and $NaTiO_2$ either order magnetically or undergo a lattice deformation on cooling [5–8].

In this context, the triangular lattice magnet $YbMgGaO_4$ was identified as a potential QSL candidate with an effective spin $J_{\mathrm{eff}} = 1/2$ [9]. $YbMgGaO_4$ contains undistorted triangular planes of magnetic Yb^{3+} with space group $R\bar{3}m$, separated by two triangular planes occupied in a disordered manner by Mg^{2+} and Ga^{3+}. The muon spin rotation (μSR) experiments indicate the absence of static magnetism down to $T = 50$ mK and neutron scattering suggests a continuum of magnetic excitations, classifying $YbMgGaO_4$ as hosting a QSL state [10–12]. However, the influence of Mg^{2+} and Ga^{3+} local disorder on this continuum of magnetic excitations remains the subject of active discussion and study.

It was recently proposed that the problem of structural disorder can be overcome in rare-earth delafossites based on Ce or Yb, in which rare-earth ions order into structurally perfect two-dimensional (2D) triangular layers. These rare-earth delafossites share the same space group of $YbMgGaO_4$ and a planar triangular spin arrangement. We have recently reported an extensive study of one of such compound, $NaYbS_2$, using a combination of thermodynamic, local-probe, and neutron spectroscopy measurements both on high-quality single crystals and polycrystalline samples [13]. These measurements clearly evidence a strongly anisotropic quasi-2D magnetism and an emerging spin-orbit entangled $S = 1/2$ state of Yb towards low temperatures, together with an absence of long-range magnetic order down to 260 mK. The clear and narrow Yb electron spin resonance (ESR) lines, together with narrow ^{23}Na nuclear magnetic resonance (NMR) lines, evidence an absence of inherent structural distortions. This identifies $NaYbS_2$ as a rather pure spin-1/2 triangular lattice magnet and a new candidate quantum spin liquid [13].

To further investigate $NaYbS_2$, particularly the nature of its static and/or dynamic ground state, we have performed detailed (μSR) experiments, both in zero field (ZF) and in longitudinal field (LF) along the initial muon polarization, in the temperature range 0.05–40 K. The main focus was in the low-temperature region ($T \to 0$) and in the longer time window (up to 20 μs) to ensure the presence or absence of any long-range static magnetic ordering, which is indispensable information to justify the presence of a QSL ground state. The present μSR studies confirm the absence of any long-range magnetic ordering down to 0.05 K. Moreover, in the low-temperature limit, muon relaxation rates (λ), which probe the dynamical/static spin susceptibility at μeV energy scales, providing vital information that is complementary to nuclear magnetic resonance and inelastic neutron scattering, are constant not only for ZF but also for LF = 50 G. This indicates the presence of a highly correlated fluctuating quantum disordered phase in $NaYbS_2$. Thus, $NaYbS_2$ is identified as a candidate material to realize a certain class of QSL ground state.

Experiment. Single-crystalline and polycrystalline samples of $NaYbS_2$ were prepared according to Ref. [13]. μSR experiments were performed at the ISIS, U.K. using the MUSR

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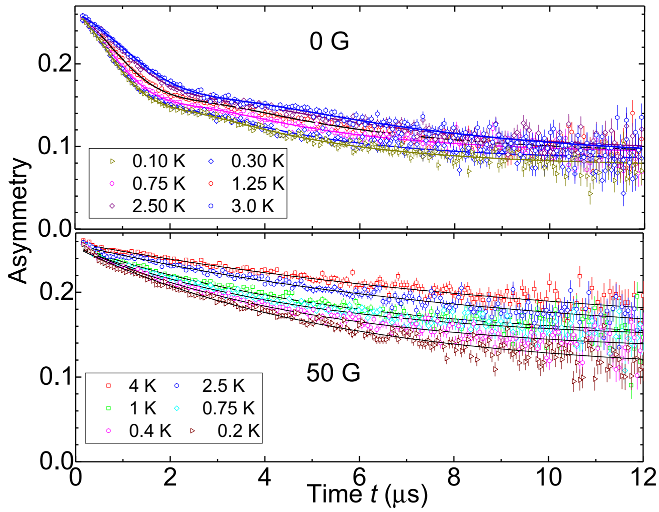

FIG. 1. Top panel: True ZF-μSR time spectra measured at ISIS. Bottom panel: μSR time spectra collected at 50 G applied longitudinal field. Lines indicate the theoretical description as detailed in the text.

instruments. For ISIS measurements, 300 mg of a powder sample, mixed with a small amount of GE varnish to ensure good thermal contact, was dispersed on a silver plate with a radius of 10 mm. The μSR data were analyzed with the free software packages MANTID.

μSR *results: Absence of long-range ordering.* Representative ZF and 50-G (LF)-μSR asymmetry spectra, measured in a wide temperature range, are shown in Fig. 1. In general, implanted muons (here positive muons) are highly sensitive to the local magnetic fields with a resolution of about $(B = \frac{2\pi}{\gamma_\mu}\nu_\mu) \approx 0.1$ mT produced by the adjacent Yb^{3+} spins [14]. This makes μSR as an ideal probe to detect the presence of any tiny static magnetism. It is clear that the present ZF-μSR spectra do not display any of the characteristic signals originating from static bulk magnetism: (1) any spontaneous coherent oscillations in the studied temperature range down to 0.05 K up to 20 μs time range (time spectra are shown up to 12 μs), (2) strong damping of the muon depolarization, or (3) a 1/3 recovery tail of the muon polarization due to a random distribution of the static field. On the contrary, the muon depolarizes faster as it cools. These points demonstrate the absence of a well-defined or disordered static magnetic field at the muon stopping site, ruling out the possibilities of any long-range ordered state of Yb^{3+} moments in $NaYbS_2$.

The ZF-time spectra can be adequately described by the following function in the whole temperature range studied,

$$A(t) = A_1 G^{\mathrm{KT}}(t, \sigma_{\mathrm{KT}}) + A_2 e^{-\lambda t} + B_{\mathrm{bg}}, \quad (1)$$

where A_1, A_2 represent the initial asymmetry, and B_{bg} is the constant background predominantly because of the muons stopped outside the sample. σ_{KT} and λ are the width of the static field distribution and muon relaxation rate, respectively. To describe the zero-field data adequately, two components are needed: One is a very small static fraction and the other one is an exponential relaxation function, respectively. The former can be easily decoupled by a small amount of longitudinal field, and the latter relates to the cooperative spin dynamics of Yb^{3+} spins. Both the contributions, as reflected in σ_{KT} and λ, increase with lowering the temperature (not shown here). We have also attempted to describe the data using a dynamic Gaussian Kubo-Toyabe $[G_Z^{\mathrm{DKT}}(t, \sigma_{\mathrm{DKT}})]$ and a product function of a Gaussian Kubo-Toyabe and exponential decay function $[G_Z^{\mathrm{KT}}(t, \sigma_{\mathrm{KT}})e^{-\lambda t}]$. However, neither of these expressions captures the behavior well across the whole temperature range.

The bottom panel of Fig. 1 shows the temperature dependence of the μSR time spectra at 50 G in LF. It appears that by applying only about 50 G LF, the static contribution is decoupled, and the μSR time relaxation spectra follow a single exponential decay, $A(t) = A_0 e^{-\lambda t} + B_{\mathrm{bg}}$. The observation of single exponential relaxation depolarization over the whole temperature range investigated evidences that $NaYbS_2$ is a dense electronic system reflecting a homogeneous electronic effect. In contrast, the $YbMgGaO_4$ system behaves differently, where the μSR time spectra were adequately described only by using a stretched exponential function, and the stretching parameter varied from 1 to 0.66 while cooling. This further supports the view that $NaYbS_2$ is electronically homogeneous.

In general, the longitudinal μ relaxation rate λ $(=1/T_1^\mu)$ can be correlated to a spin autocorrelation function by $1/T_1^\mu \sim \int_0^{+\infty} \langle \mathbf{S}(t)\mathbf{S}(0)\rangle \cos(\gamma_\mu H_{\mathrm{LF}} t)dt$, where $H_{\mathrm{LF}} = \omega/\gamma_\mu$ is the longitudinal applied magnetic field. Thus, λ depends significantly on the applied external longitudinal magnetic field and on the different correlation functions, $S(t) = \langle \mathbf{S}(t)\mathbf{S}(0)\rangle$ for the interacting Yb^{3+} spins. For an exponential correlation function, $S(t) = e^{-\nu t}$ leads to the usual Lorentzian spectral density, and this leads to $\lambda = 2\Delta^2\nu/(\nu^2 + \gamma_\mu^2 H_{\mathrm{LF}}^2)$, where Δ is the fluctuating component of the field at the muon site perpendicular to its initial polarization, ν is the fluctuation frequency, and γ_μ $(=2\pi \times 135.5$ MHz/T) is the muon gyromagnetic ratio. The field variation of λ may therefore reflect the underlying field distribution rather than the field tuned spin dynamics.

Above $T \sim 10$ K, λ $(=1/T_1^\mu)$ shows an almost temperature independent feature. Considering dimensionality two and a spin coordination number $z = 6$ for Yb^{3+} on triangular lattices, the Yb^{3+} spin fluctuation rate in the high T limit can be estimated by using $\nu = \sqrt{z}J_0 S/h \sim 1.2 \times 10^{11}$ Hz and $\sim 3.5 \times 10^{11}$ Hz while $J_{0\parallel} = (4.5$ K) and $J_{0\perp} = (13.5$ K), respectively [13,15]. Given that when the external field is zero, $\lambda = 2\Delta^2/\nu$, the internal field distributions are $\Delta_\parallel \sim 44.8\ \mu s^{-1}$ and $\Delta_\perp \sim 77.6\ \mu s^{-1}$ in the high-temperature range, which are $\ll \nu$ $(1.2 \times 10^{11}$ Hz, 3.5×10^{11} Hz). This confirms that the muon spin relaxation is in the fast fluctuation limit [10,15,16].

μSR *rate: Collective spin dynamics.* Figure 2 shows the obtained fitting parameter λ as a function of temperature for ZF and for 50 G. With lowering the temperature, λ increases constantly, and then saturates to a value of $\lambda_{\max} \approx 0.3\ \mu s^{-1}$ below 0.8 K. From the temperature dependence of the μSR investigations, at ZF three exclusive regions can be ascertained. Above 8 K, μ relaxation rates are temperature independent. This is typical for a paramagnet where spins are short time correlated. This is in agreement with the bulk magnetization data. Below 8 K, as the temperature goes down (0.8 K $< T <$ 8 K), a crossover region is evident, and there the μSR rates

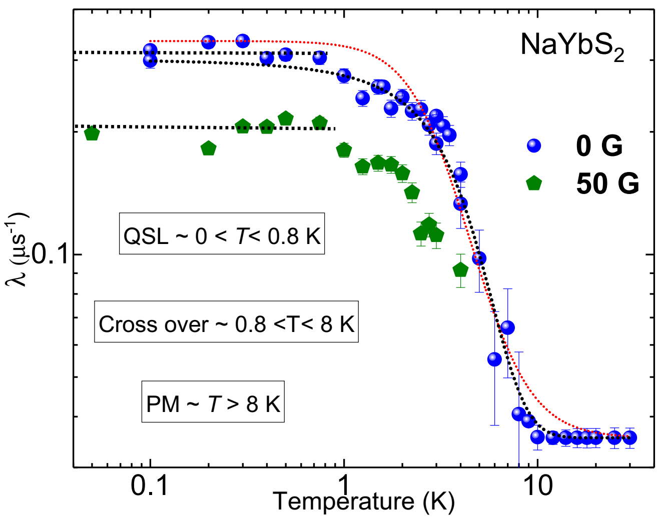

FIG. 2. Blue spheres and green pentagons: Temperature dependence of the ZF and 50-G μSR longitudinal relaxation rates of $NaYbS_2$. Dotted horizontal lines are guides to the eyes. Lines are theoretical descriptions as detailed in the main text.

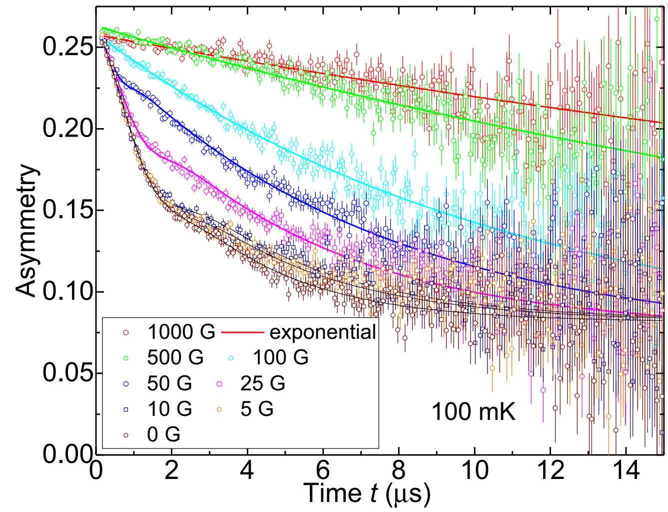

FIG. 3. Representative LF-μSR time spectra collected at 100 mK. Lines are theoretical descriptions as detailed in the main text.

are enhanced dramatically, almost 700%, which is nearly one order of magnitude higher than that which had been observed in $YbMgGaO_4$. With further lowering the temperature below 0.8 K, the μSR rates saturate to a constant value down to the lowest temperature studied. These sustained spin fluctuations are similar to the other QSL and frustrated magnets [10,15,17,18] and, in general, appear to be a signature of the QSL ground state. The red dotted line represents the logistic growth function. Clearly, this does not describe the data adequately. On the other hand, the black dotted line is the Boltzmann sigmoidal/growth functions which describe the data much better down to 0.8 K.

The next step is to study the LF effects on μ polarization to probe the nature of the spin dynamics in $NaYbS_2$. It is generally accepted that when μ depolarizes because of the presence of a static field distribution, a longitudinal field greater than the static field immediately decouples the μ depolarization. In contrast, when μ needs large LF to decouple, then this is most likely the effect of only fluctuating spins. Figure 3 shows the representative LF time spectra at 0.1 K. It is seen that even 1000 G LF is not sufficient to completely decouple the μ polarization at 0.1 K ($T \rightarrow 0$). Similar effects were also found at 4 K, i.e., in the crossover regime. In the case of $NaYbS_2$, noticeably, however, the μ signal decouples relatively smaller fields in comparison to $YbMgGaO_4$ ($S = 1/2$) or $Ba_3NiSb_2O_9$ ($S = 1$), despite the fact that in both the compounds not only the magnetic ions are located on the triangular lattice, but they also exhibit similar μ relaxation rate ($\lambda \sim 0.3\ \mu s^{-1}$) values (plateau) when approaching $T \rightarrow 0$. On the other hand, in the $NaYbO_2$ system the residual relaxation rate is $\lambda \sim 1\ \mu s^{-1}$, which is much larger than $NaYbS_2$, probably because of the larger crystalline electric fields (CEFs) [19].

For $YbMgGaO_4$ ($S = 1/2$), even 1800 G LF, and for $Ba_3NiSb_2O_9$, 8000 G LF, were not sufficient to completely decouple the muon relaxation [20]. It seems that in $NaYbS_2$ the realization of the persistent spin dynamics is rather weak. Whether or not this is a manifestation of a quantum spin liquid ground state or some other different phase is a subject of further investigations. On the other hand, it is worthwhile to consider the existence of very slow fluctuations and/or some slow freezing of the spins without any transition to a spin glass or a long-range ordered value. Thus, $NaYbS_2$ represents itself as an interesting compound which does not present any sign of ordering. In addition, this gives a demonstrable frustration parameter $(4.5/0.05) > 90$. By way of comparison, for a $NaYbO_2$ system the frustration parameter value is $(\theta_C W/0.050\ \text{K} > 200)$ [19].

The obtained λ_{LF} as a function of field ($\mu_0 H_{LF}$) for two different temperatures is shown in Fig. 4. In the low-temperature limit the simple exponential correlation function is not enough to describe λ_{LF}, and the spin dynamic correlation function should take a more general form, $S(t) \sim (\tau/t)^x e^{(-\nu t)}$, where for a simple exponential case $x = 0$ [17,21,22]. The

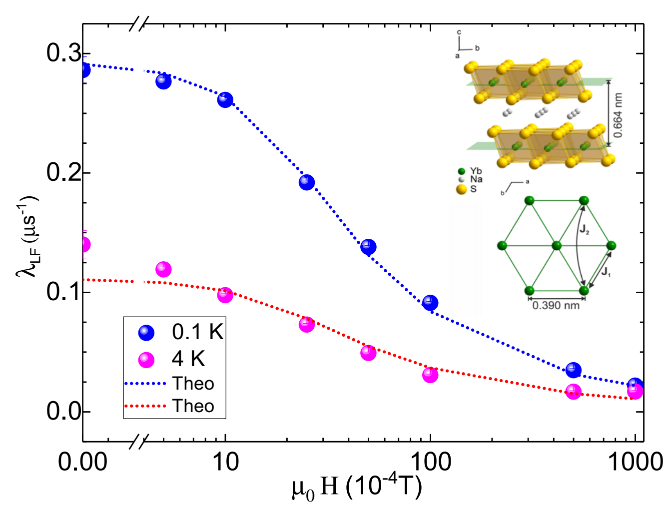

FIG. 4. Blue spheres and magenta spheres: Field dependence of the longitudinal relaxation rate (λ_{LF} at 0.1 and 4 K). The inset shows the structure of the $NaYbS_2$.

μ relaxation rate can be represented by the following equation, $\lambda(H) = 2\Delta^2\tau^x \int_0^\infty t^{-x}\exp(-\nu t)\cos(2\pi\mu_0\gamma_\mu H t)dt$. It is seen that in Fig. 4, λ_{LF} can better be described by the equation with $x \sim 0.44$ and $\nu \sim 1.7 \times 10^6$ Hz. Additionally, at 4 K, which is a representative temperature at the crossover regime, the λ_{LF} appears to be better described by $x \sim 0.49$ and $\nu \sim 1.7 \times 10^6$ Hz. This indicates much slower fluctuations with respect to the high-temperature paramagnetic state, suggesting long time spin correlations and the Yb^{3+} spins are entangled at low temperatures.

Contextually, $NaYbO_2$ has received significant attention, and is reported to be another disorder free triangular lattice QSL with a field tunable quantum disorder ground state [19,23,24]. The $NaYbO_2$ system is claimed to be an excellent system for studying the quantum disorder ground state in comparison to $YbMgGaO_4$. Very recently, we became aware that in an external field $NaYbS_2$ orders antiferromagnetically starting at 1 T [25], but in lower fields $NaYbS_2$ represents a critical QSL similar to $NaYbO_2$ [23,24]. Recent ESR and heat capacity experiments also give suggestive information about the presence of a QSL ground state in $NaYbS_2$ [25,26].

The observation of these two component features, in particular, the presence of a small amount of static component, is not new only for $NaYbS_2$ systems as for other delafossites. Recent heat capacity measurements on this compound as reported elsewhere [13] showed a maximum at around 0.8 K. This was conjectured as the emerging spin liquid phase most likely with partially gapped magnetic excitations. Another likely scenario is that the low-temperature spin dynamics are not sufficient to completely subdue magnetic order. The observed peak corresponds to a partial (probably short-range) magnetic order of a minor fraction of spins, whereas the major part remains fluctuating. This scenario cannot be completely ruled out considering the two component features of the muon time spectra. In light of this discussion, it is worthwhile to note that the other delafossites $NaYbO_2$ and $KCeS_2$ also show the same two components in their muon spectra [19,27], but they have roughly equal amplitudes. In contrast, the static contribution for $NaYbS_2$ is $< 5\%$.

Conclusions. In conclusion, a detailed μSR study on the $NaYbS_2$ system is presented. There is no sign of long-range magnetic ordering at least down to 0.05 K. μSR relaxation rate λ values below ~0.8 K are constant, suggesting a cooperative quantum disordered ground state in $NaYbS_2$. Taken all together, that is, the low dimensionality, high anisotropy, and high frustration index, the present μSR studies suggest $NaYbS_2$ to be a disorder free triangular lattice which hosts a QSL ground state. But what kind of QSL, and what kinds of excitations are relevant in $NaYbS_2$ demands further theoretical and experimental investigations.

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