

Quantum spin liquid ground state in the disorder free triangular lattice NaYbS₂

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(Dated: November 20, 2019)

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 YbMgGaO₄, which is known for considerable structural disorder due to site intermixing, NaYbS₂ delafossite realizes structurally ideal triangular layers. We present detailed μ SR studies on this regular (undistorted) triangular Yb sublattice based system with effective spin $J_{\text{eff}} = 1/2$ in the temperature range 0.05 - 40 K. Zero-field (ZF) and longitudinal field (LF) μ SR studies confirm the absence of any long range magnetic order state down to 0.05 K ($\sim J/80$). Current μ SR results together with the so far available bulk characterization data suggest that NaYbS₂ is an ideal candidate to identify QSL ground state.

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 perfect triangular lattice. Such a scenario has proved exceptionally hard to realize.^{1,2} The organic compounds κ -(BEDT-TTF)₂Cu₂(CN)₃ and EtMe₃Sb[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 analogues such as Ba₃CoSb₂O₉ and NaTiO₂ either order magnetically or undergo a lattice deformation on cooling.⁵⁻⁸

In this context, the triangular lattice magnet YbMgGaO₄ was identified as a potential QSL candidate with an effective spin $J_{\text{eff}} = 1/2$.⁹ YbMgGaO₄ contains undistorted triangular planes of magnetic Yb³⁺ with space group $R\bar{3}m$, separated by two triangular planes occupied in a disordered manner by Mg²⁺ and Ga³⁺. The μ SR experiments indicate the absence of static magnetism down to $T = 50$ mK and neutron scattering suggests a continuum of magnetic excitations classifying YbMgGaO₄ as hosting a QSL state.¹⁰⁻¹² However, the influence of Mg²⁺ and Ga³⁺ 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 2D triangular layers. These rare-earth delafossites share the same space group of YbMgGaO₄ and the planar triangular spin arrangement. We have recently reported an extensive study of one of such compounds, NaYbS₂ using a combination of thermodynamic, local-probe, and neutron spectroscopy measurements both on high quality single crystals and polycrystalline sam-

ples.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-ESR lines together with narrow ²³Na NMR lines evidence an absence of inherent structural distortions. This identifies NaYbS₂ as a rather pure spin-1/2 triangular lattice magnet and a new candidate quantum spin liquid.¹³

To further investigate NaYbS₂, particularly its nature of the static and/or dynamic ground state, we have performed detailed (μ SR) experiments, both in zero-field (ZF) and in longitudinal field (LF) along the initial muon polarisation, in the temperature range 0.05-40 K. The main focus was in the low temperature region ($T \rightarrow 0$) and in the longer time window (up to 20 μ s) to ensure the presence or absence of any long-range static magnetic ordering which is an indispensable information to justify the presence of a QSL ground state. Present μ SR 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 probes the dynamical/static spin susceptibility at μ eV 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 NaYbS₂. Thus, NaYbS₂ is identified as a candidate material to realize certain class of QSL ground state.

Experimental. Single crystalline and polycrystalline samples of NaYbS₂ were prepared according to Ref [13]. μ SR experiments were performed at the ISIS, UK using the MUSR instruments. For ISIS measurements, 300 mg of a powder sample, mixed with small amount of GE Varnish to ensure good thermal contact, was dispersed on a silver plate with a radius of 10 mm. The μ SR data were analyzed with the free software packages Mantid. *μ SR results: absence of long range ordering.* Representative

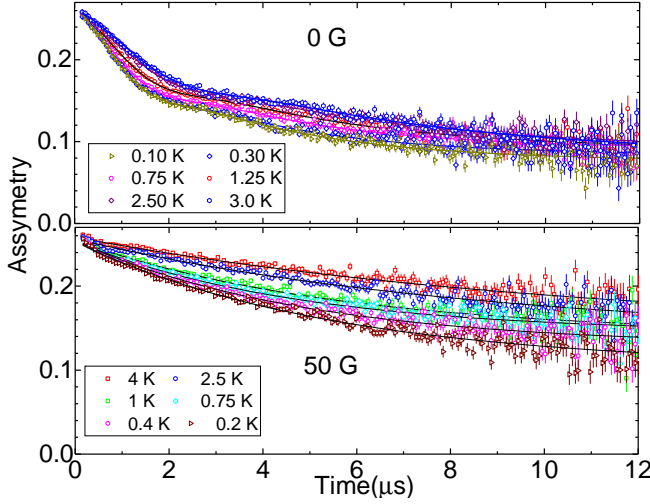


FIG. 1. Top panel: True ZF- μ SR time spectra measured at ISIS. Bottom panel: μ SR time spectra collected at 50 G applied longitudinal field. Lines indicate the theoretical description as detailed in the text.

ZF and 50 G (LF)- μ SR asymmetry spectra, measured in a wide temperature ranges, are shown in Fig. 1. In general, implanted muons (here positive muons) are highly sensitive to the local magnetic fields with a resolution about ($B = \frac{2\pi}{\gamma_\mu} \nu_\mu$) ≈ 0.1 mT produced by the adjacent Yb^{3+} spins.¹⁴ This makes μ SR as an ideal probe to detect the presence of any tiny static magnetism. It is clear that the present ZF- μ SR spectra do not display any of the characteristic signals originating from static magnetism: 1.) Any spontaneous coherent oscillations in the studied temperature range down to 0.05 K up to 20 μ s time range (time spectra are shown upto 12 μ s), 2.) Strong damping of the muon depolarization or 3.) the 1/3 recovery tail of the muon polarization due to random distribution of the static field. On the contrary muon depolarizes faster as cooling. 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:

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

where A_1 , A_2 represents the initial asymmetry, 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 very small static fraction and the other one is exponential relaxation function, respectively. The former can be easily decoupled by a small amount of longitudinal field, and the later relates to the cooperative spin dynamics of Yb^{3+} spins. Both the contributions, as reflected in σ_{KT} and λ

increase as lowering the temperature (not shown here). We have also attempted to describe the data using a Dynamic Gaussian Kubo-Toyabe ($G_Z^{\text{DKT}}(t, \sigma_{\text{DKT}})$) and a product function of Gaussian Kubo-Toyabe and exponential decay function ($G_Z^{\text{KT}}(t, \sigma_{\text{KT}})e^{-\lambda t}$). However, neither of these expressions capture the behavior well across the whole temperature range.

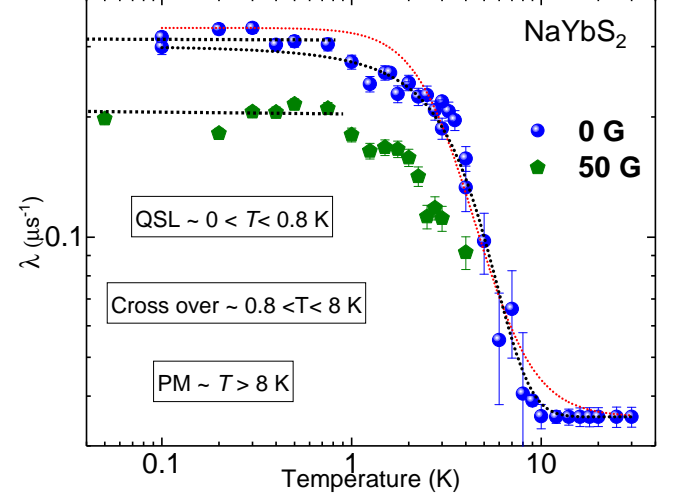


FIG. 2. (Blue spheres and green pentagons) Temperature dependence of the ZF and 50 G- μ SR longitudinal relaxation rates of NaYbS_2 . Dotted horizontal lines are guide to the eyes. Lines are theoretical descriptions as detailed in the main text.

The bottom panel of Fig. 1 shows the temperature dependence of μ SR time spectra at 50 G in LF. It appears that by applying only about 50 G LF, the static contribution is decoupled, and the μ SR time relaxation spectra follow a single exponential decay: $A(t) = A_0 e^{-\lambda t} + B_{\text{bg}}$. The observation of single exponential relaxation depolarization over the whole temperature range investigated evidences that NaYbS_2 is a dense electronic system reflecting an homogeneous electronic effect. In contrast, the YbMgGaO_4 system behaves differently, where μ SR 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 spin auto-correlation function by $1/T_1^\mu \sim \int_0^{+\infty} \langle \mathbf{S}(t) \mathbf{S}(0) \rangle \cos(\gamma_\mu H_{\text{LF}} t) dt$, where $H_{\text{LF}} = \omega/\gamma_\mu$ 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 $\lambda = 2\Delta^2 \nu / (\nu^2 + \gamma_\mu^2 H_{\text{LF}}^2)$ 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 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, Yb^{3+} spin fluctuation rate in the high T limit can be estimated by using $\nu = \sqrt{z}J_0S/h \sim 1.2 \times 10^{11}$ 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 external field is zero, $\lambda = 2\Delta^2/\nu$, gives the internal field distribution $\Delta_{\parallel} \sim 44.8 \mu\text{s}^{-1}$ and $\Delta_{\perp} \sim 77.6 \mu\text{s}^{-1}$ 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}

μ SR 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 $\lambda_{\text{max}} \approx 0.3 \mu\text{s}^{-1}$ below 0.8 K. From the temperature dependence of μ SR 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 timed correlated. This is in agreement with bulk magnetization data. Below 8 K, as the temperature goes down ($0.8 \text{ K} < T < 8 \text{ K}$) a crossover region is evident, and there μ SR rates enhance dramatically almost 700 %, which is nearly one order of magnitude higher than that had been observed in YbMgGaO_4 . With further lowering the temperature below 0.8 K, μ SR 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 QSL ground state. The red dotted line represents the logistic growth function. Clearly this doesn't describe the data adequately. On the other hand the black dotted line is the Boltzman sigmodial/growth function which describe the data much better down to 0.8 K.

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 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 case of NaYbS_2 noticeably, however, the μ signal decouples relatively smaller fields in comparison to YbMgGaO_4 ($S=1/2$) or $\text{Ba}_3\text{NiSb}_2\text{O}_9$ ($S=1$), despite that in both the compounds not only the magnetic ions are located on the triangular lattice, but also exhibit a similar μ relaxation rate ($\lambda \sim 0.3 \mu\text{s}^{-1}$) values (plateau) as approaching $T \rightarrow 0$. On the other hand, in NaYbO_2 system the residual relaxation rate is

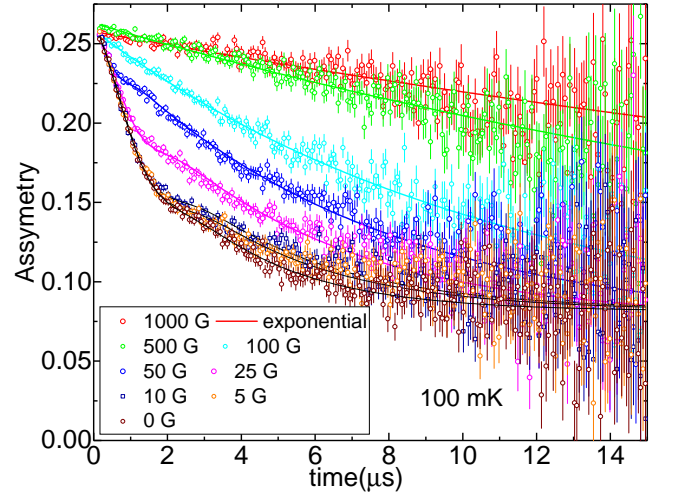


FIG. 3. Representative LF- μ SR time spectra collected at 100 mK. Lines are theoretical descriptions as detailed in the main text.

$\lambda \sim 1 \mu\text{s}^{-1}$ which is much larger than NaYbS_2 probably because of the larger CEF's.¹⁹

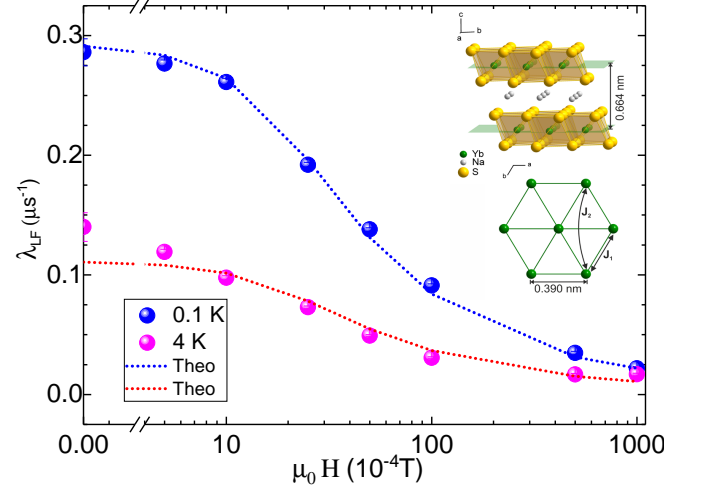


FIG. 4. (Blue spheres and magenta spheres) Field dependence of the longitudinal relaxation rate (λ_{LF} at 0.1 K and 4 K). Inset shows the structure of the NaYbS_2 .

For YbMgGaO_4 ($S=1/2$), even 1800 G LF, and for $\text{Ba}_3\text{NiSb}_2\text{O}_9$ 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 quantum spin liquid ground state or some other new phase is a subject of further investigations. On the other hand it is worthwhile to consider that the existence of very slow fluctuations and/or some slowly 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 NaYbO₂ system the frustration parameter value is $(\theta_C W/0.050 \text{ K} > 200)$.¹⁹

The obtained λ_{LF} as a function of field ($\mu_0 H_{\text{LF}}$) for two different temperatures are shown in the Fig. 4. In the low temperature limit to describe λ_{LF} the simple exponential correlation function is not enough, 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 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 $\nu \sim 1.7 \times 10^6 \text{ Hz}$. Additionally, at 4 K, which is a representative temperatures at the cross-over regime, the λ_{LF} appears to be better described $x \sim 0.49$ and $\nu \sim 1.7 \times 10^6 \text{ Hz}$. This indicates much slower fluctuations with respect to high temperature paramagnetic state suggesting a long time spin correlations and the Yb³⁺ spins are entangled at low temperatures.

Contextually NaYbO₂ 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} NaYbO₂ system is claimed to be an excellent system for studying quantum disorder ground state in comparison to YbMgGaO₄. Very recently we became aware that in external field NaYbS₂ orders antiferromagnetically starting at 1 T (Baenitz et al to be published), but in lower fields NaYbS₂ represents a critical QSL similar to NaYbO₂.^{23,24}

The observation of this two component features, in particular the presence of small amount of static component is not new only for NaYbS₂ system like other delafossites. The recent heat capacity measurements on this

compound as reported elsewhere¹³ 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 can not 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 NaYbO₂ and KCeS₂ also show the same two components in their muon spectra^{19,25}, but they have roughly equal amplitudes. In contrast, the static contribution for NaYbS₂ is $< 5\%$.

Conclusions. In conclusion, a detailed μ SR study on the NaYbS₂ system is presented. **There is no sign of long range magnetic ordering at least down to 0.05 K. μ SR relaxation rate λ values below $\sim 0.8 \text{ K}$ are constant suggesting a cooperative quantum disordered ground state in NaYbS₂.** Taken all together, that is the low dimensionality, high anisotropy, high frustration index and present μ SR studies suggest NaYbS₂ to be a disorder free triangular lattice which hosts QSL ground state. But what kind of QSL, and what kind of excitations are relevant in NaYbS₂ demands further theoretical and experimental investigations.

Acknowledgments. This work was financially supported by the Deutsche Forschungsgemeinschaft (DFG) within the SFB 1143 Correlated Magnetism From Frustration to Topology, [project-id 247310070](#) (Projects C02 and B03), GR 4667/1. We gratefully acknowledge the Science and Technology Facilities Council (STFC) for access to muon beamtime at ISIS. We thank M. Baenitz and D. Inosov for helpful discussions.

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