Muon Spin Relaxation Evidence for the U(1) Quantum Spin-Liquid Ground State in the Triangular Antiferromagnet $YbMgGaO_4$

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Muon spin relaxation (μSR) experiments on single crystals of the structurally perfect triangular antiferromagnet $YbMgGaO_4$ indicate the absence of both static long-range magnetic order and spin freezing down to 0.048 K in a zero field. Below 0.4 K, the μ^+ spin relaxation rates, which are proportional to the dynamic correlation function of the Yb^{3+} spins, exhibit temperature-independent plateaus. All these μSR results unequivocally support the formation of a gapless U(1) quantum spin liquid ground state in the triangular antiferromagnet $YbMgGaO_4$.

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Introduction.—Antiferromagnetically coupled ($J > 0$) spins on a perfect geometrically frustrated lattice, such as the triangular or kagome lattices, can preserve strong fluctuations and evade long-range order or spin freezing even at $T \ll J$. They reveal exotic phases characterized by interesting properties, such as fractionalized spin excitations, intrinsic topological order, and gapless excitations without symmetry breaking. These new phases have been proposed as quantum spin liquids (QSLs) [1–3]. The spin-$\frac{1}{2}$ triangular Heisenberg antiferromagnet, initially believed to host a resonating-valence-bond QSL ground state (GS) [4–6], develops, in fact, the 120°-type magnetic order [7–11]. However, this order is very fragile and can be melted by perturbations, such as next-nearest-neighbor couplings [12,13], spatially anisotropic interactions [14–16], and bond randomness [17]. Theoretical studies found that ring couplings destroy long-range order as well and trigger the formation of a U(1) QSL GS with a spinon Fermi surface [18–20]. On the other hand, relevant experimental systems, κ-$(BEDT\text{-}TTF)_2Cu_2(CN)_3$ [21] and $EtMe_3Sb[Pd(dmit)_2]_2$ [22], revealed linear temperature dependence of the heat capacity, in contrast to $C_v \sim T^{2/3}$ predicted for the U(1) QSL [18].

Recently, our group reported a new structurally perfect rare-earth triangular antiferromagnet $YbMgGaO_4$ [23,24]. Unlike the majority of QSL candidates, $YbMgGaO_4$ is free from magnetic defects [25–29], spatial anisotropy [29–31], and antisymmetric Dzyaloshinsky-Moriya anisotropy [32,33]. Its magnetic heat capacity reveals the $C_v \sim T^{2/3}$ behavior with almost zero residual entropy down to ~0.06 K [23] compatible with the triangular U(1) QSL GS [18]. The spin susceptibility of a U(1) QSL is expected to approach a constant value as the temperature goes down to zero [18–20]. While the divergent nature of the bulk static susceptibility of $YbMgGaO_4$ was measured down to 0.48 K [23], this experimental temperature range was certainly not low enough to probe GS properties in a system, where J_0 is as low as 1.5 K [24]. In the following, we fill this gap by probing $YbMgGaO_4$ down to 0.048 K and provide evidence for the U(1) QSL GS.

Weak magnetic couplings between the rare-earth Yb^{3+} spins render the experimental probe of the GS extremely challenging. While nuclear magnetic resonance requires an external field on the order of 1 T that inevitably perturbs such a system, inelastic neutron scattering can be performed in a zero field, but fails to detect an inelastic signal at transfer energies below 0.1 meV, owing to the contamination by the elastic signal. In this respect, muon spin relaxation (μSR) is an ideal technique that can be performed in a true zero field (ZF). ZF-μSR is an extremely sensitive probe detecting tiny internal fields on the order of 0.1 G. The μSR time window can measure magnetic fluctuation rates in the range from 10^4 to 10^{12} Hz [34].

In this Letter, we report a comprehensive μSR investigation of the GS spin dynamics of $YbMgGaO_4$ using single-crystal samples. Neither the oscillation signal nor characteristic recovery of the polarization to 1/3 is observed in the ZF measurements with the incident μ^+ polarization perpendicular and parallel to the c axis down to 0.048 K and 0.066 K, respectively, thus, ruling out any static uniform or random field exceeding 0.09 mT. The μ^+ spin relaxation rate ($\propto Yb^{3+}$ spin dynamic correlation

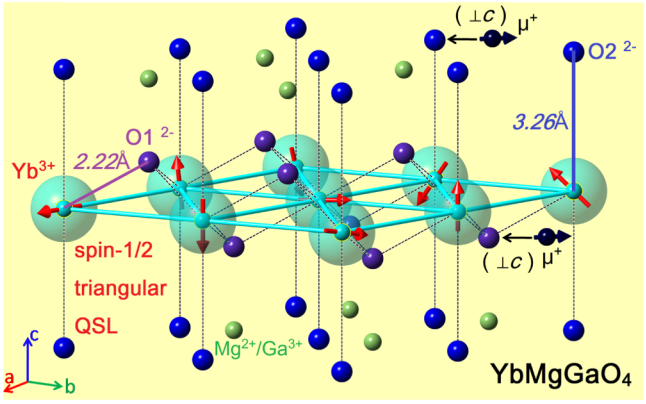

FIG. 1. Crystal structure of $YbMgGaO_4$ around the effective spin-$\frac{1}{2}$ triangular layer of Yb^{3+}. The implanted μ^+ is likely to stop near $O1^{2-}$ and $O2^{2-}$, and probes local spin dynamics of the triangular QSL.

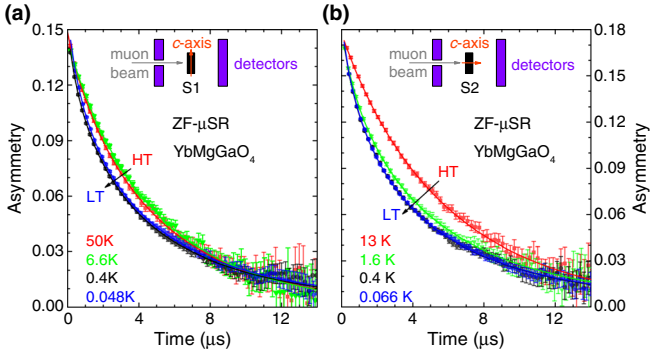

FIG. 2. Selected background-subtracted ZF-μSR signals with the incident beam (a) perpendicular ($\perp$) and (b) parallel ($\parallel$) to the c axis. The colored lines are the corresponding fits to the data using Eq. (1). The insets show relevant experimental geometries.

function) measured in a ZF exhibits a plateau below 0.4 K. These observations strongly suggest that a gapless U(1) QSL GS is formed in $YbMgGaO_4$.

Experimental technique.—Large single crystals (~1 cm) of $YbMgGaO_4$ were grown by the floating zone technique [24]. The high quality of the single crystals was confirmed by x-ray diffraction showing narrow reflections with $\Delta(2\theta) \sim 0.04°$ [35]. The crystal orientations were determined by Laue x-ray diffraction. The crystals were cut into slices along both the c axis and the ab plane with a homogeneous thickness of ~1 mm. Mosaics of slices along the c axis ($S1$) and the ab plane ($S2$) were mounted on two silver sample holders [35]. The μSR data were collected at the ISIS pulsed muon facility, Rutherford Appleton Laboratory, United Kingdom, on both samples ($S1$ and $S2$) between 0.05 and 4 K using dilution refrigerators. Additional data between 2 and 50 K were collected by transferring the sample to a ^{4}He cryostat [36]. The international system of units is used throughout this Letter, and $\langle\rangle$ represents thermal and sample average.

Absence of spin freezing.—Implanted muons are very sensitive to local magnetic fields induced by the neighboring Yb^{3+} spins (Fig. 1) [35]. Therefore, ZF-μSR is the best tool to detect long-range magnetic order or spin freezing.

Our ZF and longitudinal-field (LF) data (Figs. 2 and 3) are well fitted to a stretched exponential relaxation function

$$A^{\mathrm{ZF/LF}} = A_0 \exp[-(\lambda t)^{\beta}] + B_{\mathrm{ZF/LF}}. \quad (1)$$

Here, $A_0 \sim 0.2$ is the initial asymmetry (weakly temperature and field dependent), λ is the μ^+ spin relaxation rate, and β is the stretching exponent. The coefficient $B_{\mathrm{ZF/LF}} \sim 0.13$ is a background constant representing muons that missed the sample. As background constants depend on the sample environment (cryostat), we use background-subtracted ZF/LF data, whereas raw data can be found in the Supplemental Material [35]. For the sample $S1$, the ZF signal increases (by < 0.01) in the critical or crossover temperature range (0.4 to 4 K) at long times, but this effect is within the error bar of the data [35].

The absence of spin freezing in $YbMgGaO_4$ is supported by the following observations. (1) The ZF signals decrease continuously (see Fig. 2) without showing oscillations within the analyzed time window up to 20 μs. This continuous decrease is observed at all temperatures down to 0.048 and 0.066 K with the incident μ^+ polarization perpendicular and parallel to the c axis, respectively [35], suggesting that no static uniform local field is formed. (2) The ZF spectra also lack a recovery of the polarization to 1/3, suggesting the absence of static random fields (no spin-glass-like freezing) [37,38]. (3) The stretching exponent β gradually decreases from ~1 at high temperatures ($T \geq 4$ K $\gg J_0$) down to a constant value of ~0.6 at the lowest temperatures ($T \sim 0.1$ K $\ll J_0$), see Fig. 4(b). In contrast, for a spin glass system, β is expected to drop to 1/3 at the spin freezing temperature [39,40]. (4) The ZF μ^+ spin relaxation rate λ^{ZF} increases by 50% upon cooling from $T \gg J_0$ ($\lambda^{\mathrm{ZF}} \sim 0.2\ \mu\mathrm{s}^{-1}$) down to $T \ll J_0$ ($\lambda^{\mathrm{ZF}} \sim 0.3\ \mu\mathrm{s}^{-1}$), see Fig. 2 and Fig. 5. This indicates only a

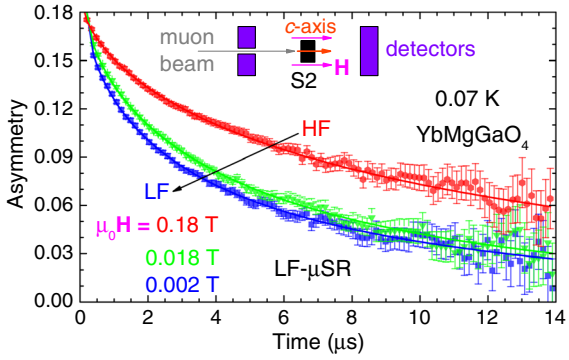

FIG. 3. Selected LF-μSR signals (background-subtracted) at 0.07 K. The colored lines are fits to the data using Eq. (1). The insets show the experimental geometries.

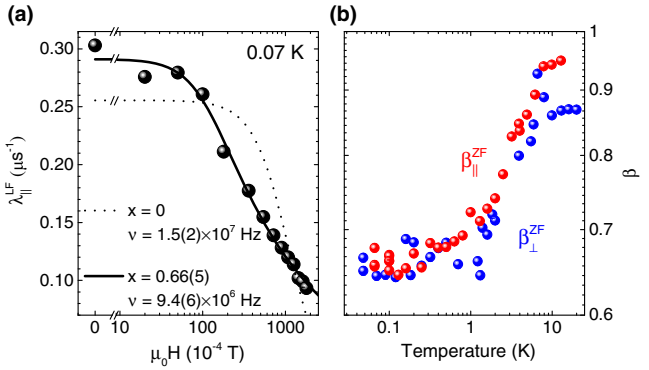

FIG. 4. (a) LF dependence of the μ^+ spin relaxation rate, $\lambda_{\parallel}^{\mathrm{LF}}(H)$, at 0.07 K. The dotted line represents the fit to the $\lambda_{\parallel}^{\mathrm{LF}}$ using Eq. (2) ($x = 0$), and the solid line is the fit using Eq. (3) with $x = 0.66(5)$. (b) Temperature dependence of the stretching exponents, β.

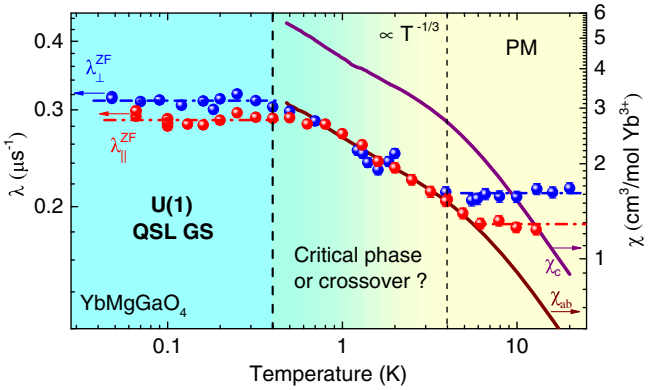

FIG. 5. Temperature dependence of the μ^+ spin relaxation rates from the zero field measurements and from the bulk static susceptibilities measured in the applied field of 0.01 T. PM represents the conventional paramagnetic phase.

weak slowing down of the Yb^{3+} spin fluctuations [41–43], whereas in a spin glass, the relaxation rate will typically increase by several orders of magnitude below the freezing point ($\lambda \sim 1$–$20\ \mu s^{-1}$) [37,38,43]. (5) The LF spectra measured at 0.07 K change gradually and only moderately when the field is increased from 0 to 0.18 T, see Fig. 3 and Fig. 4(a). This is another evidence for the dynamic relaxation and the absence of long range magnetic order down to the lowest temperature.

The upper limit of local static uniform or random fields can be estimated as $(\langle B_{\mathrm{loc}}^{ab}\rangle + \langle B_{\mathrm{loc}}^{c}\rangle)/2 < 0.09$ mT down to 0.048 K and $\langle B_{\mathrm{loc}}^{ab}\rangle < 0.09$ mT down to 0.066 K [35].

Dynamic spin correlation.—At high temperatures ($T > 4$ K), the μ^+ spin relaxations (Fig. 2) can be well fitted by Eq. (1) with a stretching exponent $\beta \sim 1$, indicating a concentrated spin system (spin-1/2 triangular lattice of Yb^{3+}) with very fast spin fluctuations [37,38]. In this case, a Gaussian distribution of local magnetic fields is expected with the width $\Delta \sim \gamma_\mu \sqrt{\langle B_{\mathrm{loc}}^2\rangle}$ and $\langle B_{\mathrm{loc}}\rangle \sim 0$, here $\gamma_\mu = 135.5$ MHz/T is the μ^+ gyromagnetic ratio. In the high-T (mean-field) limit, the Yb^{3+} spin fluctuation rate can be estimated as $\upsilon = \sqrt{z} J_0 s/h \sim 4 \times 10^{10}$ Hz [38], where $z = 6$ is the coordination number.

Above 4 K, μ^+ spin relaxation rates, both $\lambda_{\parallel}^{\mathrm{ZF}}$ and $\lambda_{\perp}^{\mathrm{ZF}}$ with the incident μ^+ polarization parallel and perpendicular to the c axis, reach temperature-independent values of 0.185(2) and 0.211(1) μs^{-1}, respectively (see Fig. 5). Using Eq. (2) with $H = 0$

$$\lambda(T > 4\mathrm{K}, H) = \frac{2\Delta^2 \nu}{\nu^2 + (\mu_0 H \gamma_\mu)^2}, \qquad (2)$$

we can estimate the distribution width of the local magnetic fields, $\Delta \sim 6 \times 10^7$ Hz $\ll \upsilon$, confirming the fast spin fluctuation limit [38,43]. The slight difference between $\lambda_{\parallel}^{\mathrm{ZF}}$ and $\lambda_{\perp}^{\mathrm{ZF}}$, as well as between $\beta_{\parallel}^{\mathrm{ZF}}$ and $\beta_{\perp}^{\mathrm{ZF}}$, is probably rooted in the anisotropic magnetic couplings due to the strong spin-orbit couplings in Yb^{3+}. At high temperatures, $(2/\nu_{ab})/(1/\nu_{ab} + 1/\nu_c) \sim 2J_{zz}/(J_{zz} + 2J_{\pm})$ [24] ~0.7 is qualitatively consistent with the measured $\lambda_{\parallel}^{\mathrm{ZF}}/\lambda_{\perp}^{\mathrm{ZF}} \sim 0.87$, assuming the isotropic nature of Δ^2 (i.e., $\Delta_c^2 \sim \Delta_{ab}^2$) ($\Delta^2$ should also be anisotropic). Note, also, that $S1$ and $S2$ are different crystals and not two different orientations of the same crystals. Therefore, some sample-related difference in the μSR signal cannot be excluded.

At the lowest temperature of our measurement, $T \sim 0.07$ K, $\lambda_{\parallel}^{\mathrm{LF}}(H)$ does not follow Eq. (2) [see Fig. 4(a)], suggesting that the spin dynamic autocorrelation function $S(t)$ should take a general form, $S(t) \sim (\tau/t)^x \exp(-\upsilon t)$, instead of the simple exponential form ($x = 0$) [41,42,44], where τ and $1/\upsilon$ are the early and late time cutoffs, respectively, and x can be defined as a critical exponent [45].

A more general expression for the μ^+ spin relaxation rate can be obtained from both semi-classical [42] and full quantum [46] treatments,

$$\lambda(H) = 2\Delta^2 \tau^x \int_0^\infty t^{-x} \exp(-\upsilon t) \cos(2\pi\mu_0 \gamma_\mu H t) dt. \qquad (3)$$

For a spin system at $T \gg J_0$, the μ^+ relaxation rates take the simple form of Eq. (2). On the other hand, at 0.07 K, $\lambda_{\parallel}^{\mathrm{LF}}(H)$ is much better described by Eq. (3) with $x = 0.66(5)$ and $\upsilon = 9.4(6) \times 10^6$ Hz [see Fig. 4(a)]. This way, at low temperatures, $S(t)$ behaves more power-law-like and attenuates much slower than $S(t) \sim \exp(-\upsilon t)$ with $\upsilon \sim 4 \times 10^{10}$ Hz at high temperatures [35,42]. It suggests the onset of long-time spin correlations at low temperatures. The Yb^{3+} spins in $YbMgGaO_4$ are strongly entangled not only in space, but also in time, yet without showing symmetry breaking or spin freezing, thus, meeting the basic requirements of a gapless quantum spin liquid.

All of the fitted stretching exponents β gradually decrease as the temperature goes down. At $T < 0.4$ K, the stretching exponent β approaches a constant value of

~0.6, indicating a distribution of the μ^+ spin relaxation rates [see Fig. 4(b)]. This decrease is a very common phenomenon in lots of frustrated magnets and may be caused by the onset of Yb^{3+} spin correlations in space [47].

Muon spin relaxation rate.—Spin relaxation rates λ provide further insight into the GS spin dynamics of the system, because a dynamic local magnetic field is induced by the effective spin-1/2 moment of Yb^{3+} on the triangular lattice. The ZF-μ^+ spin relaxation rate can be expressed as [41–43]

$$\lambda^{\mathrm{ZF}} = \gamma_\mu^2 \int_0^\infty \langle \mathbf{B}_{\mathrm{loc}}^{\perp}(t) \cdot \mathbf{B}_{\mathrm{loc}}^{\perp}(0) \rangle dt \propto S_{\omega\to 0}^{\perp}, \qquad (4)$$

where $S_{\omega\to 0}^{\perp} = \int_0^\infty \langle \mathbf{s}_i^{\perp}(t) \cdot \mathbf{s}_i^{\perp}(0) \rangle dt$ is the static spin structure factor with the sum over $\mathbf{q}$ [48].

As the temperature goes down, all of the μ^+ spin relaxation rates gradually increase by about 50% compared to the high-T limit and get saturated below 0.4 K (see Fig. 5). These observations indicate the slowing down of spin fluctuations.

Phase diagram and conclusions.—Our observations indicate that strong phase-coherent quantum fluctuations of Yb^{3+} spins survive at low temperatures. Upon changing the temperature from above 4 K to below 0.4 K and suppressing the energy of thermal fluctuations by at least 1 order of magnitude, about 2/3 of the total fluctuations at $T > 4$ K are retained due to the strong geometrical frustration [1]. The spin relaxation rate λ^{ZF} saturates below $T_s \simeq 0.4$ K (Fig. 5). This characteristic temperature of 0.4 K is nearly the same as in other QSL candidates [44,47,49–51] but is significantly higher on the relative energy scale of $YbMgGaO_4$ ($T_s/J_0 \sim 0.27$). We suggest that quantum fluctuations have a paramount effect below this temperature, regardless of the absolute scale of the magnetic couplings J_0.

Several distinct regimes of $YbMgGaO_4$ can also be inferred from the temperature evolution of the bulk static susceptibility and μ^+ spin relaxation rates, as shown in Fig. 5. At $T > 4$ K, the susceptibilities follow the Curie-Weiss law [24], and the μ^+ spin relaxation rates are constants, as is typical for the paramagnetic regime [38,41,47,49–51]. The spins are still short-time correlated with $S(t) \sim \exp(-\upsilon t)$. This high-temperature phase is compatible with the classical paramagnet. As the temperature goes down (0.4 K $< T <$ 4 K), the spin susceptibilities show an unconventional critical behavior $\chi \sim T^{-1/3}$, and the μ^+ spin relaxation rates increase by 50% following the uniform spin susceptibilities (Fig. 5).

At even lower temperatures ($T < 0.4$ K), the μ^+ spin relaxation rates approach constant values again (Fig. 5). This behavior appears to be generic for frustrated magnets [38,41,47,49–51]. The temperature-independent μ^+ spin relaxation rates suggest a constant spin correlation $S_{\omega\to 0}$ with the sum over $\mathbf{q}$, which is consistent with the triangular U(1) QSL GS [18–20].

In conclusion, we performed a comprehensive study of spin dynamics in the frustrated antiferromagnet $YbMgGaO_4$ by μSR measurements using the high-quality single crystals along both the c axis and the ab plane. No static uniform or random field is detected ($\langle B_{\mathrm{loc}} \rangle < 0.09$ mT), indicating the absence of spin freezing ($\langle \mathbf{s}_j \rangle \sim 0$) down to at least 0.048 K. The spin correlation function reaches a constant value below 0.4 K. Long-time Yb^{3+} spin correlations are developed in this temperature range. Combined with the heat-capacity data [23], our observations provide compelling evidence for the formation of gapless U(1) QSL GS in the triangular antiferromagnet $YbMgGaO_4$.

The concept of the U(1) QSL has been applied to a gamut of systems, from spin-ice pyrochlores [52,53] to high-temperature superconductors [54], but experimental observation of this state remains elusive. Spin liquids in the organic charge-transfer salts with the frustrated triangular geometry deviate from the anticipated U(1) behavior [18,21,22]. In contrast, $YbMgGaO_4$ displays several phenomenological signatures of this state: (i) the absence of long-range magnetic order and spin freezing, (ii) the constant magnetic susceptibility at low temperatures, (iii) the $C_v \simeq T^{2/3}$ power-law behavior of the specific heat [23]. Our findings pave the way for observing further emergent properties of the triangular U(1) QSL, including the $\kappa \simeq T^{1/3}$ behavior of thermal conductivity and violation of the Wiedemann-Franz law [19,55], the power-law optical conductivity inside the Mott gap [56], and surface plasmons driven by spinons [57]. Exact origin of the U(1) QSL GS of $YbMgGaO_4$ is also of interest. In contrast to the organic charge-transfer salts, where long-range magnetic order is destabilized by ring exchange, spin-orbit coupling and the ensuing magnetic anisotropy are the most likely effects that trigger strong frustration in this material [58].

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