\(\mu\)SR evidence for the U(1) quantum spin liquid ground state in the triangular antiferromagnet YbMgGaO\(_4\)

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Muon spin relaxation and rotation (\(\mu\)SR) 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 zero field. Below 0.4 K, the \(\mu^+\) spin relaxation rates, which are proportional to the dynamic correlation function of the Yb\(^{3+}\) spins, exhibit temperature-independent plateaus. Same behavior is revealed by temperature-independent local susceptibilities extracted from the Knight shifts of the \(\mu^+\) spin rotation frequencies under a transverse field of 20 Oe. All these \(\mu\)SR 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 (QSL) \cite{1,2,3}. The spin-\(\frac{1}{2}\) triangular Heisenberg antiferromagnet, initially believed to host a resonating-valence-bond QSL ground state (GS) \cite{4,5,6}, develops in fact the 120°-type magnetic order \cite{7,8,9,10,11}. However, this order is very fragile and can be melted by perturbations, such as next-nearest-neighbor couplings \cite{12,13}, spatially anisotropic interactions \cite{14,15}, and bond randomness \cite{16}. 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 \cite{17,18}. On the other hand, relevant experimental systems, \(\kappa\)-(BEDT-TTF)\(_2\)Cu\(_2\)(CN)\(_3\) \cite{21} and EtMe\(_3\)Sb[Pd(dmit)\(_2\)] \cite{22}, revealed linear temperature dependence of the heat capacity, in contrast to \(C_v \sim T^{2/3}\) predicted for the U(1) QSL \cite{18}.

Recently, our group reported a new structurally perfect rare-earth triangular antiferromagnet YbMgGaO\(_4\) \cite{23,24}. Unlike the majority of QSL candidates, YbMgGaO\(_4\) is free from magnetic defects \cite{25,26}, spatial anisotropy \cite{27,28}, and antisymmetric Dzyaloshinsky-Moriya anisotropy \cite{29,30,31}. Its magnetic heat capacity reveals the \(C_v \sim T^{2/3}\) behavior with almost zero residual entropy down to \(\sim 0.06\) K \cite{23} compatible with the triangular U(1) QSL GS \cite{18}. The spin susceptibility of a U(1) QSL is expected to approach a constant value as the temperature goes down to zero \cite{18,20}. While the divergent nature of the bulk static susceptibility of YbMgGaO\(_4\) was measured down to 0.48 K \cite{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 \cite{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 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 zero field, but fails to detect inelastic signal at transfer energies below 0.1 meV, owing to the contamination by the elastic signal. In this respect, muon spin relaxation and rotation (μSR) is an ideal technique that can be performed in 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 10⁴ to 10¹² Hz [34, 35].

In this Letter, we report a comprehensive μSR investigation of the GS spin dynamics of YbMgGaO₄ using single crystals. 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 0.9 Oe. Both the Knight shift of the μ⁺ spin rotation frequency (α local spin susceptibility) measured under a transverse field (TF) of 20 Oe and the μ⁺ spin relaxation rate (α spin dynamic correlation function) measured in ZF/TF exhibit a plateau below 0.4 K. These observations strongly suggest that a gapless U(1) QSL GS is formed in YbMgGaO₄ below 0.4 K. These observations strongly suggest that a gapless U(1) QSL GS is formed in YbMgGaO₄.

Experimental technique.—Large single crystals (~1 cm) of YbMgGaO₄ 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 Δ(2θ) ~ 0.04° [36]. The crystal orientations were determined by Laue x-ray diffraction. The crystals were cut into slices along both c-axis and 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, U.K on both samples (S1 and S2) between 0.05 K and 4 K using dilution refrigerators. Additional data between 2 K and 50 K were collected by transferring the sample to a ⁴He cryostat [37]. The SI units are used, and ⟨⟩ represents a thermal and sample average.

Absence of spin freezing.—Implanted muons are very sensitive to local magnetic fields induced by the neighboring Yb³⁺ spins (Fig. 1) [36]. Therefore, ZF-μSR is the best tool to detect long-range magnetic order or spin freezing.

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

\[ A_{ZF/LF}^{\alpha} = A_0 \exp[-(\lambda t)^{\beta}] + B_{ZF}, \]  

The raw TF data (Fig. 3b) are well fitted to a stretched exponential relaxation function with damped oscillations [37, 38]:

\[ A_{TF}^{\alpha} = A_0 \exp[-(\lambda t)^{\beta}] \cos(\omega t) + B_g \cos(\omega_0 t) + B_{TF}. \]  

Here, \( A_0 \sim 0.2 \) is the initial asymmetry (weakly temperature- and field-dependent), \( \lambda \) is the \( \mu^+ \) spin relaxation rate, and \( \beta \) is the stretching exponent. The coefficients \( B_{ZF} \sim 0.13, B_g \sim 0.12, \) and \( B_{TF} \sim 0.02 \) are background constants representing muons that missed the sample (\( B_{ZF} \)), missed the sample but stayed in the TF or stopped in the Ag-holder (\( B_g \)), and missed both the sample and the TF (\( B_{TF} \)). The frequencies \( \omega = 2\pi\gamma_\mu B_{loc} \) and \( \omega_0 = 2\pi\gamma_\mu B_0 \) are caused by the local field \( B_{loc} \) and by the external TF \( B_0 \), respectively, and \( \gamma_\mu = 135.5 \text{ MHz/T} \) is the \( \mu^+ \) gyromagnetic ratio.

The absence of spin freezing in YbMgGaO₄ is supported by the following observations:

i) The ZF asymmetries decrease continuously (see Fig. 2) without showing oscillations in the full time window, 0–20 μs. This continuous decrease is observed down to 0.048 K and 0.066 K with the incident \( \mu^+ \) field perpendicular and parallel to the c-axis, respectively [36], suggesting that no static uniform local field is formed.

ii) The ZF relaxations also lack a recovery of the polarization to 1/3, suggesting the absence of static random fields (no spin-glass-like freezing) [29, 41].

iii) The stretching exponent \( \beta \) gradually decreases from \( \sim 1 \) at high temperatures \( (T \geq 4 K \gg J_0) \) down to a constant value of \( \sim 0.6 \) at the lowest temperatures \( (T \sim 0.1 K \ll J_0) \), see the inset of Fig. 1. In contrast, for a spin glass one expects that \( \beta \) decreases from 1 at high temperatures to 1/3 at the spin freezing temperature [11, 12].

iv) The ZF \( \mu^+ \) spin relaxation rate \( \lambda_{ZF} \) increases by 50% upon cooling from \( T \gg J_0 (\lambda_{ZF} \sim 0.2 \mu s^{-1}) \) down to \( T \ll J_0 (\lambda_{ZF} \sim 0.3 \mu s^{-1}) \), see Fig. 2 and Fig. 5. This indicates only a weakly slowing down of...
the Yb$^{3+}$ spin fluctuations $^{38, 43, 44}$, whereas in a spin glass the relaxation rate will typically increase by several orders of magnitude below the freezing point ($\lambda \sim 1 - 20 \text{ ms}^{-1}$) $^{39, 40, 44}$.

Our observations indicate that the relatively 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 one 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 $\lambda_{ZF}$ saturates below $T_{c} \simeq 0.4$ K (Fig. 5). This characteristic temperature of 0.4 K is nearly the same as in other QSL candidates $^{45-49}$ and, remarkably, quite high on the relative energy scale of YbMgGaO$_{4}$ ($T_{c}/J_{0} \sim 0.27$). We suggest that quantum fluctuations have paramount effect below this temperature, regardless of the absolute scale of the exchange couplings $J_{0}$.

The upper limit of local static uniform or random fields can be estimated as $(\langle B_{\text{loc}}^{a} \rangle + \langle B_{\text{loc}}^{b} \rangle)/2 < 0.9$ Oe down to 0.048 K and $\langle B_{\text{loc}}^{o} \rangle < 0.9$ Oe down to 0.066 K $^{39, 40}$.

**Knight shift.**—The TF-μSR time spectra are shown in Fig. 3 for the TF of 20 Oe with the incident $\mu^{+}$ polarization parallel to the c-axis. The frequency of the muon spin rotation is proportional to the local field in the $ab$-plane, $\omega = \gamma_{\mu}B_{\text{loc}}^{ab}$ and the Knight shift should be proportional to the static $\mathbf{q} = 0$ susceptibility $\chi$,

$$K_{ab} = \frac{\omega - \omega_{0}}{\omega_{0}} = \frac{C_{\mu} - Y_{b} \times \chi_{ab}}{4\pi N_{A} \mu_{B}},$$  

where $C_{\mu} - Y_{b} \sim 1.4(6)$ kOe/$\mu_{B}$ represents the effective dipole magnetic coupling constant between the implanted $\mu^{+}$ spins and the Yb$^{3+}$ spins $^{48}$. $N_{A}$ and $\mu_{B}$ are Avogadro’s constant and Bohr magneton, respectively. As the temperature goes down, the Knight shift follows the bulk static susceptibility down to 0.48 K (see Fig. 4), which is the lowest temperature of the susceptibility measurement. Below 0.4 K, the Knight shift saturates at a constant value suggesting the zero-temperature spin susceptibility of $\sim 3.5$ cm$^{-1}$/mol Yb$^{3+}$, which is consistent with the U(1) QSL GS with a spinon Fermi surface $^{18}$.

**Muon spin relaxation rate.**—Spin relaxation rates $\lambda$ provide further insight into the GS spin dynamics of the system, because 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 $^{38, 43, 44}$:

$$\lambda_{ZF} = \gamma_{\mu}^{2} \int_{0}^{\infty} (B_{\text{loc}}^{ab}(t) \cdot B_{\text{loc}}^{ab}(0))dt \propto S_{\omega \rightarrow 0}^{-},$$  

where $S_{\omega \rightarrow 0}^{-} = \int_{0}^{\infty} (s_{\omega}^{+}(t) \cdot s_{\omega}^{+}(0))dt$, is the static spin structure factor with the sum over $\mathbf{q}$. It is fully compatible with the static susceptibility at $\mathbf{q} = 0$ $^{50}.$

At high temperatures ($T > 4$ K), the $\mu^{+}$ spin relaxations (Fig. 2) can be well fitted by Eq. (1) with a stretching exponent $\beta \sim 1$ (see the inset of Fig. 2b), indicating a concentrated spin system (spin-1/2 triangular lattice of Yb$^{3+}$) with very fast spin fluctuations $^{39, 40}$. In this case, a Gaussian distribution of local magnetic fields is expected with the width $\Delta \sim \gamma_{\mu} \sqrt{\langle B_{\text{loc}}^{2} \rangle}$ and $\langle B_{\text{loc}} \rangle \sim 0$. In the high-T (mean-field) limit, the Yb$^{3+}$ spin fluctuation rate can be estimated as $v = \sqrt{2} J_{0}/\hbar \sim 4 \times 10^{10}$ Hz $^{40}$, where $z = 6$ is the coordination number.

Above 4 K, experimental $\mu^{+}$ spin relaxation rates, both $\lambda_{ZF}^{ab}$ and $\lambda_{ZF}^{ab}$ with the incident $\mu^{+}$ polarization parallel and perpendicular to the c-axis, reach temperature-independent values of 0.185(2) and 0.211(1) μs$^{-1}$, respectiv-
rate can be obtained from both semi-classical \cite{43} and fluctuation limit \cite{40, 44}. The spins are still short-time correlated with $S(t) \sim \exp(-\nu t)$. 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 $\mu^+$ spin relaxation rates increase by 50\% following the uniform spin susceptibilities (Fig. 5).

As the temperature is decreased even further ($T < 0.4\, K$), the $\mu^+$ spin relaxation rates approach constant values again (Fig. 5). This behavior appears to be generic for frustrated magnets \cite{38, 40, 40, 49}. The temperature-independent $\mu^+$ spin relaxation rates suggest a constant spin correlation $S_{w=0}$ with the sum over $q$, which is consistent with the triangular U(1) QSL GS \cite{18, 20}. Furthermore, the Yb$^{3+}$ spins on the triangular lattice become long-time correlated, $S(t) \sim (\tau/t)^2 \exp(-\nu t)$, with $x = 0.66(5)$. In conclusion, we performed a comprehensive study of spin dynamics in the frustrated antiferromagnet YbMgGaO$_4$ by $\mu$SR measurements using the high-quality single crystals along both $c$-axis and $ab$-plane. No static uniform or random field is detected ($\langle B_{loc} \rangle < 0.9\, Oe$), indicating the absence of spin freezing ($\langle s_j^z \rangle \sim 0$) down to at least 0.048 K. Below 0.4 K, both local susceptibility (Knight shift) and spin correlation function reach constant values. Long-time Yb$^{3+}$ spin correlations are developed in this temperature range. Combined with the heat-capacity data \cite{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 \cite{53, 54} to high-temperature superconductors \cite{55}, 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 \cite{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 $C_\alpha \approx T^{2/3}$ power-law behavior of the specific heat \cite{23}. Our findings pave the way for observing further emergent properties of the triangular U(1) QSL, including the $\kappa \sim T^{1/3}$ behavior of thermal conductivity and violation of the Wiedemann-Franz law \cite{19, 50}, the power-law optical conductivity inside the Mott gap \cite{57}, and surface plasmons driven by spinons \cite{58}. 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 \cite{59}.
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