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Zihao Zhu,1,11 Binglin Pan,1,11 Linpeng Nie,2,11 Jiamin Ni,1,11 Yanxing Yang,1 Changsheng Chen,1 Chengyu Jiang,1 Yeyu Huang,1 Erjian Cheng,1 Yunjie Yu,1 Jianjian Miao,3 Adrian D. Hillier,4 Xianhui Chen,2,5,6 Tao Wu,2,5,6,9 Yi Zhou,7,8,9,10 Shiyan Li,1,5,10,11 and Lei Shu1,5,10,11

*Correspondence: wutao@ustc.edu.cn (T.W.); yizhou@iphy.ac.cn (Y.Z.); shiyan_li@fudan.edu.cn (S.L.); leishu@fudan.edu.cn (L.S.)
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PUBLIC SUMMARY

- Multiple techniques are used to study QSL candidate NaYbSe₂.
- The absence of long-range magnetic order is confirmed by all techniques.
- Coexistence of quasi-static and dynamic spins is observed in both μSR and NMR.
- Results of thermal conductivity suggest the absence of itinerant gapless magnetic excitations.
- A scenario of fluctuating ferrimagnetic droplets immersed in a sea of QSL is proposed.
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1State Key Laboratory of Surface Physics, and Department of Physics, Fudan University, Shanghai 200433, China
2CAS Key Laboratory of Strongly-coupled Quantum Matter Physics, Department of Physics, University of Science and Technology of China, Hefei, Anhui 230026, China
3Department of Physics, the University of Hong Kong, Hong Kong, China
4ISIS Pulsed Neutron and Muon Source, STFC Rutherford Appleton Laboratory, Harwell Campus, Didcot, Oxfordshire OX11 0QX, UK
5Collaborative Innovation Center of Advanced Microstructures, Nanjing 210093, China
6CAS Center for Excellence in Superconducting Electronics (CENSE), Shanghai 200050, China
7Institute of Physics and Beijing National Laboratory for Condensed Matter Physics, Chinese Academy of Sciences, Beijing 100190, China
8Songshan Lake Materials Laboratory, Dongguan, Guangdong 523808, China
9Kavli Institute for Theoretical Sciences and CAS Center for Excellence in Topological Quantum Computation, University of Chinese Academy of Sciences, Beijing 100190, China
10Shanghai Research Center for Quantum Sciences, Shanghai 201315, China
11These authors contributed equally

*Correspondence: wutao@ustc.edu.cn (T.W.); yizhou@iphy.ac.cn (Y.Z.); shiyan_li@fudan.edu.cn (S.L.); leishu@fudan.edu.cn (L.S.)
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The search of quantum spin liquid (QSL), an exotic magnetic state with strongly fluctuating and highly entangled spins down to zero temperature, is a main theme in current condensed matter physics. However, there is no smoking gun evidence for deconfined spinons in any QSL candidate so far. The disorder and competing exchange interactions may prevent the formation of an ideal QSL state on frustrated spin lattices. Here we report comprehensive and systematic measurements of the magnetic susceptibility, ultralow-temperature specific heat, muon spin relaxation (μSR), nuclear magnetic resonance (NMR), and thermal conductivity for NaYbSe2 single crystals, in which Yb3+ ions with effective spin-1/2 form a perfect triangular lattice. All these complementary techniques find no evidence of long-range magnetic order down to their respective base temperatures. Instead, specific heat, μSR, and NMR measurements suggest the coexistence of quasi-static and dynamic spins in NaYbSe2. The scattering from these quasi-static spins may cause the absence of magnetic thermal conductivity. Thus, we propose a scenario of fluctuating ferrimagnetic droplets immersed in a sea of QSL. This may be quite common on the way pursuing an ideal QSL, and provides a brand new platform to study how a QSL state survives impurities and coexists with other magnetically ordered states.

INTRODUCTION

Quantum spin liquid (QSL) is a highly entangled quantum state in which spins remain disordered and dynamic even down to absolute zero temperature because of strong quantum fluctuations.1–6 Such an exotic state was first proposed from the study of the triangular-lattice Heisenberg antiferromagnets in 1973 by Anderson.7 Since QSLs have potentially tight relationship with high-temperature superconductivity7–9 and quantum information applications,10 it has gained continuous attention in condensed matter physics. The QSL states are characterized by fractional spin excitations, such as spinons, and the detection of these excitations is a crucial issue for identifying QSL in real materials.2

The temperature dependence of specific heat, μSR, nuclear magnetic resonance (NMR), and ultralow-temperature thermal conductivity measurements on NaYbSe2 single crystals. The absence of magnetic order and spin glass is confirmed by different techniques down to 50 mK. With decreasing temperature in ZF, a hump followed by a linear temperature-dependent specific heat is observed. In µSR and NMR measurements, both quasi-static and dynamic spins are found clearly in NaYbSe2. Furthermore, the residual linear term of thermal conductivity at all fields is negligible, pointing to the absence of itinerant fermionic magnetic excitations in NaYbSe2. Our data reveal that NaYbSe2 hosts a ground state of fluctuating ferrimagnetic droplets immersed in a sea of QSL on Yb3+ triangular lattice.

RESULTS

The temperature dependences of magnetic susceptibility χ of NaYbSe2 in external magnetic field μ0H = 1 T in two different directions are plotted in Figure 1B. The absence of magnetic phase transition is confirmed down to 2 K. There is no splitting between ZF cooling and field cooling curves of magnetic susceptibility (Figure S2A), suggesting no spin glass in the system down to 2 K. The inset of Figure 1B presents a Curie-Weiss (CW) fit with field perpendicular to the c axis. The data above 100 K can be well fitted by CW law, giving effective moment μeff = 4.54 μB and the CW temperature θCW = −49.0 K. The value of θCW agrees with the theoretical prediction 4.54 μB for trivalent Yb3+ ion with J = 7/2. Similar to YbMgGaO4,11 magnetization M remains unsaturated but is smaller up to 7 T at 2 K in NaYbSe2 (Figure S2B). The larger absolute value of the CW temperature and smaller M indicate stronger anti-ferromagnetic (AFM) interactions in NaYbSe2.

The temperature dependence of specific heat of NaYbSe2 in various fields (H || c) from 0.05 to 20 K are shown in Figure 1C. Consistent with magnetic
susceptibility and former reports, no sharp anomaly of LRO is observed in NaYbSe$_2$ in ZF. With decreasing temperature, a broad hump of specific heat shows up, whose position shifts to a higher temperature in magnetic field. However, we do not observe the field-induced transition peak reported previously, because of the lack of the sufficiently strong field. For the ZF data, after subtracting the contributions from phonon and nuclear Schottky anomaly (Figure S3), we obtain the magnetic contribution of specific heat $C_{\text{Mag}}/T$ as shown in Figure 1D. We should emphasize that the broad hump is not caused by a crystal electric field (CEF) effect, since the energy gap of CEF is approximately 15 meV, which should appear above 100 K. As guided by the red dashed line, the temperature-independent behavior of $C_{\text{Mag}}/T$ below 0.25 K is consistent with a spinon Fermi surface. By integrating $C_{\text{Mag}}/T$, we obtain the magnetic entropy $S_{\text{Mag}}$ as shown in Figure 1D. For an effective spin-1/2 system, the theoretical magnetic entropy is $R \ln 2$, where $R$ is the gas constant. The residual entropy of NaYbSe$_2$ at 50 mK is only 5.2% of total entropy. Such little entropy remaining suggests low-temperature physics is dominated by quantum fluctuations rather than thermal fluctuations, indicating the existence of QSL.

Figure 1. Basic properties of NaYbSe$_2$ (A) The unit cell of NaYbSe$_2$. Green spheres, Na; red spheres, Yb; blue spheres, Se. (B) The temperature dependence of magnetic susceptibility at $\mu_0 H = 1$ T of NaYbSe$_2$. The inset shows the fitting result of CW law at $T > 100$ K. (C) The temperature dependence of specific heat $C$ at $\mu_0 H = 0$, 4, and 8 T. (D) The magnetic specific heat $C_{\text{Mag}}/T$ and the calculated magnetic entropy $S_{\text{Mag}}$ at ZF. The red dashed line is a guide to eyes to show that $C_{\text{Mag}}/T$ is temperature independent at low temperature.

Figure 2. ZF $\mu$SR experiment (A) Time spectra of ZF-$\mu$SR at representative temperatures. The curves are the fittings using Equation 1. (B) Temperature dependence of fraction of quasi-static spins. (C) Temperature dependence of relaxation rate caused by quasi-static and dynamic spins $\sigma$ (blue spheres) and $\lambda$ (green spheres), respectively. The error bars are statistical standard deviation in A, and are determined by the least square method in (B and C).
Both $\mu$SR and NMR, which measure spin dynamics at different frequency ranges, are powerful tools in clarifying the static and/or dynamic nature of the magnetic ground state. $\mu$SR, which uses muon as a probe, is more sensitive to local magnetic field.\textsuperscript{28,50–53} As shown in Figure 2A, the time spectra of muon polarization $P(t)$ in ZF clearly indicates that LRO, which usually induces oscillations in the spectra, is absent in ZF-$\mu$SR down to 88 mK. Similar time spectra of muon polarization $P(t)$ has been obtained previously\textsuperscript{54}; however, we find that the relaxation process of ZF-$\mu$SR can be best described by the sum of a Kubo-Toyabe (KT) term and an exponential term:

$$P(t) = fG_{KT}(\sigma,t) + (1 - f)\exp(-\lambda t),$$  \hspace{1cm}  \text{(Equation 1)}$$

where $f$ is the fraction of the KT term. The fitting function is exactly the same as the NaYbS\textsubscript{2} case.\textsuperscript{41} The KT term originates from an isotropic Gaussian distribution of randomly oriented static or quasi-static local fields, whose relaxation rate $\sigma$ is proportional to the root-mean-square width of the distribution.\textsuperscript{50} The exponential term with relaxation rate $\lambda$ originates from dynamic spins. The successful fitting with the above function strongly suggests the coexistence of distinguishable quasi-static spins and dynamic spins.

The temperature dependence of $f$, $\sigma$, and $\lambda$ are plotted in Figures 2B and 2C. At high temperatures, the value of $f$ is equal to 1, indicating a trivial paramagnetic state. The temperature-dependent NMR linewidth derived from the spectra as described in SM. The quasi-static (blue spheres) and dynamic (green spheres) components can be easily separated. (C) The temperature dependence of the spin-lattice relaxation rate $1/T_1$. The error bars are determined by the least square method in (B and C).

From 20 K to 6 K, the system gradually turns into a non-trivial paramagnetic state, in which spin correlation is established and significant spin dynamic appears. As a result, $f$ decreases continuously and the second term in Equation 1 appears. Below 6 K, the temperature-dependent $f$ stops decreasing and up-turns, while the temperature-dependent $\sigma$ also increases clearly below 6 K. These results strongly suggest the formation of quasi-static spins at low temperatures. However, both $\sigma$ and $f$ saturates to a finite value at low temperatures, and the saturation value of $f$ indicates that only 23\% of the sample becomes quasi-static at base temperature. However, both $\sigma$ and $f$ saturates to a finite value at low temperatures, and the saturation value of $f$ indicates that only 23\% of the sample becomes quasi-static at base temperature. In contrast, the temperature-dependent $\lambda$ also increases below 4 K, supporting the enhancement of spin dynamics at low temperatures. The temperature-independent behavior of $\lambda$ below 0.2 K suggests the existence of persistent spin dynamics. We would like to emphasize that these fitting parameters are fully independent and reproducible (see Figure S4 and S5 and related discussion in supplemental information.) Additional evidence for the coexistence of quasi-static and dynamic spins in NaYbSe\textsubscript{2} comes from a longitudinal field (LF) $\mu$SR, which yields that the fluctuation rate $\nu$ at 0.1 K is 2.8 MHz (Figure S4), larger than 1.7 MHz in NaYbS\textsubscript{2}.\textsuperscript{41}

Similar evidence for the coexistence of quasi-static and dynamic spins is also found in $^{23}$Na NMR experiments. As shown in Figure 3A, the three-peak structure of $^{23}$Na NMR spectra at high temperature comes from quadrupole splitting of nuclei with spin number $I = 3/2$ (Figure 3A). With temperature decreasing, a tiny asymmetry in spectrum appears below 49 K, suggesting a new component also with three-peak structure (Figures S6A and S9), which is ascribed to the quasi-static spins as observed by our ZF-$\mu$SR measurement. As shown in...
induced magnetic ordering, has been observed in previous NMR work on contrast with traditional spin glass.\textsuperscript{55,56} It should be noted that, although a similar spectral broadening at a higher magnetic field should be taken into account for linewidth broadening. However, since the sample used in the present NMR measurements has a shape of thin flake, the demagnetization effect can be neglected in the following analysis (see details in the discussion of Figure S8). The quasi-static moment can be estimated from the broad part of the NMR spectrum, yielding a small value of 0.13 $\mu_B$ (Figure S7). This result indicates that the quasi-static spins in NaYbSe$_2$ are still fluctuating, which is in sharp contrast with traditional spin glass.\textsuperscript{55,56} It should be noted that, although a similar spectral broadening at a higher magnetic field, which was ascribed to a field-induced magnetic ordering, has been observed in previous NMR work on NaYbSe$_2$\textsuperscript{44} and NaYbO$_2$.\textsuperscript{42} The lower magnetic field used in the present work only leads to the formation of magnetic droplets instead of a uniform magnetic ordering. This is also supported by the absence of a peak-like behavior in the temperature-dependent nuclear spin-lattice relaxation rate $1/T_1$. Beside the NMR spectrum, the nuclear spin-lattice relaxation also supports the coexistence of quasi-static and dynamic spins. Inhomogeneous spin dynamics are indeed observed below 2 K accompanied by the above two-component behavior in spectrum. As shown in Figure S6C, the stretching exponent $\beta$, which usually depicts the inhomogeneity of spin dynamics, decreases clearly below 2 K with the value well below 1. Especially at the lowest temperature of 0.25 K, there is a clear two-component behavior appearing in the recovery curve of $T_1$ process (Figure S6B), which is beyond a single $T_1$ fitting with stretching exponent. This is in line with the scenario proposed above with the coexistence of quasi-static and dynamic spins. Finally, the temperature dependence of the spin-lattice relaxation rate $1/T_1$, extracted from the stretched exponential fitting is plotted in Figure 3C. The broad hump feature around 50 K is usually ascribed to the development of strong spin correlation at a low temperature or CEF effect.\textsuperscript{52} The absence of magnetic order is confirmed again by the absence of any significant critical fluctuation at low temperatures. Below 2 K, $1/T_1$ saturates to a constant, coinciding with the persistent spin dynamics observed in $\mu$SR experiments. This result also excludes the possibility of a trivial spin glass phase, suggesting a novel magnetic ground state in NaYbSe$_2$.

To further check the existence of gapless magnetic excitations, we performed thermal conductivity measurements to probe the possible itinerant excitations. As for a QSL candidate, thermal conductivity measurement is highly advantageous in probing such elementary excitations, since it is only sensitive to itinerant gapless ferromagnetic excitations and phonons, respectively.\textsuperscript{57,58} Because of the specular reflections of phonons at the sample surfaces, the power $\alpha$ in the second term is typically between 2 and 3.\textsuperscript{57,58} The in-plane thermal conductivity of NaYbSe$_2$ in zero and various fields ($\mu_0H \parallel c$) are shown in Figure 4. In ZF, the fitting to the data below 0.4 K gives the residual linear term $\kappa_0/T = a = -0.038 \pm 0.007 \text{ mW K}^{-2} \text{ cm}^{-1}$ and $\alpha = 1.66 \pm 0.04$. This unusual behavior is very similar to YbMgGaO$_4$,\textsuperscript{30} both in the unphysical negative $\kappa_0/T = -0.025 \pm 0.002 \text{ mW K}^{-2} \text{ cm}^{-1}$ and $\alpha = 1.85 \pm 0.04$ for YbMgGaO$_4$.\textsuperscript{30} For comparison, we also measured the thermal conductivity of the non-
magnetic NaLuSe$_2$ single crystal, as plotted in Figure S10. There is no magnetic field effect on the thermal conductivity of NaLuSe$_2$, and its thermal conductivity shows a typical phonon behavior in the boundary scattering limit, with $\kappa_0/T = -0.008 \pm 0.010$ mW K$^{-2}$ cm$^{-1}$ and $\alpha = 2.57 \pm 0.05$. More important, the magnitude of thermal conductivity for NaYbSe$_2$ is lower than that of NaLuSe$_2$. Again, this is a similar situation to YbMgGaO$_4$, which has a lower magnitude than non-magnetic LuMgGaO$_4$. We note that the magnitude of $\kappa/T$ is comparable between NaYbSe$_2$ and YbMgGaO$_4$ in ZF. Previously, we estimated the upper limit of the spinon (if it exists) means a free path with $k_0/T = 0.041 \pm 0.001$ mW K$^{-2}$ cm$^{-1}$ and $\alpha = 1.97 \pm 0.05$. Therefore, $\kappa_0/T$ is virtually zero in all fields, indicating the absence of itinerant gapless magnetic excitations in NaYbSe$_2$, and its thermal conductivity is mainly contributed by phonons. Figure 4A, the fitting to the data of $\mu H = 5$ T gives $k_0/T = -0.041 \pm 0.001$ mW K$^{-2}$ cm$^{-1}$ and $\alpha = 1.97 \pm 0.05$. Therefore, $\kappa_0/T$ is virtually zero in all fields, indicating the absence of itinerant gapless magnetic excitations in NaYbSe$_2$, and its thermal conductivity is mainly contributed by phonons. Figure 4B plots the field dependence of the $\kappa/T$ at 0.2, 0.3, and 0.4 K. For $\mu H < 3$ T, $\kappa/T$ is independent of fields. With increasing fields, the spins are increasingly polarized, thus reducing the scattering of phonon, leading to the rapid enhancement of thermal conductivity from 3 to 5 T, as observed in YbMgGaO$_4$. 

**DISCUSSION**

We now turn to discuss the ground state of NaYbSe$_2$. The absence of LRO and spin glass in NaYbSe$_2$ is confirmed down to 50 mK. Both $\mu$SR and NMR experiments point out that a minority of quasi-static spins and a majority of dynamic spins co-exist in NaYbSe$_2$ down to the base temperature. In fact, our specific heat measurements also hint at this picture. The broad hump of $C_{\text{total}}/T$ at approximately 0.8 K comes from the correlations of quasi-static spins, while the temperature-independent behavior below 0.25 K suggests the existence of well-defined magnetic excitations, which is an essential feature of gapless QSLs.

Comparing our results in ZF, these two characteristic temperatures coincide with our $\mu$SR data astonishingly. We note that the external field applied in the NMR experiments could affect the ground state, and the experimental principles of NMR and $\mu$SR are different, so the results of NMR cannot be compared with $\mu$SR or specific heat directly. As for the thermal conductivity measurements, the dynamic spins should result in a finite residual linear term $k_0/T$ in NaYbSe$_2$.

However, the gapless spinons may be strongly scattered by the quasi-static spins, leading to a negligible $k_0/T$.

We propose here a possible picture of a mixed state of fluctuating short-range up-down ferrimagnetic droplets and QSL. As for the minority quasi-static spins, there are no long-range but at least short-range correlations between them. Since there is AFM interaction on a triangular lattice, the total moment cannot be canceled. Therefore, we take the state as an up-up-down ferrimagnetic state. They are not static like spin glass, and our NMR result suggests that they are still fluctuating. They only take 23% of the total spins, suggesting they distribute in the system like droplets. Such droplets might come from the 5% wrong occupation at Na sites, which is difficult to avoid. Note that such 5% site-mixing (Na-Yb) ratio is obtained by single-crystal X-ray refinements, while the powder neutron scattering refinement suggests an upper limit of 10% of Yb at the Na site. From our powder X-ray refinements (data not shown), this ratio is approximately 11.6%. When it comes to the dynamic spins, they remain disordered and fluctuating down to our base temperature, exactly matching the definition of QSL.

The evidence for spinon Fermi surface was also given by our specific heat measurements. Additionally, there is only < 5.2% residual entropy at zero temperature, also indicating the presence of QSL. The same scenario was also proposed for NaYbS$_2$.

Now it brings us to why other methods like magnetic susceptibility and neutron scattering do not observe such ferrimagnetic droplets. For the magnetic susceptibility technique, it is more sensitive to slower fluctuations, whose limit is about 10$^5$ Hz, while NMR and $\mu$SR are more sensitive to faster fluctuations. Hence a state could be dynamic in magnetic susceptibility measurements, but quasi-static in NMR and $\mu$SR. In inelastic neutron scattering experiments, such fluctuating droplets could also mimic spinon continuum because of its randomness, making it difficult to differentiate.

The ferrimagnetic droplets immersed in a sea of QSLs are illustrated in Figure 5, on which an up-up-down magnetic structure forms within each droplet in accordance with field-induced AFM orders in Yb$^{3+}$ compounds on triangular lattice. It is natural to assume that such a ferrimagnetic ordering state has a slightly higher energy than the QSL state, which allows the nucleation of ferrimagnetic droplet around a defect. Meanwhile, the thermal fluctuation of these magnetic droplets will give rise to the residual entropy. The ratio between the residual entropy and the total magnetic entropy is estimated to be $s_{\text{fl}}/s_{\text{SR}}$, where $s_{\text{fl}}$ is the volume fraction of droplets and each droplet carries m Yb$^{3+}$ ions. It is expected that the size of the fluctuating droplets will be enhanced by an external magnetic field, resulting in a long-ranged AFM order in bulk when the applied magnetic field exceeds some threshold.

Other remaining issues include why an impurity favors a $Q = (1/3, 1/3)$ magnetic droplet in such a QSL and what kind of QSLs can host this possibility. These issues can be addressed by assuming a nested (or nearly nested) spinon Fermi surface as illustrated in Figure 6A, which indicates the instability of the spinon Fermi surface. Thus, a spinon density wave, that is, a magnetic ordered structure of the wave vector $Q = (1/3, 1/3)$ will be energetically favored. An impurity in a
QSL background can be viewed as a hole of local magnetic moments. It will generate a Ruderman-Kittel-Kasuya-Yosida-like (RKKY) oscillation that is characterized by the spin susceptibility function $\chi(\mathbf{q}, \omega = 0)$, provided that there exists a spinon Fermi surface. If the spinon Fermi surface becomes nested, the spin susceptibility $\chi(\mathbf{q}, \omega = 0)$ will diverge at the nesting wave vector $\mathbf{q} = \mathbf{Q}$, resulting in a magnetic ordering in short range, so that the nucleation of a $\mathbf{Q} = (1/3, 1/3)$ magnetic droplet comes into being without the impurity.

However, an unusual spin rotationally invariant system, such a nested Fermi surface, leads to a 1/3-filling spinon band instead of the 1/2-filling spinon band (see Figure 6A) required by the Mottness. This seeming contradiction can be resolved by considering the spin-orbit interaction that comes from the buckling of Se atoms and spoils the spin rotational symmetry. Because of the buckling, triangular layers YbSe$_2$ has a $D_{3d} = D_3 \times I = C_{3v} \times I$, but not $C_3$ rotational symmetry. To solve the problem, we consider an effective Hamiltonian as follows,

$$H_{\text{eff}} = -t \sum_{\langle \mathbf{m},n \rangle,a} e^{\pm \phi_{mn}} c_{\mathbf{m}a}^\dagger c_{\mathbf{n}a},$$

(3.3)

where $(mn)$ denotes a pair of nearest neighbor sites $m$ and $n$ on a triangular lattice, $c_{\mathbf{m}a}$ creates a fermionic spinon, $s = \pm 1$ refers to up (down) spin (or to precise, pseudospin), and $\theta_{mn}$ are phases that give rise to staggered gauge fluxes $\pm \Phi$ on elementary triangles (see Figure 6B). Note that both $C_{3v}$ and the time-reversal symmetry are respected by the Hamiltonian $H_{\text{eff}}$, because the net gauge flux through a unit cell (containing two triangles) is zero. However, the spatial inversion symmetry is broken. When $\Phi = 0$, the spin-rotational symmetry is restored, together with the spatial inversion symmetry, yielding a circular Fermi surface at 1/2-filling. When $\Phi \neq 0$, up and down spinons possess two different Fermi surfaces at 1/2-filling. These two Fermi surfaces can be transformed to each other under a time-reversal or a reflection. In particular, when $\Phi = \pi/2$ (or $-\pi/2$), as illustrated in Figure 6A, the two spinon Fermi surfaces become two perfect triangles, and their edges are nested by a wave vector $\mathbf{Q} = (1/3, 1/3)$ or its $C_3$ rotations. Therefore, we suggest that the QSL depicted by the effective Hamiltonian $H_{\text{eff}}$ in Equation 3 is a promising candidate for the magnetic phase bordering a $\mathbf{Q} = (1/3, 1/3)$ magnetic ordering state in NaYbSe$_2$. The QSL to magnetic ordering phase transition occurs at $\Phi = \pm \pi/2$.

CONCLUSION

We present specific heat, $\mu$SR, NMR, and thermal conductivity measurements on triangular-lattice compound NaYbSe$_2$ single crystals to figure out its ground state. The absence of long-range magnetic order and spin glass is confirmed down to 50 mK. Specific heat, $\mu$SR, and NMR measurements all find a majority of dynamic spins and a minority of quasi-static spins mixed in NaYbSe$_2$, which is further supported by thermal conductivity measurements. The ground state of NaYbSe$_2$ can be regarded as a mixed state with both QSL and fluctuating short-range ferrimagnetic droplets, providing a platform to study how disorder influences the QSL state.

MATERIALS AND METHODS

Sample preparation

High-quality NaYbSe$_2$ single crystals were grown by a modified flux method following Schleid and Lissner. Analytically pure Yb powder, Se powder, and NaCl as flux in a molar ratio of 2:3:1 were sealed in a quartz tube and heated to 950°C for 7 days, followed by a maintaining at 950°C for 7 days. The mixture was slowly cooled down to 600°C at a rate of 50°C per day. In the end, red-black platelets with largest size of 7–8 mm, as shown in the inset of Figure S1, were separated by dipping in water. The large natural surface was determined to be the (001) plane by X-ray diffraction, as illustrated in Figure S1 and no impurity phases were observed, indicating a relatively high crystallization quality.

Magnetic measurements

The magnetic susceptibility measurements were performed in commercial SQUID and the specific heat was measured in the physical property measurement system (PPMS, Quantum Design) by the relaxation method.

$\mu$SR measurements

In a $\mu$SR experiment, a beam of nearly 100% spin polarized muon is implanted into the sample. Muon spin precesses and relaxes because of an inhomogeneous local magnetic field. One can measure the spectra of muon spin polarization, and the relaxation process can reveal the distribution of local field. Besides, muon is extremely sensitive to small fields, which is a powerful technique to check the essence of magnetic order. ZF and LF $\mu$SR measurements were performed down to 88 mK on a modified $\mu$SR spectrometer at ISIS, Rutherford Appleton Laboratory, Chilton, UK. Single crystals of NaYbSe$_2$ were aligned so that the $c$ axis is normal to the sample’s planar surface and parallel to the initial muon spin polarization and mounted onto a silver holder covering a circle area of 1 inch in diameter and 3 mm in thickness. $\mu$SR data were analyzed using the MANTID PROJECT and MUSRFIT software packages. Subtracting the constant background signal due to silver sample holder, ZF muon spin polarization spectra $P(t)$ can be described by the formula $P(t) = P_0 (t) + (1 - f) \exp(-\lambda t)$, where $\lambda$ is the recovery rate 1/T of the nuclear magnetization $M(t)$ is fitted with the function $1 - \frac{MT}{M_0} = I_0 \left[0.1 \exp \left(-\left(\frac{MT}{M_0}\right)^6\right) + 0.9 \exp \left(-\left(\frac{MT}{M_0}\right)^{12}\right)\right]$. The error bars are defined by the least squares method.

Thermal conductivity measurements

The single crystal selected for the thermal conductivity measurements was a rectangular shape of dimensions 5.38 × 1.30 mm$^2$ in the ab plane, with a thickness of 0.04 mm along the c axis. The thermal conductivity was measured in a dilution refrigerator, using a standard four-wire steady-state method with two RuO$_2$ chip thermometers, calibrated in situ against a reference RuO$_2$ thermometer. Magnetic fields were applied along the c axis for specific heat and thermal conductivity measurements and perpendicular to the heat current in the thermal conductivity measurements.

Residual entropy

Assuming each ferrimagnetic droplet carries m spin-1/2 (U$_{m} = 1/2$ local moment) and the fractional volume of the ferrimagnetic droplets is $r$, then the effective spin of each droplet is $3 + s_{3} + \frac{m}{2}$, which gives rise to the upper bound of the residual entropy $R(n l + 2S_n) = R(n l + m/3)$. Thus, the ratio between the residual entropy and the total magnetic entropy at high temperatures has an upper bound of $\frac{R(n l + m/3)}{R(n l + 2S_n)}$.

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AUTHOR CONTRIBUTIONS

L.S. and S.L. planned the project. B.P. synthesized the sample, and characterized the sample with C.J. Z.Z., Y.Yang, and S.C. carried out the µSR experiments with experimental assistance from A.D.H. L.N. carried out the NMR experiments: B.P., J.N., Y.H., E.C., and Y.Yu performed the thermal conductivity measurements. L.S., S.L., T.W., Z.B., B.P., L.N., and J.N. analyzed the data. Y.Z. and J.M. provided the theoretical explanation. L.S., S.L., T.W., Y.Z., X.C., Z.Z., B.P., and L.N. wrote the paper.

DECLARATION OF INTERESTS

The authors declare no competing interests.

DATA AVAILABILITY

All data needed to evaluate the conclusions in the paper are present in the main text or the supplemental information.

SUPPLEMENTAL INFORMATION

It can be found online at https://doi.org/10.1016/j.xinn.2023.100459.

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