

Emergent spin-singlet pairing in the frustrated kagome metal $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$

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The metallic kagome compound $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$ has attracted attention as a candidate platform where geometric frustration and itinerant electrons may cooperate to stabilize a quantum-disordered magnetic ground state. Here, we combine bulk thermodynamic probes, low-noise focused-ion-beam-device transport, and comprehensive ^{55}Mn nuclear magnetic resonance (NMR) measurements to elucidate the low-temperature spin dynamics of this system. The bulk data reveal strongly reduced magnetic entropy, a negative magnetoresistance arising from spin scattering, and field-dependent transport indicates the spin fluctuations, while showing no signatures of long-range magnetic order. NMR provides a direct local view of the correlated Mn moments: the nuclear spin-spin relaxation time T_2 exhibits a pronounced low-temperature enhancement driven by an indirect internuclear coupling through electronic spin fluctuations, whose temperature and distance dependencies point to partially gapped low-energy spin excitations. The spin-lattice relaxation rate T_1^{-1} displays a broad peak near 10 K that may originate from spin-singlet pairing and coincides with the resistivity crossover and a subtle heat-capacity anomaly. Together, our results suggest that $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$ hosts an unconventional correlated state dominated by frustrated, gapped spin dynamics, placing it among the rare metallic kagome systems proximate to a quantum spin liquid.

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I. INTRODUCTION

Quantum spin liquids (QSLs) are exotic states of matter in which geometric frustration and quantum fluctuations prevent spins from developing long-range magnetic order, even at the lowest temperatures [1–6]. The kagome lattice, a planar network of corner-sharing triangles, provides an ideal stage for realizing such states, as vividly demonstrated in insulating compounds with Cu^{2+} (spin-1/2) layers, where fractionalized excitations and continuum spin dynamics have been established [7–12]. In metallic systems with kagome layers, the situation is even more intriguing: flat bands, Dirac dispersions, and strong spin correlations intertwine with itinerant electrons, enabling unconventional charge transport coupled to frustrated magnetism [13]. For kagome metals, such as FeSn and AV_3Sb_5 ($A = \text{Rb}, \text{K}, \text{Cs}$), it has been elucidated how lattice geometry can stabilize novel correlated states, from topological magnetism to charge order [14,15]. The recently reported compound HoAgGe , a distorted kagome material, is claimed to host a spin-ice state arising from frustration of Ising-like Ho moments on the kagome network [16]. A

central challenge, however, remains to identify disorder-free metallic kagome systems in which geometric frustration can stabilize short-range spin-singlet correlations, the metallic analog of the singlet physics often associated with insulating quantum spin liquids [17]. Such singlet formation in a metal is rare, yet theoretically anticipated, motivating the search for kagome metals where frustration suppresses classical magnetism and allows spin-singlet or gapped-fluctuation states to develop.

The kagome metal $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$ has recently been discussed as a candidate for Mn-based QSL [18]. Initial work established its synthesis and crystal structure, together with bulk magnetic and transport properties that hinted at correlated behavior [19]. Magnetic, heat capacity, and neutron scattering measurements established the absence of long-range magnetic order and revealed a markedly reduced Mn moment, consistent with an itinerant magnetic character on the kagome network, whereas subsequent theoretical and spectroscopic works suggested orbital-selective flat bands and concomitant ferromagnetic fluctuations [18–21]. These features naturally raise the question of whether short-range spin singlets or a gapped spin-fluctuation regime may form at low temperature in this metal.

Here, we present a comprehensive investigation of high-quality single crystals, combining thermodynamic measurements with electrical transport and Hall effect on a device cut by focused ion beam (FIB), as well as electron spin resonance (ESR) and a detailed ^{55}Mn nuclear magnetic resonance (NMR) study. Our results demonstrate suppressed magnetic entropy, a field-dependent resistivity crossover coincident with a heat capacity anomaly, negative

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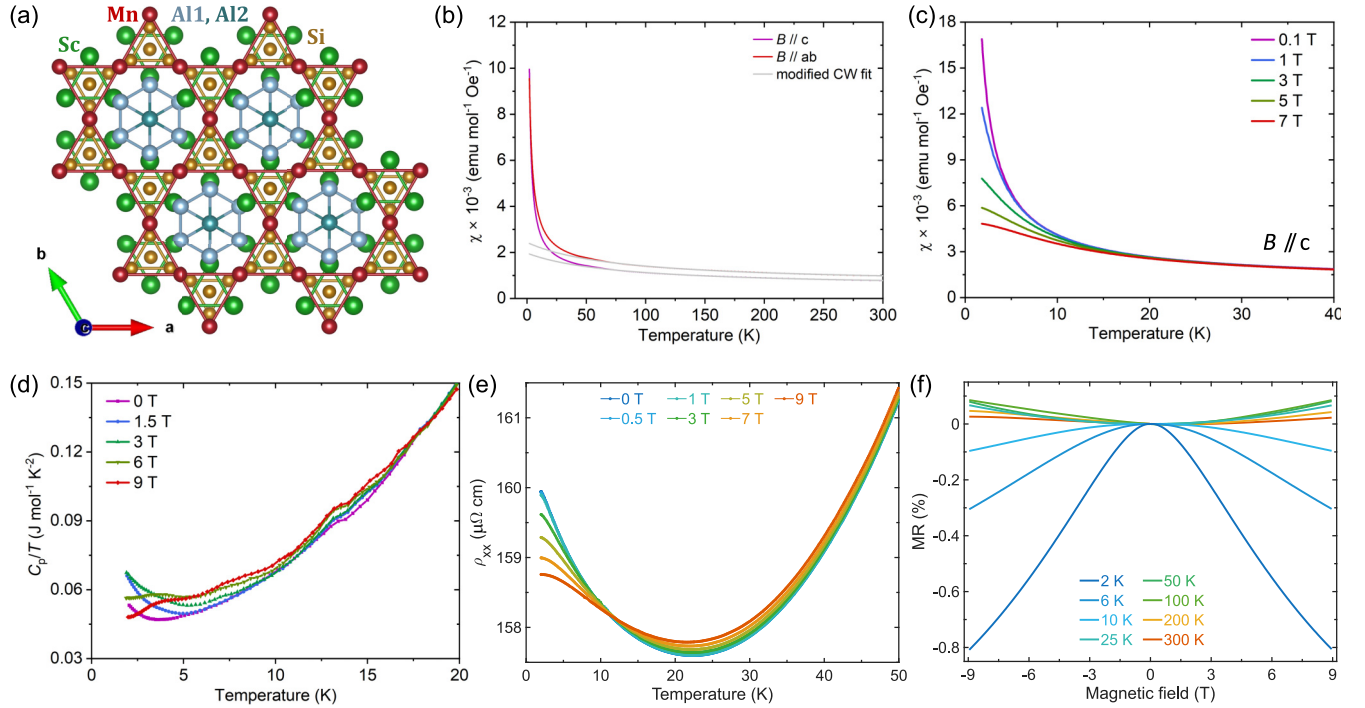


FIG. 1. (a) Crystal structure of $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$. Note that Al1 and Al2 reside on chemically nonequivalent lattice sites yielding different NMR resonance lines with an intensity ratio of 6:1. (b) Temperature-dependent dc susceptibility $\chi(T)$ and modified CW fits (grey lines) with the field along c and ab . (c) Temperature-dependent dc susceptibility $\chi(T)$ at different fields for $B \parallel c$. (d) C_p/T versus T in the presence of various fields. (e) Details of the temperature-dependent longitudinal resistivity (ρ_{xx}) at various fields. See Fig. S4 in the Supplemental Material [22] for the full temperature range. (f) Field dependence of magnetoresistance $\text{MR}(\%) = 100 \times (\rho - \rho(0))/\rho(0)$ at various temperatures for $I \parallel ab$ and $B \parallel c$.

magnetoresistance (MR) driven by spin scattering, and field-dependent transport signatures of strong spin fluctuations. While ESR remains silent, nuclear spins are indirectly coupled via electronic spin fluctuations yielding a unique nuclear spin-spin relaxation below about 50 K besides evidence of a Hebel-Slichter coherence peak implying a temperature-dependent spin-gap and spin-singlet formation. These findings establish $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$ as a compelling metallic kagome system in which geometric frustration and itinerancy cooperate to generate short range singlet physics.

II. RESULTS AND DISCUSSION

$\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$ crystallizes in a hexagonal lattice with $P6_3/mmc$ (No. 194) space group [18]. The crystal structure consists of Mn atoms forming a planar kagome network separated by Sc and Al/Si layers [Fig. 1(a)], providing a geometrically frustrated framework for magnetic interactions. The dc magnetic susceptibility $\chi(T)$ reveals no signature of long-range magnetic order down to 2 K [Figs. 1(b) and 1(c)]. Instead, $\chi(T)$ increases strongly upon cooling, with the divergence most pronounced at low fields and progressively suppressed by the applied field. A modified Curie-Weiss analysis to $\chi(T)$, as shown in Fig. 1(b), yields $\chi_0 = 4.8 \times 10^{-4} \text{ emu mol}^{-1} \text{ Oe}^{-1}$, an effective Mn moment of $\mu_{\text{eff}} = 1.0(1) \mu_B/\text{Mn}$, a Curie-Weiss temperature $\theta_{\text{CW}} = -76(2) \text{ K}$ for $B \parallel c$ and $\chi_0 = 6.1 \times 10^{-4} \text{ emu mol}^{-1} \text{ Oe}^{-1}$, $\mu_{\text{eff}} = 1.0(1) \mu_B/\text{Mn}$, and $\theta_{\text{CW}} = -77(2) \text{ K}$ for $B \parallel ab$. The

temperature-independent susceptibility χ_0 is sizable for both orientations, comparable to previous reports [18,19]. This likely reflects enhanced Pauli or Van Vleck contributions arising from the flat-band-enhanced density of states (DOS). Other factors like tiny amounts of localized impurities and weak ferromagnetic moments cannot entirely be ruled out within the present study. The Curie-Weiss response is essentially isotropic, in agreement with earlier work. The small effective Mn moment indicates that the Mn 3d electrons are strongly hybridized and the magnetism is predominantly itinerant in nature. The negative θ_{CW} indicates dominant antiferromagnetic interactions or correlated magnetism [19,21]. The absence of long-range magnetic ordering despite large θ_{CW} points to the strongly fluctuating character of spin interactions on the Mn kagome lattice. After subtracting the linear $M(B)$ background at 20 K, the isothermal magnetization at 2 K (Fig. S2 in the Supplemental Material [22]) reveals a weak nonlinear contribution that saturates by $\sim 5 \text{ T}$ for both $B \parallel c$ and $B \parallel ab$. The low value of the saturated magnetization signals the presence of fluctuating spins. The nearly isotropic response and absence of hysteresis indicate the polarization of fluctuating short-range correlated Mn moments rather than the onset of long-range order.

The specific heat $C_p(T)$ of $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$ shows no sharp anomaly down to 2 K, ruling out the presence of conventional long-range magnetic order [Figs. 1(d) and S3(a) [22]]. To estimate the magnetic contribution $C_{\text{mag}}(T)$, the lattice contribution is modeled using a combined Debye-Einstein

expression, which is a standard approach for complex intermetallics in the absence of a nonmagnetic analog. Although such a phonon model is necessarily approximate, it provides a reasonable baseline from which to assess the overall scale of magnetic contributions. The resulting magnetic entropy per formula unit, obtained by integrating $C_{\text{mag}}(T)$, remains well below $1 \text{ J mol}^{-1} \text{ K}^{-1}$ up to 50 K, which is less than 1% of the full theoretical value $R \ln(2S + 1)$ expected for localized Mn^{3+} moments ($S = 2$). Regardless of the exact phonon parametrization, only a very small fraction of the expected magnetic entropy is released at low temperatures. Such strong entropy suppression is characteristic of systems where magnetic degrees of freedom are either strongly fluctuating and itinerant. At the lowest temperatures, C_p/T exhibits an upturn that changes characteristically with field and is captured by a Schottky term, consistent with nuclear contributions rather than intrinsic order. Surprisingly, a subtle shoulder appears near $T^* \approx 12 \text{ K}$, as shown in Fig. 1(d), which is not observed previously [19]. This feature, coincident with anomalies in both resistivity and NMR relaxation, which will be discussed later, likely reflects a crossover in the spin excitation spectrum. At low temperatures, the C_p/T versus T^2 data exhibits a linear regime between the Schottky anomaly and the characteristic temperature T^* . A fit in this range gives a linear coefficient $\gamma_e = 40 \text{ mJ mol}^{-1} \text{ K}^{-2}$ [see also Fig. S3(c) in the Supplemental Material [22]], indicative of substantial low-energy electronic correlations, and a phonon coefficient $\beta = 0.276 \text{ mJ mol}^{-1} \text{ K}^{-4}$, corresponding to a Debye temperature $\theta_D \sim 500 \text{ K}$. The coexistence of a finite γ_e with suppressed entropy suggests a correlated metallic state, in which Mn moments remain fluctuating rather than ordering.

Electrical transport was measured exclusively on microstructured devices prepared using FIB technique [see Fig. S4(a) in the Supplemental Material [22]]. The resistivity $\rho(T)$ decreases monotonically upon cooling, consistent with poor metallic behavior at high temperatures. A large residual resistivity ρ_0 , consistent with earlier reports [18,19], likely reflects intrinsic alloy scattering in this multicomponent metallic system. Interestingly, $\rho(T)$ develops an upturn below $\sim 40 \text{ K}$ [Figs. 1(e) and S4(b) [22]]. This low temperature upturn may be due to enhanced scattering from spin fluctuations, which act to impede charge transport in the absence of long-range magnetic order. Application of magnetic fields systematically suppresses the upturn and reveals a characteristic crossover near $T^* \sim 12 \text{ K}$, coincident with the anomaly observed in heat capacity [Fig. 1(e)]. The negative MR seen at 2, 6, 10, and 15 K points to the suppression of spin-disorder scattering by field-induced spin polarization [Fig. 1(f)]. At higher temperatures, the MR changes signs and becomes positive, reflecting the dominance of conventional orbital contributions once magnetic fluctuations are less effective scatterers. Hall effect measurements further support this picture [Fig. S4(c) in the Supplemental Material [22]]. While both the carrier density and mobility remain nearly constant below 40 K, they exhibit pronounced variations above this temperature, consistent with a change in the dominant scattering mechanism [Fig. S4(d) in the Supplemental Material [22]]. The combined evolution of resistivity, magnetoresistance, and Hall response underscores the role of fluctuating Mn moments in controlling transport and highlights the strong interplay between spin correlations and conduction electrons in $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$.

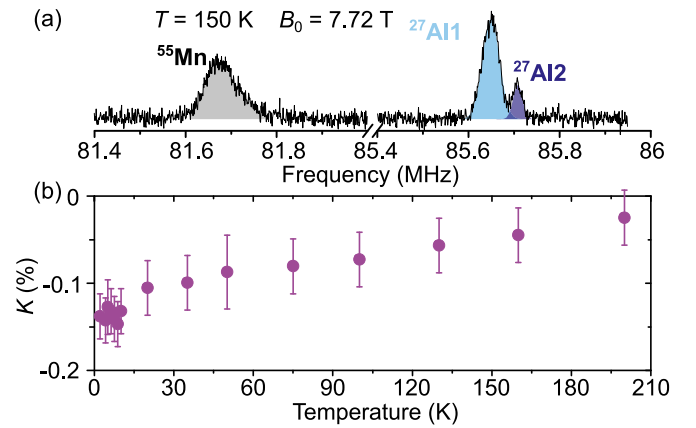


FIG. 2. (a) ^{55}Mn and ^{27}Al NMR spectra of the respective central transitions (CTs) obtained at 7.72 T and 150 K. (b) The ^{55}Mn Knight shift as a function of temperature.

To directly probe these spin dynamics and test their microscopic origin, we employ ^{55}Mn NMR and ESR as local spin probes. ESR measurements were performed at 9.4 GHz (X band), corresponding to a resonance field of the Mn spins at around 0.3 T, assuming $g_{\text{Mn}} = 2$. Interestingly, we found no resonance absorption in the whole temperature range investigated (3–300 K). We suspect that this ESR silence is due to a large resonance-linewidth originating from strong Mn-spin fluctuations which are present in the whole temperature range as indicated by susceptibility and specific heat. The effect of spin fluctuations on the ESR linewidth has been exemplified, for instance, for the weak itinerant ferromagnet MnSi on the basis of Moriya's theory for spin fluctuations in itinerant electron magnets [23,24].

NMR of $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$ has so far only been reported in research articles for ^{27}Al nuclei, while there is no account of ^{55}Mn NMR, even though such measurements offer a unique microscopic probe of the intrinsic spin fluctuations responsible for the correlated behavior in this kagome metal [19,21]. It has to be noted, though, that ^{55}Mn NMR of $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$ is mentioned in conference abstracts of the Spring Meetings of the Physical Society of Japan 2023 and 2024 but we are not aware of a subsequent publication [25,26]. In the current report, we focus on ^{55}Mn —a quadrupole nucleus ($I = 5/2$) of 100% natural abundance (NA)—the central transition (CT) of which is shown in Fig. 2(a) on the left, obtained using selective spin echoes at 7.72 T and 150 K. On the right, the two CTs of Al1 and Al2 ($I = 5/2$ and NA = 100%) are shown, reflecting the two chemically non-equivalent Al sites in the $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$ crystal structure. The resonances were normalized with respect to the stoichiometry, i.e., the Mn CT was divided by 3, while the two Al CTs spectrum was divided by 7. After correction for the transverse relaxation (T_2), the areas under the resonance lines (signal intensity) of Mn and Al match perfectly (gray vs blue), while the light blue Al1 signal intensity is 6 times that of the dark blue Al2. This finding corroborates the sample's stoichiometry and crystal structure, cf. also Ding *et al.* [21].

Mn nuclei are subject to a large and temperature-independent quadrupolar interaction of more than 5 MHz in terms of the quadrupole splitting frequency. Such a large

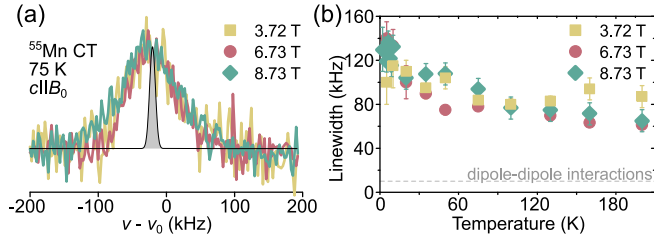


FIG. 3. (a) The ^{55}Mn central transition (CT) in frequency units (kHz) at 75 K for three different magnetic fields applied parallel to the c axis. The gray line shows the approximate line broadening as expected from direct dipole-dipole interactions (~ 10 kHz) [29]. (b) Temperature dependent linewidth (full width at half maximum) in frequency units of the ^{55}Mn CT for three different fields.

quadrupole coupling gives rise to a substantial field dependent shift from second order quadrupole effects ($\propto B_0^{-1}$). We thus measured the temperature dependent ^{55}Mn resonance frequency for several external fields to extract the Knight shift. The procedure is documented in the Supplemental Material [22]. The results are shown in Fig. 2(b). Similar to Al NMR reported in the literature [19,21], the Mn Knight shift shows a weak and approximately linear temperature dependence. Clearly, there is no unusual behavior at low temperatures for any measured field as observed in the magnetic susceptibility [Figs. 1(b) and 1(c)]. This implies, as already suggested by Ding *et al.* [21], that the steep upturn of the magnetic sus-

ceptibility for low temperatures and fields may reflect a small number of localized moments from impurities that remain invisible for the majority of nuclear spins.

In Fig. 3(a), we show the Mn CT for three different fields. The spectra are provided in frequency units (kHz), the respective carrier frequencies of the measurements have been subtracted. The significance of the spectra in Fig. 3(a) lies in their large linewidths (80–90 kHz). The linewidth increases with decreasing temperatures [Fig. 3(b)] and remains essentially independent of the external field over the full temperature range. Field independent line broadening cannot be assigned to defects and corresponding distributions of the magnetic ($\propto B_0$) and the 2nd order electric quadrupole shift ($\propto B_0^{-1}$). Typically, field-independent broadening stems from direct dipole-dipole interactions, which, however, are much too small to account for the observed broadening. Therefore, nuclei must be coupled indirectly via electronic excitations [27,28]. Through such an indirect internuclear coupling, contributions from neighboring nuclear magnetic moments to the local magnetic field seen by a nucleus are significantly amplified, yielding the observed broadening.

Consequently, the Mn resonance is governed by a substantial inhomogeneous broadening due to the fields of near neighbors, including ^{45}Sc (natural abundance $NA = 100\%$), ^{27}Al ($NA = 100\%$), and other Mn nuclei (less so ^{29}Si due to the low $NA < 5\%$). This allows us to limit the number of excited nuclei during a measurement. The pulse in an NMR experiment acts as a band pass filter with a narrow frequency

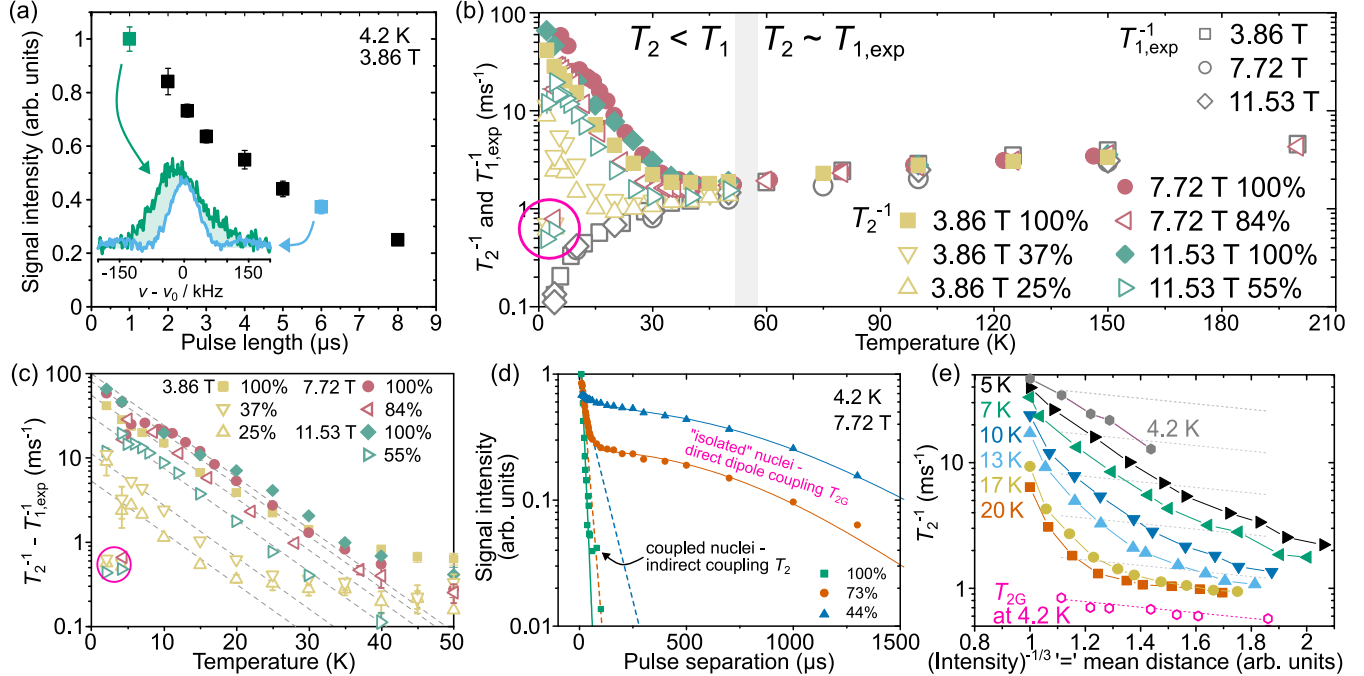


FIG. 4. (a) Signal intensity (T_2 corrected) for various pulse lengths (excitation band widths) and two example resonances for short (1 μs) and long (6 μs) pulses. (b) The spin-spin relaxation rate (T_2^{-1}) and inverse CT lifetime ($T_{1,\text{exp}}^{-1}$) as functions of temperature for the three external fields 3.86 T, 7.72 T, and 11.53 T. The grey bar separates the two temperature regimes (see text for details). The very small relaxation rates at the lowest temperatures (pink circle) denote isolated spins that decay via direct dipolar couplings. (c) Spin-spin relaxation rate after subtraction of the corresponding CT lifetime. Grey parallel lines are guides to the eye, corresponding to an exponential function with $\tilde{T} = 7.1$ K. (d) T_2 measurement (signal intensity as function of pulse delay) for 4.2 K, 7.73 T, and various excitation conditions (100%, 73%, and 44% excited nuclei). Solid lines are fittings using a sum of a single exponential (T_2) and a Gaussian decay (T_{2G}). (e) The spin-spin relaxation rates as a function of the mean distance between Mn nuclei for various temperatures.

window inversely proportional to the pulse length [30]. As shown in Fig. 4(a), by increasing the exciting pulses' lengths, we systematically narrow down the observed resonance line and thus, the corresponding signal intensity (area under the curve) which is proportional to the number of excited nuclei. As will be shown next, this severely affects the low temperature spin-spin relaxation (T_2) of Mn nuclei, the timescale of phase coherence of the nuclear precession

In Fig. 4(b), we present (T_2^{-1}) of ^{55}Mn nuclei for a variety of external fields and excitation conditions. Also shown in the figure and denoted by the gray open symbols is the temperature-dependent inverse CT lifetime $T_{1,\text{exp}}^{-1}$, i.e., the effective recovery rate which is directly related to the system's spin-lattice relaxation rate T_1^{-1} [31]. We identify two regimes. For temperatures above about 50 K, T_2^{-1} equals approximately $T_{1,\text{exp}}^{-1}$. This implies that the very short lifetime sets a natural limit for the phase coherence of the nuclear precession in which case one expects that $T_2 = 2T_1$, or, as related to the ESR silence, fast and isotropic spin fluctuations govern both, spin-spin as well as spin-lattice relaxation. For $T < 50$ K, the T_2^{-1} values deviate strongly from $T_{1,\text{exp}}^{-1}$, implying the spin-spin relaxation to be driven by a different mechanism. An effective spin-echo decay (T_2) is observed if coupled nuclei are flipped simultaneously during a measurement, because this will change their mutual local field causing a loss of coherence. Therefore, a strong indirect coupling as reflected in the extensive line broadening will cause fast spin-spin relaxation (as in cuprates [32,33]). The temperature dependence implies that the indirect coupling requires virtual transition across a very small gap and is thus of a Bloembergen-Rowland rather than a Ruderman-Kittel-Kasuya-Yoshida-type, which involves transitions between degenerate bands [27,28]. Thermal excitations interfere with the virtual transitions and lead to the exponential suppression of T_2^{-1} with temperature as can be seen in Fig. 4(c). We find $\tilde{T} \approx 7.1$ K as the characteristic temperature from a simple exponential fit, corresponding to 0.61 meV (148 GHz) for the spin excitations.

We repeated the T_2 measurements under various excitation conditions. While the solid symbols in Figs. 4(b) and 4(c) represent measurements where the full resonance line was excited with sufficiently short pulses (broad excitation width), i.e., 100% of the CT nuclei, the open triangles denote measurements with longer pulses/reduced bandwidths and thus fewer excited nuclei. The effect on T_2^{-1} is substantial. While the external field seems not to have any significant impact on the low temperature T_2^{-1} behavior [solid symbols in Figs. 4(b) and 4(c)], reducing the number of excited nuclei shifts the upturn of T_2^{-1} systematically to lower temperatures (open triangles), from initially ~ 50 K for 100% of the nuclei to less than 25 K for about 25% (yellow triangles). Reducing the number of excited nuclei during an experiment for a given temperature [triangles in Figs. 4(b) and 4(c)] means to look at an increasingly dilute spin system with the mean distance between excited spins increasing, because any fraction of excited nuclei will fill the same volume. And since the underlying electronic correlation decays rapidly in real-space, the corresponding internuclear coupling is increasingly hampered, yielding T_2^{-1} values to fall as the spin system is further diluted. In other words, by reducing the number of excited nuclei, we gradually decouple them from each other which is reflected in

a longer spin-spin relaxation. The fact that we can manipulate the spin-echo decay by changing the excitation width, i.e., slowing down an otherwise rapid relaxation, proves that we observe true spin-spin relaxation with the driving mechanism being a strong coupling among the nuclear spins. The only difference to the ordinary behavior is the indirect coupling instead of dipole-dipole interactions. This finding challenges the prevailing argument of ferromagnetic fluctuations to be the source for the spin-echo decay as suggested previously [19,21]. The reason is that an external source like electronic spins cannot be manipulated by a weak rf field as applied during an NMR experiment. In particular, the amplitude at a nucleus of interest or the correlation time of the corresponding field fluctuations that can give rise to transverse relaxation [34] do not depend on the rf-pulse length/power, yielding spin-echo decay independent of the NMR excitation conditions.

At the lowest temperatures (2.1 and 4.2 K), up to 25–30% of the Mn nuclei can even be entirely decoupled, yielding the very small decay rates highlighted by pink circles in Figs. 4(b) and 4(c) (see also Supplemental Material [22]). This slow decay [Gaussian T_{2G} in Fig. 4(d)] stems from direct dipole-dipole interactions that are weak but longer ranging ($\sim r^{-3}$) than the indirect coupling. This observation proves the indirect coupling to decay very rapidly in real space. To track the distance dependence of the indirect coupling, we conducted systematic temperature and excitation dependent measurements and plot T_2^{-1} vs. the inverse cube root signal intensity, cf. Fig. 4(e). The latter is related to the mean distance between coupling partners for an isotropic system in three dimensions. At higher temperatures, the decay rapidly levels off due to the lifetime of the nucleus and its (unlike) neighbors offering efficient relaxation channels. At low temperatures these contributions freeze-out, leaving the distance dependence to reflect only the indirect coupling. The decay is much steeper than that of the dipolar component at 4.2 K (pink data points and gray dashed lines) and together with the occurrence of decoupled nuclei implies a short range, exponential distance as expected for gapped spin fluctuations.

Finally, we address the spin-lattice relaxation (T_1) of ^{55}Mn nuclei. In metallic systems, one typically analyzes the DOS to derive the temperature dependence of the corresponding spin-lattice relaxation. A peak in the Mn DOS as expected from a flat band near the Fermi level gives rise to a slight downturn in the corresponding $(T_1T)^{-1}$ for high temperatures, but is too far from E_0 (~ -80 meV) to affect the low temperature behavior of the spin-lattice relaxation, as shown in Fig. 5 by the blue line. Instead, we observe a clear peak in $(T_1T)^{-1}$ near 10 K, typically suggesting a magnetic phase transition in magnetic materials, as in the case of the itinerant ferromagnet ZrZn_2 [35], but it lacks the inverse field dependence. The spin-lattice relaxation data shown by Ihara *et al.* shows a similar peak around 10 K and an even more pronounced drop in $(T_1T)^{-1}$ below 10 K [25]. In the light of strong electronic correlations and the absence of long-range order from bulk measurements, this peak could indicate spin-singlet pairing, a situation similar to the s -wave pairing in superconductors, where the opening of the superconducting gap yields the Hebel-Slichter peak in the spin-lattice relaxation rate [36]. Ihara *et al.* also concluded a

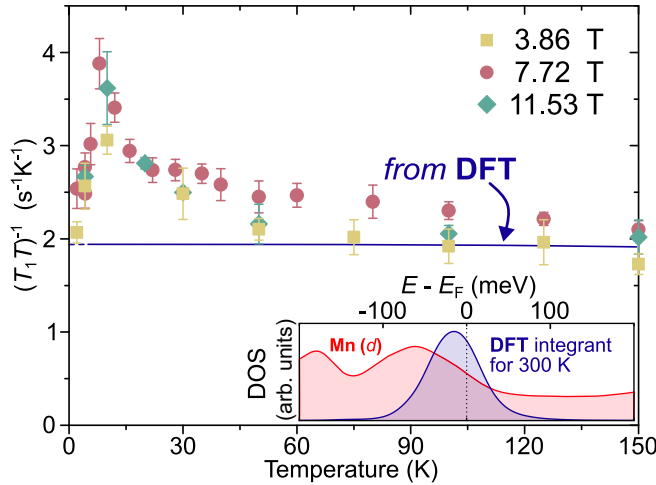


FIG. 5. The spin-lattice relaxation rate divided by the temperature $(T_1T)^{-1}$ as a function of temperature for three different magnetic fields, including the calculated behavior from the DOS (blue). Inset: The density of states (DOS) for Mn d orbitals (red) near the Fermi level. To extract the temperature dependence of the spin-lattice relaxation, we calculated $T_1^{-1} \propto \int f(E - \mu_c)[1 - f(E - \mu_c)]D(E)^2dE$, using the Fermi function $f(E - \mu_c)$ for various temperatures. The blue line denotes the integrant for 300 K, representing the range of thermally excited states available for nuclear relaxation.

band-dependent gap from their site-selective relaxation data [26]. This scenario is further supported by the small gap implied by the temperature and distance dependence of T_2^{-1} due to an indirect nuclear coupling of Bloembergen-Rowland type. Our peak, however, clearly differs from the sharp feature in superconductors, suggesting a continuous singlet formation spread over a large temperature range rather than a well-defined phase transition. Similar arguments, e.g., the gradual emergence of spin-singlets with spatially varying gaps or spin-singlet formation associated with nodal gaps as those found in anisotropic superconductors have repeatedly been discussed in the context of quantum spin liquids and anomalies in the corresponding spin-lattice relaxation rates [17,37]. Notably, the T_1^{-1} peak coincides with the subtle anomaly in the heat capacity at $T^* \sim 12$ K and with the resistivity crossover, reinforcing a unified picture of evolving singletlike correlations at low temperature. The low-temperature part of the peak in $(T_1T)^{-1}$ points to a constant value rather than displaying an exponential behavior as observed in gapped systems. This may be related to the gradual emergence of spin-singlets, an anisotropic gap, or an underlying metallic contribution from the considerable density of states. A detailed investigation of this low-temperature region should be the subject of future studies.

III. CONCLUSIONS

In summary, we present a comprehensive investigation of the kagome metal $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$ through bulk thermodynamic and transport measurements together with local spin probes. The NMR spin-spin relaxation T_2 reveals a pronounced low-temperature enhancement arising from an indirect internuclear coupling via electronic spin fluctuations, whose temperature and spatial dependence indicate that

gapped spin excitations become relevant below ~ 50 K. A corresponding peak in the spin-lattice relaxation rate around 10 K, coincident with subtle anomalies in heat capacity and resistivity, points to the formation of short range spin-singlet correlations in the absence of long-range magnetic order. Rather than undergoing a conventional phase transition, $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$ thus enters a quantum-disordered regime dominated by frustrated, slowly fluctuating spin singlets. Our results highlight the need for a theoretical framework for the indirect nuclear coupling in the presence of strong spin fluctuations and demonstrate that T_2 relaxation may provide a sensitive tool to uncover hidden magnetic dynamics in correlated quantum materials.

IV. METHODS

Crystal synthesis and characterization: Single crystals of $\text{Sc}_3\text{Mn}_3\text{Al}_7\text{Si}_5$ were successfully grown using the Al-self flux method with a ratio of Sc:Mn:Al:Si $\sim 3:3:30:5$. All the elements were of high purity and, thus, mm-size single crystals were grown. They were found to be stable in the air and moisture. The magnetization measurements were performed using a Quantum Design MPMS-3. Resistivity, magnetoresistance, and Hall measurements were carried out on a micro device sculptured by focused ion beam lithography with a Thermo Scientific Helios 5 CX. The device contained eight contacts, and all were used at the same time. The size of the Hall bar was $76 \times 9.8 \times 4.7 \mu\text{m}^3$. The heat capacity was measured using the HC option of a quantum design PPMS.

NMR: Measurements were carried out on JANIS sweepable 9 T and 14 T magnets using a TECMAG APOLLO NMR console. Two flat ($\sim 1 \times 1 \times 0.3 \text{ mm}^3$) and hexagonal samples were cut from a large single crystal and placed in a high (S1: eight turn) and a low (S2: 20 turns) frequency coil. All experiments were carried out with $c \parallel B_0$. Echo experiments ($\pi/2 - \tau - \pi$) were used to excite individual transitions, their τ dependence was studied for transverse (T_2) and their recovery following a $\pi/2$ -saturation pulse for longitudinal relaxation (T_1). The shift was obtained by referencing the respective spin species via the rf-coil's ^{63}Cu resonance line and the second reference method [38].

ESR: Electron spin resonance studies on platelet and needle shaped crystals (1.25 mg and 11.5 mg) were performed at X-band frequency, 9.4 GHz, using a standard continuous-wave spectrometer for temperatures between 3.5 K and 300 K.

Numerical calculations: The simulated results were obtained by using *ab initio* calculations in the framework of density-functional theory (DFT), as implemented in the program VASP [39]. In this code, augmented plane waves are used as a basis set together with pseudopotentials. To describe the exchange-correlation potential, the generalized-gradient approximation [40] was used.

The self-consistent calculations were carried out on a $20 \times 20 \times 10$ k mesh. Convergence for total energy and DOS was carefully checked. For the DOS calculations, a k mesh of $60 \times 60 \times 30$ was used.

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P.Y. grew the single crystals. P.Y. and A.K.S. performed all bulk thermodynamic measurements (magnetization and heat capacity), and carried out the magnetotransport experiments with assistance from R.K., W.S., and C.S., and analyzed the corresponding data. A.K.S. did the FIB device

fabrication for transport measurements. R.G. conducted and analyzed NMR experiments with assistance from M.B., J.S. carried out ESR experiments, J.N. carried out DFT calculations, C.F. lead the project, P.Y. planned, executed, and wrote the bulk properties section of the manuscript with the help of A.K.S. R.G. prepared the overall manuscript with contributions from all authors and substantial input from P.Y.

The authors declare no competing interests.

DATA AVAILABILITY

The data that support the findings of this article are openly available [41].

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