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Magnetic transition, long-range order, and moment fluctuations in the pyrochlore iridate $\text{Eu}_2\text{Ir}_2\text{O}_7$

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Muon spin rotation and relaxation experiments in the pyrochlore iridate $\text{Eu}_2\text{Ir}_2\text{O}_7$ yield a well-defined muon spin precession frequency below the metal-insulator/antiferromagnetic transition temperature $T_M = 120$ K, indicative of long-range commensurate magnetic order and thus ruling out quantum spin liquid and spin-glass-like ground states. The dynamic muon spin relaxation rate is temperature-independent between 2 K and $\sim T_M$ and yields an anomalously long Ir^{4+} spin correlation time, suggesting a singular density of low-lying spin excitations. Similar behavior is found in other pyrochlores and geometrically frustrated systems, but also in the unfrustrated iridate BaIrO_3 . $\text{Eu}_2\text{Ir}_2\text{O}_7$ may be only weakly frustrated; if so, the singularity might be associated with the small-gap insulating state rather than frustration.

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Geometrical frustration of collinear near-neighbor spin interactions is a consequence of the corner-shared tetrahedral structure of pyrochlore transition-metal oxides, and has motivated considerable study of these materials.¹ Compounds in the pyrochlore iridate family $R_2\text{Ir}_2\text{O}_7$, where R is a trivalent lanthanide, are particularly interesting: Ir^{4+} ($5d^5$) is expected to be a low-spin $S = 1/2$ ion, and the behavior of the Ir-derived conduction band is unusual.² For $R = \text{Pr}$, Nd , Sm , and Eu these compounds exhibit metallic behavior at high temperatures, while for $R = \text{Gd}$, Tb , Dy , Ho , Er , Yb , and Y they are semiconducting. This crossover was attributed to reduction of the width of the Ir^{4+} -derived band as the R ionic radius decreases across the rare-earth series.³

In early studies⁴ spin-glass-like ordering was reported for $R = \text{Y}$, Lu , Sm , and Eu on the basis of bifurcation of field-cooled (FC) and zero-field-cooled (ZFC) magnetizations and little or no specific heat anomaly at a transition temperature T_M . ^{151}Eu Mössbauer studies of $\text{Eu}_2\text{Ir}_2\text{O}_7$ ^{4,5} found no long-range magnetic ordering down to 4.2 K. Subsequently, metal-insulator (MI) transitions at T_M with small specific heat anomalies were reported² for $R = \text{Nd}$, Sm , and Eu , and an exotic chiral spin-liquid metallic ground state⁶ was found in $\text{Pr}_2\text{Ir}_2\text{O}_7$. The MI transitions were attributed to Ir^{4+} $5d$ electrons, with complex antiferromagnetic (AFM) ordering.

Y^{3+} and Lu^{3+} are nonmagnetic, as is Eu^{3+} in the Hund's-rule ground state $J = 0$ ($L = S$),^{2,3} so that only Ir^{4+} $5d$ electrons contribute to magnetism in these compounds.⁴ Magnetic ordering of localized Ir^{4+} ions has been observed in a number of insulating iridates outside the pyrochlore family⁷ and is quite anomalous, because overlap of the large Ir^{4+} wave functions should result in metallic conduction via Ir-derived bands. In the case of (unfrustrated) Sr_2IrO_4 a detailed treatment⁸ involving strong spin-orbit coupling leads to the possibility of a Mott transition, however, and suggests an effective angular momentum $J_{\text{eff}} = 1/2$. Alternatively, a Slater transition, as found in the pyrochlore $\text{Cd}_2\text{Os}_2\text{O}_7$,⁹ is suggested by the second-order nature of the transition.

Thus $\text{Eu}_2\text{Ir}_2\text{O}_7$ is a potential example of a geometrically frustrated system with “spin” = $1/2$, and as such is of considerable fundamental interest.¹ This Rapid Communication reports results of muon spin rotation and relaxation (μSR) experiments¹⁰ on a polycrystalline sample of this compound. A well-defined muon-spin precession frequency is observed below T_M , indicating a uniform internal field and thus ruling out significant disorder; the magnetic order is commensurate and long-ranged. The dynamic muon-spin relaxation rate λ_d reflects anomalously slow spin fluctuations and remains constant to low temperatures. We speculate that this behavior might not be due solely to geometrical frustration, but may signal new low-lying spin excitations associated with a small-gap insulating state. The data show no critical slowing down of magnetic fluctuations as $T \rightarrow T_M$ from above, suggesting a mean-field-like transition.

Polycrystalline samples of $\text{Eu}_2\text{Ir}_2\text{O}_7$ were fabricated using a solid-state reaction technique.¹¹ dc magnetization data (not shown) are consistent with previous results.^{4,12} μSR experiments were carried out at the M20 beam line at TRIUMF, Vancouver, Canada, using standard time-differential μSR .¹⁰ A weak (25-Oe) magnetic field was applied parallel to the initial muon polarization, to decouple¹³ muon spins from nuclear dipolar fields in the paramagnetic state. Data were taken in a ^4He gas-flow cryostat over the temperature range 2–200 K.

Representative early-time asymmetry (signal amplitude) data $A(t)$ are shown in Fig. 1. Damped oscillations are observed below 120 K, due to precession of the muon spins in a quasistatic¹⁴ component $\langle \mathbf{B}_{\text{loc}} \rangle$ of the local field \mathbf{B}_{loc} at muon sites. This confirms the magnetic transition found from the dc magnetization measurements. The oscillation is weakly damped except for the initial half cycle, indicating that $\langle \mathbf{B}_{\text{loc}} \rangle$ is relatively homogeneous. The late-time asymmetry data (not shown) exhibit exponential relaxation, due solely to dynamic (thermal) fluctuations of \mathbf{B}_{loc} .¹³ This relaxation is much slower than the oscillation damping rate, indicating that the latter reflects a quasistatic distribution of $\langle \mathbf{B}_{\text{loc}} \rangle$.

The data were fit using the two-component asymmetry function

$$A(t) = A_s \exp[-(\Lambda_s t)^K] \cos(\omega_\mu t + \theta) + A_d \exp(-\lambda_d t). \quad (1)$$

The subscripts s and d denote (quasi)static and dynamic components, respectively. The first term models the damped oscillation, with frequency ω_μ and spectrometer-dependent initial phase θ . Neither simple exponential damping nor a Bessel function (expected for an incommensurate spin density wave) gave good fits; the phenomenological stretched-exponential damping form of Eq. (1) was used instead, with relaxation rate Λ_s and stretching power $K < 1$. The second term describes the late-time dynamic relaxation, which was well fit by a single exponential with rate λ_d .

The results of these fits are shown in Fig. 1. The data yield a single well-defined frequency (as does the Fourier transform, not shown), consistent with a commensurate magnetic structure and only one muon stopping site. The total initial asymmetry $A(0) = A_s + A_d$ was found to be ≈ 0.21 independent of temperature and applied field.

The temperature dependence of $\omega_\mu/2\pi$ and Λ_s from the fits are shown in Figs. 2(a) and 2(b), respectively. The abrupt onset of ω_μ and hence $\langle \mathbf{B}_{\text{loc}} \rangle$ below 120 K indicates a magnetic transition at this temperature. At $T = 2$ K $\omega_\mu/2\pi = 13.32(3)$ MHz, corresponding to $\langle B_{\text{loc}} \rangle = \omega_\mu/\gamma_\mu = 987(2)$ G. A rough estimate of the static Ir^{4+} moment μ_{Ir} is given by equating this value to the internal field $4\pi\mu_{\text{Ir}}/v_{\text{Ir}}$ of a uniform Ir^{4+} magnetization, where v_{Ir} is the volume per Ir ion. This yields $\mu_{\text{Ir}} \approx 1.1\mu_B$, of the order of the moment expected for $J_{\text{eff}} = 1/2$.⁸ The estimate is very crude, however, because neither the Ir^{4+} magnetic structure nor the muon stopping site is known.

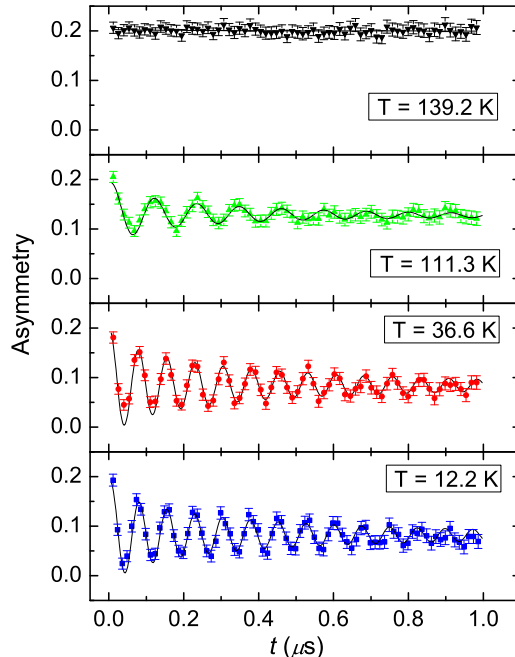


FIG. 1. (Color online) Representative early-time asymmetry data $A(t)$ in $\text{Eu}_2\text{Ir}_2\text{O}_7$, longitudinal field = 25 Oe. Solid curves: fits using Eq. (1).

As shown in the inset to Fig. 2(a), the late-time fraction $\eta_d = A_d/(A_s + A_d)$ approaches 1 as $T \rightarrow T_M$ from below. This is due to the disappearance of $\langle \mathbf{B}_{\text{loc}} \rangle$, and is consistent with the behavior of $\omega_\mu(T)$. At 2 K $\eta_d = 0.39(1)$, close to the value $1/3$ expected from a randomly-oriented $\langle \mathbf{B}_{\text{loc}} \rangle$.¹³ The increase of η_d as $T \rightarrow T_M$ is smooth rather than abrupt, suggesting a distribution of transition temperatures in the sample.

The temperature dependence of Λ_s is given in Fig. 2(b).¹⁵ The cusp at $\sim T_M$ is probably an artifact of the distribution of T_M noted above rather than a critical divergence, since as discussed below there is no sign of critical slowing down in the dynamic relaxation rate λ_d . The fractional width Λ_s/ω_μ of the spontaneous field distribution, also plotted in Fig. 2(b), is small (0.05–0.07) at low temperatures and then increases rapidly as $T \rightarrow T_M$. Thus the local field is nearly uniform except in the neighborhood of T_M ; this, like the behavior of η_d noted above, suggests a distribution of T_M .

The stretching power K for the quasistatic damping, shown in the inset of Fig. 2(b), parameterizes the shape of the distribution of $\langle \mathbf{B}_{\text{loc}} \rangle$: for small K the wings of the distribution become more prominent.¹⁶ The value of K is temperature-independent (~ 0.55) at low temperatures and increases as $T \rightarrow T_M$.

The simple exponential form of the late-time relaxation data indicates that the dynamic muon spin relaxation, like $\langle B_{\text{loc}} \rangle$ (but unlike T_M), is homogeneous. The temperature dependence of the dynamic relaxation rate λ_d is given in Fig. 3. We note two features: (i) $\lambda_d = 0.029(3) \mu\text{s}^{-1}$ is constant below ~ 100 K, and (ii) with decreasing temperature there is an unusual step-like increase in λ_d below T_M but no sign of the paramagnetic-state divergence that is often found in frustrated and unfrustrated magnets^{17–20} due to critical slowing down of dynamic fluctuations. This absence suggests a mean-field-like transition.

The relation between dynamic muon relaxation and the moment fluctuations that cause it is generally complex. Limiting cases are (A) quasistatic (slow) fluctuations of $\langle \mathbf{B}_{\text{loc}}(t) \rangle$ with zero long-time average, where the relaxation time is essentially the correlation time of $\langle \mathbf{B}_{\text{loc}}(t) \rangle$,¹³ and (B) fluctuations $\delta \mathbf{B}_{\text{loc}}$ about a nonzero static $\langle \mathbf{B}_{\text{loc}} \rangle$, i.e., $\mathbf{B}_{\text{loc}}(t) = \langle \mathbf{B}_{\text{loc}} \rangle + \delta \mathbf{B}_{\text{loc}}(t)$;²¹ here the relaxation rate depends on the magnitude and stochastic properties of $\delta \mathbf{B}_{\text{loc}}(t)$. Case A describes dynamic relaxation in a paramagnet with extremely slow spin dynamics, and yields a fluctuation rate $\sim \lambda_d \approx 3 \times 10^4 \text{ s}^{-1}$. The data cannot rule this scenario out in $\text{Eu}_2\text{Ir}_2\text{O}_7$ but it seems quite unlikely, given the phase-transition-like behavior of the muon spin precession frequency (Fig. 2) and the fact that a kilohertz fluctuation rate would be many orders of magnitude lower than any other frequency in the system. We therefore assume Case B in further discussion of the dynamic relaxation.

In the motional narrowing limit $\omega_f \tau_c \ll 1$ $\lambda_d \approx \omega_f^2 \tau_c$, where $\omega_f = \delta B_{\text{loc}}/\gamma_\mu$ is the fluctuating field amplitude in frequency units and τ_c is the correlation time of the fluctuations. Assuming a maximum ω_f of the order of the full

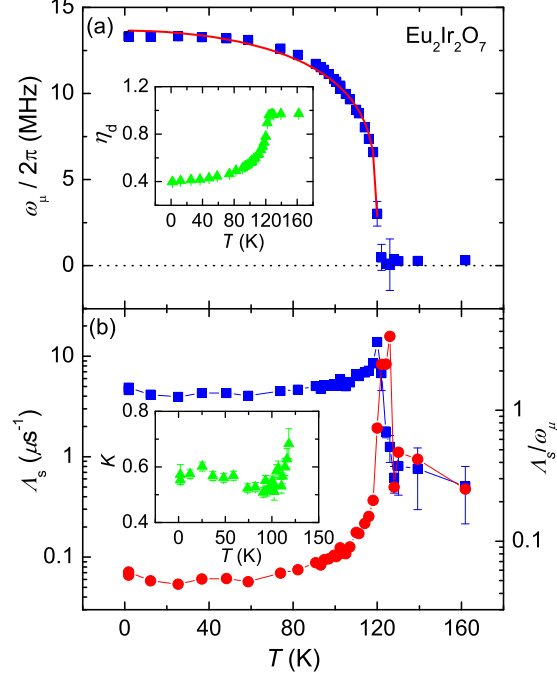


FIG. 2. (Color online) (a) Temperature dependence of muon spin precession frequency $\omega_\mu/2\pi$ in $\text{Eu}_2\text{Ir}_2\text{O}_7$. Inset: late-time fraction η_d . The curve is a guide to the eye. (b) Temperature dependence of quasistatic muon spin relaxation rate Λ_s (squares, left axis) and fractional width of field distribution Λ_s/ω_μ (circles, right axis). Inset: stretching power K .

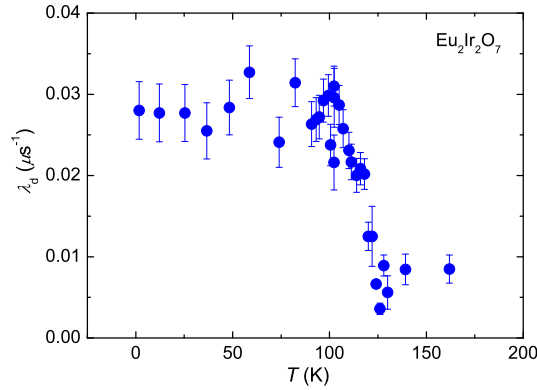


FIG. 3. (Color online) Temperature dependence of the dynamic muon spin relaxation rate λ_d in $\text{Eu}_2\text{Ir}_2\text{O}_7$.

quasistatic field in frequency units ($\omega_f \lesssim \omega_\mu \approx 8.5 \times 10^7 \text{ s}^{-1}$), this yields an upper bound $\tau_c^{-1} \lesssim 2.5 \times 10^{11} \text{ s}^{-1}$, or $\hbar/k_B\tau_c \lesssim 2 \text{ K}$. In ordinary antiferromagnets $\hbar/k_B\tau_c$ is of the order of the Néel temperature T_N for $T \lesssim T_N$.²² For $\text{Eu}_2\text{Ir}_2\text{O}_7$, with $T_N = T_M = 120 \text{ K}$, τ_c is therefore at least two orders of magnitude longer than expected.

The combination of a well-defined muon spin precession frequency (Fig. 1), i.e., homogeneous magnetic order, and the persistence of λ_d to low temperatures (Fig. 3) is unexpected. In conventional ordered magnets nuclear or muon spin relaxation below the ordering temperature is due to thermal spin-wave excitations, and λ_d decreases with decreasing temperature as the thermal population of such excitations decreases. Such a conventional scenario seems to be ruled out in $\text{Eu}_2\text{Ir}_2\text{O}_7$.

Persistent low-temperature muon spin relaxation is observed in a number of geometrically frustrated systems.^{18,23–26} It indicates an enormously enhanced and possibly singular density of low-lying excitations, but is not well understood.

In compounds containing non-Kramers rare-earth ions with nonmagnetic crystal-field ground states, fluctuations of hyperfine-enhanced nuclear magnetism can couple to muon spins and lead to persistent relaxation.²⁷ This mechanism requires rare-earth ions with magnetic Hund's-rule ground states. A similar effect is associated with the low-lying Eu^{3+} spin-orbit-split $J \geq 1$ multiplets; this, however, results in reduction rather than enhancement of Eu nuclear moments.^{28,29} The persistent spin dynamics in $\text{Eu}_2\text{Ir}_2\text{O}_7$ must therefore be electronic in origin and associated with Ir^{4+} magnetism.

The relatively high transition temperature of $\text{Eu}_2\text{Ir}_2\text{O}_7$ suggests that the AFM exchange constant is not much larger than T_M , in which case $\text{Eu}_2\text{Ir}_2\text{O}_7$ is a weakly frustrated material.³⁰ Noting that the unfrustrated iridate BaIrO_3 also exhibits persistent muon spin relaxation,³¹ we consider the possibility that frustration may not be the primary cause of persistent relaxation in $\text{Eu}_2\text{Ir}_2\text{O}_7$ and we look for another mechanism.

In iridate compounds, frustrated or unfrustrated, the large Ir $5d$ wave functions are expected to weaken the on-site repulsion relative to the width of the $5d$ conduction band. If an AFM state associated with a metal-insulator transition is nevertheless retained (perhaps because of strong spin-orbit coupling⁸) but the electrons are not well localized, the gap energy Δ_g can be comparable to $k_B T_M$. The resistivity of single-crystal $\text{Eu}_2\text{Ir}_2\text{O}_7$ in fact yields a maximum gap value $\approx 10 \text{ meV} \approx k_B T_M$.¹² We speculate that charge fluctuations³² and accompanying spin fluctuations over this gap might be involved in the enhanced density of spin excitations. Topological Mott insulating states have been proposed for some of these systems³³, but spin effects in a 3D topological insulator are confined to the sample surface and seem unlikely to contribute to the bulk muon spin relaxation. A spectroscopic study of low-lying fluctuations and Δ_g in $\text{Eu}_2\text{Ir}_2\text{O}_7$ would elucidate the situation, as would μSR experiments on the frustrated hyperkagomé iridate $\text{Na}_4\text{Ir}_3\text{O}_8$ ³⁴ and the (unfrustrated) weak Mott insulator Sr_2IrO_4 .⁸

In summary, the uniform spontaneous local field observed at muon sites below the MI/AFM transition indicates that $\text{Eu}_2\text{Ir}_2\text{O}_7$ exhibits long-range magnetic order, ruling out both quantum-spin-liquid (at least within the μSR time window) and spin-glass ground states. The magnetic structure cannot be obtained from μSR experiments alone, and neutron scattering in iridates is prohibitively difficult because of the high neutron absorption cross-sections of Ir nuclei. Resonant x-ray magnetic diffraction would be a useful alternative.

The dynamic muon spin relaxation rate $\lambda_d(T)$ shows no sign of critical slowing down above T_M , suggesting a mean-field-like transition, and in the ordered state $\lambda_d(T)$ reveals an anomalous persistence of slow Ir^{4+} spin fluctuations to low temperatures. Although geometric frustration may play a role in this behavior, the weakness of frustration in $\text{Eu}_2\text{Ir}_2\text{O}_7$, evidenced by the relatively large transition temperature, leads us to speculate that low-lying excitations associated with small-gap insulating behavior may be involved. Studies of other iridates, frustrated and unfrustrated, are clearly desirable.

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