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Magnetic transition, long-range order, and moment fluctuations in the pyrochlore iridate $Eu_2Ir_2O_7$

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Muon spin rotation and relaxation experiments in the pyrochlore iridate $Eu_2Ir_2O_7$ 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 iridates $BaIrO_3$ and $Sr_2Ir_2O_4$. $Eu_2Ir_2O_7$ 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 $R_2Ir_2O_7$, 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 = Pr, Nd, Sm, and Eu these compounds exhibit metallic behavior at high temperatures, while for R = 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 studies[4] spin-glass-like ordering was reported for R = 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 $Eu_2Ir_2O_7$[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 reported[2] for R = Nd, Sm, and Eu, and an exotic chiral spin-liquid metallic ground state[6] was found in $Pr_2Ir_2O_7$. 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 family[7] 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 treatment[8] involving strong spin-orbit coupling leads to the possibility of a Mott transition, however, and suggests an effective angular momentum $J_{\rm eff} = 1/2$. Alternatively, a Slater transition, as found in the pyrochlore $Cd_2Os_2O_7$,[9] is suggested by the second-order nature of the transition.

Thus, $Eu_2Ir_2O_7$ 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) experiments[10] 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 $T \to T_M$ from above, suggesting a mean-field-like transition.

Polycrystalline samples of $Eu_2Ir_2O_7$ 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 decouple[13] muon spins from nuclear dipolar fields in the paramagnetic state. Data were taken in a ^{4}He 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 quasistatic[14] component $\langle \mathbf{B}_{\rm loc} \rangle$ of the local field $\mathbf{B}_{\rm loc}$ 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 $\langle \mathbf{B}_{\rm loc} \rangle$ is relatively homogeneous. The late-time asymmetry data (not shown) exhibit exponential relaxation, due solely to dynamic (thermal) fluctuations of $\mathbf{B}_{\rm loc}$.[13] This relaxation is much slower than the oscillation damping rate, indicating that the latter reflects a quasistatic distribution of $\langle \mathbf{B}_{\rm loc} \rangle$.

The data were fit using the two-component asymmetry function

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

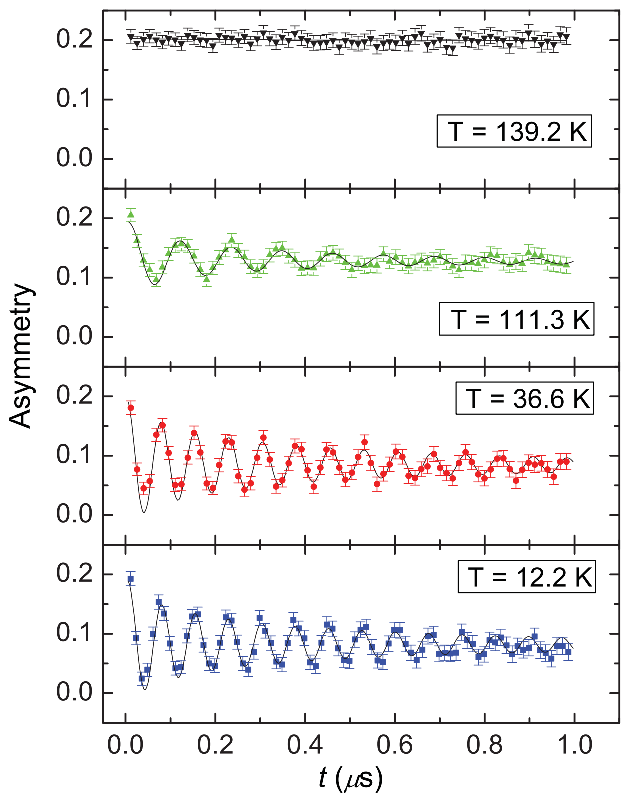

FIG. 1. (Color online) Representative early-time asymmetry data $A(t)$ in $Eu_2Ir_2O_7$; longitudinal field = 25 Oe. Solid curves: fits using Eq. (1).

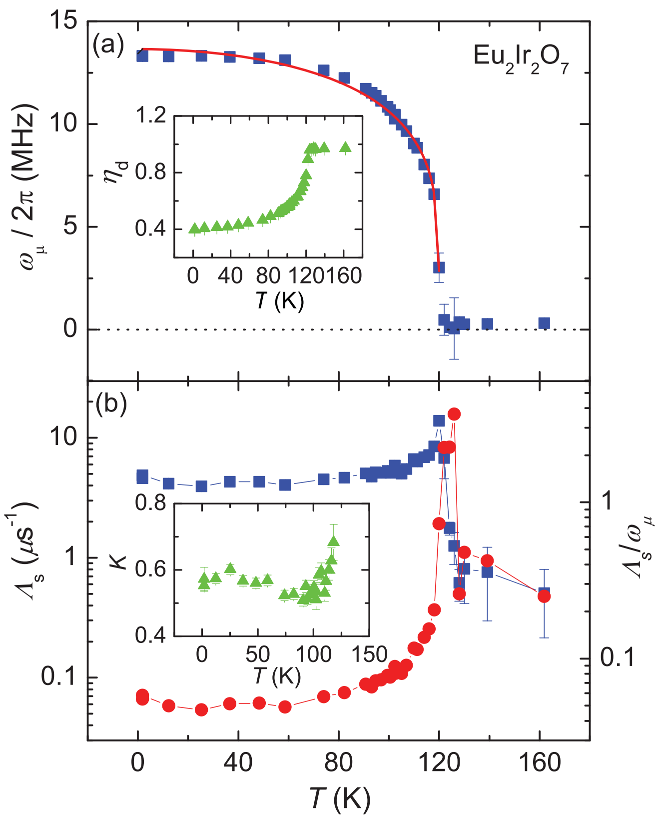

FIG. 2. (Color online) (a) Temperature dependence of muon spin precession frequency $\omega_\mu/2\pi$ in $Eu_2Ir_2O_7$. Inset: Late-time fraction η_d. The curve is a guide for the eye. (b) Temperature dependence of quasistatic muon spin relaxation rate Λ_s (squares, left axis) and fractional width of field distribution Λ_s/ω_μ (circles, right axis). Inset: Stretching power K.

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.

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

As shown in the inset to Fig. 2(a), the late-time fraction $\eta_d = A_d/(A_s + A_d)$ approaches 1 as $T \to T_M$ from below. This is due to the disappearance of $\langle \mathbf{B}_{\rm loc} \rangle$, 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 $\langle \mathbf{B}_{\rm loc} \rangle$.[13] The increase of η_d as $T \to T_M$ 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 $T \to T_M$. 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 $\langle B_{\rm loc} \rangle$: For small K the wings of the distribution become more prominent.[16] The value of K is temperature-independent ($\sim$0.55) at low temperatures and increases as $T \to T_M$.

The simple exponential form of the late-time relaxation data indicates that the dynamic muon spin relaxation, like $\langle B_{\rm loc} \rangle$ (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) $\lambda_d = 0.029(3)\,\mu s^{-1}$ is constant below ∼100 K, and (ii) with decreasing temperature there is an unusual steplike 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 $\langle \mathbf{B}_{\rm loc}(t)\rangle$ with zero long-time average, where the relaxation time is essentially the correlation time of $\langle \mathbf{B}_{\rm loc}(t)\rangle$ (Ref. 13), and (B) fluctuations $\delta\mathbf{B}_{\rm loc}$ about a nonzero static $\langle \mathbf{B}_{\rm loc}\rangle$, that is, $\mathbf{B}_{\rm loc}(t) = \langle \mathbf{B}_{\rm loc}\rangle + \delta\mathbf{B}_{\rm loc}(t)$;[21] here the relaxation rate depends on the magnitude and stochastic properties of $\delta\mathbf{B}_{\rm loc}(t)$. Case A describes dynamic relaxation in a paramagnet with extremely slow spin dynamics and yields a fluctuation rate $\sim\lambda_d \approx 3 \times 10^4\ {\rm s}^{-1}$. The data cannot rule this scenario out in $Eu_2Ir_2O_7$ 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 $\omega_f = \delta B_{\rm loc}/\gamma_\mu$ 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 quasistatic field in frequency units ($\omega_f \lesssim \omega_\mu \approx 8.5 \times 10^7\ {\rm s}^{-1}$), this yields an upper bound $\tau_c^{-1} \lesssim 2.5 \times 10^{11}\ {\rm s}^{-1}$ or $\hbar/k_B\tau_c \lesssim 2$ 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 $Eu_2Ir_2O_7$, with $T_N = T_M = 120$ 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), that is, 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 $Eu_2Ir_2O_7$.

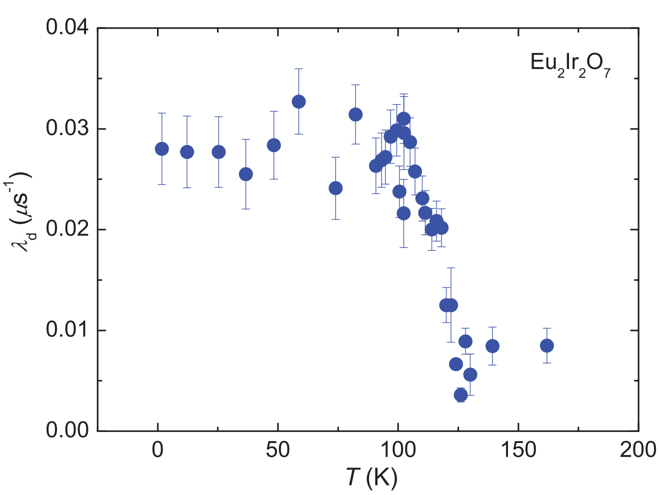

FIG. 3. (Color online) Temperature dependence of the dynamic muon spin relaxation rate λ_d in $Eu_2Ir_2O_7$.

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 $J \geqslant 1$ multiplets; this, however, results in reduction rather than enhancement of Eu nuclear moments.[28,29] The persistent spin dynamics in $Eu_2Ir_2O_7$ must therefore be electronic in origin and associated with Ir^{4+} magnetism.

The relatively high transition temperature of $Eu_2Ir_2O_7$ suggests that the AFM exchange constant is not much larger than T_M, in which case $Eu_2Ir_2O_7$ is a weakly frustrated material.[30] Noting that the unfrustrated iridates $BaIrO_3$ and Sr_2IrO_4 both exhibit persistent muon spin relaxation,[31] we consider the possibility that frustration may not be the primary cause of persistent relaxation in $Eu_2Ir_2O_7$ 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 MI transition is nevertheless retained (perhaps because of strong spin-orbit coupling[8]) but the electrons are not well localized, the gap energy Δ_g can be comparable to k_BT_M. The resistivity of single-crystal $Eu_2Ir_2O_7$ in fact yields a maximum gap value ≈10 meV ≈ k_BT_M.[12] We speculate that charge fluctuations[32] 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 systems,[33] 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 $Eu_2Ir_2O_7$ would elucidate the situation, as would μSR experiments on the frustrated hyperkagomé iridate $Na_4Ir_3O_8$.[34]

In summary, the uniform spontaneous local field observed at muon sites below the MI/AFM transition indicates that $Eu_2Ir_2O_7$ exhibits long-range magnetic order, ruling out both quantum-spin-liquid (at least within the μSR time window) and spin-glass-like 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 $Eu_2Ir_2O_7$, 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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[15]The signal amplitude associated with the nonzero Λ_S above T_M is very small [$\eta_d \approx 1$; cf. inset to Fig. 2(a)] and is either an instrumental artifact or due to a few percent of ordered second phase in the sample.

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