Long-range order and moment fluctuations in the pyrochlore iridate $Eu_2Ir_2O_7$

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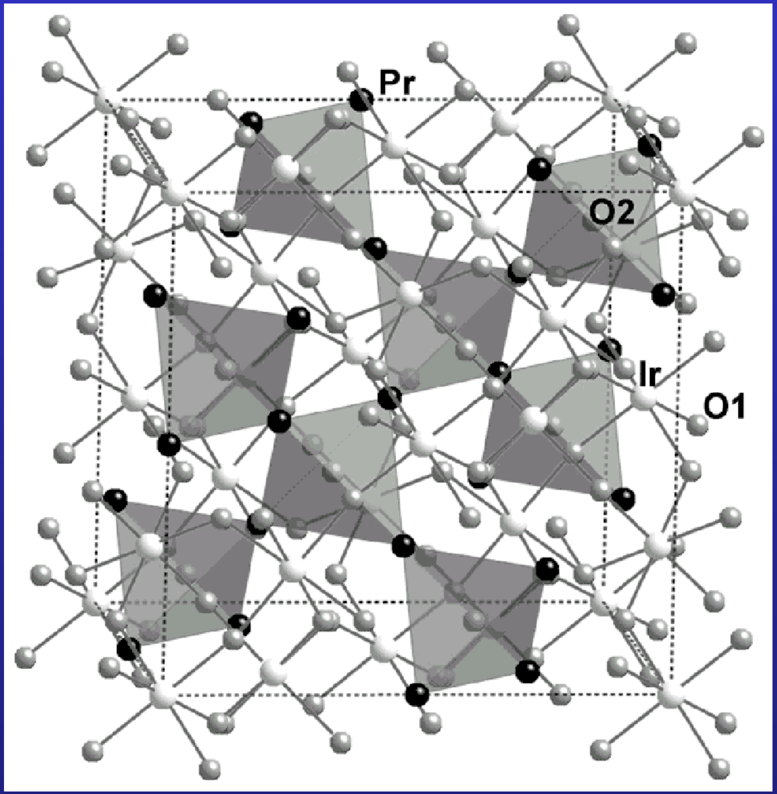
Outline

- Pyrochlore rare-earth *iridates* $R_2Ir_2O_7$: metal-insulator (MI) transition across rare-earth series.
- $Eu_2Ir_2O_7$: Eu^{3+} *nonmagnetic* (Hund's-rule $L = S$, $J = 0$). Only Ir^{4+} ($5d^5$, low-spin $S = 1/2$) magnetism.
- Metal-insulator transition, $T_M = 120$ K; "complex" antiferromagnetic ordering at $T_N = T_M$. Large Ir 5*d* overlap usually ⇒metallic conduction. A *weak Mott insulator*.
- Is $Eu_2Ir_2O_7$ a geometrically frustrated "spin 1/2" system?

wLF-µSR:

- *Well-defined frequency below* T_N. Commensurate long-range order.
- Dynamic relaxation *rela`tively fast*, and *persists to low temperatures*
  - $\Rightarrow$ *singular* density of low-lying excitations.
  - Observed in other pyrochlores & frustrated systems, but also in *unfrustrated* $BaIrO_3$ and Sr_2IrO_4.
- $Eu_2Ir_2O_7$ is only *weakly* frustrated (Ramirez frustration parameter $J_{\mathrm{ex}}/T_N \approx$ 1).
- Persistent relaxation due to *small-gap Mott behavior* rather than frustration?

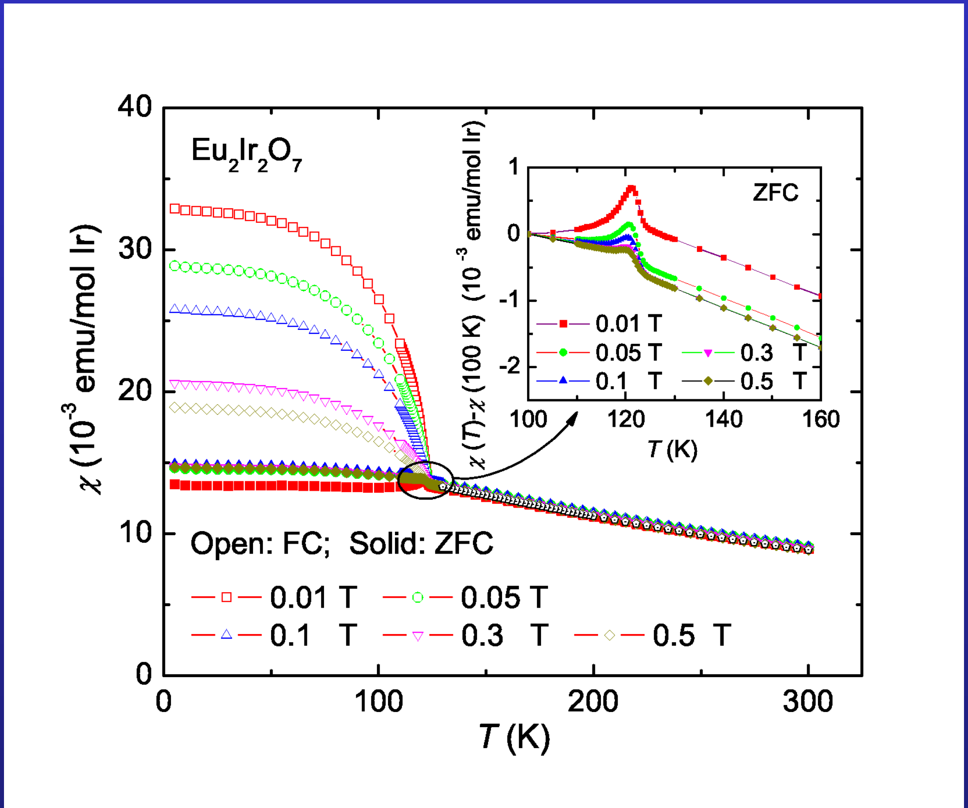
$Eu_2Ir_2O_7$



$Eu_2Ir_2O_7$: pyrochlore structure.

Independent Eu and Ir sublattices of corner-sharing tetrahedra.

Magnetic Susceptibility



Clear signature of transition at 120 K.

Large bifurcation between field-cooled (FC) and zero-field-cooled (ZFC) data. Spin glass? Complex antiferromagnet?

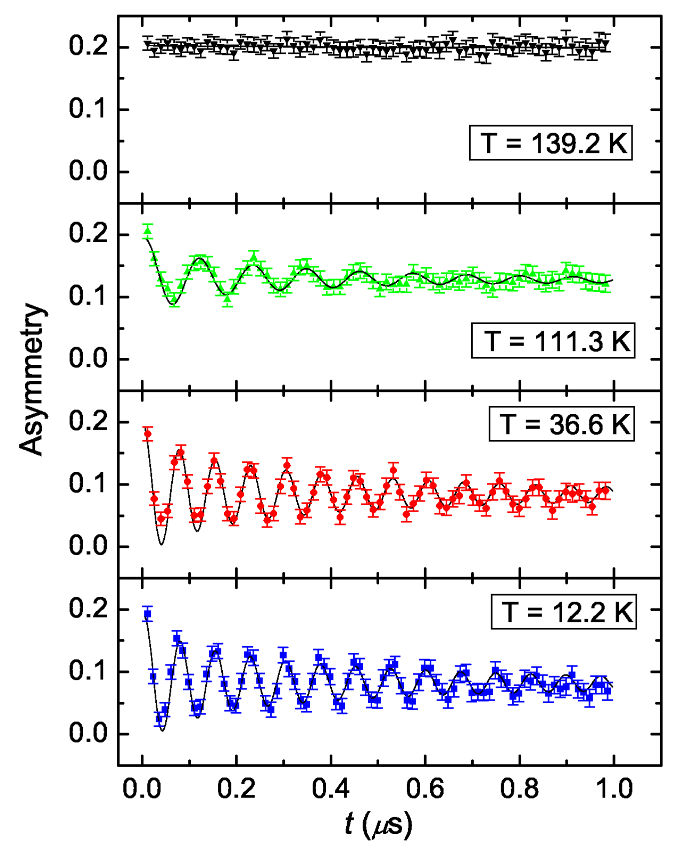
Weak longitudinal-field μSR in $Eu_2Ir_2O_7$

(Weak longitudinal field to decouple nuclear dipolar field above T_N.)

Weakly damped oscillations below 120 K. *Homogeneous* local field.

Damping not exponential; best fit by "stretched" exponential $\exp[-(\Lambda_s t)^K]$, $K < 1$.

Late-time dynamic relaxation (not shown): *single exponential*.



Static properties: frequency, asymmetries

(a): Frequency ω_μ (local static field $B_{\mathrm{loc}} = \omega_\mu/\gamma_\mu$) sets in sharply at T_N. A magnetic transition.

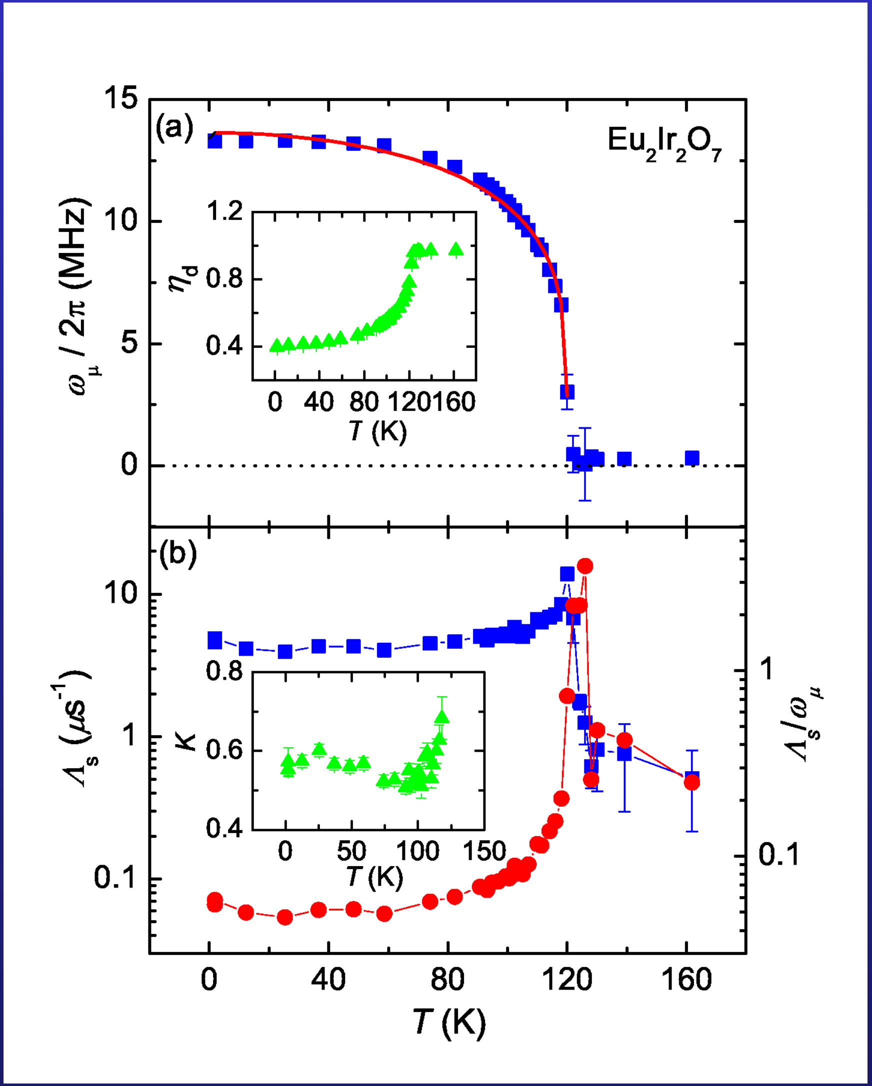
At $T = 2$ K $B_{\mathrm{loc}} \approx 990$ G. Crude estimate of ordered moment: ~1 μ_B/Ir ion.

Temperature dependence ⇒ small "critical" exponent $\beta < 1/3$.

Insert: late-time (dynamic) asymmetry fraction $\eta_d = A_d/(A_s + A_d)$.

Expect $\eta_d = 1/3$ in ordered state (powder sample); 1 above T_N.

Smooth variation ⇒ *distribution* of T_N.



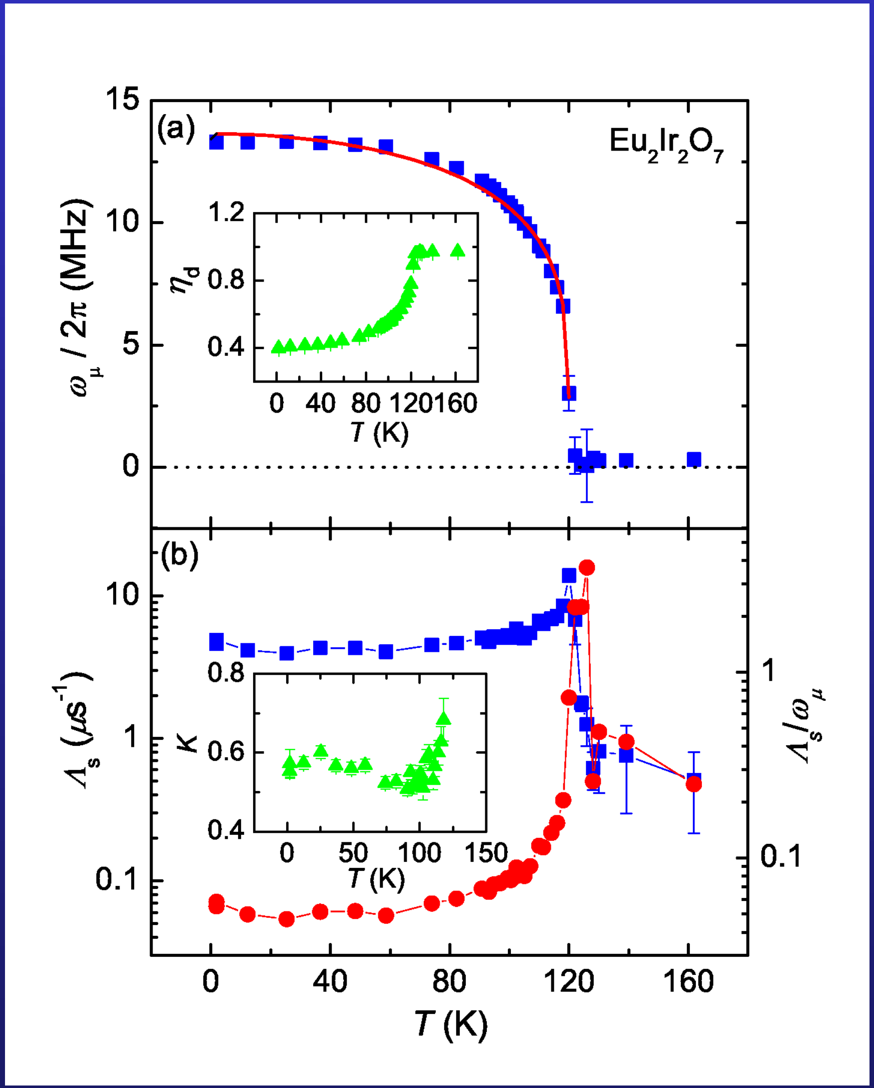
Static properties: damping, inhomogeneity

(b): Static damping rate Λ_s from spread in local field.

(Above T_N η_d nearly 1; data either instrumental artifact or second phase.)

Damping is relatively weak: $\Lambda_s/\omega_\mu \approx$ 5–7% at low temperatures.

Inset: stretching power $K \approx 0.55$ at low temperatures; increases near T_N. Increase probably due to distribution of transition temperatures.



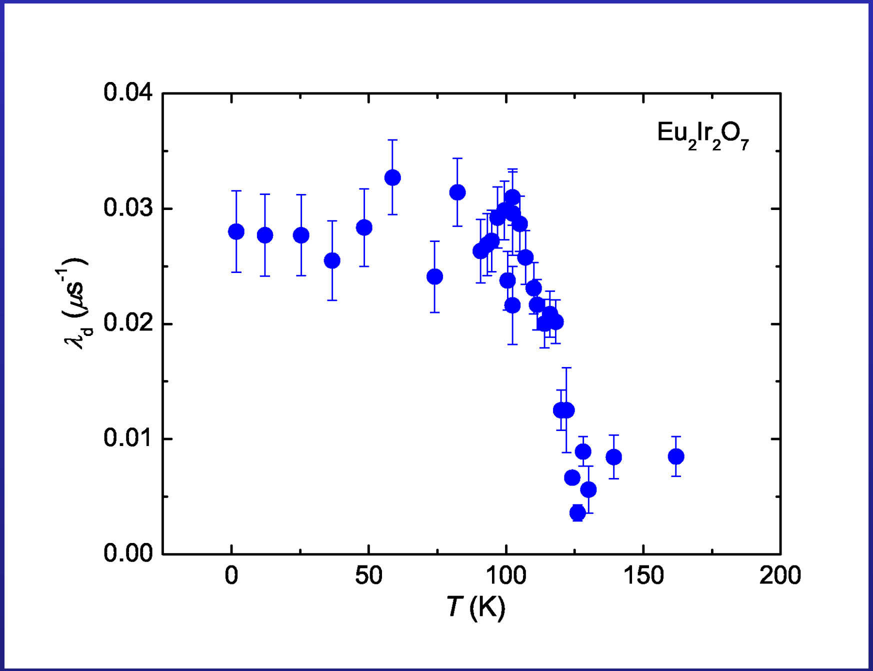
Dynamic relaxation

Late-time dynamic (spin-lattice) relaxation exponential; rate λ_d

- *is constant* [0.029(3) μs^{-1}] below ~100 K,
- shows step below T_N but no critical divergence. Mean-field-like transition.

Assume *motional narrowing* limit (quasistatic flucts. very unlikely):

- $\lambda_d \approx \omega_f^2 \tau_c$, ω_f = rms fluctuating field in freq. units, τ_c = correlation time.
- Yields $1/\tau_c < 2.5 \times 10^{11}$ s^{-1}, or ~2 K.

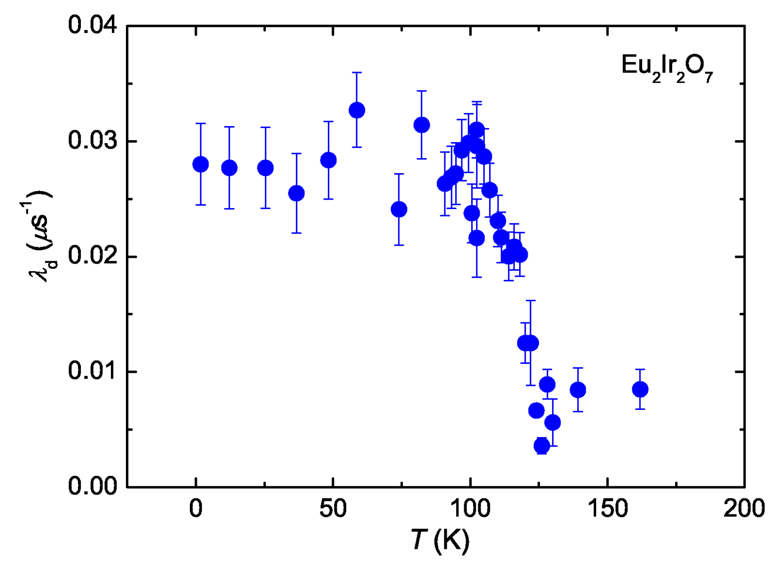


Persistent low-temperature relaxation and homogeneous magnetic order *unexpected*.

In ordinary antiferromagnets λ_d is due to thermal spin-wave excitations; decreases at low temperatures.

Fluctuation rate $1/\tau_c$ expected to be $\sim T_N$ near the transition; measured value two orders of magnitude smaller.

In $Eu_2Ir_2O_7$ fluctuations are *slow*, but relaxation rate *does not decrease at low temperatures*.



Discussion: whence persistent relaxation?

Persistent relaxation *often seen* in geometrically frustrated systems.

Indicates strongly enhanced (singular) density of low-lying excitations.

Mechanisms?

- For rare-earth non-Kramers ion with nonmagnetic CEF ground state (e.g., Pr^{3+} in filled skutterudite $PrOs_4Sb_{12}$) *hyperfine-enhanced nuclear magnetism* can couple to muon spin.
- Similar hyperfine effect from Eu^{3+} spin-orbit-split $J > 0$ multiplet, but effective Eu nuclear moment is *reduced*. No other candidate nuclei in $Eu_2Ir_2O_7$.
- $\Rightarrow$mechanism must be *electronic in origin*, associated with Ir^{4+} magnetism.

Is $Eu_2Ir_2O_7$ highly frustrated?

Ramirez frustration parameter J_{ex}/T_N: *large* in highly frusrated materials.

- Usually estimate exchange constant J_{ex} from paramagnetic Curie-Weiss temperature.
- But $\chi(T)$ *not Curie-Weiss* in metallic state (no local moments).
- But T_N is relatively high, and susceptibility is relatively large.

⇒unlikely that $J_{\mathrm{ex}} >> T_N$. $Eu_2Ir_2O_7$ appears to be *weakly frustrated*.

Unfrustrated iridates $BaIrO_3$ and Sr_2IrO_4 also exhibit persistent relaxation.

Conclusion: *frustration might not be mechanism for persistent relaxation in* $Eu_2Ir_2O_7$. Look for other candidates.

Weak Mott insulator ⇒ new dynamics?

Ir-based materials: large Ir 5*d* wave functions weaken on-site repulsion.

- Even if MI transition & AFM retained (e.g., strong S-O coupling), 5*d* electrons *not well localized*.
- ⇒ gap energy $\Delta_g(T) \approx k_B T_N$. In $Eu_2Ir_2O_7$ Δ_g(max.) ≈ 10 meV from transport measurements.

Topological Mott insulating states? Unlikely; spin effects in 3D topological insulators confined to sample surface.

Speculation: charge/spin fluctuations over gap might be involved in slow spin excitations. *New mechanism* for persistent dynamics?

Conclusions

Uniform B_{loc} in AFM $Eu_2Ir_2O_7$ ⇒ *homogeneous* long-range order.

- Rules out quantum spin liquid, spin-glass-like ground states.
- Magnetic structure not determined. (Ir nuclei capture thermal neutrons; probably need resonant x-ray magnetic Bragg diffraction.)

Dynamic muon spin relaxation:

- *persistent* low-temperature spin fluctuations,
- frustration probably *weak*.

Low-lying excitations associated with *weak Mott insulating state*?

Studies of other iridates, frustrated and unfrustrated, desirable.

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