Spin-liquid/spin-glass-like behavior in pyrochlore Pr$_2$Ir$_2$O$_7$

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Pr$_2$Ir$_2$O$_7$ is a metallic compound with a well-isolated Pr$^{3+}$ $T_3$ magnetic doublet CEF ground state. Transport properties suggest a Kondo effect and partial screening of the moments, even below a RKKY interaction temperature $T^* \sim 20$ K; geometrical frustration was suggested as a mechanism for suppression of magnetic order. The magnetic susceptibility exhibits an unusual $-\ln T$ temperature dependence over more than a decade of temperature below $\sim 2$ K, below which there is evidence for partial spin freezing. μSR experiments in powder samples of Pr$_2$Ir$_2$O$_7$ for temperatures in the range 0.02–20 K yield two-component relaxation functions in low applied fields similar in shape to that observed in canonical spin glasses. This is strong evidence for freezing of a dilute concentration $c$ of randomly sited frozen Pr$^{3+}$ moments, presumably associated with lattice defects. The static relaxation rate $a$ at low temperatures corresponds to $c \sim 0.01–0.03$. The spin-freezing process therefore leaves $\geq 97\%$ of the Pr$^{3+}$ spins in a spin-liquid state. Relaxation at later times is dynamic in origin.

The temperature dependencies of $a$ and the dynamic rate $\lambda$ give a glass temperature $T_g \approx 25$ K $\approx T^*$, which is a completely unexpected result, as previous bulk measurements had given no hint of spin freezing above $\sim 0.1$ K. Furthermore, freezing of a small fraction $c$ of spins should occur at a much lower temperature ($\sim cT^*$ for a $1/r^5$ RKKY interaction). The temperature dependence of $a$ (which represents the order parameter in spin glasses) is non-mean-field-like, continuing to increase well below $T_g$. Perhaps the most surprising aspect of our initial results is the absence of a peak in $\lambda(T)$ at $T_g$. Such a peak is expected due to critical slowing down of ‘pre-formed’ local moments, and is observed in spin glasses and the pyrochlore antiferromagnet Y$_2$Mo$_2$O$_7$. A mean-field-like transition would narrow the critical region and make observation of the peak difficult, but the non-mean-field behavior of $a(T)$ makes this scenario unlikely. Alternatively, the localized moments themselves may exist only below $T_g$, similar to Cooper pairs in conventional superconductors. Further work is clearly needed to understand this very unusual behavior.