**Fig. S2.** EPR spectrum (solid line) and simulation (dashed line) 46 mM [PhBP(Pr)3]FePMe3 in toluene. Simulation parameters: $S = 3/2$, $D = +20 \text{ cm}^{-1}$, $E/D = 0.16$, and $g = 2.2$. Spectral parameters: microwaves, 9.65 GHz ($B_1 \perp B$), 0.2 mW, $T = 15 \text{ K}$.

**Fig. S3.** Isofield parallel magnetization ($\chi_T$, dots) for a powder sample of 4 at the magnetic fields listed on the plot. The simulations (solid lines) use: $S_A = S_B = 3/2$, $J = +4 \text{ cm}^{-1}$, $D_A = D_B = -53 \text{ cm}^{-1}$, $E/D_A = E/D_B = 0.10$, $g_A = g_B = 2.35$. The Brillouin curve ($D = 0$, $g = 2.0$) is for an $S = 3$ state at 0.5 T.