Supporting Information for:

Enhancing the Magnetic Anisotropy of Linear Cr(II) Chain Compounds Using Heavy Metal Substitutions

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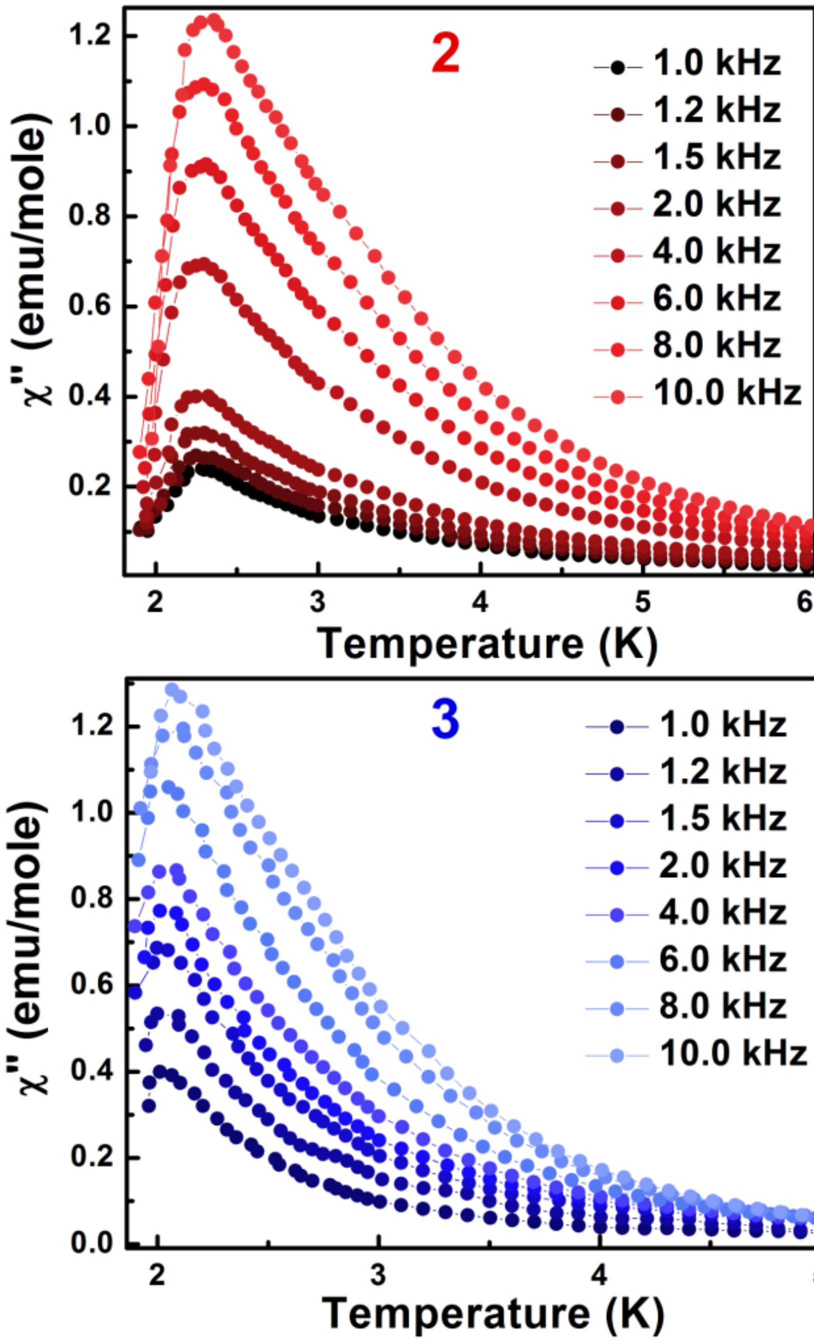

Figure S1. Variable-temperature, variable-frequency *ac* magnetic susceptibility measurements of **2** (top) and **3** (bottom) under an applied *dc* field of 0.2 Tesla. Only a small frequency dependence in χ'' was observed for these compounds.

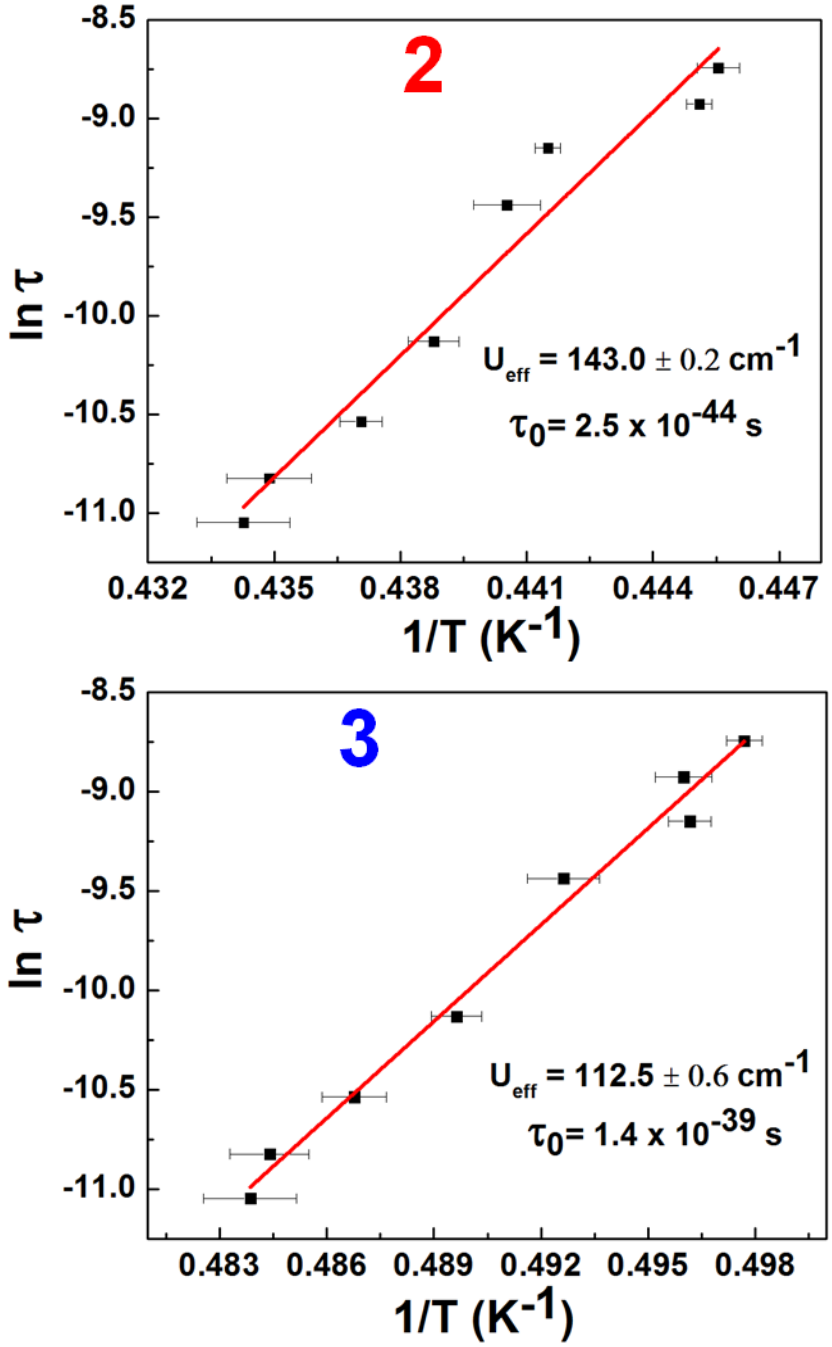

Figure S2. Arrhenius plot of the relaxation rate vs. inverse temperature for **2 (top)** and **3 (bottom)**. The frequency dependence was too small to be meaningful for the Arrhenius analysis.

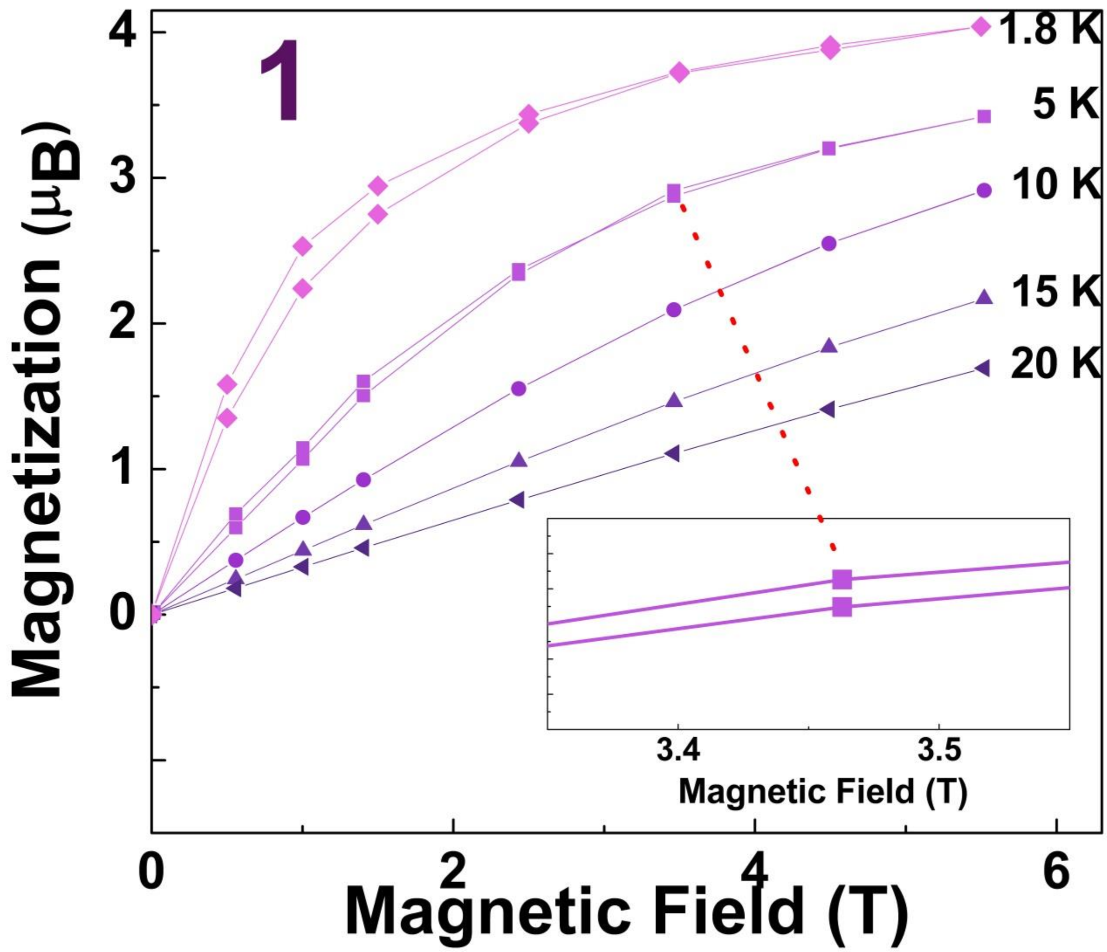

Figure S3. Magnetization of **1** with zoomed-in inset showing the highest temperature at which hysteresis is observed.

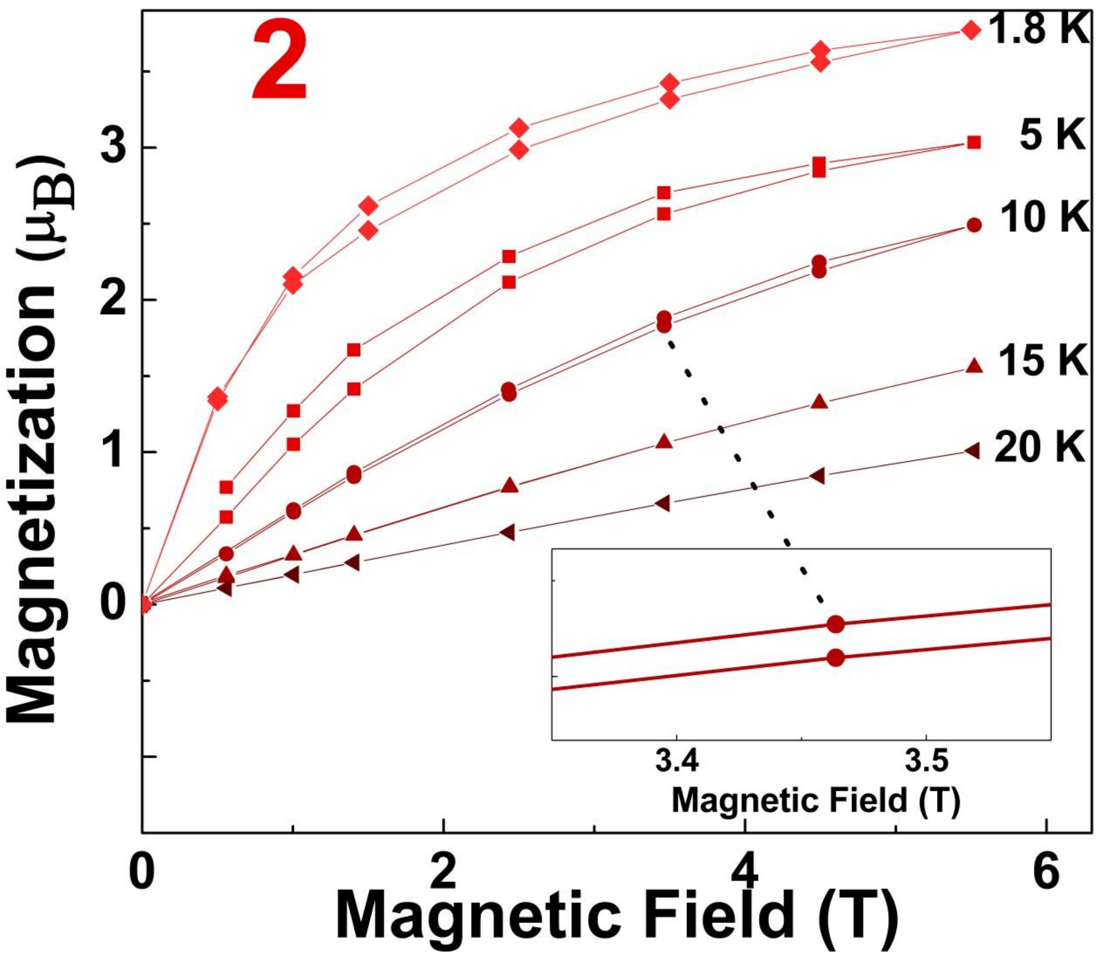

Figure S4. Magnetization of **2** with zoomed-in inset showing the highest temperature at which hysteresis is observed.

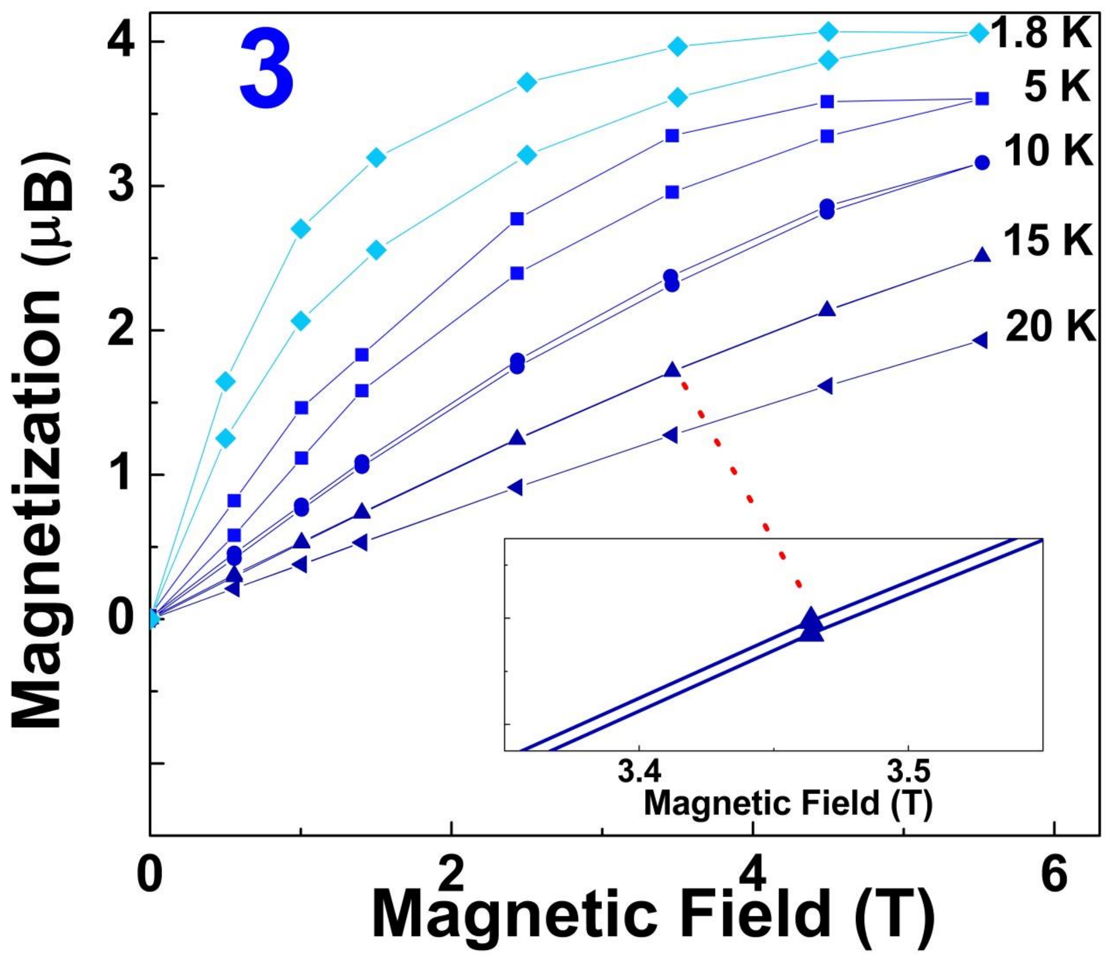

Figure S5. Magnetization of **3** with zoomed-in inset showing the highest temperature at which hysteresis is observed.

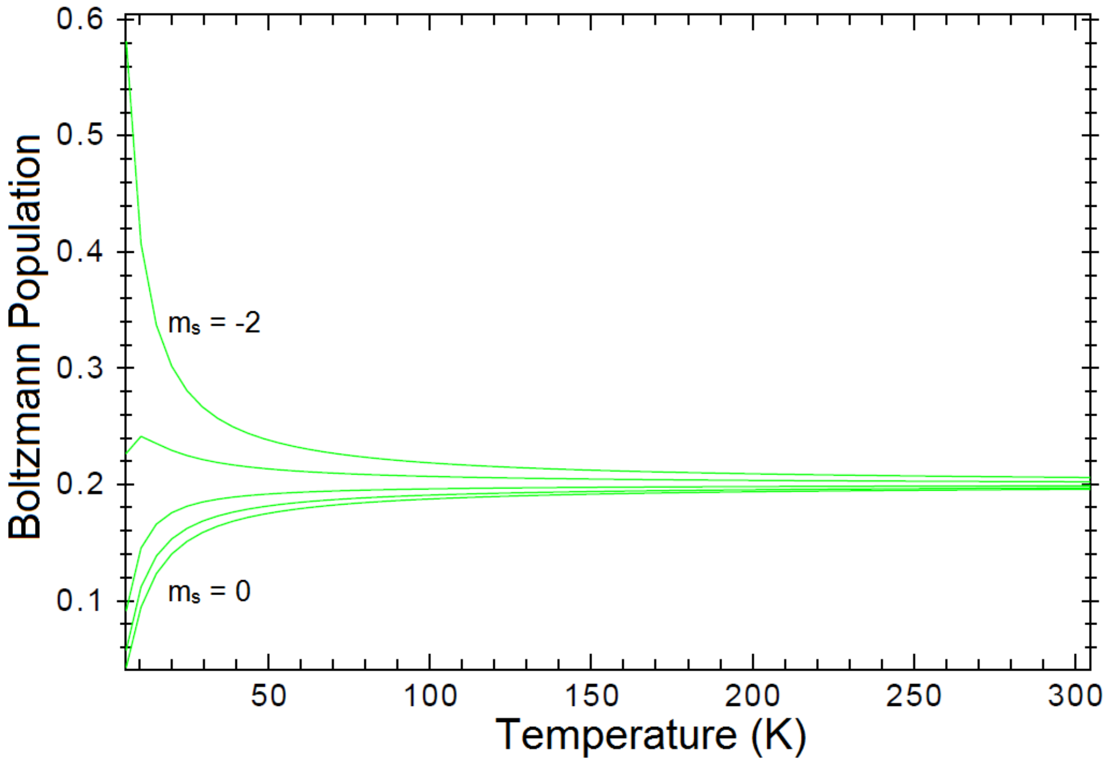

Figure S6. The Boltzmann population for **1** is calculated using S = 2, D = -1.643 cm^{-1}, E/D = 0.021, and isotropic g = 1.99. The black numbers represent the M_s quantum numbers in the high-field limit. At low temperatures (< 20 K) it is seen that the ± 2 states are the most populated. Variable-temperature HF-EPR allows for definitive assignment of the sign of D by observing changes in the EPR peak intensities upon cooling and then comparing these changes with the simulated energy level diagram and the calculated Boltzmann population.