

A Triangular Frustrated Eu^{II} Magnetic Framework for Sub-Kelvin Refrigeration

A.S. Manvell,¹ M.A. Dunstan,¹ D. Gracia,² J. Hrubý,³ M. Kubus,¹ J.N. McPherson,¹ E. Palacios,² H. Weihe,⁴ S. Hill,³ J. Schnack,⁵ M. Evangelisti,² K.S. Pedersen¹
1. TU Denmark, Denmark; 2. CSIC & Universidad de Zaragoza, Spain; 3. NHMFL, FSU; 4. University of Copenhagen, Denmark; 5. Universität Bielefeld, Germany



Funding Grants: K. M. Amm (NSF DMR-2128556); K. S. Pedersen (Villum Fonden, Sapere Aude, Carlsberg Foundation); M. Evangelisti (European Union, Gobierno de Aragón)

Attaining sub-kelvin temperatures is technologically challenging and often relies on the scarce resource ³He, unless employing adiabatic demagnetization refrigeration. Herein, the coolant typically consists of weakly coupled paramagnetic ions, whose magnetic interaction strengths are comparable in energy to the relevant temperature regime of cooling. Such interactions depend strongly on inter-ion distances, fundamentally hindering the realization of dense coolants for sub-kelvin refrigeration.

In this investigation, a magnetically concentrated triangular coordination network, Eu_{0.9}Ba_{0.1}I₂(pyrazine)₃, is reported featuring the large $s = 7/2$ spin moment of Eu^{II}. High-field electron paramagnetic resonance (EPR), magnetization, and heat capacity measurements indicate weak antiferromagnetic correlations between the Eu^{II} spins and a dominant easy-plane magnetic anisotropy. In particular, EPR measurements at high magnetic fields and relatively high temperatures (80K – see Figure) are essential to quantifying the magnetic anisotropy independently from the inter-ion antiferromagnetic exchange interaction energy.

This interdisciplinary collaboration has demonstrated a perfect combination of weak magnetic interactions and geometric frustration in the low-dimensional organic Eu_{0.9}Ba_{0.1}I₂(pyrazine)₃ framework, which prevents entropy-annihilating magnetic ordering down to at least 0.17K; large magnetic entropy is a critical requirement for adiabatic demagnetization refrigeration. These factors, combined with the large spin moment and high Eu^{II} concentration, open up exciting possibilities for efficient on-chip cryogenic refrigeration down to remarkably low (sub-kelvin) temperatures, especially in scenarios where established inorganic refrigerants cannot be utilized.

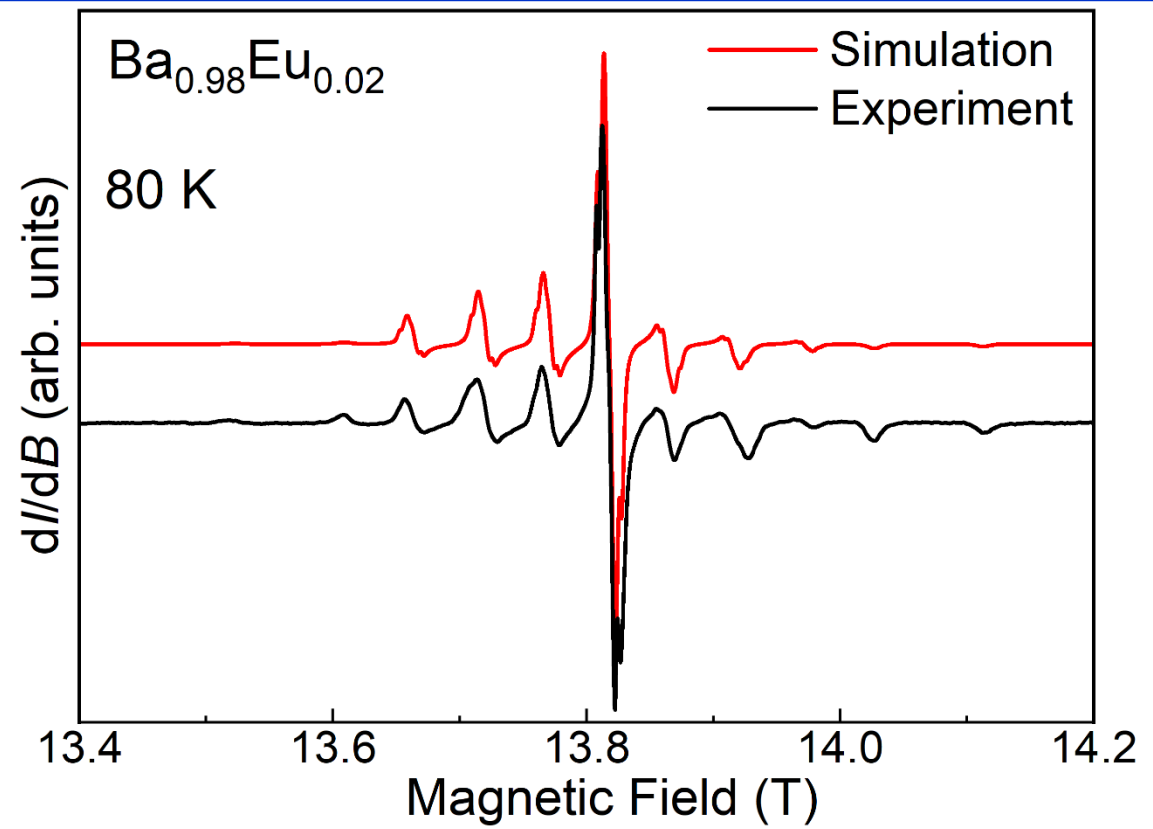


Figure 1. High-field EPR spectrum (Experiment) recorded at a frequency of 385GHz along with the corresponding simulation, from which quantitative information about the Eu^{II} magnetic anisotropy can be deduced.

Facilities and instrumentation used: Electron Magnetic Resonance (EMR) Facility – 15/17 Tesla Superconducting Magnet System.

Citation: Manvell, A.S.; Dunstan, M.A.; Gracia, D.; Hrubý, J.; Kubus, M.; McPherson, J.N.; Palacios, E.; Weihe, H.; Hill, S.; Schnack, J.; Evangelisti, M.; Pedersen, K.S., *A Triangular Frustrated Eu(II)–Organic Framework for Sub-Kelvin Magnetic Refrigeration*, **Journal of the American Chemical Society**, **147** (9), 7597-7603 (2025) doi.org/10.1021/jacs.4c17003 - [Data Set](#)