Barocaloric, topological and heavy-fermion systems – current research and the potential of neutron techniques in their extension

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1. **Ordinary MCE** - DREAM
2. **Rotational MCE (RMCE) –** MAGiC+DREAM
3. **Topological semimetals, short- and long-range order –** MAGiC+DREAM
4. **Inelastic scattering, crystal field levels, HF systems** – MIRACLES + MAGiC
5. **Summary**

Prof. dr. hab. Tomasz Toliński, Head of group
Prof. dr. hab. Bogdan Idzikowski
Dr. hab. Andrzej Kowalczyk, Prof. IFM PAN
Dr. hab. Eng. Zbigniew Śniadecki, Prof. IFM PAN
Dr. hab. Eng. Michał Falkowski
Dr. hab. Eng. Karol Synoradzki
Dr. Mieszko Kołodziej
Dr. Eng. Andrzej Musiał
Dr. Eng. Przemysław Skokowski

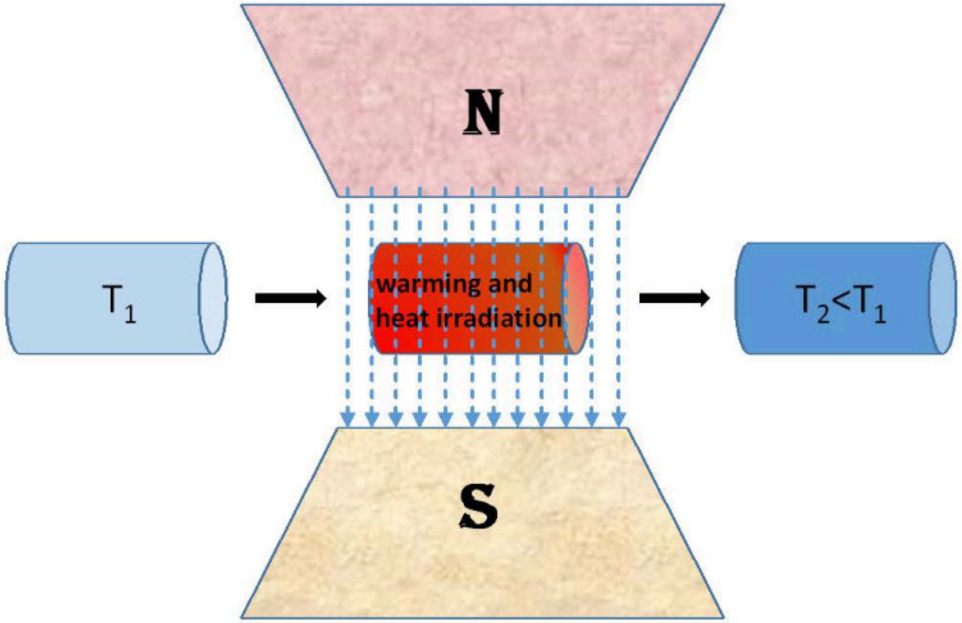
PhD students
M.Sc. Valentin Anière

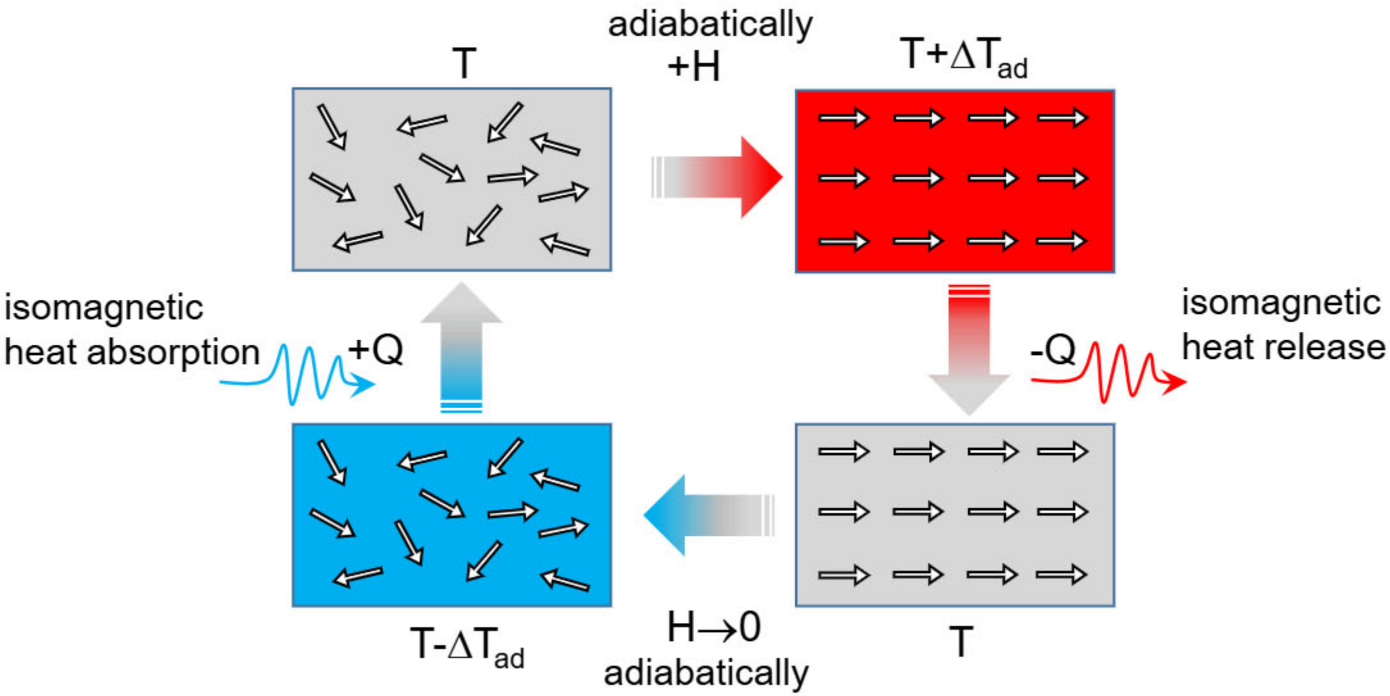
Topics

- intermetallic compounds
- amorphous and nanocrystalline alloys
- magnetocaloric effect
- topological insulators and semimetals
- thermoelectric power
- hard magnetic materials
- Kondo lattices, heavy fermions

DREAM
Bispectral Powder Diffractometer

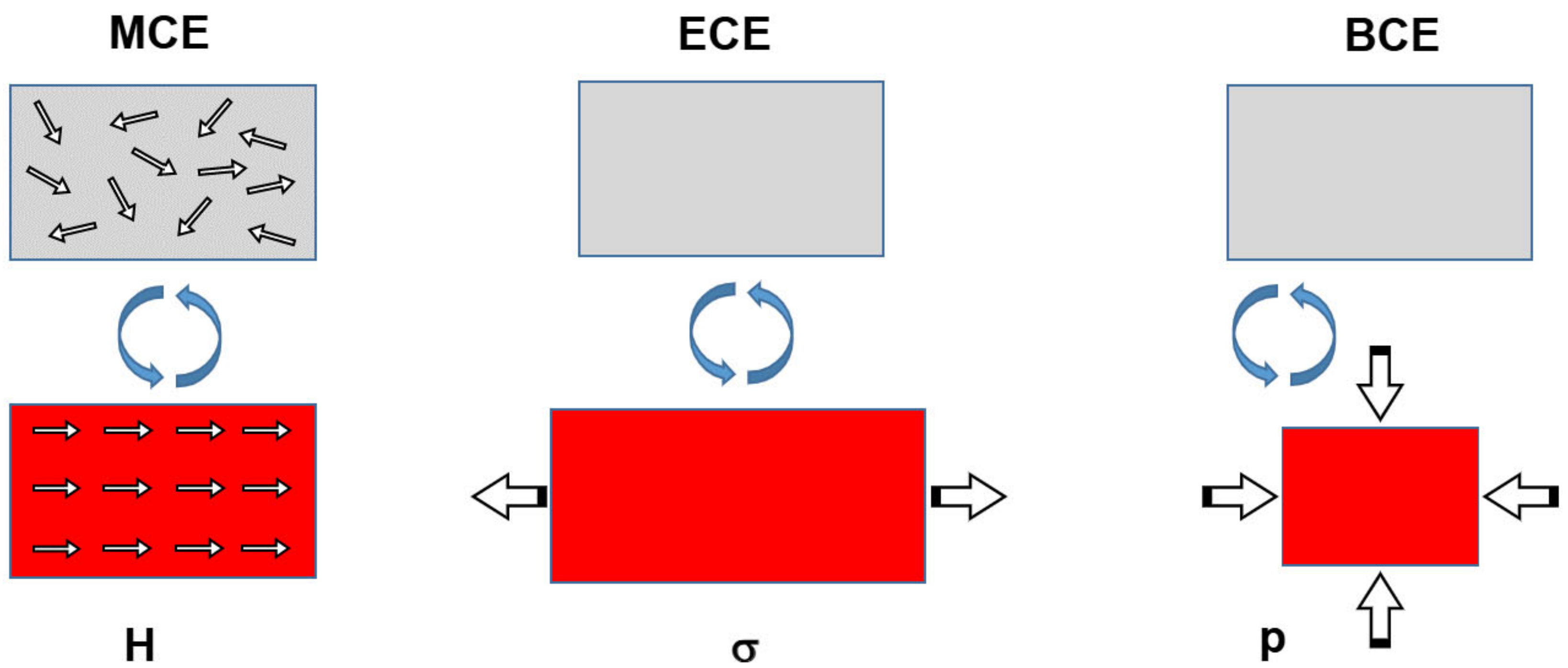
Magnetism: studies of complex magnetic structures, weak magnetic moments, multiferroics, magneto-elastic coupling, interplay between magnetic order and superconductivity, quantum criticality, magnetism at high pressure, orbital and charge ordering.


N
T1
warming and
heat irradiation
T2<T1
S



$$S\,(T,H) = S_m(T,H) + S_{lat}(T) + S_{el}(T)$$

Caloric effects



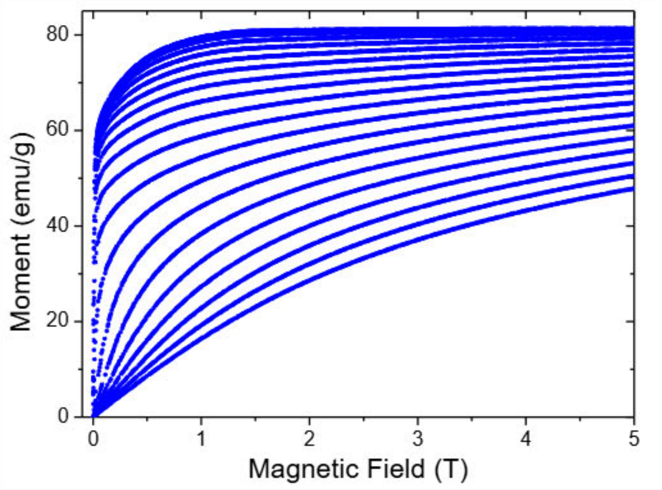
Elastocalorics

- Strain-driven changes in sample volume
- Large values at martensitic phase changes
- Hysteresis

Barocalorics

- Similar, but 3D hydrostatic pressure applied
- MCE materials can also show a BCE

MCE from M-H isotherms

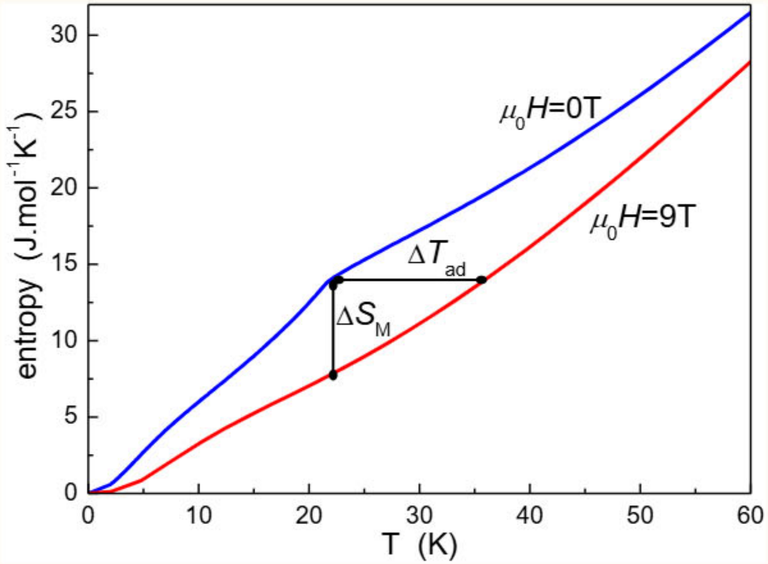


$$\left(\frac{\partial S(T,H)}{\partial H}\right)_T = \left(\frac{\partial M(T,H)}{\partial T}\right)_H$$

$$\Delta S_M(T) \approx \frac{\mu_0}{\delta T}\left[\int_0^{H_{max}} M(T+\delta T, H)dH - \int_0^{H_{max}} M(T,H)dH\right]$$

MCE from specific heat

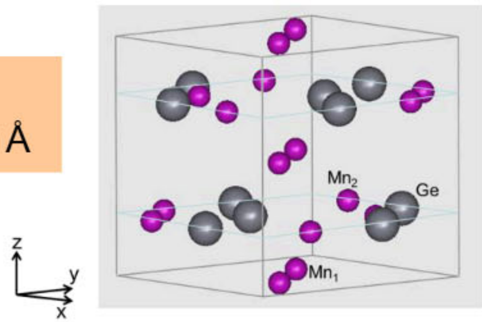
$$S(T)_{H=0} = \int_0^{H_{max}} \frac{C(T)_{p,H=0}}{T} dT \qquad S(T)_{H\neq 0} = \int_0^{H_{max}} \frac{C(T)_{p,H\neq 0}}{T} dT$$

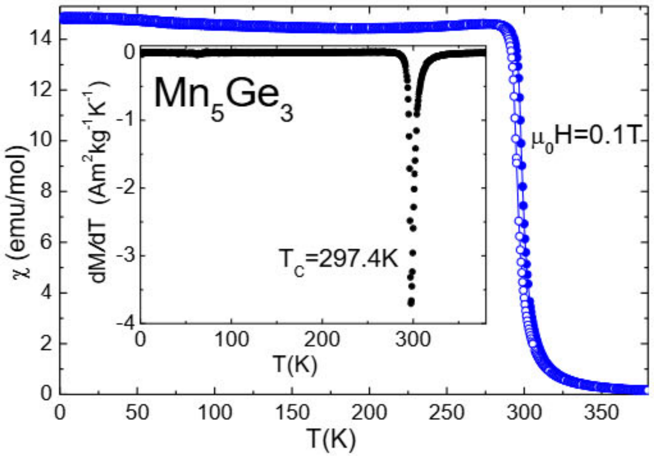


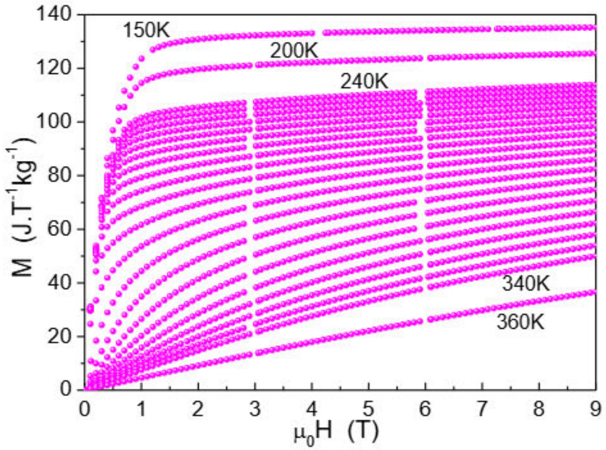
$$\Delta S_M(T) = S(T,H) - S(T,0)$$
$$\Delta T_{ad}(T) = T(S,H) - T(S,0)$$

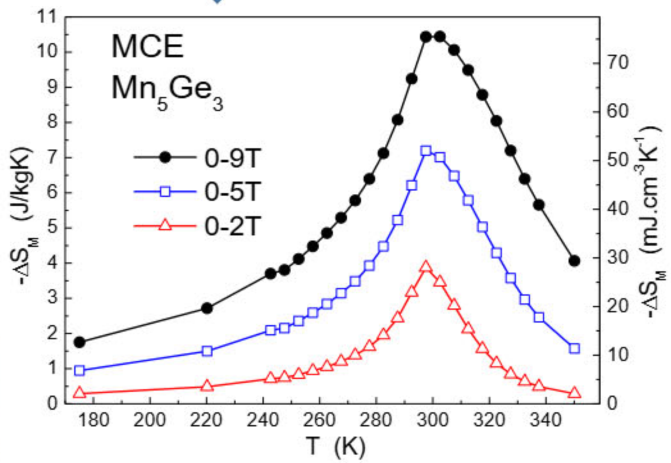
MCE determination

heksagonal, $P6_3/mcm$,
a = 7.197 Å and c = 5.036 Å









$$\Delta S_M(T) \approx \frac{\mu_0}{\delta T}\left[\int_0^{H_{max}} M(T+\delta T, H)\,dH - \int_0^{H_{max}} M(T, H)\,dH\right]$$

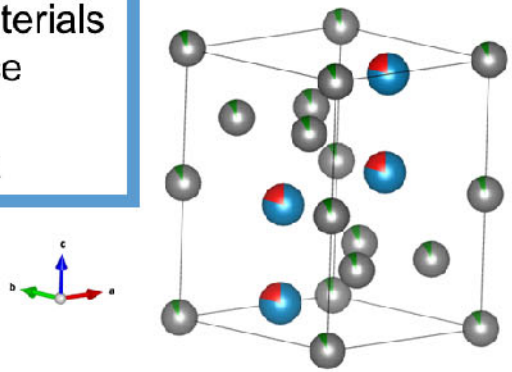
RE-free magnetoelastic materials for efficient and environmentally friendly cooling

$A_{1-x}B_x(Fe_{1-y}T_y)_2$

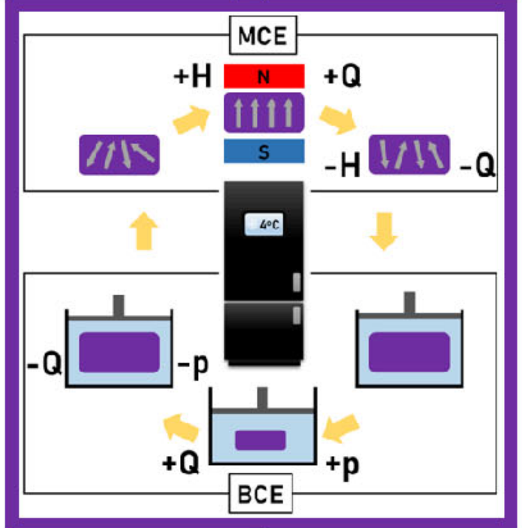
A = Sc, Hf ; B = Ti, Ta, ... ; T = Mn, Co,...

Why RE-free?

- non critical materials
- high abundance
- low price
- low supply risk



hexagonal structure of A_1Fe_2



Magnetocaloric effect

$$\Delta S_M(T)_{\Delta H} = \int_{H_i}^{H_f} \left(\frac{\partial M(T,H)}{\partial T}\right)_H dH \qquad \Delta T_{ad}(T)_{\Delta H} = -\int_{H_i}^{H_f} \left(\frac{T}{C(T,H)}\right)_H \left(\frac{\partial M(T,H)}{\partial T}\right)_H dH$$

Barocaloric effect

$$\Delta S_T(T)_{\Delta p} = \int_{p_i}^{p_f} \left(\frac{\partial V(T,p)}{\partial T}\right)_p dp \qquad \Delta T_{ad}(T)_{\Delta p} = -\int_{p_i}^{p_f} \left(\frac{T}{C(T,p)}\right)_p \left(\frac{\partial V(T,p)}{\partial T}\right)_p dp$$

isobaric thermal expansion: $\alpha = \frac{1}{V}\left(\frac{\partial V(T,p)}{\partial T}\right)_p$

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Bispectral Powder Diffractometer

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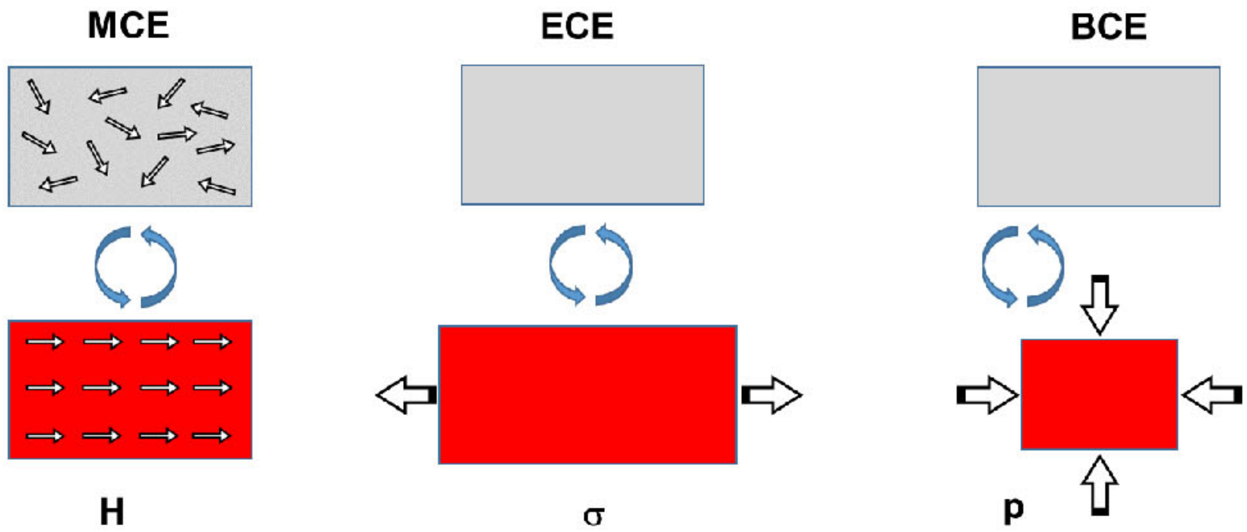
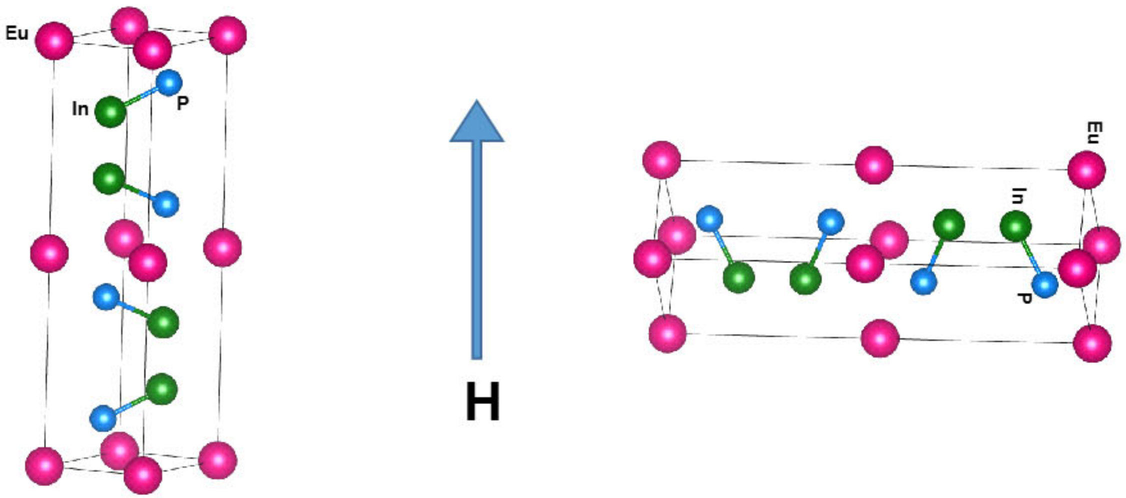


maybe also for MCE materials …

MAGiC
Magnetism Single-Crystal Diffractometer

…complex interplay of magnetism and superconductivity in unconventional superconductors (pnictides, cuprates, heavy-fermion intermetallics and organic superconductors), multifunctionality in magnetically-induced ferroelectrics, and frustrated or low-dimensional magnetic materials featuring strong magnetic correlations or spin-liquid-like quantum coherent ground states. Some of these materials cannot be studied at present due to the lack of large enough single-crystalline materials and/or weakness of the magnetic contribution to the diffraction pattern.



Anisotropic (Rotational) MCE (RMCE)

$$\Delta S_{diff}(T) = S_M(T, H_{ab}) - S_M(T, H_c) =$$

$$[S_M(T, H_{ab}) - S_M(T,0)] - [S_M(T, H_c) - S_M(T,0)] =$$

$$\boxed{\Delta S_M(T, H_{ab}) - \Delta S_M(T, H_c)}$$

MAGiC? (+DREAM)

project: *Extraordinary electronic transport in magnetic topological insulators and semimetals*

Principal Investigator - prof. Dariusz Kaczorowski
Institute of Low Temperature and Structure Research, Polish Academy of Sciences, Wrocław, Poland

IFM PAN coordinator – prof. Tomasz Toliński
Institute of Molecular Physics, Polish Academy of Sciences, Poznań, Poland

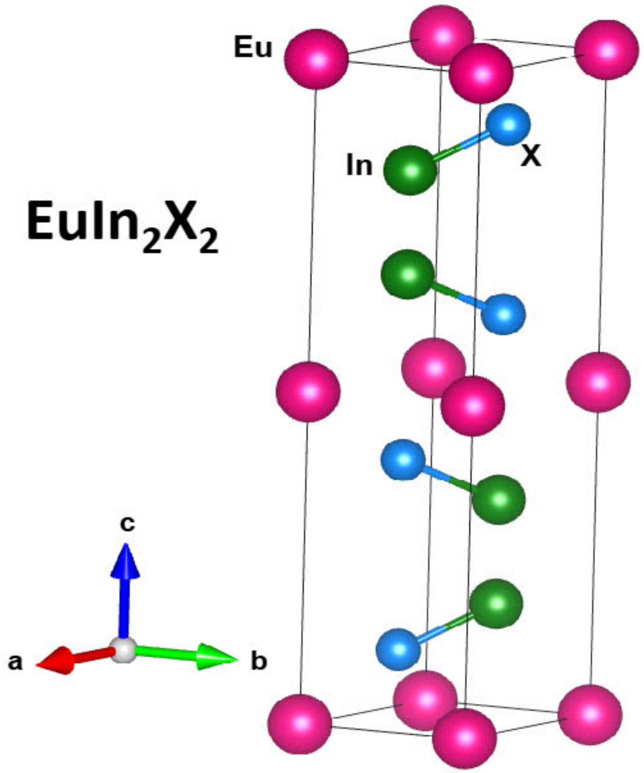
supported by the National Science Centre (Poland) under research grant 2021/41/B/ST3/01141

MAGiC? (+DREAM)

$EuIn_2As_2$

- hexagonal unit cell (space group P6_3/*mmc*)
- lattice parameters: *a*=b=4.207Å, *c*=17.889Å
- it shows AFM order below T_N=16.1K with the Eu magnetic moments aligned ferromagnetically within the *ab* plane but antiparallel in respect to adjacent Eu layers along the *c*–axis
- it was predicted to be an axion insulator for magnetic moments within the *ab* plane, and a HOTI for easy magnetization direction along the *c* axis

$EuIn_2X_2$



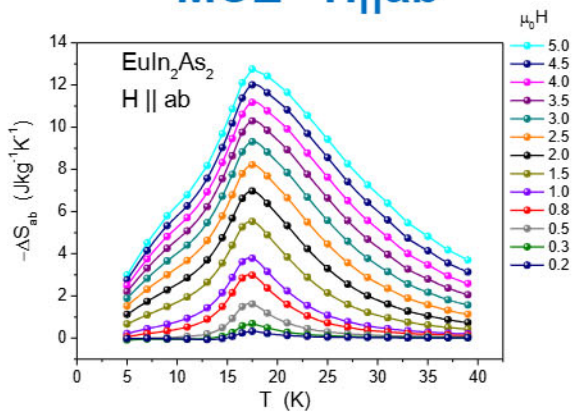
$EuIn_2P_2$

- hexagonal unit cell (space group P6_3/*mmc*)
- lattice parameters: *a*=*b*=4.083Å, *c*=17.595Å
- it orders magnetically at T_C=24K with the Eu magnetic moments aligned ferromagnetically within the *ab* plane but tilted alternately along the *c*–axis
- *ab initio* simulation: it can be a semimetal with a Weyl point for in-plane magnetization and with a Weyl nodal-line for magnetization oriented along *c*-axis)

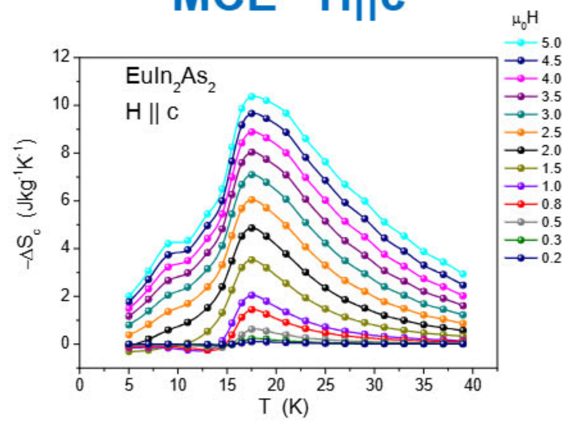
$EuIn_2As_2$

MAGiC? (+DREAM)

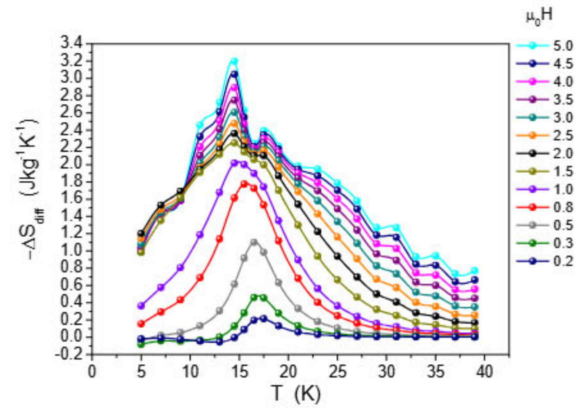
MCE H||ab

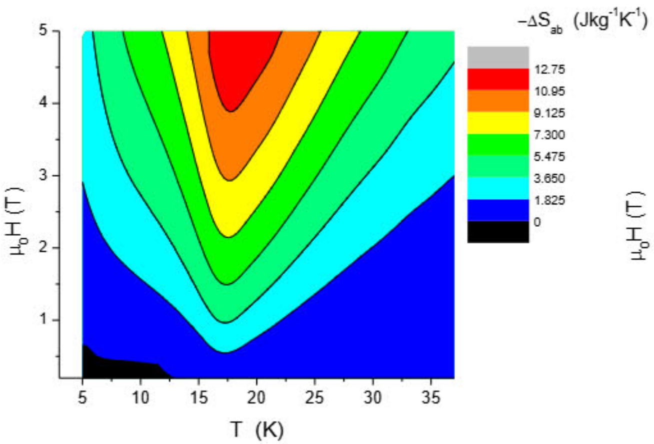


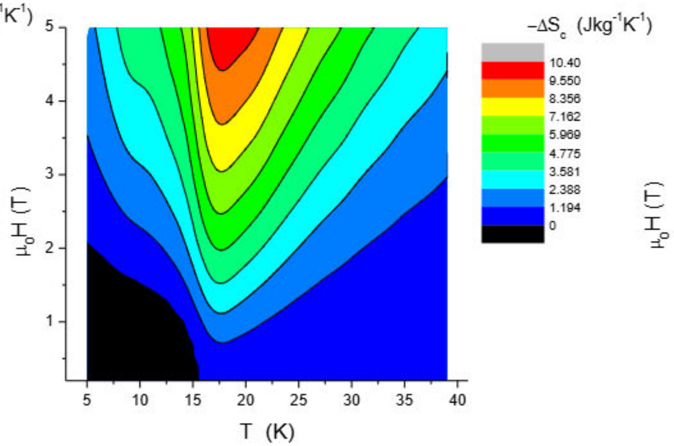
MCE H||c

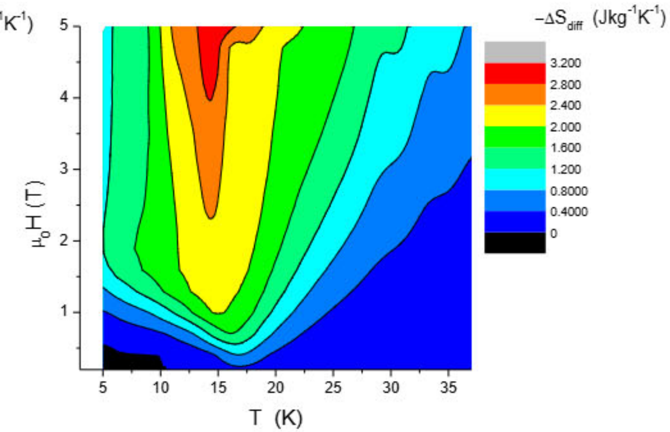


RMCE



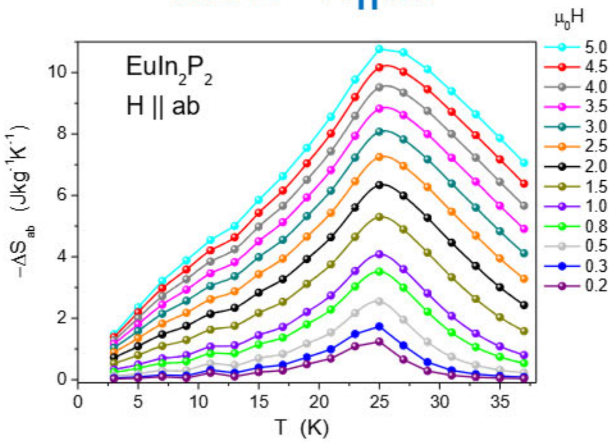




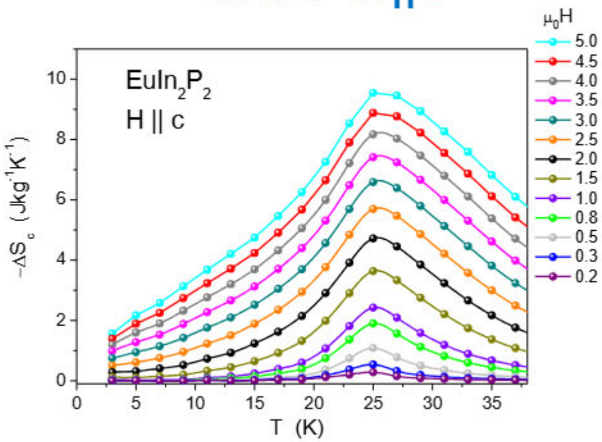


MCE vs. RMCE $EuIn_2P_2$ MAGiC? (+DREAM)

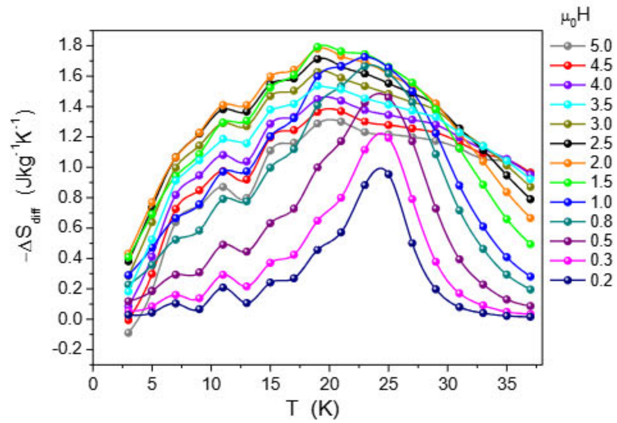
MCE H||ab

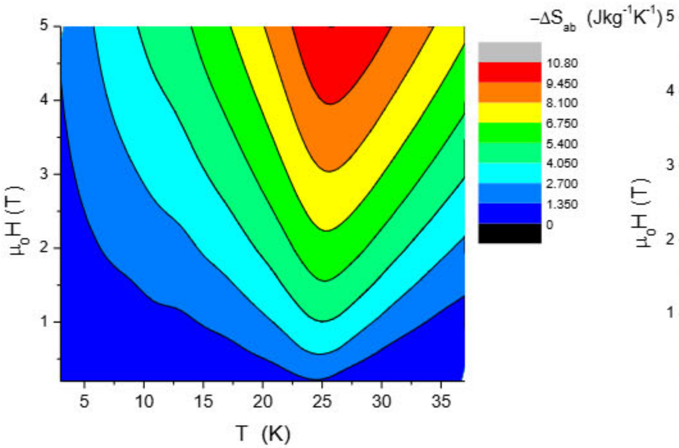


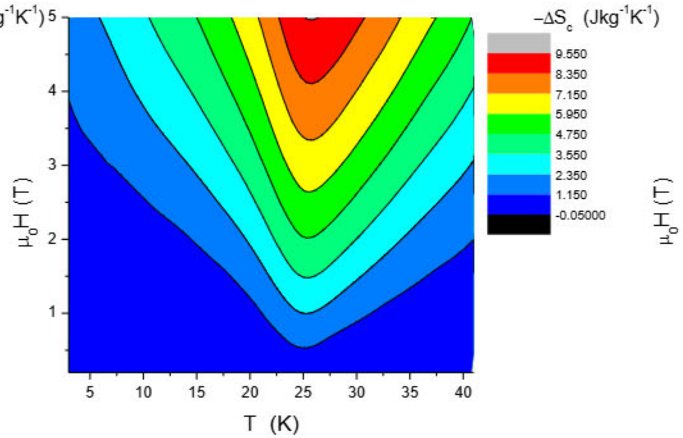
MCE H||c

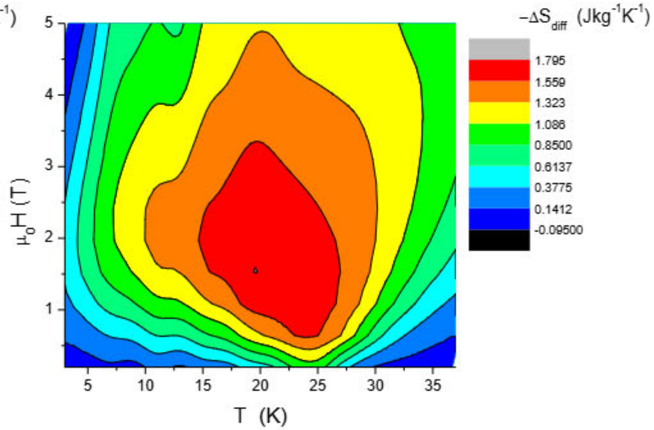


RMCE



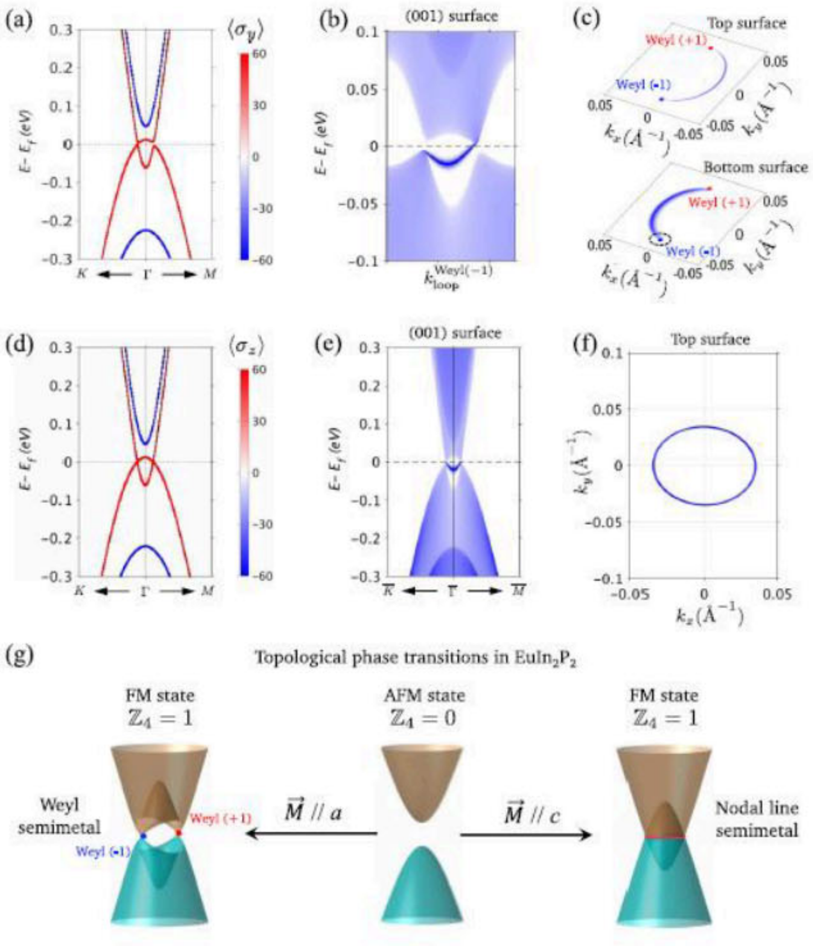




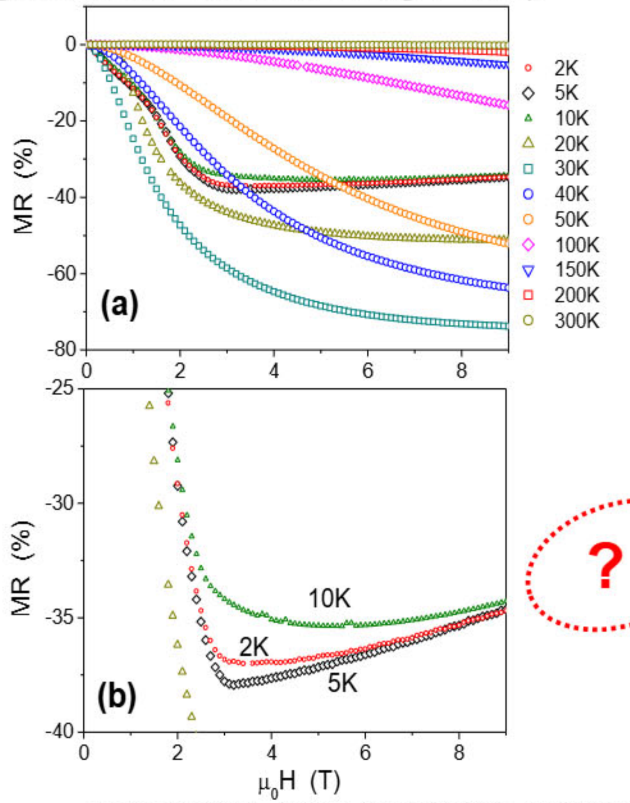


$EuIn_2P_2$ - small crystals, MAGiC? (+DREAM)

ab initio simulation: it is a trivial insulator with $\mathbb{Z}_4$=0 but magnetic field can transform it to Weyl fermion with $\mathbb{Z}_4$=1 (it can be a Weyl point for in-plane magnetization and a Weyl nodal-line for magnetization oriented along *c*-axis)



Sarkar et al., Phys. Rev. Mat. 6, 044204 (2022)



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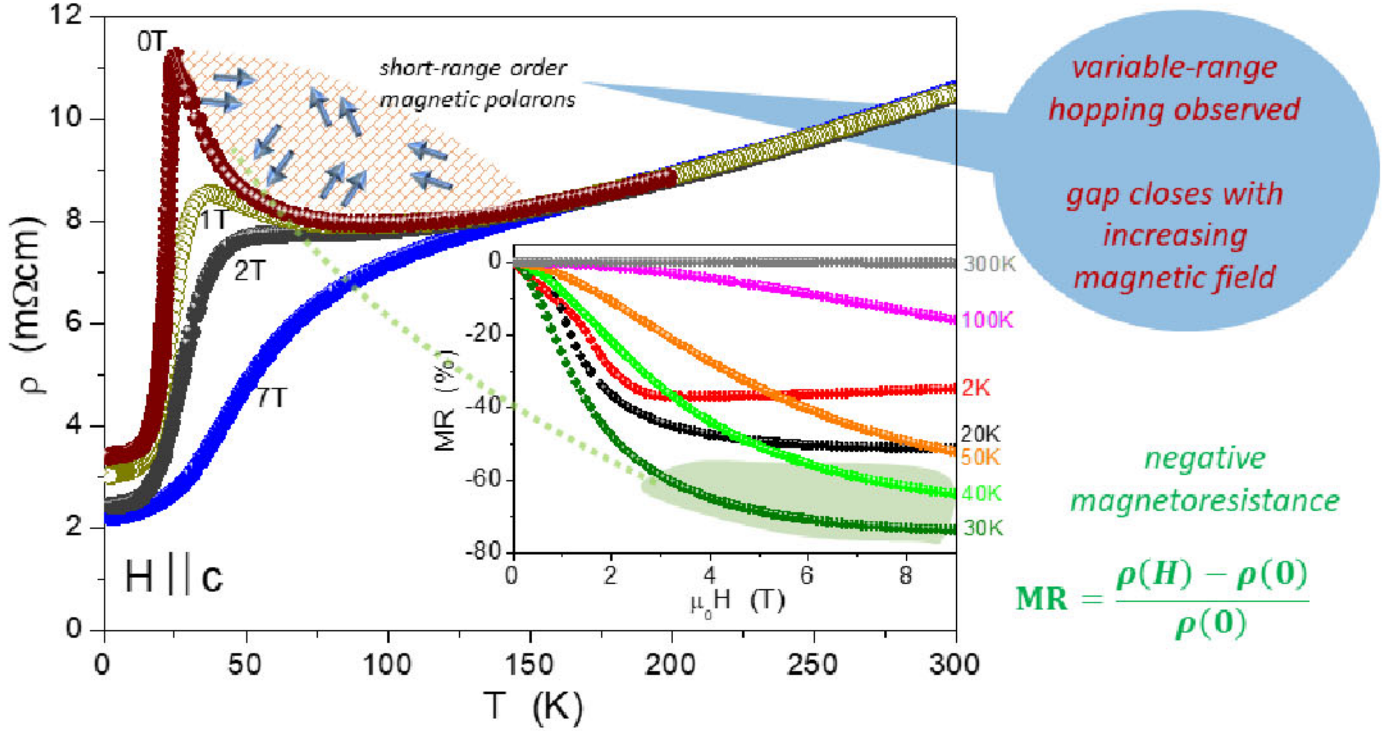
Toliński et al., Phys. Rev. B 110, 174425 (2024)

Short- and long-range order in Eu-based compounds



$$\mathbf{MR} = \frac{\rho(H) - \rho(0)}{\rho(0)}$$

T. Toliński, Q. Ain, K. Synoradzki, A. Łapiński, S. Zięba, T. Romanova, K. Singh, O. Pavlosiuk, P. Wiśniewski, D. Kaczorowski, Phys. Rev. B 110, 174425 (2024)

MIRACLES
Backscattering Spectrometer

The configuration of the long time-of-flight primary spectrometer and the large backscattering secondary spectrometer will give unprecedented energy resolution in accelerator-driven neutron source spectrometers, with an outstanding performance and versatility due to the flexibility of tuning energy resolution from a large dynamic range and performing quasielastic and inelastic neutron scattering experiment (QENS and INS) in a wide collection of operation modes.

Inelastic Neutron Scattering – $CeCoAl_4$

Journal of Magnetism and Magnetic Materials 345 (2013) 243–248

Crystal field manifestation in inelastic neutron scattering, magnetic susceptibility and specific heat of the antiferromagnetic $CeCoAl_4$

T. Toliński [a,*], K. Synoradzki [a], A. Hoser [b], S. Rols [c]

[a] Institute of Molecular Physics, Polish Academy of Sciences, Smoluchowskiego 17, 60-179 Poznań, Poland
[b] Helmholtz-Zentrum, Glienicker Straße 100, D-14109 Berlin, Germany
[c] Institut Laue Langevin, 6 rue Jules Horowitz B.P. 156 F-38042, Grenoble Cedex 9, France

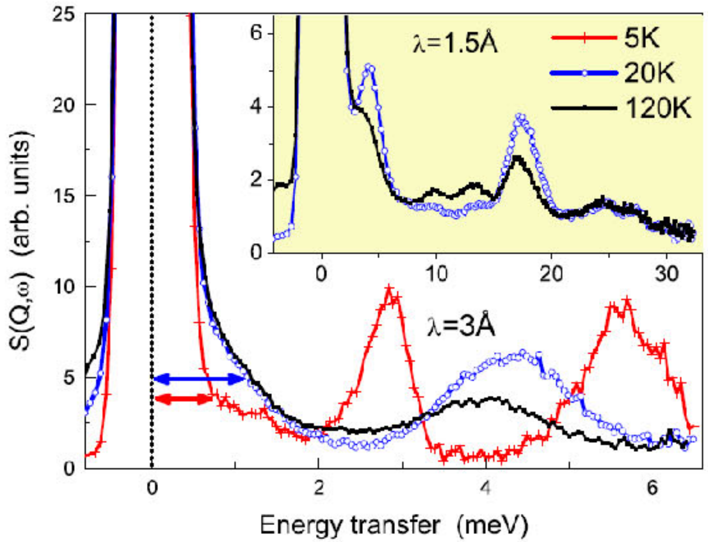
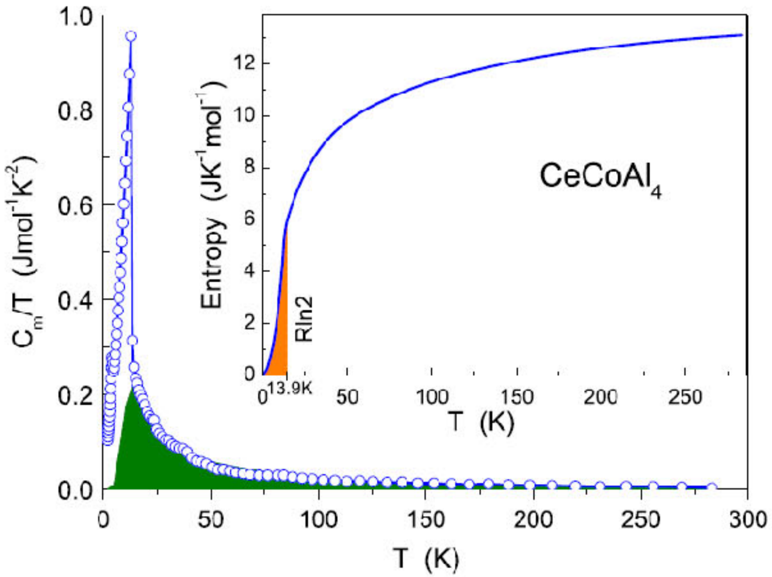


Fig. 2. The temperature effect on the INS spectra of $CeCoAl_4$ at a wavelength of $\lambda_I = 3$ Å and $\lambda_I = 1.5$ Å (inset). Arrows show schematically the drop of the quasielastic contribution below the ordering temperature.



The disappearance of the quasielastic contribution below T_N and its small linewidth Γ=2 suggest that $T_K < T_N$, therefore the Kondo interactions may be not relevant in the compounds studied. The analysis of the magnetic specific heat has shown that the magnetic entropy reaches the theoretical value of Rln2 close to T_N=13.5K, which would be not expected for strong Kondo interactions.

Inelastic Neutron Scattering – $CeCoAl_4$

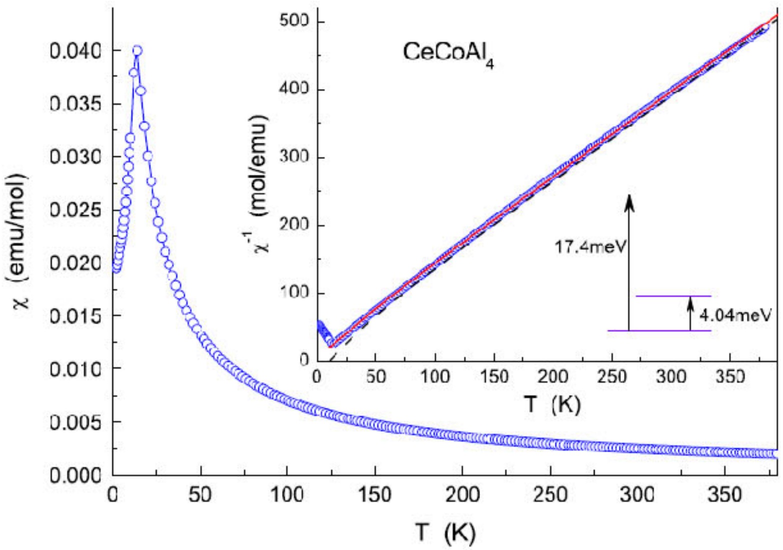


Fig. 4. Zero field cooled magnetic susceptibility of $CeCoAl_4$ measured in $H=1$ kOe. Inset: Inverse magnetic susceptibility (circles) and a calculation with the CEF model (solid line). The dashed line corresponds to the molecular field parameters obtained from the analysis of the INS spectra at 5 K (see text).

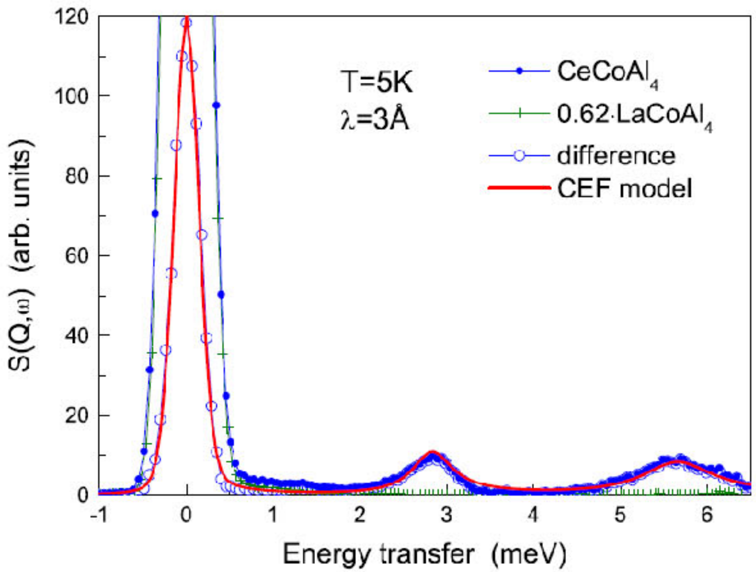


Fig. 3. The INS spectrum of $CeCoAl_4$ at 5 K, before and after subtraction of the spectrum of the nonmagnetic reference compound $LaCoAl_4$. The solid line drawn through the open symbols represents a fit with a CEF model discussed within the text.

$$H_{CEF} = B_2^0 O_2^0 + B_2^2 O_2^2 + B_4^0 O_4^0 + B_4^2 O_4^2 + B_4^4 O_4^4$$

$$\chi_{\mathrm{CEFi}} = \frac{N(g_J\mu_B)^2}{Zk_BT}\left(\sum_{\substack{n,m\\E_n=E_m}} |\langle n|J_i|m\rangle|^2 e^{-E_n/k_BT} + 2k_BT\sum_{\substack{n,m\\E_n\neq E_m}} \frac{|\langle n|J_i|m\rangle|^2}{E_m-E_n} e^{-E_n/k_BT}\right)$$

$$S(Q,\varepsilon) = f^2(Q)\frac{\varepsilon}{1-\exp(-\varepsilon/kT)}\left(\sum_{\substack{n,m\\E_n=E_m}} \chi_C^{nm} F(\varepsilon,\Gamma_{nm}) + \sum_{\substack{n,m\\E_n\neq E_m}} \chi_{VV}^{nm} F(\varepsilon-\varepsilon_{nm},\Gamma_{nm})\right)$$

Identification of the crystal field levels in intermetallics like:
$PrTGe_3$, $NdTGe_3$ (T – transition metal), $R_3Co_4Ge_{13}$, $R_5Co_4Ge_{13}$, and Pr_2Rh_3Ge.
For example, for Pr_2Rh_3Ge, the intriguing and still unexplained question remains the reason for the increase in the effective electron mass. The most probable cause of the formation of the heavy-fermionic state in Pr_2Rh_3Ge can be a mechanism related to dynamic crystal field interactions.

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+

MAGiC
Magnetism Single-Crystal Diffractometer

…complex interplay of magnetism and superconductivity in unconventional superconductors (pnictides, cuprates, heavy-fermion intermetallics and organic superconductors), multifunctionality in magnetically-induced ferroelectrics, and frustrated or low-dimensional magnetic materials featuring strong magnetic correlations or spin-liquid-like quantum coherent ground states. Some of these materials cannot be studied at present due to the lack of large enough single-crystalline materials and/or weakness of the magnetic contribution to the diffraction pattern.

SUMMARY

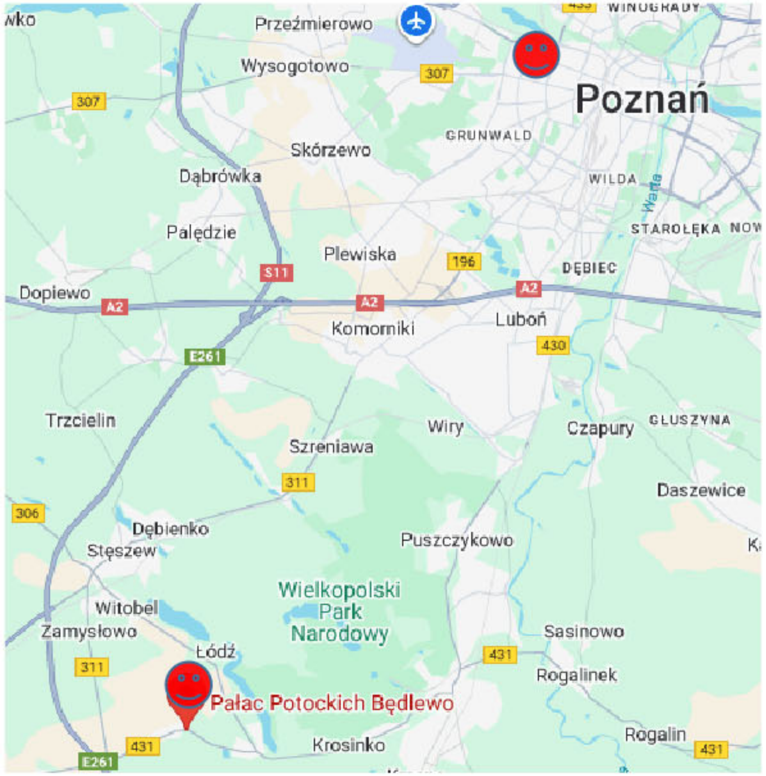
INSTRUMENT	INSTRUMENT FEATURES	POSSIBLE EXPERIMENTS
DREAM **Bispectral Powder Diffractometer**	Magnetism: studies of complex magnetic structures, weak magnetic moments, multiferroics, magneto-elastic coupling, interplay between magnetic order and superconductivity, quantum criticality, magnetism at high pressure, orbital and charge ordering.	Laves Phases and other MCE/BCE/ECM materials: Importance of simultaneous information on the crystalline and magnetic structure
MAGiC (+ DREAM) **Magnetism Single-Crystal Diffractometer**	…heavy-fermion intermetallics and …. materials featuring strong magnetic correlations …. Some of these materials cannot be studied at present due to the lack of large enough single-crystalline materials and/or weakness of the magnetic contribution to the diffraction pattern.	Magnetic structures of MCE and anisotropic MCE materials; Nodal line semimetals (e.g. $EuIn_2P_2$); Short range order in topological materials; non-colinear magnetic order
MIRACLES (+MAGIC) **Backscattering Spectrometer**	versatility due to the flexibility of tuning energy resolution from a large dynamic range and performing quasielastic and inelastic neutron scattering experiment (QENS and INS) in a wide collection of operation modes.	Inelastic scattering to determine CEF levels scheme; Kondo physics ($PrTGe_3$, $NdTGe_3$, $R_3Co_4Ge_{13}$, $R_5Co_4Ge_{13}$, and Pr_2Rh_3Ge, …)

Neutron Scattering and Complementary Methods in Modern Research

27 June 2026 to 1 July 2026

Research and Conference Centre of the Institute of Mathematics of the Polish Academy of Sciences in Będlewo near Poznań

Neutron Scattering
and Complementary Methods
in Modern Research



The European Conference

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Celebrating 50th Anniversary

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University of Gothenburg
*Gothenburg, **Sweden***

Saroj Prasad Dash
Chalmers University of Technology
*Gothenburg, **Sweden***

Massimiliano Esposito
University of Luxembourg
*Esch-sur-Alzette, **Luxembourg***

Peter Fischer
Lawrence Berkeley National Lab
*Berkeley, California, **USA***
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*Santa Cruz, California, **USA***

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*Ostrava, **Czech Republic***

Amalia Patanè
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