MA 13: Focus Session: New Perspectives for Adiabatic Demagnetization Refrigeration in the Kelvin and sub-Kelvin Range (joint session TT/MA)

Efficient cooling into the Kelvin and sub-Kelvin range is a long-standing challenge relevant to both fundamental research and future quantum technologies. The standard cooling cycle based on vapor compression exploits expensive and rare helium. Low-temperature physicists world-wide are presently looking for cheaper and accessible alternatives, not to mention the need of compact cooling technology for desktop quantum technology, or special requirements for applications such as space missions and scanning tunneling microscopes. One of the key candidates is adiabatic demagnetization refrigeration (ADR). ADR is based on magnetic solids with a huge magnetocaloric effect and requires no helium. Even if paramagnetic salts are known and used for ADR applications for almost a century, there is an ongoing quest for materials with better magnetocaloric and mechanical properties, thermal conductivity, and vacuum compatibility. In this symposium, new fundamental ideas and the recent successful design and characterization of quantum materials for improved ADR will be highlighted. These materials exploit collective phenomena in correlated electron systems, such as the concept of geometrically frustrated magnetism to push the entropy to low temperatures, as well as heavy-fermion, and quantum-critical states.

Organizers: Andreas Honecker (CY Cergy Paris Université) and Jürgen Schnack (Universität Bielefeld)

Time: Tuesday 9:30–13:15 Location: HSZ 03

Invited Talk MA 13.1 Tue 9:30 HSZ 03

Self-cooling molecular spin quantum processors — •Marco Evangelisti[1], Fernando Luis[1], Elias Palacios[1], David Aguilà[2], and Guillem Aromí[2] — [1]INMA, CSIC & Universidad de Zaragoza, Spain — [2]Dept. Química Inorgànica, Universidad de Barcelona, Spain

Cryogenic refrigeration is crucial for a wide range of emerging applications in the field of quantum technologies. Indeed, thermal energy must be minimized to avoid the excitation of vibrational motions that could disturb quantum operations. Synthetic chemistry provides a sophisticated methodology for the design and synthesis of materials displaying a wide variety of properties. Molecular materials are capable of excellent and unique characteristics that can be exploited either for caloric cooling[1] or spin-based quantum computing[2]. However, these features are not yet being implemented as such to act together within the same material, that is, at the molecular scale. Here, we show that a spin qubit (or qudit) can be brought into proximity with a spin centre that acts as a cooler. To this end, we make use of rare-earth-based asymmetric molecular dimers. A chemically engineered structural asymmetry introduces different coordination environments for each metal ion, operating similarly as for molecular quantum gates reported by some of us[3]. This strategy allows selecting individually both constituent ions, leading to e.g. the direct observation of the cooling of a single Er(III) ion qubit, or a Tm(III) electronuclear spin qudit, driven chiefly by the demagnetization of a single Gd(III) ion located within the same molecule.

[1] Dalton Trans. 39, 4672 (2010)
[2] Nat. Chem. 11, 301 (2019)
[3] Phys. Rev. Lett. 107, 117203 (2011)

Invited Talk MA 13.2 Tue 10:00 HSZ 03

Triangular rare-earth borates for milli-Kelvin adiabatic demagnetization refrigeration — •Philipp Gegenwart — Experimental Physics VI, Center for Electronic Correlations and Magnetism, University of Augsburg

Adiabatic demagnetization refrigeration (ADR) is a classical cooling technique with renewed recent attention as alternative to costly and elaborate $^3He/^4He$ dilution refrigeration. Established water containing ADR salts suffer from chemical instability which requires delicate treatment to avoid degradation and ensure good thermal contact. Water-free $KBaYb(BO_3)_2$ is an excellent alternative with high entropy density that allows ADR to below 20 mK [1]. Sintered pellets with silver powder admixture to ensure good thermal coupling are easy to manufacture, inexpensive and long-term stable even upon heating up to 700°C, enabling also ultra-high vacuum applications. $KBaYb(BO_3)_2$ belongs to a family of rare-earth-based borates with triangular arrangement of magnetic moments. We discuss the impact of geometrical frustration and structural randomness on its low-temperature properties and demonstrate the enormous tunability of cooling power and operating temperature by chemical substitution.

[1] Y. Tokiwa, S. Bachus, K. Kavita, A. Jesche, A.A. Tsirlin, and P. Gegenwart, Commun. Mater. 2 (2021) 42.

Invited Talk MA 13.3 Tue 10:30 HSZ 03

A millikelvin scanning tunnelling microscope in ultra-high vacuum with adiabatic demagnetisation refrigeration — •Ruslan Temirov — Peter Grünberg Institut (PGI-3), Forschungszentrum Jülich, Germany — University of Cologne, Institute of Physics II, Cologne, Germany

Scanning tunnelling microscopes (STMs) operating in ultra-high vacuum (UHV) and low-temperature conditions are used widely for imaging and precise manipulation of surface nanostructures. A growing interest in studies of quantum-coherent phenomena in such nanostructures stimulates the development of STMs that operate at very low millikelvin temperatures. This contribution presents the design of a first-ever UHV STM cooled by adiabatic demagnetisation refrigeration (ADR) to below 30 mK. The use of ADR makes the STM design modular and helps it to reach a remarkable degree of mechanical stability. Tunnelling spectra collected on an atomically clean superconducting Al(100) surface reveal that the electronic temperature of the tunnelling junction is less than 80 mK. The inelastic electron tunnelling spectroscopy of an individual electron spin performed in magnetic fields of up to 8 Tesla validates the STM capabilities for quantum nanoscience research.

15 min. break

Invited Talk MA 13.4 Tue 11:15 HSZ 03

ADR cryostats in low temperature physics and their applications — •Doreen Wernicke — Entropy GmbH, Gmunder Str. 37a, 81379 München

Entropy GmbH is a company founded in 2010 in Munich, Germany, specializing in the development and manufacture of low temperature cryostats. All Entropy cryostats are based on closed-cycle pre-cooling to temperatures below 3K. Further cooling stages such as ADR units, Joule-Thomson stages, and dilution refrigerators including electronics and software are proprietary developments. The modular design of all cryostats offers the possibility of adaptation to many different experiments and applications. One of Entropy's most common products are the ADR cryostats. The presentation will explain the principle of ADR cooling and features such as base temperature and holding time at operating temperature. Applications for low temperature device operation such as various types of superconducting detectors (TES, MKIDs, SQUIDs, SNSPDs) and Qubit characterization will be presented to demonstrate the performance and limitations of adiabatic demagnetization refrigeration.

Invited Talk MA 13.5 Tue 11:45 HSZ 03

Frustrated dipolar materials for low-temperature magnetic refrigeration — •Mike Zhitomirsky — Institute of Interdisciplinary Research, CEA-Grenoble, France

Low-temperature refrigeration is crucial for emergent quantum-information technologies and other scientific applications that outstretch from space telescopes to medicine. This growing demand fuels an interest in alternative low-temperature techniques including the adiabatic demagnetization refrigeration. The existing ADR technologies for the sub-Kelvin range utilize dilute paramagnetic salts of Cr and Fe magnetic ions, which have limited efficiency at higher temperatures. I shall discuss general directions of the ongoing search of prospective refrigerant materials by exploring collective effects in systems of interacting magnetic moments as opposed to noninteracting moments in paramagnetic salts. Specifically, I focus on geometrically frustrated magnets with a residual ground-state degeneracy as well as on dipolar magnets. I present new experimental and theoretical results obtained recently in Grenoble for two dipolar materials: $Yb_3Ga_5O_{12}$, which is a spin-1/2 dipolar ferromagnet on a hyper-Kagome lattice, and $GdLiF_4$, which exhibits a hidden magnetic frustration. The striking properties of the latter material including a fractional magnetization plateau demonstrate importance of new magnetocaloric materials not only for applied but also for basic research in magnetism.

MA 13.6 Tue 12:15 HSZ 03

ADR based sub-Kelvin cryostats for applied quantum technologies — •Pau Jorba[1], Felix Rucker[1], Steffen Säubert[1], Alexander Regnat[1], Jan Spallek[1], and Christian Pfleiderer[2] — [1]kiutra GmbH, Flößergasse 2, D-81369 München, Germany — [2]Physik-Department, Technische Universität München, D-85748 Garching, Germany

In view of the increasing demand for the cooling of quantum electronic devices, the development of scalable cooling solutions that provide low temperatures independent of rare helium-3 will be mandatory for the adoption and commercial use of next-generation quantum technologies. We present novel ADR based sub-Kelvin cryostats[1] specifically developed for the characterization and operation of quantum devices. We address how known challenges of ADR systems such as limited hold time and magnetic stray fields can be overcome. Specifically, we describe how continuous sub-Kelvin cooling and wide-range temperature control can be achieved by combining multiple ADR units and mechanical thermal switches. We also present a novel sample loader mechanism[2] that allows taking advantage of the solid-state nature of ADR and to cool samples from room temperature to 100 mK in less than 3 hours.

[1] Regnat et al. (2018) Cryogen-free cooling apparatus (EP 3163222). European Patent Office.
[2] Spallek et al. (2022) System and method for inserting a sample into a chamber (EP 3632560). European Patent Office.

MA 13.7 Tue 12:30 HSZ 03

ADR below the ordering temperature in triangular $KBaGd(BO_3)_2$ — •Noah Winterhalter-Stocker[1], Alexander Bellon[1], Fabian Hirschberger[1], Sebastian Bachus[1], Sebastian Erdmann[1], Alexander Tsirlin[1,2], Yoshifumi Tokiwa[1,3], Anton Jesche[1], and Philipp Gegenwart[1] — [1]Experimental Physics VI, Center for Electronic Correlations and Magnetism, University of Augsburg, D-86159 Augsburg, Germany — [2]Felix Bloch Institute for Solid-State Physics, Leipzig University, D-04103 Leipzig, Germany — [3]Advanced Science Research Center, Japan Atomic Energy Agency, Tokai, Ibaraki 319-1195, Japan

Compared to the triangular ADR magnet $KBaYb(BO_3)_2$ [1] the isotructural sister compound $KBaGd(BO_3)_2$ with spin 7/2 moments has a three times enhanced magnetic entropy density of 192 $mJK^{-1}cm^{-3}$. We report a low-temperature magnetic and thermodynamic investigation of polycrystalline $KBaGd(BO_3)_2$ down to 50 mK. Specific heat indicates an antiferromagnetic phase transition at 263 mK, strongly broadened due to randomness and frustration, that becomes suppressed beyond 0.5 T. Further increase of magnetic field shifts the available entropy of R log 8 towards high temperatures. Interestingly, ADR of a pellet utilizing the same setup as used in [1] reveals a minimal temperature if $T_{\min}$=122 mK that is more than twice below T_N along with a hold time of more than 8 hours. The combination of minimal temperature and entropy density in $KBaGd(BO_3)_2$ is outstanding among known ADR materials.

[1] Y. Tokiwa *et al.*, Communications Materials **2.1**, 1-6 (2021)

MA 13.8 Tue 12:45 HSZ 03

Magnetocaloric properties of $(RE)_3Ga_5O_{12}$ (RE=Tb, Gd, Nd, Dy) — Markus Kleinhans[1], Klaus Eibensteiner[1,2], Jon Leiner[1], •Christoph Resch[1], Lukas Worch[1], Marc Wilde[1], Jan Spallek[1,2], Alexander Regnat[1,2], and Christian Pfleiderer[1] — [1]Physik Department, Technical University Munich, D-85748 Garching, Germany — [2]kiutra GmbH, Rupert-Mayer-Str. 44, D-81379 Munich, Germany

We report the characteristic magnetic properties of several members of the rare earth garnet family, $Gd_3Ga_5O_{12}$ (GGG), $Dy_3Ga_5O_{12}$ (DGG), $Tb_3Ga_5O_{12}$ (TGG), and Nd_3Ga5O_{12} (NGG), and compare their relative potential utility for magnetocaloric cooling, including their minimal adiabatic demagnetization refrigeration (ADR) temperatures and relative cooling parameters. A main objective of this work was to find potential improvements over the magnetocaloric properties of GGG for use in low temperature ADR cryostats. Using Tb^{+3} and Dy^{+3} in the RE-site oers, in principle, a higher saturation magnetization and Nd^{+3} gives a lower de Gennes factor and therefore potentially low transition temperature. Our results show that $Dy_3Ga_5O_{12}$ yields an optimal relative cooling parameter (RCP) at low applied fields and a low transition temperature, which would allow for the design of more efficient ADR cryostats.

[1] M. Kleinhans et al., arXiv/2204.01752; Phys. Rev. Appl. in press (2022).

MA 13.9 Tue 13:00 HSZ 03

Study of the large rotational magnetocaloric effect in $Ni(en)(H_2O)_4SO_4{\cdot}2H_2O$ — •Róbert Tarasenko, Petro Danylchenko, Erik Čižmár, Vladimír Tkáč, Alexander Feher, Alžbeta Orendáčová, and Martin Orendáč — Institute of Physics, Faculty of Science, Pavol Jozef Šafárik University, Park Angelinum 9, 041 54 Košice, Slovakia

The title compound $Ni(en)(H_2O)_4SO_4{\cdot}2H_2O$ (*en* = ethylenediamine) has been identified as a spin-1 paramagnet with the nonmagnetic ground state introduced by the easy-plane anisotropy $D/k_B = 11.6$ K with $E/D = 0.1$ and negligible exchange interactions $J \approx 0$. We present an experimental study of the rotational magnetocaloric effect (MCE) in single crystals at temperatures above 2 K, associated with adiabatic crystal rotation between the easy plane and hard axis in magnetic fields up to 7 T. The experimental observations are completed with *ab* initio calculations of the anisotropy parameters. Theoretical simulations of the rotational MCE in the $S = 1$ paramagnet were performed and the simulations were compared with experimental data. A large rotational magnetic entropy change ≈16.9 $Jkg^{-1}K^{-1}$ has been achieved in 7 T. The adiabatic rotation of the crystal in 7 T starting at the initial temperature of 4.2 K leads to the cooling of the sample down to 0.34 K, which suggests the application of this material in low-temperatures cooling. Our simulations show that $S = 1$ Ni(II)-based systems with easy-plane anisotropy can have better rotational magnetocaloric properties than costly materials containing rare-earth elements.

Supported by project No. APVV-18-0197.