MA 51: Caloric Effects in Ferromagnetic Materials

Time: Friday 9:30–13:15 Location: H 2013

Invited Talk MA 51.1 Fri 9:30 H 2013
Functional and microstructural design of multicaloric Heusler alloys — •Franziska Scheibel[1], Lukas Pfeuffer[1], Andreas Taubel[1], Christian Lauhoff[2], Philipp Krooss[2], Thomas Niendorf[2], and Oliver Gutfleisch[1] — [1]TU Darmstadt, Darmstadt, Germany — [2]University of Kassel, Kassel, Germany

Due to their first-order magnetostructural transition (FOMST), Ni-Mn-based Heusler alloys exhibit a variety of exploitable phenomena, such as magneto-, elasto-, and multicaloric effects. Functional performance and FOMST can be tailored by compositional and microstructural design. Besides a high cyclic caloric effect as a key requirement for solid-state refrigeration, shaping, functional fatigue, and scalability are key criteria when the material is to reach application. The mechanical strength and cyclic stability can be improved by grain refinement or precipitate formation through doping. By combining the two, it was even possible to achieve a cycle stability of more than 16000 cycles. [1]. Powder-based techniques (such as additive manufacturing (AM), spark plasma sintering, or hot compaction), can be used to achieve scalability and formability. We investigated the entire processing chain from gas-atomized, spherical powder to post-processed parts. In particular, this is important for AM to optimize microstructure, FOMST, and caloric effect [2,3]. We thank the ERC Adv. Grant "CoolInnov", and the CRC/TRR 270 "HoMMage" for funding.

[1] L. Pfeuffer et al., Acta Mater. 221, 117390 (2021)
[2] F. Scheibel et al., Adv. Eng. Mater., 2200069 (2022)
[3] F. Scheibel et al., MTLA 29, 101783 (2023)

MA 51.2 Fri 10:00 H 2013
Impact of spin-entropy on the thermoelectric properties of a 2D magnet — •Alessandra Canetta — Institute of Condensed Matter and Nanosciences, Université catholique de Louvain (UCLouvain), 1348 Louvain-la-Neuve, Belgium

The heat-to-charge conversion efficiency of thermoelectric materials is closely linked to the entropy per charge carrier. Therefore, finding ways to increase this entropy could lead to new design strategies for highly efficient energy harvesters or spot-cooling devices. To this end, magnetic materials are a promising choice because their carrier entropy is boosted by a spin degree of freedom. To study this capability, a model magnetic system with high tunability of its magnetic order is required. The A-type antiferromagnet CrSBr fits these requirements and we investigate how spin entropy impacts heat-to-charge conversion in this 2D layered magnet. We perform simultaneous measurements of electrical conductance and thermocurrent while changing magnetic order using temperature and magnetic field as tuning knobs. We find a 10% decrease of the Seebeck coefficient when establishing antiferromagnetic order. We further reveal a drastic reduction of the Seebeck coefficient by one order of magnitude below a temperature of 40 K, which we attribute to a drop in spin entropy due to a spin-freezing process. Our results highlight the sizeable impact of spin entropy on thermoelectric properties and suggest thermoelectric measurements as a sensitive tool to study magnetic phase transitions in low-dimensional magnets.

MA 51.3 Fri 10:15 H 2013
A temperature model for the Magnetic Hyperthermia — •Viorica Monica Moisiuc and Iordana Astefanoaei — Faculty of Physics, Alexandru Ioan Cuza University of Iasi, Romania

Magnetic hyperthermia is an important medical technique which uses therapeutic heat from the magnetic nanoparticles when an external high frequency magnetic field was applied. The temperature developed within tumoral tissues by the nanoparticles can be controlled by the fine control of the magnetic field parameters: i) frequency and ii) the amplitude of the field. In the last years, this topic receives a special attention in all theoretical and experimental studies. The spatial distribution of magnetic nanoparticles in the tumoral tissue significantly influences the temperature during this treatment. In this work, a concentric tissue geometry (tumoral tissue surrounded by the healthy tissue) was considered. The magnetic nanoparticles with different properties are distributed non-uniformly in the tumor tissue. The thermal response of the tumoral tissue heated by the magnetic nanoparticles in a high-frequency electromagnetic field was analyzed by the analytical and numerical computations. Bioheat transfer equation was solved to obtain the temperature field within tissues. The spatio-temporal evolution of the temperature in correlation with the nanoparticles volume fraction was analyzed for different types of magnetic particles injected into the tumor tissue. The mathematical model allows the computation of the hyperthermic temperature for nanoparticle systems with the following particularities: different concentrations, sizes and magnetic and thermal properties (maghemite, magnetite).

MA 51.4 Fri 10:30 H 2013
Measuring the Anomalous Ettingshausen effect in the magnetic Weyl semimetal Co_2MnGa — •Mohammadali Razeghi and Pascal Gehring — IMCN/NAPS, Université Catholique de Louvain, Louvain-la-Neuve, Belgium

Magnetic Weyl semimetals (MWSMs) have been praised for their exceptional electrical transport properties. Often overlooked, energy and heat transport properties remain to be investigated. Especially coupled properties, such as heat to charge conversion, are expected to be enhanced by topological effects in MWSMs. The Heusler alloy Co_2MnGa has been reported to show MWSM state at room temperature with large anomalous Hall and Nernst coefficients.

In this work, we explore the cooling capability of Co_2MnGa devices using Scanning Thermal Microscopy (SThM). SThM allows us to locally map temperature variations and to trigger temperature driven electrical transport effects in micropatterned Co_2MnGa devices. We explore thermomagnetic and thermoelectric effects and their interplay with geometry, charge density and magnetic field. In particular we observe a pronounced anomalous Ettingshausen effect and reveal a strong contribution from band topology to its magnitude. We furthermore quantify the cooling and heating efficiency of the topological alloy and assess its potential for realization of heat management devices.

MA 51.5 Fri 10:45 H 2013
Multicaloric effect of powder-in-tube Heusler material — •T. Niehoff[1,2], L. Beyer[3,4], C. Salazar Mejia[1], T. Gottschall[1], and J. Wosnitza[1,2] — [1]Hochfeld-Magnetlabor Dresden (HLD-EMFL), HZDR, Dresden, Germany — [2]Institut für Festkörper- und Materialphysik, TU Dresden, Germany — [3]Leibniz Institute for Solid State and Materials Research, Dresden, Germany — [4]TU Bergakademie Freiberg, Germany

Research on magnetocaloric materials with first-order phase transitions typically focuses on reducing their hysteresis. However, in this presentation, we will discuss an alternative approach by capitalizing on the width of the hysteresis to enhance the cooling performance through the synergistic utilization of multiple caloric effects. The fine tuning of the hysteresis is achieved through the rational substitution of appropriate quantities of Fe and Co within the Ni-Mn-Sn Heusler alloy. In addition, the powder-in-tube method is employed to further enhance performances such as mechanical stability, hysteresis, and the magnetocaloric effect itself. To comprehensively investigate the properties and mechanical stability of the material, we conducted stress-strain measurements and adiabatic temperature changes for different initial temperatures. This analysis was performed under varying uniaxial loads within pulsed magnetic fields, reaching up to 50 T.

MA 51.6 Fri 11:00 H 2013
Direct measurements of the adiabatic temperature change in a holmium single crystal using high magnetic fields — •E. Bykov[1], C. Salazar Mejia[1], T. Gottschall[1], J. Wosnitza[1,2], M. D. Kuz'min[3], Y. Mudryk[4], and D. L. Schlagel[4] — [1]Hochfeld-Magnetlabor Dresden (HLD-EMFL), HZDR, Dresden, Germany — [2]Technische Universität Dresden, Dresden, Germany — [3]Aix-Marseille Université, IM2NP, Marseille, France — [4]Ames Laboratory, U.S. Department of Energy, Iowa State University, Ames, USA

Holmium stands out as an extraordinary magnetocaloric substance due to one of the most substantial magnetic moments found in the periodic table. Its magnetic behavior under temperature and magnetic field variation is intricate, as reflected in a complex phase diagram featuring paramagnetic, helicoidal, conical, fan, spin-slip, and ferromagnetic phases. Even with field variations of 5 T along its easy axis, holmium exhibits a robust magnetocaloric effect, showcasing ΔT_{ad} of approximately 5 K and $\Delta S_T \approx$ -10 J kg^{-1} K^{-1} across a wide temperature span from 20 to 100 K. This positions holmium as a promising material for refrigeration in an active magnetic regenerator system, particularly for applications such as natural gas and hydrogen liquefaction. We present the results of a comprehensive study of single-crystalline holmium in fields up to 60 T.

15 min. break

MA 51.7 Fri 11:30 H 2013

High throughput approach for finding magnetocaloric materials — Rafael Martinho Vieira[1], Sagar Ghorai[2], Olle Eriksson[1], and •Heike C. Herper[1] — [1]Department of Physics and Astronomy, Uppsala University, Sweden — [2]Functional Materials, TU Darmstadt, Germany

After several decades of research, the pool of materials suitable for room-temperature refrigeration is still very limited. Materials with magnetostructural phase transition (MST) are possible candidates for magnetocaloric materials. MnNiSi-based systems undergo a structural phase transition and have the potential for MST. We show that an MST can be reached by Fe and Al doping and for Mn1-xFexNiSi0.95Al0.5 we observe a giant magnetocaloric effect at room temperature. [(https://doi.org/10.48550/arXiv.2307.00128)] By combining experiments with ab initio theory and spin dynamics methods we could show that the key to the large magnetocaloric response lies in the coexistence of the two magnetically very different phases at low temperatures. Based on the findings for MnFeNiSiAl we designed a high-throughput search method to identify candidate phases with MST. Big data searches were combined with first-principles calculations and spin dynamics simulations. [1] About 20 systems with several polymorphs were found. Out of this group, several candidates show potential for an MST near room temperature. Their magnetic properties have been studied in our theoretical approach and will be discussed as well as the expected magnetocaloric performance.

[1] Journal of Alloys and Compounds 857, 157811(2021)

MA 51.8 Fri 11:45 H 2013

Ab initio Quantification of Electronic and Magnetoelastic Mechanisms of First-order magnetic phase transitions — •Eduardo Mendive Tapia[1], Lluís Mañosa[1], Aleix Abadia-Huguet[1], Enric Stern Taulats[1], Marius Costache[1], Benedikt Eggert[2], Mehmet Acet[2], Mihai Sturza[3], Holger Kohlmann[3], Christopher Patrick[4], Tilmann Hickel[5], Jörg Neugebauer[5], and Julie Staunton[6] — [1]University of Barcelona, Spain — [2]Universität Duisburg-Essen, Germany — [3]Universität Leipzig, Germany — [4]University of Oxford, UK — [5]MPIE, Düsseldorf, Germany — [6]University of Warwick, Coventry, UK

While magnetovolume coupling is a well-known mechanism driving first-order magnetic phase transitions [1], purely electronic sources [2,3] have a long, subtle history and remain poorly understood. We present an ab initio disordered local moment theory to quantify electronic and magnetoelastic effects [4] underlying the magnetic phase transitions of two different caloric materials: the famous La-Fe-Si compound [3] and a van der Waals Cr2Fe2Te6 crystal [5]. Results in very good agreement with experiment explaining the ab initio origin of the first and second-order nature of their transitions will be shown, together with recent experiments focused on multicaloric effects in Cr2Fe2Te6.

[1] C. P. Bean and D. S. Rodbell, Phys. Rev. 126, 104 (1962)
[2] E. P. Wolfarth and P. Rhodes, Phil. Magazine, 7:83, 1817 (1962)
[3] A. Fujita et al., Phys. Rev. B 65, 014410 (2001)
[4] E. Mendive-Tapia, et al., J. Phys. Energy 5, 034004 (2023)
[5] C. Gong, et al., Nature 546, 265 (2017)

MA 51.9 Fri 12:00 H 2013

Magnetocaloric effect in (La, Ce)(Fe, Si, Mn)$_{13}$ with tunable, low transition temperature — •M. Strassheim[1,2], C. Salazar-Mejia[1], L. Beyer[3,4], J. Wosnitza[1,2], and T. Gottschall[1] — [1]Hochfeld-Magnetlabor Dresden (HLD-EMFL), HZDR, Dresden, Germany — [2]TU Dresden, Germany — [3]Leibniz-Institut für Festkörper- und Werkstoffforschung, Dresden, Germany — [4]TU Bergakademie Freiberg, Germany

The upcoming technology of magnetocaloric cooling has several advantages depending on the working temperatures. At room temperature, one can avoid environmentally harmful refrigerants and at cryogenic temperatures - for instance in hydrogen liquefaction - the process itself has a much higher Carnot efficiency than the conventional one. In both cases, a challenge is the availability of magnetocaloric materials: They usually involve large amounts of of costly rare earths to show a significant magnetocaloric effect. The ternary La(Fe,Si)$_{13}$ solves this issue, but has a transition temperature at 200 K. Recently, we showed that this transition temperature can be tuned down by introducing both Ce and Mn to the system. In this work, we show characterization results of the powder produced in sizes of 25-75 μm as characterized with electron microscopy and x-ray diffraction. In the end, we discuss the possible applicability in 3D printing techniques such as selective laser melting.

MA 51.10 Fri 12:15 H 2013

RCo2Hx magnetocaloric compounds for cryogenic gas liquefaction — •Allan Doering, Imants Dirba, Konstantin Skokov, and Oliver Gutfleisch — Technical University of Darmstadt, Darmstadt, Germany

Magnetic cooling, based on the magnetocaloric effect, is a new refrigeration technology that has been intensively developed for room temperature in recent decades. This technology could play also an important role at cryogenic temperatures to liquefy hydrogen, aiming for a future green economy and carbon-neutral societies. Typically, to use the magnetocaloric cycle, the hydrogen is pre-cooled with liquid nitrogen down to 77 K, and then magnetocaloric refrigeration is operated in the temperature range from 77 to 20 K to liquefy the H2 gas. Obviously, this new technology requires new materials. Since the maximum MCE takes place near the Curie temperature (TC), it is critical to use materials with a TC between 20 K and 77 K. The RCo2 family (where R stands for rare earth elements) are potential materials for this application due to their large MCE, however, some of these compounds have TC higher than 77 K. In our work, we tune TC of the RCo2 family to temperatures below 77 K using hydrogen as interstitial atoms. Samples were synthesized by arc-melting and annealed afterward. The quality of samples was determined by X-ray diffraction and scanning electron microscopy. Magnetization and heat capacity were measured to determine magnetocaloric effect of obtained alloys (magnetic entropy change and adiabatic temperature change. We acknowledge the HyLICAL and CRC 270 projects for the funding of this research.

MA 51.11 Fri 12:30 H 2013

Highly reversible magnetocaloric effect in $Gd_5Si_{0.25}Ge_{3.75}$ in moderate magnetic fields for hydrogen liquefaction — •Wei Liu[1], Eduard Bykov[2], Tino Gottschall[2], Konstantin Skokov[1], and Oliver Gutfleisch[1] — [1]Materials Science, Technical University of Darmstadt, Darmstadt, Germany — [2]High Magnetic Field Lab, Helmholtz-Zentrum Dresden-Rossendorf, Dresden, Germany

Magnetocaloric (MC) hydrogen liquefaction could be a 'game changer' for the liquid hydrogen industry due to its potentially higher efficiency than the conventional Joule-Thomson-expansion-based liquefaction technologies [1]. However, second-order MC materials exhibit less excellent MCEs near the nitrogen condensation point (77 K), where the hydrogen starts to be cooled by a magnetic cooling cycle to reach its condensation point (20 K) [2]. First-order MC materials can achieve much higher MCEs, but their poor reversibility due to their significant thermal hysteresis is a problem which makes their applications in real devices more complex [3]. In this work, we demonstrate that $Gd_5Si_{0.25}Ge_{3.75}$ exhibits a highly reversible MCE near 77 K in a magnetic field of 5 T despite its significant thermal hysteresis. Considering that a magnetic field of 5 T is often proposed to be used in a practical active magnetic regenerator, this work shows that first-order MC materials with significant thermal hysteresis can also be promising candidates for hydrogen liquefaction.

[1] Liu et al., J. Phys. Energy 3 (2023), 034001 [2] Liu et al., Appl. Mater. Today 29 (2022), 101624 [3] Gutfleisch et al., Phil. Trans. R. Soc. A 374 (2016), 20150308

MA 51.12 Fri 12:45 H 2013

Tuning magnetic properties in (RE,La)(Fe,Si)13 with 4f alloying — •Johanna Lill[1], Benedikt Eggert[1], Benedikt Beckmann[2], Olga N. Miroshkina[1], Konstantin Skokov[2], Jose R. Mardegan[3], Damian Günzing[1], Simon Rauls[1], Philipp Klassen[1], Tom Helbig[1], Sonia Francoual[3], Richard A. Brand[1], Kurt Kummer[4], Markus E. Gruner[1], Oliver Gutfleisch[2], Katharina J. Ollefs[1], and Heiko Wende[1] — [1]Faculty of Physics and CENIDE, UDE, Duisburg, Germany — [2]Material Science, TU Darmstadt, Darmstadt, Germany — [3]DESY, PETRA III, Hamburg, Germany — [4]ESRF, Grenoble, France

Magnetic refrigeration in comparison to gas-compression refrigerators reduces green house gas emissions drastically. Magnetocaloric (MC) materials are alloyed to 1) shift phase transition temperatures accord-

ing to the need for specific applications and 2) to increase the MC effect. The size of the MC effect is proportional to the magnetic field integral of $\frac{\delta M}{\delta T}$. Therefore, the response will be stronger if the magnetic material shows a stronger magnetisation change. Here, we analyse 4f elements in the well-known material system $La(Fe,Si)_{13}$, using Ce, Nd and Pr on the La site with up to 30%. With 4f alloying, the saturation magetization strongly increases while the Fe magnetic moment, usually responsible for the overall magnetic behaviour, does not respond as detected in Mössbauer studies. We performed XMCD studies on their elements to determine the rare earths contribution to the total magnetisation. We acknowledge funding of the DFG through CRC 270 HoMMage and thank the ESRF and DESY for allocating beamtime.

MA 51.13 Fri 13:00 H 2013

Influence of hydrostatic pressure on the magnetocaloric effect of $La_{0.7}Ce_{0.3}Fe_{11.6}Si_{1.4}$ — •Benedikt Beckmann[1], Lukas Pfeuffer[1], Johanna Lill[2], Benedikt Eggert[2], David Koch[1], Barbara Lavina[3,4], Jiyong Zhao[4], Thomas Toellner[4], Esen E. Alp[4], Katharina Ollefs[2], Konstantin P. Skokov[1], Heiko Wende[2], and Oliver Gutfleisch[1] — [1]Institute of Materials Science, TU Darmstadt, Darmstadt, Germany — [2]Faculty of Physics and CENIDE, UDE, Duisburg, Germany — [3]Center for Advanced Radiation Sources, UChicago, Chicago, USA — [4]Advanced Photon Source, Argonne National Laboratory, Argonne, USA

Magnetocaloric liquefaction of hydrogen, which is a key enabler for the successful transition towards a carbon neutral society based on renewable energies, is a crucial and promising future technology. However, the current dependence on highly-critical, heavy rare-earth based magnetocaloric materials is a major disadvantage for the global usage of the technology. In this work, we aim to mitigate this limitation by using a multi-stimuli approach, utilizing isotropic pressure and magnetic field as external stimuli to tailor and induce the phase transition associated with a large caloric effect. We use non-toxic, low-cost, and low-criticality $La_{0.7}Ce_{0.3}Fe_{11.6}Si_{1.4}$, achieving phase transitions in the temperature range from 200 K down to liquid hydrogen temperatures with large isothermal entropy changes up to 28 $J(kgK)^{-1}$.

We acknowledge financial support from DFG through CRC 270 HoMMage (Project-ID 405553726) and thank the Argonne National Laboratory for allocating beamtime.