

MA 61: Caloric Effects

Time: Friday 9:30–13:15

Location: HSZ 101

MA 61.1 Fri 9:30 HSZ 101

On the exploration of mechanical and magnetic properties of Ni-Mn-In Heusler alloy system doped with Gd — ●WEI LIU, FRANZISKA SCHEIBEL, LUKAS PFEUFFER, ANDREAS TAUBEL, KONSTANTIN SKOKOV, and OLIVER GUTFLEISCH — Funktional Materials, Technische Universität Darmstadt

NiMn- based Heusler alloys can be used in a six-step multistimuli cooling cycle which combines the magnetocaloric effect and the elastocaloric effect. This multistimuli cooling cycle has been demonstrated to be able to exploit the thermal hysteresis to achieve a fully reversible caloric effect [1]. However, the brittleness of NiMn- based Heusler alloys is problematic for cyclic application with stress. A possible method to strengthen them is precipitation hardening. By doping with Gd we introduced Gd-rich precipitates to the Ni-Mn-In system. With a slight increase of the hysteresis and a slight decrease of the transition steepness, the magnetic entropy change of the doped sample of $9 \text{ J kg}^{-1} \text{ K}^{-1}$ can be largely retained, compared to the undoped sample with a value of $11 \text{ J kg}^{-1} \text{ K}^{-1}$. An adiabatic temperature change of 4 K is measured in the Gd-doped sample under 2 T, whereas the value for the undoped sample is 5 K. The compressive tests show that the compressive stress is increased by almost 3 times by doping, which is beneficial for the multi-stimuli cooling cycle. The work is supported by the ERC (Project 'Cool Innov') and DFG (Grant No. SPP 1599). [1] T. Gottschall, A. Gràcia-Condal, M. Fries, A. Taubel, L. Pfeuffer, L. Mañosa, A. Planes et al., A multicaloric cooling cycle that exploits thermal hysteresis, *Nature materials* 17 (2018) 929*934.

MA 61.2 Fri 9:45 HSZ 101

Magnetocaloric materials for multi-stimuli cooling cycle - required functional properties and potential materials — ●FRANZISKA SCHEIBEL¹, TINO GOTTSCHALL², LUKAS PFEUFFER¹, ANDREAS TAUBEL¹, DOMINIK OHMER¹, KONSTANTIN SKOKOV¹, BAIXIANG XU¹, and OLIVER GUTFLEISCH¹ — ¹Technische Universität Darmstadt, Darmstadt, Germany — ²Dresden High Magnetic Field Laboratory (HLD-EMFL), HZDR, Germany

Materials with a first-order magnetostructural phase transition (FOMST) exhibit a large magnetocaloric effect. However, due to the thermal hysteresis coming along with FOMST the cyclic performance of that materials is limited. The origins of the hysteresis are quite complex but this complexity can also be utilized to tune it [1,2]. So far, a lot of research has been directed towards reducing the hysteresis. Our new cooling concept actually attempts to exploit hysteresis in a multi-stimuli approach [3]. Herein, a second stimulus (uniaxial load) besides the magnetic field stimulus is used to overcome hysteresis and enable a fully reversible FOMST and therefore increase the cyclic performance of the magnetocaloric material. For the multi-stimuli approach a variety of new functional properties are required and we present here a first summary of experimental and theoretical findings on tailored Ni-Mn-X Heusler alloys as potential candidates. This work was supported by the ERC Advanced Grant "Cool Innov" and the SFB-TRR270 "Hommage". [1] O. Gutfleisch et al., *Phil. Trans. R. Soc. A* 374: 20150308 (2016) [2] F. Scheibel et al., *Energy Technol.* 6, 1397 (2018) [3] T. Gottschall et al., *Nat. Mater.* 17, 929 (2018)

MA 61.3 Fri 10:00 HSZ 101

Optimizing the magnetocaloric effect in all-d-metal Ni-Co-Mn-Ti Heusler alloys — ●BENEDIKT BECKMANN, ANDREAS TAUBEL, LUKAS PFEUFFER, FRANZISKA SCHEIBEL, KONSTANTIN SKOKOV, and OLIVER GUTFLEISCH — Institut für Materialwissenschaft, TU Darmstadt, 64287 Darmstadt, Germany

Magnetocaloric refrigeration is a promising cooling technology which could be an environmentally friendly and more energy efficient alternative to conventional vapor compression refrigeration. Among magnetocaloric materials, Ni-Mn based Heusler alloys, showing a first-order magnetostructural phase transition, are promising candidates. In this study, we carried out a systematic analysis of all-d-metal $\text{Ni}_{50-x}\text{Co}_x\text{Mn}_{50-y}\text{Ti}_y$ Heusler alloys. Due to their enhanced mechanical stability and large volume change, these alloys could be used for cooling cycles that use magnetic field and pressure as external stimuli for inducing the phase transition. A systematic heat treatment optimization is carried out, resulting in a substantial decrease of the transition width down to only 4 K. The microstructural differences between the

as-cast and differently annealed alloys are analyzed in detail by *in-situ* Kerr microscopy. As a result, large isothermal entropy changes of up to $38 \text{ J kg}^{-1} \text{ K}^{-1}$ in 2 T are achieved. The adiabatic temperature change is measured directly for this material system and values of up to -3.8 K for the first field application and -0.8 K under cyclic conditions are obtained in moderate magnetic field changes of 2 T.

We acknowledge financial support from ERC (Advanced Grant "Cool Innov", No. 743116) and DFG (CRC/TRR 270).

MA 61.4 Fri 10:15 HSZ 101

Time-dependence of the martensitic transformation in Ni-Mn-In Heusler compounds — ●LUKAS PFEUFFER¹, TINO GOTTSCHALL², TOM FASKE¹, ANDREAS TAUBEL¹, FRANZISKA SCHEIBEL¹, KONSTANTIN SKOKOV¹, and OLIVER GUTFLEISCH¹ — ¹TU Darmstadt, Institut für Materialwissenschaft, 64287 Darmstadt, Germany — ²Dresden High Magnetic Field Laboratory, Helmholtz-Zentrum Dresden-Rossendorf, 01328 Dresden, Germany

Ni-Mn-In-(Co) Heusler compounds exhibit very promising magnetocaloric properties upon the first-order transition between low temperature, low magnetization martensite to high temperature, high magnetization austenite. Thereby, the phase transformation is characterized by a shear lattice distortion, which takes place via nucleation and growth. We studied the transition process of Ni-Mn-In by simultaneous adiabatic temperature change and strain measurements in pulsed magnetic fields. By different field-sweep rates, kinetical effects of the martensitic transformation were investigated. Moreover, the influence of the initial sample temperature on critical transformation field, field hysteresis and reversibility was analyzed and correlated with isofield measurements of magnetization, strain and resistivity.

The work was supported by the European Research Council (ERC) under the European Union's Horizon 2020 research and innovation programme (grant no. 743116*project Cool Innov). We thank the HLD at HZDR, member of the European Magnetic Field Laboratory (EMFL) and the Helmholtz Association for funding via the Helmholtz-RSF Joint Research Group (Project No. HRSF-0045)

MA 61.5 Fri 10:30 HSZ 101

Thermomagnetic properties of Co or Cu doped Ni-Mn-Ga films for micro energy harvesting — ●LUKAS FINK^{1,2}, ANETT DIESTEL¹, KORNELIUS NIELSCH^{1,2}, and SEBASTIAN FÄHLER¹ — ¹Leibnitz IFW Dresden, Institute for Metallic Materials, D-01171 Dresden, Germany — ²TU Dresden, Institute of Materials Science, D-01062 Dresden, Germany

Recovery of waste heat is decisive for a more efficient use of primary energy. Furthermore, at the micro scale it enables the Internet of Things without the need for wiring. However, except for thermoelectrics, there is hardly any technology available to harvest low temperature waste heat. As an alternative approach recently first thermomagnetic microsystems were developed, which use magnetocaloric materials as active material. The high surface-to-volume ratio of thin films enables a fast heat transfer, resulting in high cycling frequencies and power densities compared to bulk devices.

For this application we prepare Ni-Mn-based Heusler films by sputter deposition. We focus on Ni-Mn-Ga-X films (X= Cu, Co) and examine the role of the doping elements and chemical order on 1) the transition temperature, 2) hysteresis losses, and 3) difference of magnetization at the martensitic transition, which is the decisive property for thermomagnetic harvesting of low temperature waste heat. We discuss the suitability of these alloys for thermomagnetic harvesting of low temperature waste heat in comparison to magnetocaloric refrigeration.

This work is funded by the DFG (FA453/14).

MA 61.6 Fri 10:45 HSZ 101

Orbital Nernst Effect of Magnons — ●LI-CHUAN ZHANG^{1,2}, FABIAN R. LUX^{1,2}, JAN-PHILIPP HANKE¹, PATRICK M. BUHL³, SERGI GRYTSIUK¹, STEFAN BLÜGEL¹, and YURIY MOKROUSOV^{1,3} — ¹Peter Grünberg Institut and Institute for Advanced Simulation, Forschungszentrum Jülich and JARA, 52425 Jülich, Germany — ²Department of Physics, RWTH Aachen University, 52056 Aachen, Germany — ³Institute of Physics, Johannes Gutenberg University Mainz, 55099 Mainz, Germany

In the past, magnons have proven to mediate thermal transport of spin

in various systems (see references in e.g. [1]). We will reveal that the fundamental coupling of the scalar spin chirality, inherent to magnons, to the electronic degrees of freedom in the system can result in the generation of sizeable orbital magnetization and thermal transport of orbital angular momentum. We will demonstrate the emergence of the latter phenomenon of the orbital Nernst effect by referring to the spin-wave Hamiltonian of Kagome ferromagnets and predict that in a wide range of systems the transverse current of orbital angular momentum carried by magnons in response to an applied temperature gradient can overshadow the accompanying spin current [2]. We suggest that the discovered effect fundamentally correlates with the topological Hall effect of fluctuating magnets, and suggest that it can be utilized in magnonic devices for generating magnonic orbital torques. We acknowledge funding from DFG through SPP 2137 "Skyrmionics", DARPA TEE Program and CSC (No. [2016]3100). [1] L.Zhang *et al.*, arXiv:1901.06192 (2019); [2] L.Zhang *et al.*, arXiv:1910.03317 (2019)

MA 61.7 Fri 11:00 HSZ 101

Magnonic Analogue of Edelstein Effect in Antiferromagnetic Insulators — BO LI¹, ●ALEXANDER MOOK², ALDO RAELIARIJAONA¹, and ALEXEY KOVALEV¹ — ¹Department of Physics and Astronomy and Nebraska Center for Materials and Nanoscience, University of Nebraska, Lincoln, Nebraska 68588, USA — ²Department of Physics, University of Basel, CH-4056 Basel

The electronic Edelstein or inverse spin-galvanic effect comprises a nonequilibrium spin density as response to a charge current [1]. Herein [2], we investigate the nonequilibrium spin polarization due to a temperature gradient in antiferromagnetic insulators, which is the magnonic analogue of the Edelstein effect.

We derive a linear response theory of a temperature-gradient-induced spin polarization in collinear and noncollinear antiferromagnets and present selected examples in one, two, and three dimensions. Assuming a realistic temperature gradient of 10 K/mm, we find two-dimensional spin densities of up to $\sim 10^6$ h/cm² [e.g., in the noncollinear kagome antiferromagnet KFe₃(OH)₆(SO₄)₂] and three-dimensional bulk spin densities of up to $\sim 10^{14}$ h/cm³, encouraging an experimental detection.

[1] A. Aronov and Y. B. Lyanda-Geller, JETP Lett. **50**, 431 (1989); V. Edelstein, Solid State Commun. **73**, 233 (1990); [2] B. Li, A. Mook, A. Raeliarijaona, A. Kovalev, preprint arXiv:1910.00143

MA 61.8 Fri 11:15 HSZ 101

Non-hysteretic first-order ferromagnetic transitions by itinerant electron feedback and Fermi surface topology change — ●EDUARDO MENDIVE TAPIA^{1,2}, DURGA PAUDYAL³, LEON PETIT⁴, and JULIE STAUNTON² — ¹Max-Planck Institut für Eisenforschung, Düsseldorf, Germany — ²Dept of Physics, University of Warwick, Coventry, UK — ³The Ames Laboratory, U.S. Dept of Energy, Iowa State University, USA — ⁴Daresbury Laboratory, Warrington, UK

Refrigeration and air conditioning are crucial in modern life and in adapting to climate change. Discontinuous magnetic phase transitions have great promise for new, energy efficient and environmentally friendly solid-state cooling technology. Huge exploitable entropy and temperature changes typically result from the coupling between a material's spin polarized interacting electrons and the crystal structure. Such magnetostructurally driven cooling, however, is nearly always degraded by hysteresis. We present an *ab-initio* theory which can find mechanisms for first-order magnetic phase transitions that are purely electronic in origin [1], thus avoiding the need for magnetostructural effects. We show that this electronic mechanism arises from an itinerant electron feedback to magnetic order. In particular, it is demonstrated that a topological change of the Fermi surface explains the hysteresis-free giant cooling properties recently measured in Eu₂In [2].—This work is funded by the EPSRC (UK) and the U.S. Dept of Energy, and forms part of the PREMAG project (University of Warwick).

[1] E Mendive-Tapia and J Staunton, Phys. Rev. B **99**, 144424 (2019) [2] F Guillou *et al.*, Nat. Comm. **9**, 2925 (2018)

MA 61.9 Fri 11:30 HSZ 101

Utilizing the Thermomagnetic Response of the Magnetocaloric alloy Energy Harvesting Application — ●DEEPAK KAMBLE^{1,2} and RAJU V. RAMANUJAN¹ — ¹School of Materials Science and Engineering, Nanyang Technological University, Singapore 639798 — ²Present address: Leibniz IFW Dresden, Institute for Metallic Materials, Helmholtzstraße 20, 01069 Dresden, Germany

Thermal management is important to convert waste heat to useful electricity. Here we present an approach based on magnetocaloric ma-

terials, which allows us to harvest electricity from waste heat. As a first step we synthesized and characterized (MnNiSi)_{1-x}(Fe₂Ge)_x alloys exhibiting 1st order magnetostructural transition. The alloys have a highly tunable transition range near room temperature with promising magnetocaloric parameters [K. Deepak et al., J. Alloys Compd. **743** (2018) 494-505]. As a second step the thermomagnetic response of the alloy with suitable transition temperature was utilized as working material to develop a novel hybrid thermomagnetic oscillator for electricity harvesting using waste heat from a heat load. The thermomagnetic alloy (TMA) was suspended in a quartz tube between heat load at the top and heat sink at the bottom resulting in oscillation of TMA due to the thermomagnetic response. The continuous oscillation gives rise to a magnetic flux change in the coil to generate electricity as well as transfers the heat from the heat load to the heat sink [K. Deepak et al., Applied Energy **233-234** (2019) 312-320].

MA 61.10 Fri 11:45 HSZ 101

Optimizing a thermomagnetic generator with flux reversal by finite element calculations — ●DIETMAR BERGER¹, DANIEL DZEKAN¹, DEEPAK KAMBLE^{1,2}, KORNELIUS NIELSCH¹, and SEBASTIAN FÄHLER¹ — ¹IFW Dresden, Helmholtzstraße 20, D-01069 Dresden, Germany — ²School of Materials, Science and Engineering, Nanyang Technological University, Singapore 639798

Climate protection and the efficient use of primary energy are increasingly gaining the focus of public interest. Against this background, the generation of electrical energy from environmentally friendly and regenerative energy sources is increasingly important. In particular the huge amount of low temperature waste heat below 100 °C is almost not reusable.

The discovery of the giant magnetocaloric effect by Pecharsky and Gschneidner in 1997 [1] led to the development of new materials exhibiting a sharp change in magnetization in vicinity of room temperature. These materials also enabled the inverse energy conversion process: thermomagnetic harvesting of low temperature waste heat.

In this presentation we use our recent thermomagnetic generator with flux reversal as starting point [2]. The focus of our current work is to increase both, the conversion efficiency and the cycle frequency. By using FEM simulations we analyze the drawbacks of the present demonstrator and sketch improvements for the next generation of thermomagnetic generators.

[1] V. K. Pecharsky and K. A. Gschneidner, Jr., Phys. Rev. Lett. **78**, 4494 (1997) [2] A. Waske et al., Nature Energy **4**, 68-74 (2019)

MA 61.11 Fri 12:00 HSZ 101

Evaluation of magnetocaloric materials for thermomagnetic energy harvesting — ●DANIEL DZEKAN^{1,2}, ANJA WASKE³, KORNELIUS NIELSCH^{1,2}, and SEBASTIAN FÄHLER² — ¹Technical University Dresden, Institute for Material Science — ²Leibniz IFW Dresden, Institute for Metallic Materials — ³Bundesanstalt für Materialforschung und -prüfung (BAM), Berlin, Germany

Nowadays recovering waste heat becomes decisive to use the limited primary energy most effective. A promising way to do so is the usage of thermomagnetic systems. Within these, heat is first converted into magnetic energy and then into electricity. Early device concepts had been suggested by Edison and Tesla but only few of them were realized. This changed due to the emerging field of magnetocaloric cooling and the development of new materials. These exhibit a steep change of magnetisation in a narrow temperature range, which paved the way for several thermomagnetic demonstrators and prototypes. Recently we presented a thermomagnetic generator with a novel design of the magnetic circuit, which increased the power output, frequency and efficiency significantly [1]. Here we identify and analyse the material requirements for a more energy and economic efficient conversion. We describe the influence of magnetisation change and heat capacity on thermodynamic efficiency, as well as the consequences of thermal conductivity on power density. [1] Waske et al., Nature Energy **4**, 68-74 (2019)

MA 61.12 Fri 12:15 HSZ 101

The impact of hydrogenation and chemical substitution on the itinerant electron metamagnetism in La-Fe-Si-based magnetocaloric materials — ●MARKUS ERNST GRUNER — Faculty of Physics and Center for Nanointegration, CENIDE, University of Duisburg-Essen, Germany

State-of-the-art magnetocaloric materials like La-Fe-Si or FeRh are characterized by an intricate coupling between the electronic structure, magnetism and lattice, which is responsible for the large entropy

change occurring at a first-order metamagnetic transition. Recent investigations prove that first-principles calculations in the framework of density functional theory (DFT) are in excellent agreement with element-resolved experimental techniques such as nuclear resonant inelastic X-ray-scattering, X-ray absorption or Mössbauer spectroscopy and can thus help to disentangle the interplay of the different degrees of freedom [1]. This contribution reports on recent DFT-based advances regarding the impact of interstitial doping with hydrogen and chemical substitution with transition metals and main group elements on magneto-elastic coupling and the functional properties of La-Fe-Si-based materials.

Funding from DFG in the framework of SPP 1599 and TRR 270 is gratefully acknowledged.

[1] F. Scheibel *et al.*, Energy Technology **6**, 1397 (2018).

MA 61.13 Fri 12:30 HSZ 101

Tailoring the Phase Transition of FeRh by Nickel Doping

— •TOBIAS LOJEWSKI¹, BENEDIKT EGGERT¹, ALISA CHIRKOVA², KONSTANTIN SKOKOV², ILIYA RADULOV², OLGA SHULESHOVA³, MARKUS E. GRUNER¹, KATHARINA OLLEFS¹, IVAN KABAN³, OLIVER GUTFLEISCH², and HEIKO WENDE¹ — ¹Faculty of Physics and CENIDE, University of Duisburg-Essen — ²Functional Materials, TU Darmstadt — ³IFW Dresden

FeRh in the B2-crystal structure shows a substantial magnetocaloric effect during the metamagnetic phase transition from the AFM to the FM phase, at a transition temperature around 400 K. This phase transition can be tuned using Ni as dopant, shifting the phase transition to lower temperatures [1] and making the system more relevant for room temperature applications. As such, the effects of nickel doping were studied in a Fe₄₉Rh₅₀Ni₁ sample using Conversion Electron Mössbauer Spectroscopy (CEMS) to obtain a microscopic picture of the ⁵⁷Fe-nuclei surroundings during the phase transition. Here a reduced transition temperature of about 327 K can be observed. From the CEMS measurements, a possible electron transfer from nickel to iron is indicated by a variation in the isomer shift. Additionally, a similar hyperfine field can be observed in both FeRh and FeRhNi for the identical phases, suggesting no change of the iron magnetic moments. The microscopic magnetic structure is then compared to macroscopic magnetic properties obtained from magnetometry data. Financial support by DFG(WE2623/14-1) and CRC/TRR 270 is acknowledged.

[1] Baranov NV, Barabanova EA. J All Comp 1995;219:139

MA 61.14 Fri 12:45 HSZ 101

Experimental determination of the thermal conductivity of oxide barriers in magnetic tunnel junctions

— HYEJIN JANG¹, LUCA MARNITZ², TORSTEN HUEBNER², JOHANNES KIMLING¹, ULRIKE MARTENS³, JAKOB WALOWSKI³, MARKUS MÜNZENBERG³, ANDY THOMAS⁴, GÜNTER REISS², DAVID CAHILL¹ and •TIMO KUSCHEL² —

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The tunnel magneto-Seebeck (TMS) effect in magnetic tunnel junctions (MTJs) has large potential for future nanoelectronic devices [1]. However, quantitative determination of the TMS coefficients requires knowledge of the temperature drop across the tunnel barrier and, thus, of the thermal conductivity of the oxide barrier material. Here, we present two new approaches to obtain the barrier's thermal conductivity, which is usually difficult to access experimentally. For the first approach, we utilize laser-induced TMS in combination with finite-element modeling extracting values of the thermal conductivity of 0.7 W/(K·m) for MgAl₂O₄ and 5.8 W/(K·m) for MgO [2]. The second method uses ultrafast thermoreflectance and magneto-optic Kerr effect thermometry and provides values of the thermal conductivity of 0.4-0.6 W/(K·m) for both oxide barrier materials [3]. These results are in nice agreement with theoretical predictions for ultra-thin oxide barriers [4].

[1] Kuschel et al., J. Phys. D: Appl. Phys. **52**, 133001 (2019)

[2] Huebner et al., J. Phys. D: Appl. Phys. **51**, 224006 (2018)

[3] Jang, Marnitz, Huebner, Kimling, Kuschel, Cahill, under review

[4] Zhang et al., Phys. Rev. Lett. **115**, 037203 (2015)

MA 61.15 Fri 13:00 HSZ 101

Magnetic phase diagram of the magnetocaloric compound Mn₃Fe₂Si₃

— •MOHAMMED AIT HADDOUCH¹, JÖRG VOIGT¹, KAREN FRIESE¹, ANDREAS EICH¹, JÖRG PERSSON¹, ARMAND BUDZIANOWSKI², NICOLÒ VIOLINI¹, FABIANO YOKAICHIYA³, DEVASHIBHAI ADROJA⁴, and THOMAS BRÜCKEL¹ — ¹Jülich Centre for Neutron Science (JCNS-2) and PGI-4, Forschungszentrum Jülich, Germany — ²National Centre for Nuclear Research, Otwock, Poland — ³Helmholtz-Zentrum Berlin für Materialien und Energie, Germany — ⁴ISIS Facility, Rutherford Appleton Laboratory, U.K

We have studied the magnetic phases of single-crystalline Mn₃Fe₂Si₃ by neutron diffraction and magnetization measurements. Within the series Mn_{5-x}Fe_xSi₃, an inverse magneto-caloric effect (MCE) has been observed for x=0, while for x=4 a moderately high direct MCE occurs [1]. Similarly to the parent compound Mn₅Si₃, Mn₃Fe₂Si₃ exhibits two antiferromagnetic phase transitions to an AF1 and AF2 phase, respectively. The transition from AF1 → AF2 gives rise to an inverse MCE, i.e. the magnetic entropy is increased by the application of a magnetic field, albeit with complex field and temperature dependences. We discuss these changes in light of the preferential replacement of Mn by Fe on one of the two distinct lattice sites of the crystal structure (space group P6₃/mcm at RT). This leads to an increase in the transition temperatures and critical fields when compared to Mn₅Si₃. In addition, we find hints for ferromagnetic short-range correlations, which persist at temperatures twice as high as the Neel temperature. [1] Songlin et al, J. Alloys Compd, **334**, 249–252 (2002)