MA 10: Caloric Effects in Ferromagnetic Materials

Time: Monday 15:00–18:30 Location: HSZ 04

Invited Talk MA 10.1 Mon 15:00 HSZ 04

Magnetoelastic coupling and lattice dynamics in magnetocaloric materials — •Markus Ernst Gruner[1], Werner Keune[1], Michael Wolloch[2], Peter Mohn[2], Oliver Gutfleisch[3], Heiko Wende[1], and Rossitza Pentcheva[1] — [1]University of Duisburg-Essen — [2]TU Vienna — [3]TU Darmstadt

Solid state cooling concepts based on magnetocaloric materials offer an energy-efficient alternative to the conventional gas-compressor scheme. Large magnetocaloric effects are observed near magnetic first order transitions which involve a simultaneous change in volume or lattice symmetry and are subject to hysteresis losses. In metamagnetic Heusler alloys, lattice vibrations and magnetic order provide competing contributions to the entropy change, which further reduces the performance [1]. In metamagnetic FeRh [2] and conventional magnetocaloric La-Fe-Si-based compounds [3], lattice and magnetic degrees of freedom contribute cooperatively to the free energy. Comparing recent results from first-principles calculations and nuclear resonant inelastic X-ray scattering, we will review the relevant mechanisms for the favorable coupling of magnetism and lattice degrees of freedom in these two systems and discuss strategies to reduce the intrisic causes of hysteresis.

Contributions from researchers of Argonne National Laboratory, the universities of Bochum, Darmstadt, Duisburg-Essen and Vienna, IFW Dresden, KIT and the SPP 1599 are gratefully acknowledged.

[1] T. Gottschall *et al.*, Phys. Rev. B 93, 184431(2016)
[2] M. Wolloch *et al.*, Phys. Rev. B 94, 174435 (2016)
[3] M. E. Gruner *et al.* Phys. Rev. Lett. 114, 057202 (2015)

MA 10.2 Mon 15:30 HSZ 04

Simulation of complex magnetic intermetallics for ferroic cooling — •Peter Entel and Markus Gruner — Faculty of Physics, University Duisburg-Essen, 47048 Duisburg, Germany

We discuss the structural and magnetic properties of magnetic Heusler intermetallics like Ni-Mn-(Ga, In, Sn) doped with Co, which undergo a magnetostructural phase transition if rapidly quenched. This leads to a large inverse magnetocaloric effect. The coupling of structural and magnetic degrees of freedom is an intrinsic effect and can be explained by a microscopic itinerant band model where electrons are coupled to tetagonal distortions. The model explains why martensitic instability and high-spin state (ferromagnetic order) are mutually exclusive and only distortion and low-spin state (antiferromagnetic order) may coexist. Under magnetic field we observe kinetic arrest phenomena. For the materials which are less rapidly quenched, this magnetostructural phase transition shows time-dependent effects and starts to fade away and phase segregation into cubic ferromagnetic Heusler phase and tetragonal antiferromagnetic Ni-Mn phase occurs for all Mn excess Heusler alloys in agrement with experimental findings [1].

[1] A. Cahir, M. Acet, M. Farle, Sci. Rep. 6, 29831 (2016)

MA 10.3 Mon 15:45 HSZ 04

Substitutional influences on the magnetocaloric properties of the MnNiGe-system — •Andreas Taubel, Tino Gottschall, Konstantin Skokov, and Oliver Gutfleisch — Alarich-Weiss-Straße 16, 64287 Darmstadt

Magnetocaloric materials exhibit a temperature change when placed within a magnetic field under adiabatic conditions. They are studied intensively in order to make it a competitive and more energy efficient cooling technology besides gas compression [1]. The MM'X material system of Mn-Ni-Ge attracts recently significant interest for magnetocalorics because of a sharp and nicely tunable phase transition. The transition temperature can be tuned by Fe substitution for Ni or Mn, which additionally induces ferromagnetic interactions and establishes large magnetization changes.

In our work, the influence of elemental substitutions on the Ge site is studied in detail. The substitution of Si for Ge reduces the amount of expensive Ge and enhances the ferromagnetic behavior leading to a favored transition behavior in low fields. Moreover the mechanical stability of the samples is enhanced compared to brittle MnNiGe, which is important towards utilization of the material for cooling cycles. The high volume change during phase transition is responsible for the initial instability but enables significant pressure tuning, especially for the Si substituted samples. Further substitutions with Al and Sn can be done to completely renounce expensive Ge.

This work was supported by DFG (Grant No. SPP1599).

[1] O. Gutfleisch et al., Adv. Mater. 23, 821-842 (2011)

MA 10.4 Mon 16:00 HSZ 04

Spin dynamics of magnetocaloric compound Mn5Si3 — •Nikolaos Biniskos[1,2], Karin Schmalzl[1], Stephane Raymond[2], Sylvain Petit[3], and Thomas Brueckel[4] — [1]JCNS, Forschungszentrum Juelich GmbH, Outstation at ILL, Grenoble, France — [2]CEA-Grenoble, INAC MEM, 38054 Grenoble, France — [3]CEA-CNRS UMR 12, IRAMIS LLB, 91190 Gif-sur-Yvette, France — [4]JCNS and PGI, JARA-FIT, Forschungszentrum Juelich GmbH, 52425 Juelich, Germany

The magnetocaloric effect (MCE) refers to a change of entropy of a magnetic material exposed to a magnetic field change. A large MCE at room temperature and low magnetic field for a material with abundant and environmentally friendly elements opens the way for magnetic cooling devices. Inelastic neutron scattering (INS) studies might point out ingredients that may favor large MCE. From the Mn5-xFexSi3 series the parent compound Mn5Si3 exhibits positive and negative magnetic entropy change in relation with two distinct antiferromagnetic phase transitions at TN1=66K (AF1: non-collinear, non-coplanar structure) and TN2=99K (AF2: collinear structure), respectively. Experiments performed on a single crystal with unpolarized INS in the paramagnetic (PM) state and in the AF2 and AF1 phases revealed that AF1 is characterized by sharp spin-waves, but AF2 is characterized by a diffuse signal that resembles the one of the PM state, indicating strong spin fluctuations.

MA 10.5 Mon 16:15 HSZ 04

Strain-dependent magnetization measurements of epitaxial Ni-Mn-Ga-Co thin films on piezoelectric substrates — •Benjamin Schleicher[1,2], Stefan Schwabe[1], Robert Niemann[1], Anett Diestel[1], Ruben Hühne[1], Kornelius Nielsch[1], Ludwig Schultz[1,2], and Sebastian Fähler[1,2] — [1]IFW Dresden, P.O Box 270116, D-01171 Dresden, Germany — [2]TU Dresden, Institute of Solid State Physics, D-01062 Dresden, Germany

Heusler alloys such as Ni-Co-Mn-X (X=Ga, In, Sb, Sn), which show an inverse magnetocaloric effect, are promising materials for solid state cooling. Additionally to high external magnetic fields, the phase transition can also be induced by the application of mechanical stress. This can be realized with an electric field when the magnetocaloric thin film is deposited on a piezoelectric substrate. Thin films are of particular interest since their high surface-to-volume ratio allows fast heat transfer and high cycling frequencies, which leads to higher cooling power using less material. We present an investigation of sputtered epitaxial Ni-Mn-Ga-Co thin films on piezoelectric PMN-PT substrates [1]. Temperature dependent texture and magnetic measurements show the structural and magnetic phase transition in the material. The influence of temperature, magnetic field and mechanical stress on the magnetization of the Ni-Mn-Ga-Co thin film was investigated with a dedicated SQUID setup. In particular, the stress variation was achieved by applying an electric field to the multiferroic stack. This work is supported by DFG through SPP 1599 www.FerroicCooling.de. [1] B. Schleicher et al., J. Appl. Phys. 118 053906 (2015)

MA 10.6 Mon 16:30 HSZ 04

Reversible magnetocaloric effect in Ni-Mn based Heusler alloys — •Parul Devi[1], Mahdiyeh Ghorbani-Zavareh[1,2], Sanjay Singh[1], and Claudia Felser[1] — [1]Max Planck Institute for Chemical Physics of Solids, Dresden, Germany — [2]Dresden High Magnetic Field Laboratory, Helmholtz-Zentrum Dresden Rossendorf, Dresden, Germany

Magnetic refrigeration technology based on the magnetocaloric effect (MCE) has higher refrigeration efficiency and environment friendly, make it an edge over the others. In comparison to other Magnetocaloric materials, Ni-Mn based Heusler alloys are the subject of special interest as they do not involve toxic and rare earth elements. Heusler alloys show higher value of MCE due to the large magnetization change at their first order magneto-structural transition. Although, Heusler alloys show large MCE, the irreversibility of first order phase transition with respect to the magnetic field cycles is a major issue for practical applications. We show here an enhanced reversibility of MCE in a Ni-Mn-Ga and Ni-Mn-In Heusler alloys. The reversible MCE in these alloys is linked with the volume conversion and geometrical compatibility (co-factor) condition of martensite phase transition.

15 min. break

MA 10.7 Mon 17:00 HSZ 04

Influence of particle orientation on the effective thermal conductivity of magnetocaloric composites — •Kai Sellschopp[1,2,3], Bruno Weise[2,3], Marius Bierdel[2,3], Alexander Funk[2,3], Manfred Bobeth[3], Maria Krautz[2], and Anja Waske[3] — [1]TU Hamburg-Harburg — [2]IFW Dresden — [3]TU Dresden

Plates of epoxy-bonded La-Fe-Si-type particles display improved mechanical stability compared to sintered La-Fe-Si-type plates. The magnetocaloric properties are maintained very well in these composites and they are easy to process. However, the effective thermal conductivity (ETC) is decreased because of the low thermal conductivity of the epoxy. Our idea to overcome this problem without changing the volume fraction of magnetocaloric material is to produce an anisotropic ETC which prefers the out-of-plane direction, thereby enhancing heat transport to the heat exchange fluid. Using the anisotropic shape of the particles this can be achieved by orienting the particles' largest dimension in the out-of-plane direction.

Based on a real 3D data set obtained from X-ray computed tomography (XCT) several particle orientation distributions were modeled using equivalent ellipsoids for the description of particle shape. The ETC tensor was calculated from FEM simulations for all scenarios to study the influence of particle orientation on ETC. The simulations showed that orientation of the particles in the out-of-plane direction can increase the out-of-plane ETC by over 25%.

MA 10.8 Mon 17:15 HSZ 04

Magnetocaloric test bench with a new permanent magnet source — •Dimitri Benke, Jonas Wortmann, Tino Gottschall, Andreas Taubel, Konstantin Skokov, Iliya Radulov, and Oliver Gutfleisch — TU Darmstadt, Funktionale Materialien, Deutschland

Due to the rising demand in energy for cooling applications[1], and the rising problems caused by global warming, it is necessary to find ways to contain it to a bearable level. One possible way is the development of magnetocaloric cooling as an alternative cooling technology with the prospect of being more efficient than regular gas-compression systems [2].

To drive this development, we have built a magnetocaloric testbench that is able to create a siginificant thermal span which allows us to compare different magnetocaloric materials in real-life environments. To improved the testbench, we optimized especially the permanent magnet source. This lead to an increased active volume, higher magnetic field change, lower magnet mass and higher measured thermal span in the testbench.

The dependancy on rare-earths is a problem of magnetocaloric cooling. To show that it is possible to decrease that dependancy, the permanent magnet was manufactured from recycled material in cooperation with Urban Mining Co. This work was supported by the Darmstadt Graduate School of Excellence Energy Science and Engineering.

[1] M. Isaac, D. P. van Vuuren, Energy Policy 37 (2009) 507
[2] C. Zimm et al. (1998) Adv. Cryog. Eng

MA 10.9 Mon 17:30 HSZ 04

Examination of Gd/Gd-Y alloys stacks with different Curie temperatures in a magnetocaloric test bench — •Jonas Wortmann, Dimitri Benke, Tino Gottschall, Marc Pabst, Andreas Taubel, Konstantin Skokov, Iliya Radulov, and Oliver Gutfleisch — TU Darmstadt, Funktionale Materialien, Deutschland

Refrigeration using the magnetocaloric effect has the potential to overtake classical gas compression techniques in regards to efficiency and environmental friendliness [1]. To come closer to the goal of building a working refrigerator a linear magnetocaloric test bench with an average magnetic field change of 1T was built.

This article investigated the options of maximizing the temperature span and lowest achievable temperature by stacking Gd and Gd-Y alloys with different Curie temperatures. The temperatures between the stacks were investigated to estimate the necessary mass of specific of stacks and identify possible efficiency drops inside of the magnetocaloric material. Furthermore, the effect of the frequency, pumped water volume, angle between magnetic field and water flow as well as the utilization of a heat sink were examined.

To evaluate the applicability of the magnetocaloric effect the cooling power and the COP were determined as well as the behavior of the magnetocaloric material when exposed to a heat source were studied.

MA 10.10 Mon 17:45 HSZ 04

Magneto-optical imaging of magnetocaloric particle ensembles — •Anja Waske[1], Alexander Funk[1], Cecilia Bennati[2,3], Francesco Laviano[2], and Rudolf Schäfer[1] — [1]IFW Dresden — [2]Politecnico of Torino — [3]Istituto Materiali Elettronica e Magnetismo del Consiglio Nazionale delle Ricerche, Italy

A combination of 3D structural information obtained by X-ray computed tomography (XCT) with data of 2D methods like magneto-optical imaging or electron backscatter diffraction (EBSD) can expand the scope of these techniques by providing information in the third dimension, like e.g. the size and position of minority phases and defects hidden beneath the observed surface. For magnetocaloric materials, the nucleation and growth of the ferromagnetic phases was observed to strongly depend on such microstructural features [1]. We show that for La(Fe,Co,Si)13, alpha-Fe inclusions and cracks beneath the observed surface can have an impact on the magnetocaloric transition determined at the surface by e.g. magneto-optical imaging. Furthermore, the behavior of magnetocaloric particle ensembles can be tracked down to single particle behavior.

This work is supported by DFG through SPP 1599 Ferroic Cooling and BASF New Business.

1.Bennati, C., et al., JMMM 2016. 400, 339-343.

MA 10.11 Mon 18:00 HSZ 04

Study of magnetic anisotropy in single crystalline Mn1.9Co0.1Sb — •Mamuka Chikovani, Karen Friese, Jörg Voigt, Jörg Persson, and Thomas Brückel — Forschungszentrum Jülich GmbH, JCNS-2/PGI-4

According to the literature, Co-modified compounds in the Mn2Sb system exhibit three different magnetically ordered states [Kanomata at el 1990]. Among them, the transition from the ferrimagnetic to the antiferromagnetic state features a steep M(T) dependence, making it a promising candidate for magnetocaloric applications. We report on the study of the magnetic anisotropy of a single crystal of Mn1.9Co0.1Sb. We synthesized a polycrystalline sample as precursor material for the single crystal in a cold crucible using induction melting procedure, which is followed by the single crystal growth employing the Czochralski method. To check the quality of the samples, we compared x-ray powder diffraction from the precursor material and from a crushed piece of the single crystal, respectively. In addition, we oriented a single crystal by x-ray Laue diffraction to probe the temperature dependent magnetization along the tetragonal c-direction and in a,b-plane using the vibrating sample magnetometer option of a *Quantum Design PPMS. The magnetic response is distinctly different for the directions under study. Along the c-direction, the response is rather antiferromagnetic, while for a field perpendicular to c, the response seems to be ferro/ferri magnetic. Eventually, based on the results, the additional transition can be explained by a powder average of the single crystal response and lead to a new magnetic phase diagram for this compound.

MA 10.12 Mon 18:15 HSZ 04

Vibrational density of states in magnetocaloric, hydrogenated La(FeSi)$_{13}$-based compounds — •Alexandra Terwey[1], Joachim Landers[1], Soma Salamon[1], Werner Keune[1], Valentin Brabänder[2], Oliver Gutfleisch[2], Michael Y. Hu[3], Jiyong Zhao[3], E. Ercan Alp[3], Markus E. Gruner[1], and Heiko Wende[1] — [1]Faculty of Physics and CENIDE, University of Duisburg-Essen, Germany — [2]TU Darmstadt, Germany — [3]APS, Argonne National Laboratory, USA

By employing ^{57}Fe nuclear resonant inelastic X-ray scattering as in [1], we determined the Fe-projected vibrational density of states (VDOS) of hydrogenated $LaFe_{11.4}Si_{1.6}H_y$ and non-hydrogenated Mn-doped $La_{1.06}Fe_{11.4}Si_{1.5}Mn_{0.1}$ as well as hydrogenated and Mn doped $La_{1.06}Fe_{11.4}Si_{1.5}Mn_{0.1}H_y$. The hydrogenated compounds have potential use as refrigerants due to a first-order magneto-structural phase transition near room temperature. Distinct differences between the VDOS of non-hydrogenated and hydrogenated samples were discovered. This finding is in qualitative agreement with preliminary results from DFT calculations. Vibrational thermodynamic quantities were calculated from the VDOS. The change of vibrational entropy and specific heat across the phase transition was found to be smaller for the hydrogenated than for the non-hydrogenated samples. Our results demonstrate that knowledge of the VDOS is important to understand intrinsic thermodynamics of these magnetocaloric materials. Funding by the DFG (SPP1599) and U.S. DOE (DE-AC02-06CH11357) is acknowledged. [1] M.E. Gruner et al., Phys.Rev.Lett. 114, 057202 (2015)