MM 21: Transport in Materials: Thermal transport

Time: Wednesday 10:15–13:00 Location: H45

MM 21.1 Wed 10:15 H45

Computational modeling of extremely anisotropic van der Waals thermal conductors — Shi En Kim[1], Fauzia Mujid[1], Akash Rai[2], •Fredrik Eriksson[3], Joonki Suh[1], Preeti Poddar[1], Ariana Ray[4], Chibeom Park[1], Erik Fransson[3], Yu Zhong[1], David A. Muller[4], Paul Erhart[3], David G. Cahill[2], and Jiwoong Park[1] — [1]Uni. of Chicago, USA. — [2]Uni. of Illinois UC, USA. — [3]Chalmers, Sweden. — [4]Cornell Uni., USA.

Materials with thermal conduction anisotropy can provide innovative thermal management strategies in integrated circuits. However, artificially engineered material lacks the anisotropy ratios seen in nature. Here, we report extremely anisotropic thermal conductors based on large-area van der Waals thin films with random interlayer rotations, which produce a room-temperature thermal anisotropy ratio close to 900 in MoS_2, one of the highest ever reported. This is enabled by the interlayer rotations that impede the through-plane conductivity, while the long-range intralayer crystallinity maintains high in-plane onductivity. In this contribution we will present the computational analysis of the measured ultralow thermal conductivities in the through-plane direction for MoS_2 (57 ± 3 mW m^{-1} K^{-1}). Using molecular dynamics simulations we quantitatively explain these values and reveal a one-dimensional glass-like thermal transport. Conversely, the in-plane thermal conductivity in these MoS_2 films is close to the single-crystal value. Our work establishes interlayer rotation in crystalline layered materials as a new degree of freedom for engineering-directed heat transport in solid-state systems. Nature 597, 660-665 (2021)

MM 21.2 Wed 10:30 H45

Searching and Finding Thermal Insulators via ab initio Green-Kubo Simulations — •Florian Knoop[1,2], Thomas A.R. Purcell[1], Matthias Scheffler[1], and Christian Carbogno[1] — [1]The NOMAD Laboratory at the FHI-MPG and HU, Berlin, Germany — [2]Department of Physics, Chemistry and Biology (IFM), Linköping University, Sweden

We present a first-principles search for thermal insulators in material space, covering hundreds of compounds in seven space groups, including simple rocksalt and zinc blende structures up to complex perovskites. Using the high-throughput framework *FHI-vibes* [1] and a recently developed measure for the strength of anharmonicity, [2] we identify 120 experimentally known materials with potential for ultralow thermal conductivity <2 W/mK at room temperature, i.e., comparable to those of thermoelectrics such as SnSe. For the 60 most promising candidates, we perform non-perturbative, fully anharmonic *ab initio* Green-Kubo simulations [3] to include all anharmonic effects. Besides revealing seven ultra-insulating compounds, these calculations shed light on the importance of strong anharmonic effects not accessible in perturbative phonon formalisms, e.g., short-lived metastable configurations and precursors of phase transitions.

[1] F. Knoop *et al.*, *J. Open Source Softw.* **5**, 2671 (2020).
[2] F. Knoop *et al.*, *Phys. Rev. Mater.* **4**, 083809 (2020).
[3] C. Carbogno, R. Ramprasad, and M. Scheffler, *Phys. Rev. Lett.* **118**, 175901 (2017).

MM 21.3 Wed 10:45 H45

A spatially resolved optical method to measure thermal diffusivity — •Fei Sun[1], Simli Mishra[1], Philippa H McGuinness[1], Zuzanna h Filipiak[1], Igor Markovic[1], Dmitry A Sokolov[1], Naoki Kikugawa[2], Joseph W Orenstein[3,4], Sean A Hartnoll[5], Andrew P Mackenzie[1,6], and Veronika Sunko[1,3] — [1]Max Planck Institute, CPfS, Dresden, Germany — [2]NIMS, Ibaraki, Japan — [3]UC Berkeley, California, USA — [4]LBNL, California, USA — [5]Univ. of Cambridge, Cambridge, UK — [6]Univ. of St Andrews, St Andrews, UK

We introduce an optical method to directly measure thermal diffusivity across a broad range of temperatures. Two laser beams are used, one as a source to locally modulate the temperature, and the other as a probe of the reflectivity. Thermal diffusivity is obtained from the phase delay between two beams. Combining the technique with a microscope setup allows for spatially resolved measurements. The in-plane diffusivity can be obtained when overlapping the two laser beams instead of separating them in the traditional way, which further enhances the spatial resolution. We demonstrate on two ruthenates: $Sr_3Ru_2O_7$ and $Ca_3Ru_2O_7$. The spatial resolution allowed us to study the diffusivity in single domains of the latter, and we uncovered a temperature-dependent in-plane diffusivity anisotropy. Finally, we used the enhanced spatial resolution to study the Ti-doped $Ca_3Ru_2O_7$. We observed large variations of transition temperature over the same sample, originating from doping inhomogeneity, and pointing to the power of spatially resolved techniques in accessing inherent properties.

MM 21.4 Wed 11:00 H45

Investigation of temperature dependent thermal transport in Sr_2RuO_4 and $Sr_3Ru_2O_7$ over a wide temperature range — Fei Sun[1], •Simli Mishra[1], Ulrike Stockert[1], Ramzy Daou[2], Naoki Kikugawa[3], Robin S Perry[4], Sean A Hartnoll[5], Andrew P Mackenzie[1,6], and Veronika Sunko[1,7] — [1]Max Planck Institute - CPfS, Dresden, Germany — [2]CRISMAT,ENSICAEN, UNICAEN, Normandie Université, Caen, France — [3]NIMS, Ibaraki , Japan — [4]LCN, University College London, London, UK — [5]DAMTP, University of Cambridge, Cambridge, UK — [6]School of Physics and Astronomy, University of St. Andrews, UK — [7]Department of Physics, University of California, Berkely, USA

We use optics to study thermal transport by modifying a typical laser pump-probe technique. A direct measurement of thermal conductivity is very challenging at high temperatures. In contrast, our method measures temperature dependent diffusivity over a wide range of temperature (10K up to 330K). As thermal diffusivity is the ratio of the thermal conductivity to the heat capacity, we can access the higher temperature thermal conductivity by an optical measurement of diffusivity and standard heat capacity measurement. We have used this technique to measure thermal diffusivity of two strongly correlated metals, Sr_2RuO_4 and $Sr_3Ru_2O_7$. This temperature dependent thermal transport combined with the resistivity measurements offers an insight into the electronic and phononic contributions to the quasiparticle scattering.

MM 21.5 Wed 11:15 H45

Simultaneous measurement for the complete characterization of thermoelectric materials - real experiment and its digital twin — •Severin Kopatz[1], Eckhard Müller[1,2], and Pawel Ziolkowski[1] — [1]Institute of Materials Research, German Aerospace Center, Cologne, Germany — [2]Institute of Inorganic and Analytical Chemistry, Justus Liebig University Gießen, Gießen, Germany

Thermoelectric (TE) materials directly convert thermal energy into electric energy. Their efficiency depends on the figure of merit zT (zT = $S^2\sigma/\kappa$). Commonly, the individual transport properties, mainly the electrical conductivity (σ), the Seebeck coefficient (S), and the thermal conductivity (κ) are measured in separate measurement set-ups. Here, we report on the 'Combined ThermoElectric Measurement' (CTEM) apparatus, which provides temperature-dependent measurements of the aforementioned transport properties on a single TE sample such as Co-doped $FeSi_2$ simultaneously. The measurements are carried out in a temperature range between room temperature and 600 °C. Since experimental CTEM results deviate from reference values, we introduce a digital twin of the CTEM. In combination with the real experiment, the numerical model allows to study possible error sources. One of the examples discussed in this talk concerns radiative effects which have to be considered especially at high temperatures above 300 °C and affect the measurement of the thermal conductivity in particular.

15 min. break

MM 21.6 Wed 11:45 H45

Appearance of non-equilibrium grain boundaries in additively manufactured high-entropy CoCrFeMnNi alloy — •Nuri Choi[1,2], Vladislav Kulitckii[1], Josua Kottke[1], Bengü Tas[1], Jungho Choe[3], Ji Hun Yu[3], Sangsun Yang[3], Joo Hyun Park[2], Jai Sung Lee[2], Gerhard Wilde[1], and Sergiy Divinski[1] — [1]Institute of Material Physics, University of Münster, Germany — [2]Dep. of Mat. Sci. & Chem. Eng., Hanyang University, South Korea — [3]Center for 3D Printing Materials Research, KIMS, South Korea

Additive manufacturing process with laser melting includes the repetitive melting/solidification, which generates residual thermal stresses in the bulk material, alongside with creation of various defects, including

point defects, dislocations and numerous grain boundaries. How far the kinetic properties of these interfaces are modified by the processing remains an open issue, that is of fundamental importance for such phenomena as creep, phase transformation and precipitation. In the present study, grain boundary diffusion in additively manufactured CoCrFeMnNi high-entropy alloys is measured using the radiotracer technique. Since the additive manufacturing results in a hierarchical microstructure, grain boundary diffusion is examined in different samples prepared via different scan/build strategies. A non-equilibrium state of a fraction of high-angle grain boundaries is discovered. The non-equilibrium state is shown to relax after annealing at low temperatures without measurable microstructural changes. The grain boundary diffusivities of the 3D printed alloys are discussed with respect to those for fully homogenized cast or severe plastically deformed alloys.

MM 21.7 Wed 12:00 H45

Grain boundary diffusion and segregation of Cr in high-purity Ni bi-crystals with a Σ11 grain-boundary — •D. Gaertner[1], S. V. Sevlikar[1], G. M. Muralikrishna[1], D. Cozlin[2], D. Irmer[2], D. Schreiber[3], B. Tas[1], M. Vaidya[1], T. Brink[4], V. E. Esin[2], C. Duhamel[2], G. Dehm[4], V. Razumoskii[3], G. Wilde[1], and S. V. Divinski[1] — [1]Institute of Materials Physics, University of Münster, Münster, Germany — [2]MINES ParisTech, PSL University, Centre des Matériaux, Évry, France — [3]Materials Center Leoben Forschung GmbH (MCL), Austria — [4]Max-Planck-Institut für Eisenforschung GmbH, Düsseldorf, Germany

Grain-boundary diffusion of Cr in a Ni near Σ11(113)[110] bi-crystal is measured in an extended temperature interval between 503 K and 1303 K using the radiotracer technique. The grain boundary diffusion coefficients, D_{gb}, and the triple products, P are determined in the C- and B-type kinetic regimes, observing a strong deviation from the otherwise linear Arrhenius-type temperature dependence above 1000 K. The present results substantiate that the segregation factor of Cr in Ni is about unity, being in agreement with the preliminary findings in polycrystalline counterparts measured by SIMS and are fully supported by DFT-based calculations. Extensive MD simulations with empirical interatomic potentials substantiate an extreme stability of the Σ11 grain-boundary structure in Ni from 0 K up to melting point. The non-linear Arrhenius temperature dependence is interpreted in terms of pronounced anharmonic contributions to defect formation at elevated temperatures.

MM 21.8 Wed 12:15 H45

grain boundary melting phase transition in Ni-Bi system — •Baixue Bian[1], Boris Straumal[2], Sergiy Divinski[1], and Gerhard Wilde[1] — [1]Institute of Materials Physics, University of Münster, Wilhelm-Klemm-Str. 10, 48149 Münster, Gemany — [2]Chernogolovka

Grain boundary segregation may drastically decrease the strength of structural materials. The intermediate temperature embrittlement of Ni-based alloys is a classical example of a catastrophic matrix degradation caused by solute segregation, in particular by Bi. In this work, the impact of grain boundary segregation of Bi in dilute polycrystalline Ni-Bi alloys on Ni grain boundary diffusion was systematically studied as function of temperature and composition. The radiotracer technique was used applying the 63Ni radioisotope and measuring grain boundary diffusion in both B- and C-type kinetic regimes after Harrison*s classification. An abrupt increase of the grain boundary diffusivities by about two orders of magnitude was observed at certain Bi contents which are unequivocally in the single-phase region of the corresponding bulk phase diagram. The present results unambiguously prove the occurrence of a pre-melting grain boundary phase transition in the Ni-Bi system.

MM 21.9 Wed 12:30 H45

Investigation of Phase Transitions in Polycrystalline Tungsten Trioxide Films during Ion Insertion and Extraction by in Situ Transmission and Raman Spectroscopy — •Markus S. Friedrich[1,2], Alexander G. Strack[1,2], Paul K. Tuchecker[1,2], Jan L. Dornseifer[1,2], and Peter J. Klar[1,2] — [1]Institute of Experimental Physics I, Giessen, Germany — [2]Center for Materials Research, Giessen, Germany

The International Energy Agency stated in its "European Union 2020 Energy Policy Review" that in 2016 in the European Union alone 152 TWh where consumed for air conditioning (AC), despite only six percent of the global stock of AC units is operated in the EU. So called "smart windows", e.g. windows based on electrochromic materials, such as tungsten trioxide, are promising candidates to reduce this kind of energy consumption. The coloration and bleaching of the EC material is based on the reversible insertion and extraction of small ions inside the material. Ion diffusion plays a key role in this process and therefore needs to be understood in depth in order to enable the optimization of future devices. Burkhardt et al. found, that the diffusion coefficient in this material is dependent on the concentration of already incorporated ions. We suggest, that this originates from changes in the crystal structure of the thin films. To substantiate this suggestion spatially and temporally resolved in situ transmission and Raman spectroscopic experiments were performed during potentiostatic ion insertion and extraction to correlate the diffusion of small ions inside the material with changes of its crystal structure.

MM 21.10 Wed 12:45 H45

Self-diffusion in ge2sb2te5 thin films — •qingmei gong, sergiy divinski, and gerhard wilde — University of Münster, Institute of Materials Physics, Wilhelm-Klemm-str. 10, 48149 Münster, Germany

Phase change memory devices (PCM) are considered as one of the most mature technologies among the emerging non-volatile memories and are based on the reversibility of the amorphous-to-crystalline transition within a nanosecond timescale. Ge2Sb2Te5 is the most widely studied composition. In this work, the self-diffusion in Ge2Sb2Te5 thin films is measured using secondary ion mass spectroscopy (SIMS) and applying the highly enriched natural 122Te isotope. A 200 nm thick layer of Ge2Sb2Te5 was deposited on a Si substrate by DC magnetron sputtering at room temperature. Subsequently, a thin layer of 122Te was deposited on as-prepared Ge2Sb2Te5/Si samples using physical vapor deposition. In the as-deposited state, Ge2Sb2Te5 was amorphous as confirmed by XRD and transmission electron microscopy. The Te diffusion coefficients in Ge2Sb2Te5 were estimated from SIMS measurements after annealing at different temperatures below crystallization onset.