MM 6: Phase Transformations

Time: Monday 15:45–18:30 Location: H10

MM 6.1 Mon 15:45 H10

Phase transitions in 2D halide perovskites using machine learned potentials — •Erik Fransson, Julia Wiktor, and Paul Erhart — Chalmers University of Technology, Gothenburg, Sweden

Hybrid halide perovskites are a promising class of materials for various applications, including high-efficiency solar cells, lasers, and light-emitting diodes. So-called two-dimensional (2D) halide perovskites, composed of a small number of perovskite layers stacked on top of each other and separated by organic cations that act as spacers, have much improved stability compared to their 3D counterparts. Here, we focus on the prototypical perovskite methylammonium lead halide (MAPI), and demonstrate that the dimensionality of these 2D materials and the choice of organic linker molecules can have a strong impact on phase transitions in these systems. This is investigated through large-scale molecular dynamics simulations using machine-learned potentials. We analyze the phase transition temperatures and characteristics with varying numbers of perovskite layers to understand how the transition properties change as a function of the system's dimensionality. For a larger number of perovskite layers, the 3D bulk phase transition temperature is recovered, whereas, for only a few perovskite layers, the phase transition temperature shifts up by about 100 K. Additionally, we observe surface effects, such as the surface layers (closest to the organic linker) exhibiting stronger octahedral tilting and undergoing phase transitions at higher temperatures (about 100 K) compared to the interior bulk layers.

MM 6.2 Mon 16:00 H10

Quantum phase diagram of sulfure hydride — •Marco Cherubini and Michele Casula — IMPMC, CNRS, Paris, France

In the recent rush towards room temperature superconductivity, hydrogen-based materials are the most prosiming candidates. Sulfulr hydride exhibits a maximum superconductive critical temperature of about 200K at 150 GPa. To our knowledge, a comprehensive theoretical characterization of the phase diagram of sulfur hydride in a wide temperature range is still missing in literature. To address this, we performed path integral molecular dynamics simulations (PIMD). The description of the low temperature regime has been made feasible by the use a machine learning potential trained on ab-initio data. We found three different regimes. At high pressure, the phase diagram is dominated by a paraelectric phase, with symmetric hydrogen bonds. Reducing the pressure, we observed first a regime characterized by finite local dipole moments and finally, at even lower pressures, the ferroelectric regime. Quantum simulations show that the formation of finite local moments is temperature-independent, unlike the transition to the ferroelectric regime. Classical simulations, by contrast, showed stronger temperature dependence for both the transitions and significantly higher critical pressures, highlighting the impact of quantum nuclear fluctuations.

MM 6.3 Mon 16:15 H10

Magnetic-field induced phase transition crossover in the triangular-lattice antiferromagnet: Ba_3CoSbO_9 — •Sanjay Kumar[1], Koushik Chakraborty[2], Aditi Agrawal[2], Shalini Mishra[3], M. P. Saravanan[2], Arvind Kumar Yogi[2], Satyapal S. Rathore[4], and Rashi Nathawat[1] — [1]Functional Ceramics and Smart Materials Lab, Department of Physics, Manipal University Jaipur, Jaipur - 303007, India — [2]UGC-DAE Consortium for Scientific Research, University Campus, Khandwa Road, Indore-452001, India — [3]Department of Physics, Govt. Holkar Science College, Indore (M.P.) 452001, India — [4]Department of Physics, Cluster University of Jammu, Jammu * 180001, India

We report magnetic and structural properties of triangular-lattice antiferromagnet Ba_3CoSbO_9 by means of x-ray diffraction (XRD), magnetic susceptibility, specific heat, x-ray photoelectron spectroscopy (XPS), and dielectric measurements. Thermodynamic measurements show a long-range ordered (LRO) state at Néel temperature $T_N = 3$ K which was found to be shift at higher temperatures at about $T_N = 4.1$ K under the higher magnetic fields. Moreover, we have found higher Curie-Weiss temperature θ_{CW} ∼-133.2 K from the inverse susceptibility fit which reveals frustration parameter about 44, suggesting magnetic lattice is highly frustrated. Further, a spin-glass state signature was evident at around 6.5 K, which was found to be fully suppressed at a higher magnetic field (H ∼16 T). Interestingly, Ba_3CoSbO_9 exhibits a broad maximum at around T_{max} ∼5 K which becomes pronounced as the magnetic field is increased.

MM 6.4 Mon 16:30 H10

Anomalous spin-lattice coupling in the quasi-one-dimensional spin-1 corrugated skew-chain antiferromagnet: $Ni_2V_2O_7$ — •Arvind Kumar Yogi[1], Hemant Singh Kunwar[1], Koushik Chakraborty[1], Aditi Agrawal[1], Binoy Krishna De[1], Pragati Sharma[1], Shalini Mishra[2], D. T. Adroja[3], Mayanak Kumar Gupta[4], R. Mittal[4], R. Venkatesh[1], and V. G. Sathe[1] — [1]UGC-DAE Consortium for Scientific Research, University Campus, Khandwa Road, Indore-452001, India — [2]Department of Physics, Govt. Holkar Science College, Indore (M.P.) 452001, India — [3]ISIS Neutron and Muon Facility, STFC, Rutherford Appleton Laboratory, Chilton, Oxfordshire OX11 0QX, United Kingdom — [4]Solid State Physics Division, Bhabha Atomic Research Centre, Trombey, Mumbai-400005, India

We report spin-lattice coupling through detailed structural, magnetic, and lattice-dynamics studies of the $S = 1$ quasi-one-dimension lattice $Ni_2V_2O_7$ compound. Our susceptibility χ (T) and heat capacity measurements (C_P/T) measurement conclusively show that the antiferromagnetic transition occurs at T_{N1} = ∼6.7 K and T_{N2} = ∼5.8 K. From detailed lattice dynamics, two Raman mode showed anomalous lattice softening below ∼100 K due to Ni dimerization as supported by ordering of the J_1 exchange interaction. Importantly, the spin-lattice coupling has been established below ∼100 K and the spin-lattice coupling constant (λ_{sp}) for various Raman modes has been deduced which shows multiferroic behaviour below T_{N1}. In addition, the detailed study of lattice dynamics by first principle calculation is presented.

MM 6.5 Mon 16:45 H10

Chirality in the Kagome Metal CsV_3Sb_5 — H.J. Elmers[1], O. Tkach[1], Y. Lytvynenko[1], P. Yogi[1], M. Schmitt[2,3], D. Biswas[2], J. Liu[2], S.V. Chernov[4], Q. Nguyen[5], M. Hoesch[4], D. Kutnyakhov[4], N. Wind[4,6], L. Wenthaus[4], M. Scholz[4], K. Rossnagel[4,6], A. Gloskovskii[4], C. Schlueter[4], A. Winkelmann[7], A.-A. Haghighirad[8], T.-L. Lee[2], M. Sing[3], R. Claessen[3], M. Le Tacon[8], J. Demsar[1], G. Schönhense[1], and •O. Fedchenko[1] — [1]JGU Mainz, Germany — [2]DIAMOND, Didcot, United Kingdom — [3]Physikalisches Institut Würzburg, Germany — [4]DESY, Germany — [5]SLAC, Menlo Park, USA — [6]CAU Kiel, Germany — [7]University of Krakow, Poland — [8]KIT, Karlsruhe, Germany

Using x-ray photoelectron diffraction (XPD) and angle-resolved photoemission spectroscopy, we study photoemission intensity associated with the changes in the geometric and electronic structure of the kagome metal CsV_3Sb_5 upon transition to an unconventional charge density wave (CDW) state. The XPD patterns reveal the presence of a chiral atomic structure in the CDW phase. Using circularly polarized x-rays, we have found a pronounced non-trivial circular dichroism in the angular distribution of the valence band photoemission in the CDW phase, indicating a chirality of the electronic structure. This observation is consistent with the proposed orbital loop current order. The results suggest an antiferromagnetic coupling of the orbital magnetic moments along the c-axis.

[1] H.J. Elmers et al., e-print on arXiv, 2408.03750 (2024).

15 min. break

MM 6.6 Mon 17:15 H10

Pressure-temperature phase diagram of calcium using quantum-accurate finite-temperature free energies — •Raynol Dsouza[1], Marvin Poul[1], Liam Huber[2], and Jörg Neugebauer[1] — [1]Max Planck Institute for Sustainable Materials, Düsseldorf, Germany — [2]Grey Haven Solutions, Vancouver, Canada

Pure calcium has been experimentally shown to exhibit several stable phases across a range of high pressures. While many of these phases have been theoretically predicted using T=0 K ab inito calculations [1], the relative stability of the distorted simple cubic phases above 30 GPa at finite temperatures remains unclear. To address this ambiguity, we developed an Atomic Cluster Expansion (ACE) potential [2] for calcium, fitting it to a structural dataset generated using the ASSYST

methodology outlined in [3]. Quantum-accurate finite-temperature free energies were determined using the Temperature Remapping Approximation (TRA) [4]. The resulting pressure-temperature phase diagram offers new insights into the phase stability of calcium at elevated pressures and temperatures.

[1] Ishikawa et al., https://doi.org/10.1103/PhysRevB.81.092104 [2] Bochkarev et al., https://doi.org/10.1103/PhysRevMaterials.6.013804 [3] Poul et al., https://doi.org/10.21203/rs.3.rs-4732459/v1 [4] Dsouza et al., https://doi.org/10.1103/PhysRevB.105.184111

MM 6.7 Mon 17:30 H10

Phase-stability Study of the Marcasite-Structure Solid Solutions (Fe,TM)Sb_2 (TM = Cr, Ni) Synthesized via Combinatorial Co-Deposition and Antimonization — •Martin Kostka[1], Luqman Mustafa[1], Maik Golombiewski[1], Jill Fortmann[2], Aurelija Mockute[2], Alan Savan[2], Alfred Ludwig[2], Adreas Kreyssig[1], and Anna Böhmer[1] — [1]Experimental physics IV, Ruhr University Bochum, 44801 Bochum — [2]Materials Discovery and Interfaces, Institute for Materials, Ruhr University Bochum, 44801 Bochum

Transition-metal di-pnictides of the pyrite-marcasite family are model systems for crystal chemistry. We investigate phase formation and stability of transition-metal di-antimonides in the orthorhombic marcasite structure. We employ a two-step synthesis of granular films, using combinatorial co-deposition of the transition metals and subsequent antimonization at different temperatures. This technique allows efficient exploration of substitution ranges and lattice parameter evolution within the marcasite structure. We investigate the formation and crystal structures of the (Fe,Ni)Sb_2 and (Fe,Cr)Sb_2 substitution series. We evidence the continuous substitution of $FeSb_2$ (class A marcasite) with Ni up to $Fe_{0.5}Ni_{0.5}Sb_2$, and a clear phase separation between $Fe_{0.5}Ni_{0.5}Sb_2$ and a Ni-rich $Ni_{1-z}Fe_zSb_2$ phase (class B marcasite). Cr-substituted $FeSb_2$ shows a subtle phase separation into an Fe-rich $Fe_{1-y}Cr_ySb_2$ and a Cr-rich $Cr_{1-z}Fe_zSb_2$-phase (both class A marcasite) when synthesized at 500°C, but not at higher Temperatures. We acknowledge support from DFG (TRR288,A02).

MM 6.8 Mon 17:45 H10

Investigation of glass formation in Pd40Ni40P20 metallic glass via fast scanning calorimetry — •Hongshuai Li, Sher Bahadur Raut, and Gerhard Wilde — Institute of Materials Physics, University of Münster, Wilhelm-Klemm-Str. 10, 48149 Münster, Germany

Pd-Ni-P metallic glasses (MGs) exhibit exceptional glass-forming ability; in fact, Pd40Ni40P20 was the first bulk metallic glass-forming alloy discovered. Understanding glass formation in these materials requires exploring factors that impede correlated atomic motions in glass-forming liquids. This study investigates the atomic relaxation and thermal history of Pd40Ni40P20 MG, utilizing data from multiple cycles of heating around the glass transition temperature, activation energy determined by flash differential scanning calorimetry (DSC), and critical cooling rates. Additionally, we examine the effects of annealing time on the DSC signals at high heating rates of up to 1000 K/s. Our results demonstrate that adjustments in cooling rates and annealing duration significantly influence glass transition behavior and thermal stability. These findings enhance our understanding of the mechanisms underlying glass formation in Pd-Ni-P MGs and their potential applications in advanced materials.

MM 6.9 Mon 18:00 H10

Tuning excitonic transition by Cr doping and associated lattice softening in the vdW chalcogenide: Ta2NiSe5 — •Isha Isha[1], Koushik Chakraborty[1], Aditi Agrawal[1], M. Isobe[2], and Arvind Kumar Yogi[1] — [1]UGC-DAE Consortium for Scientific Research, University Campus, Khandwa Road, Indore-452001, India — [2]Max-Planck-Institut für Festkörperforschung, Heisenbergstr. 1, D-70569 Stuttgart, Germany

We have successfully grown defect free ultra-high quality single crystals of Cr doped Ta2NiSe5, maximum of 10% at Ni site in the lattice. Our preliminary STM results unambiguous provide clear signature of CDW phase, vdW layers and the vdW gap for Ta2NiSe5 which was found to be ~3.5 Å. We found a clear anomaly at the critical temperature Tc = 326 K in our transport measurement over rod shaped crystals - a transition into an excitonic insulator ground state as reported for parent Ta2NiSe5 compound. It is noteworthy to highlight our finding that Cr doping significantly suppresses the insulating ground state and the 10% doping reduces the resistivity of three orders. In addition, our detailed high-temperature Raman scattering for parent as well as Cr doping reveals sharp Raman modes and significant shift and suppression for 230 cm-1 phonon mode which might be associated with electron-phonon coupling, which may helps in resistivity drop as evident in our transport data. In addition, our detailed Raman scattering study suggests the persistence of lattice softening which may be due to strong electron-lattice coupling, and the origin is presumably due to strong excitonic fluctuations.

MM 6.10 Mon 18:15 H10

Magnetic Imaging of the Local Insulator to Metal Transition in CaRuO by NV Magnetometry — •Hayden Binger[1], Cissy Suen[2], Elina Zhakina[1], Luke Turnbull[1], Yejin Lee[1], Young-Gwan Choi[1], Max Krautloher[2], Bernard Keimer[2], Claire Donnelly[1], and Uri Vool[1] — [1]Max Planck Institute for the Chemical Physics of Solids,Dresden, Germany — [2]Max Planck Institute for Solid State Research, Stuttgart, Germany

The current-driven insulator to metal transition in $Ca_2Ru_2O_4$ is a fascinating phenomenon where increasing current driven across a sample causes a smaller voltage difference to develop. While this transition has been well studied in bulk $Ca_2Ru_2O_4$ through transport, and recently investigated by ARPES, little is known about the local character of the transition - especially in nanoscale devices. In this work we utilize scanning NV magnetometry to measure the local magnetic field induced by the local current in 100 nm thick lamellas of $Ca_2Ru_2O_4$. We observe a nonuniform field distribution that reveals the local current channels, allowing us to image the local character of the insulator to metal transition in $Ca_2Ru_2O_4$.