Materials Physics in Space and ground-based Research with X-rays and Neutrons

Andreas Meyer*

Fan Yang, Elke Sondermann, Dirk Holland-Moritz, Florian Kargl, Thomas Voigtmann, Jörg Drescher et al.

Institute of Materials Physics in Space
German Aerospace Center DLR, Cologne

*since 10/2021 seconded to ILL

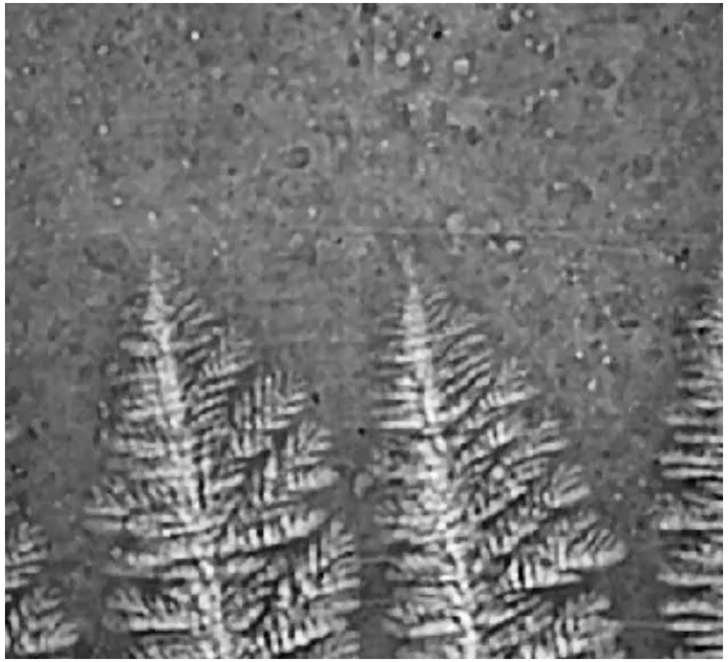
DLR

HELMHOLTZ
RESEARCH FOR GRAND CHALLENGES

Materials Design from the Melt

solidification and microstructure formation in alloys

- develop an in-depth understanding of morphological transitions by in-situ *experiments under well defined conditions* on ground and in space

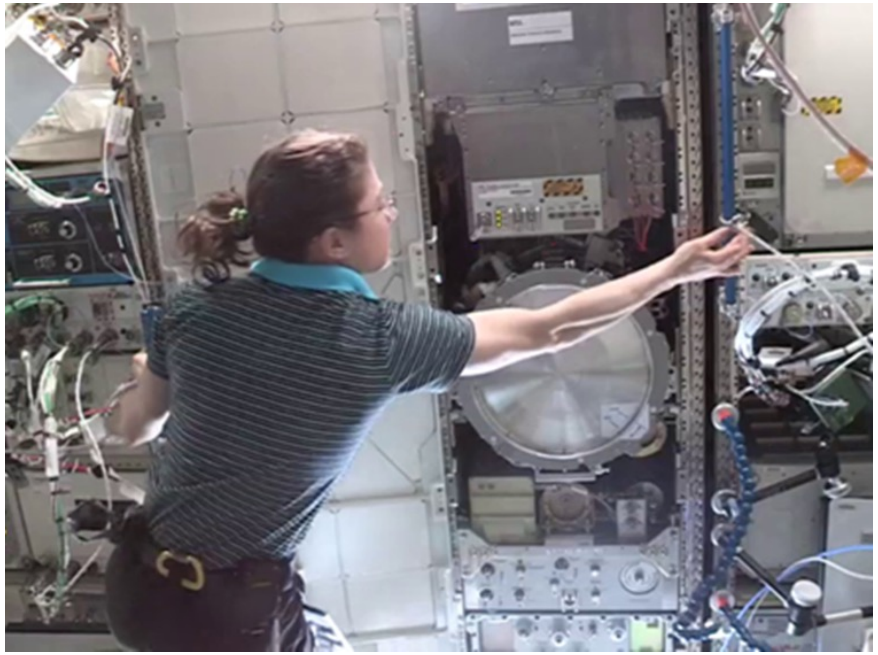


dendrite growth in Al-Cu at ESRF:
R. Mathiesen et al. NTNU

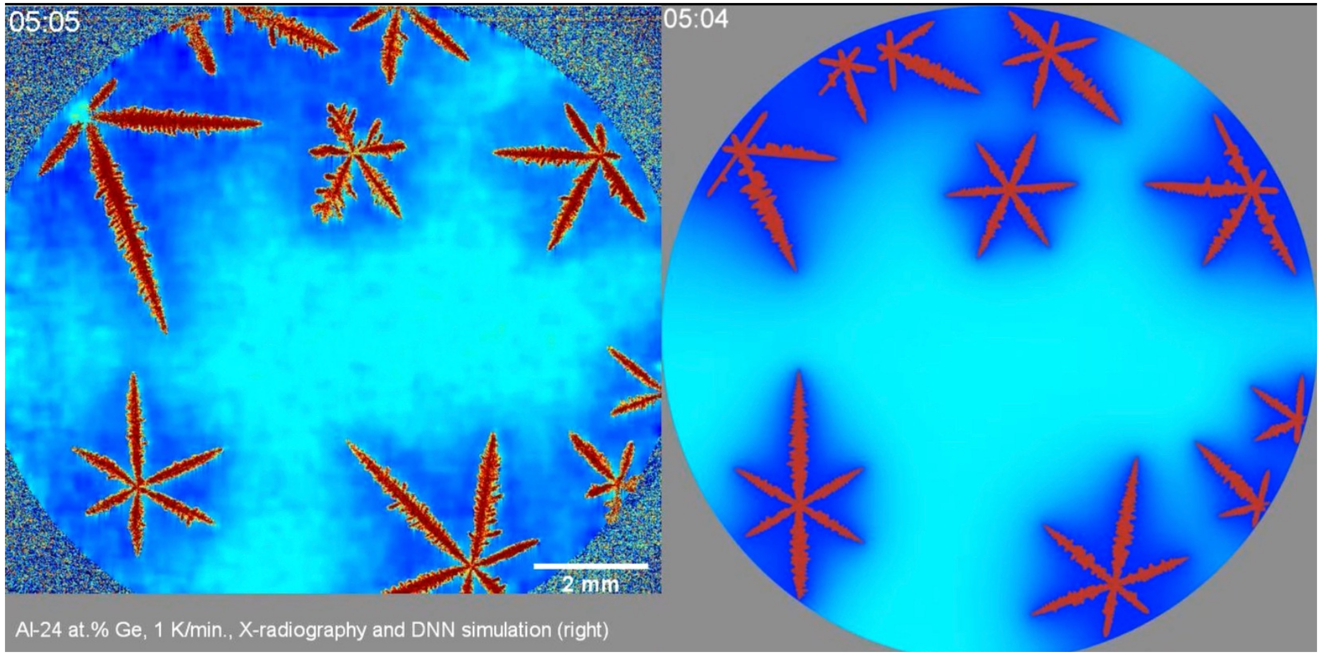
Materials Design from the Melt

solidification and microstructure formation in alloys

- develop an in-depth understanding of morphological transitions by in-situ *experiments under well defined conditions* on ground and in space
- requires an understanding of the liquid state including measurements of *accurate transport coefficients*



Materials Science Lab (ESA-MICAST – ISS)

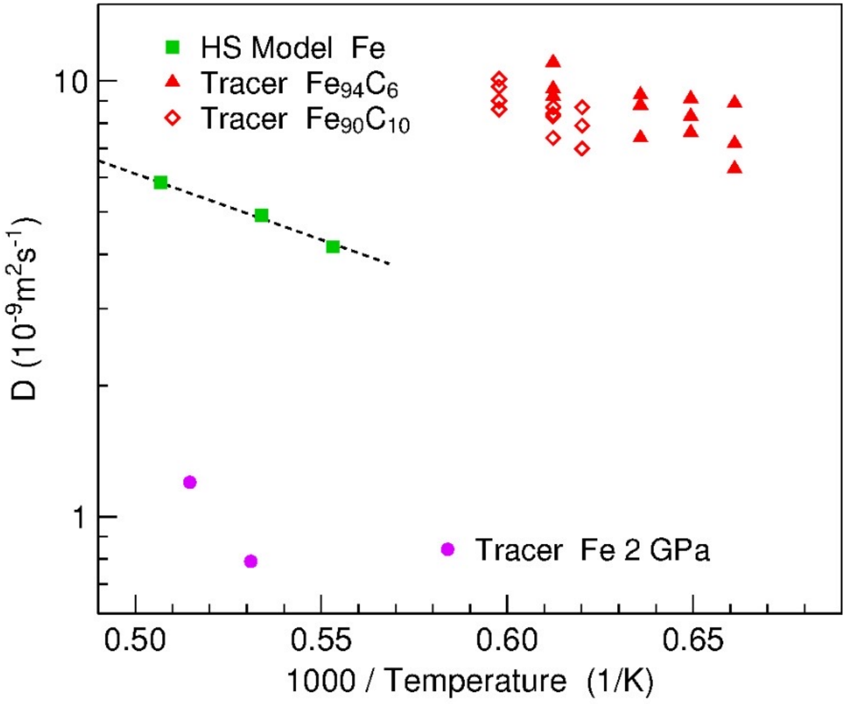


X-ray radiography and modeling of diffusion controlled growth

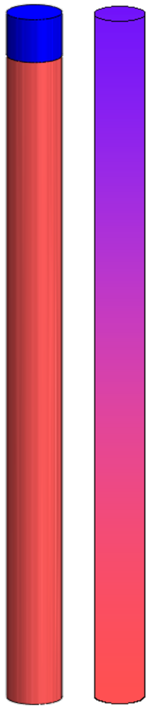
Becker, Sturz, Bräuer, Kargl, Acta Mater (2020)

Measurement of Self- and Interdiffusion Coefficients

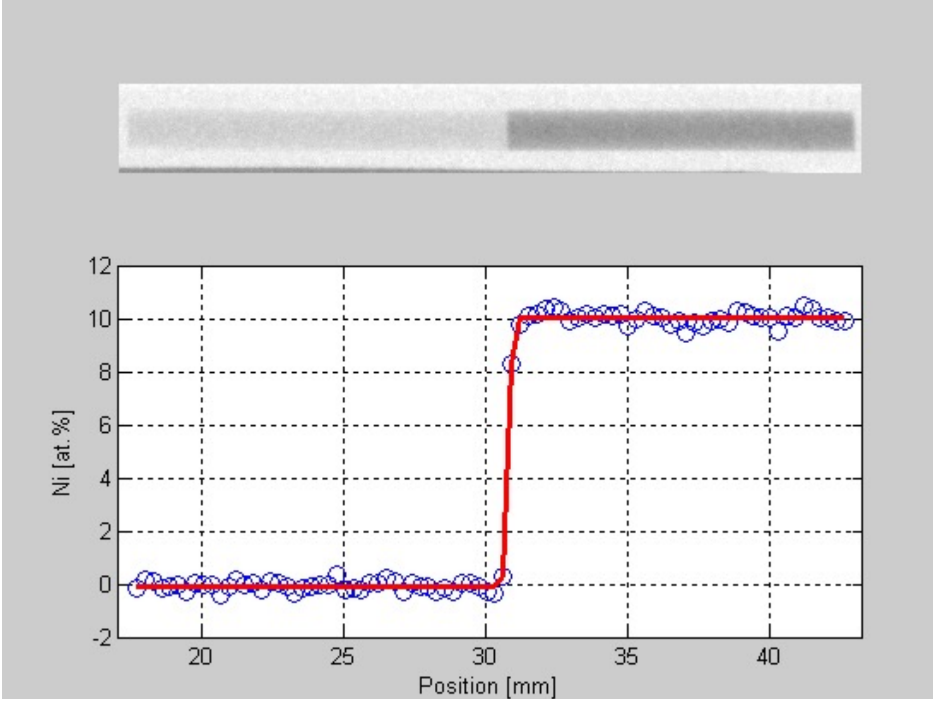
long-capillary set-up with ex-situ analysis versus in-situ experiments



^{59}Fe tracer



Yang et al.
Trans. AIME
1956



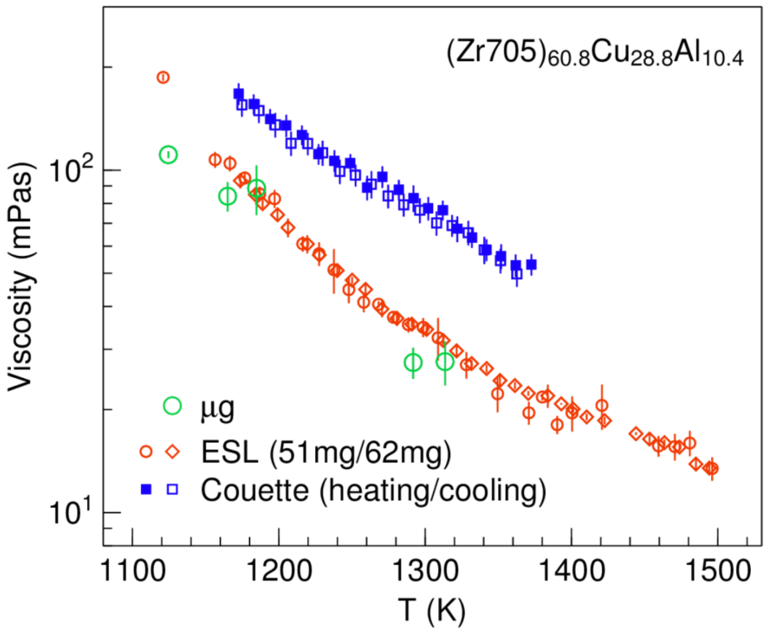
Zhang, Griesche, Meyer, Phys. Rev. Lett. (2013)

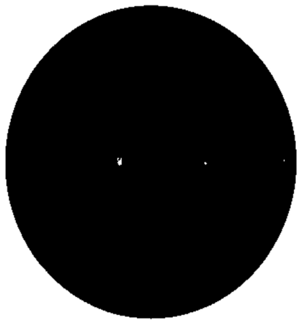
- classical long-capillary experiments: data hampered by melting, convection, solidification
- *accurate values with in-situ techniques*: quasielastic neutron scattering (QENS) and X-ray radiography

Meyer, Hennig, Unruh, Kargl, J. Phys.: Cond. Matter (2019)

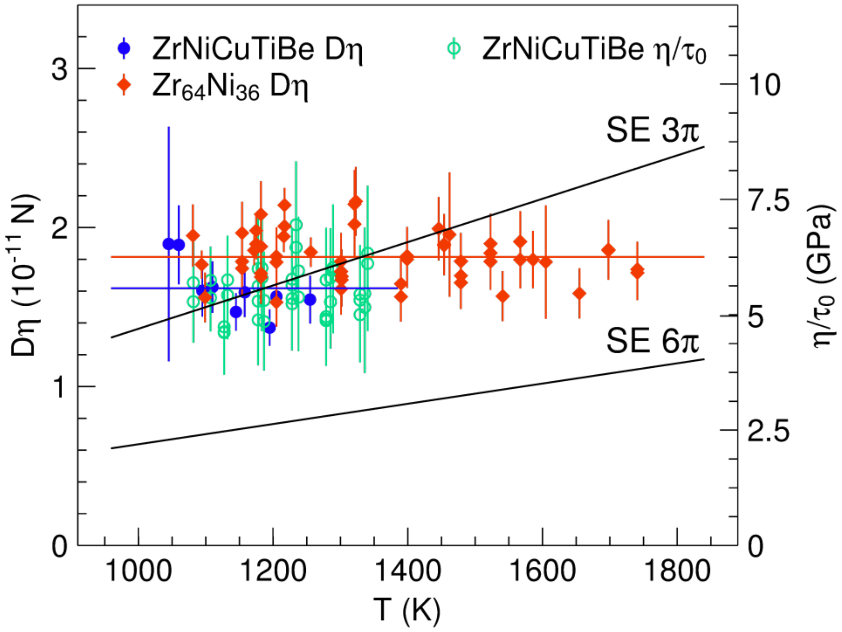
Accurate Viscosity Data of Liquid Alloys

ElectroStatic (ESL) and ElectroMagnetic (EML) Levitation





freely suspended
oscillating droplet



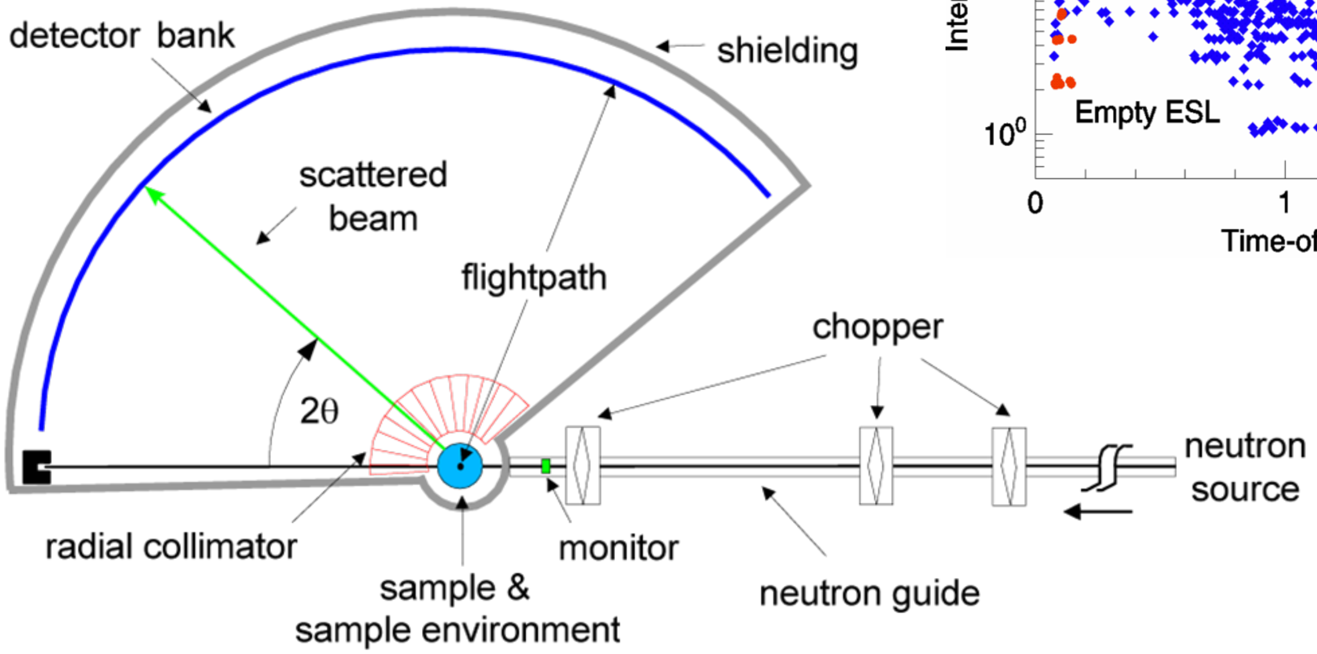
- new ESL technique → accurate viscosity data; μg EML experiments to validate technique
- accurate data: self diffusion D times viscosity η ~ const. not $D \cdot \eta \propto kT$
- *Stokes-Einstein relation not valid* in dense metallic liquids:
  → structural relaxation dominates diffusion D and viscosity η

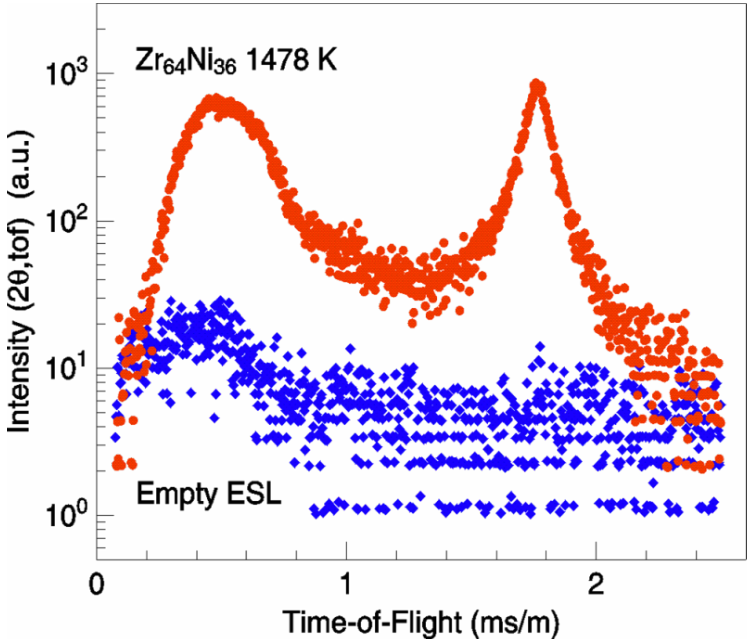
Jonas, Yang, Hambree, Busch, Meyer, Appl. Phys. Lett. (2018); Yang, Unruh, Meyer, EPL (2014); Brillo, Pommrich, Meyer, Phys. Rev. Lett. (2011)

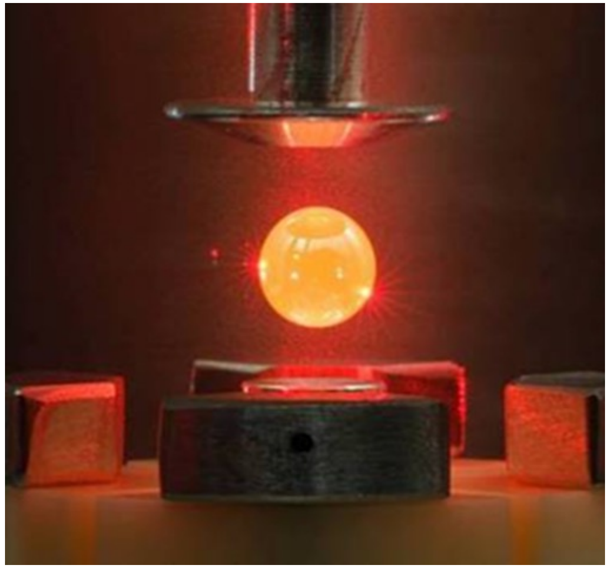
Quasielastic Neutron Scattering on Levitated Droplets

time-of-flight spectroscopy

measurement of atomic dynamics
on an *absolute scale*







electrostatic levitation

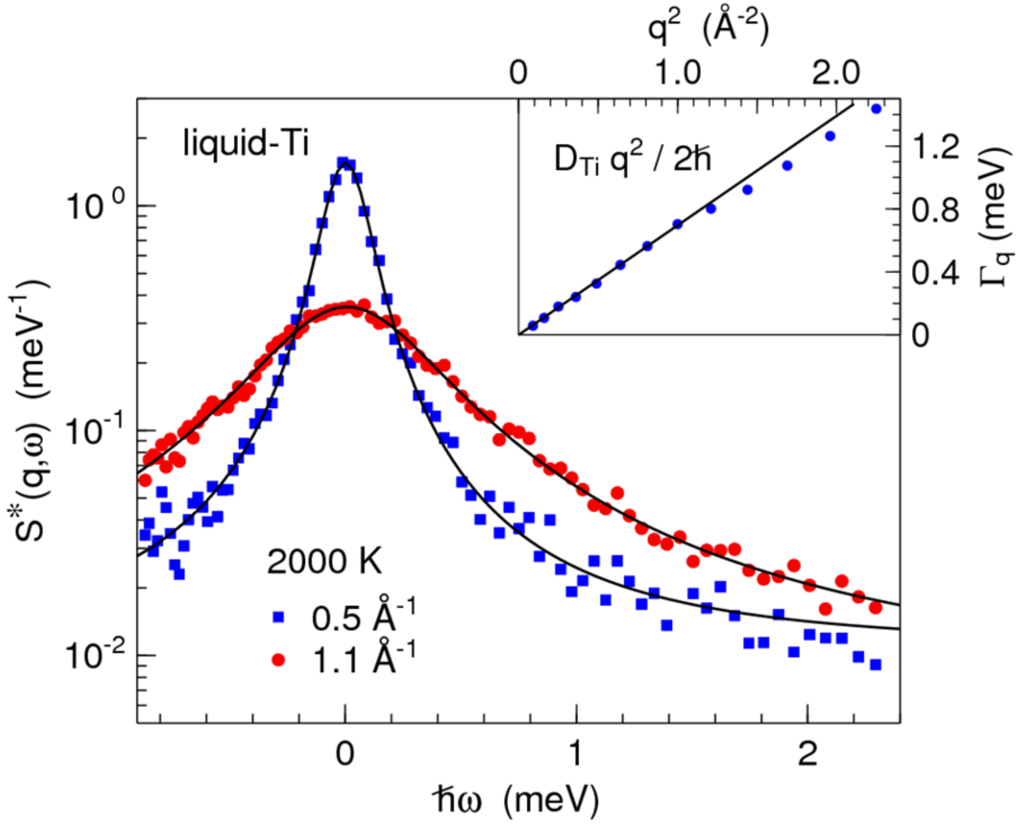
access to:

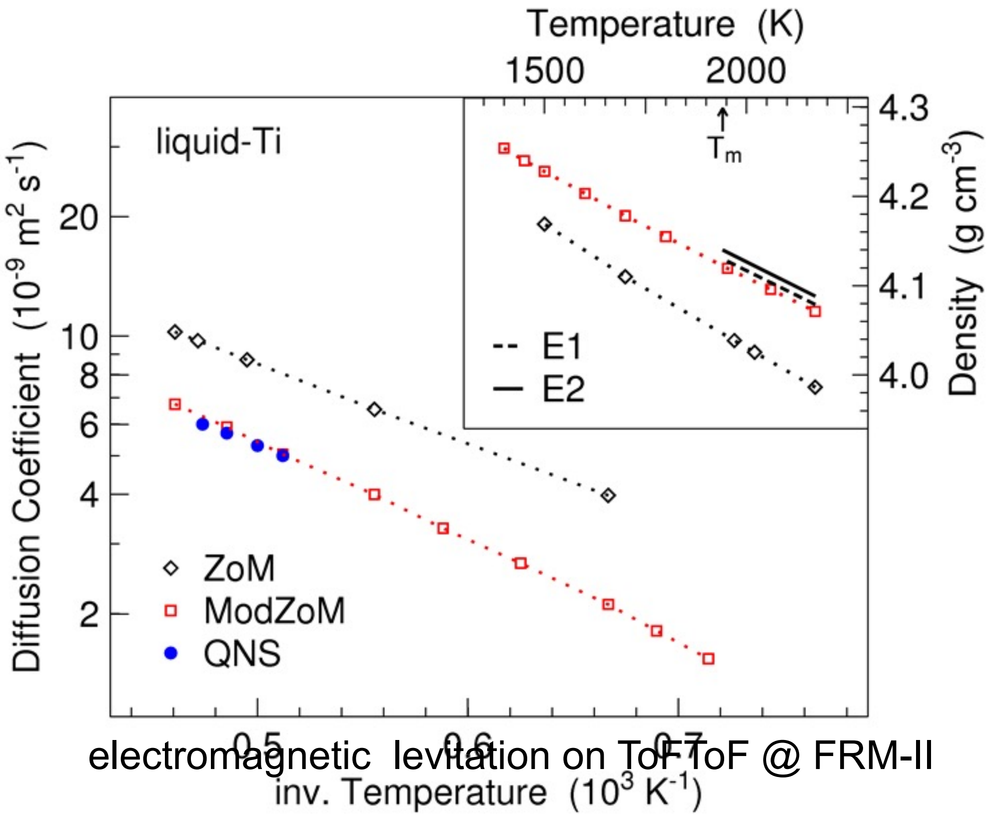
- chemical reactive samples
- extreme temperatures
- undercooling

ESL / ToFToF FRM-II: Kordel, Klein, Yang, Holland-Moritz, Peters, Meyer, Phys. Rev. B (2011)

Quasielastic Neutron Scattering on Levitated Droplets

time-of-flight spectroscopy





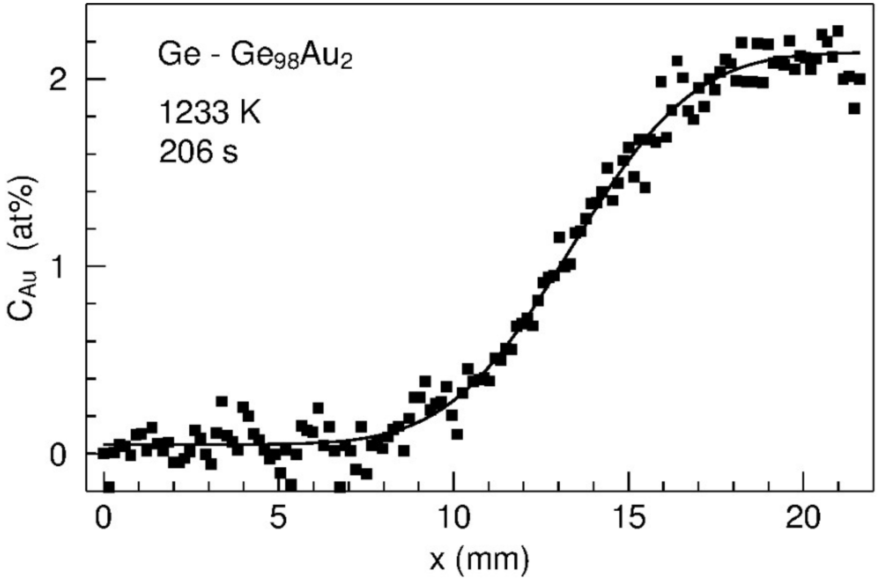
electromagnetic levitation on ToF-ToF @ FRM-II

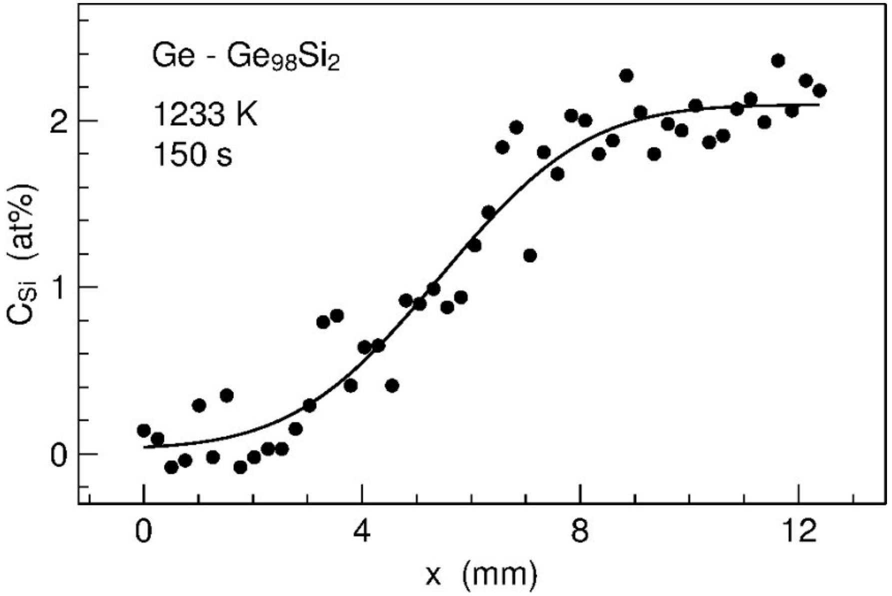
- small q: QENS signal dominated by *incoherent* scattering
- *rescaling* of MD interaction potential with diffusion data:
  good agreement with density, thermal expansion, melting point

Horbach, Rozas, Unruh, Meyer, Phys. Rev. B (2009)

Diffusion of Minor Additions to Liquid Germanium

QENS, X-ray radiography, sounding rocket experiment



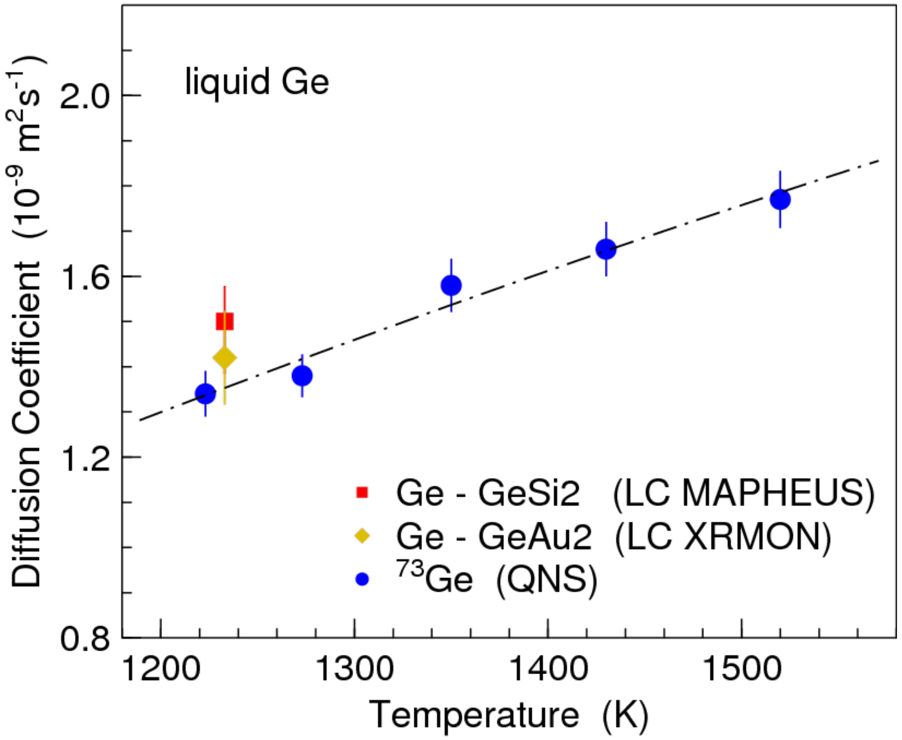


- crystalline Ge and Si: diffusion coefficients of minor additions differ by orders of magnitude
- behavior in loosely packed liquid Ge and Si ?
- *kinetic theories* of diffusion in the liquid state: size and *mass dependence*
- stable density layering suppresses buoyancy driven convection in Ge-Au (LC with X-ray radiography)
  ➡ does not hold for Ge-Si (MAPHEUS experiment)

Weis, Kargl, Koza, Unruh, Meyer, J. Phys.: Cond. Matter (2019)

Diffusion of Minor Additions in Liquid Germanium

QENS, X-ray radiography, sounding rocket experiment



Darken:
$D_{AB} = (N_A D_B + N_B D_A)\,\Phi$

toward small concentration N_A :
D_{AB} *similar equal* D_A

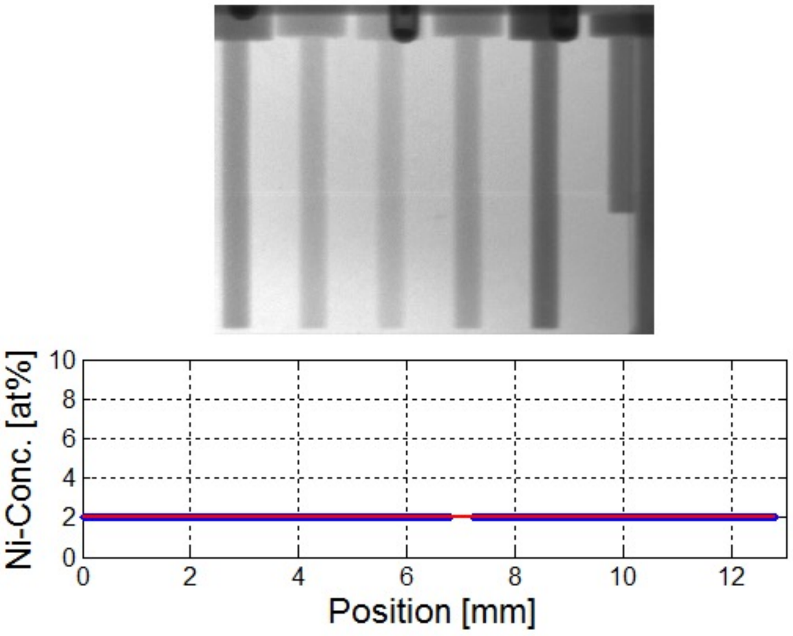
- pure liquid Germanium: fast diffusion on 10^{-8} m^2s^{-1} and small activation energy (0.16 eV)
- Ge_{98} – $Si(Au)_2$ alloys: similar diffusion coefficients for Ge self diffusion and diffusion of Si or Au
- *no dependence* of mass (difference of factor seven) and size (within error bars)

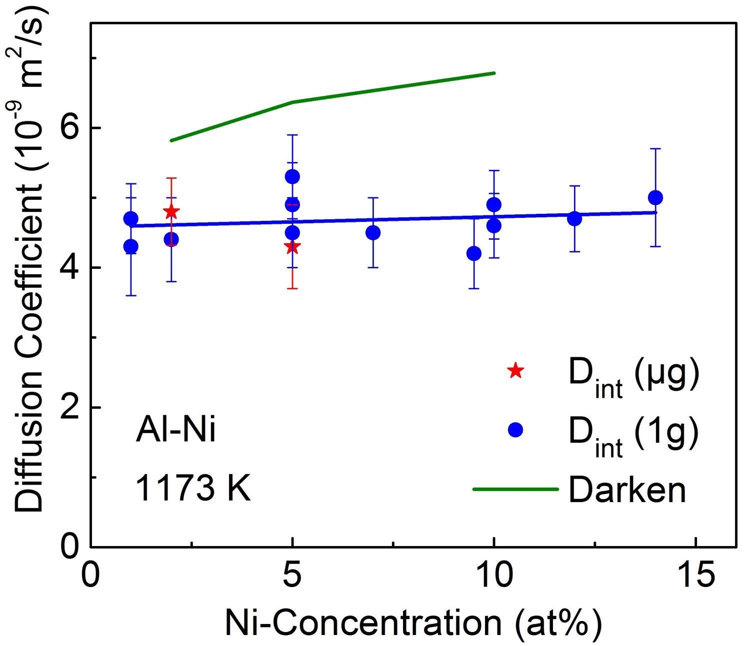
Weis, Kargl, Koza, Unruh, Meyer, J. Phys.: Cond. Matter (2019)

Accurate Diffusion Data by in-situ Techniques

Darken‘s Equation for self- and interdiffusion

compact and X-ray transparent
1500°C shear-cell furnace



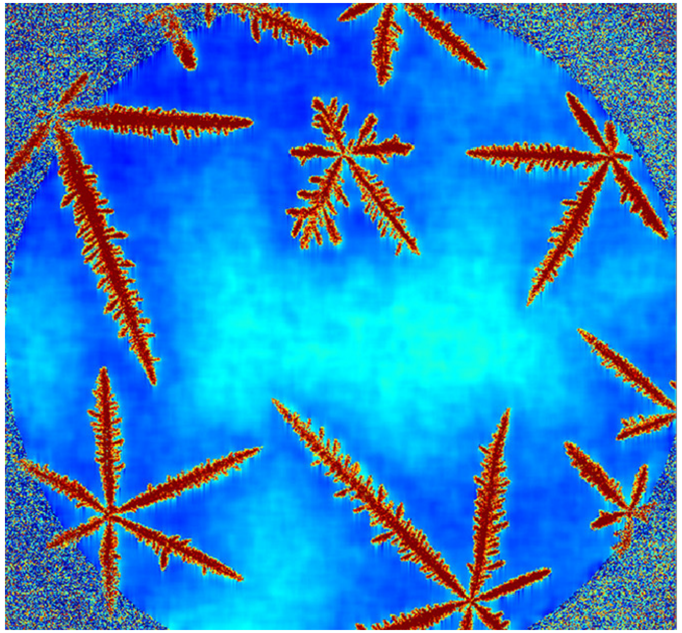


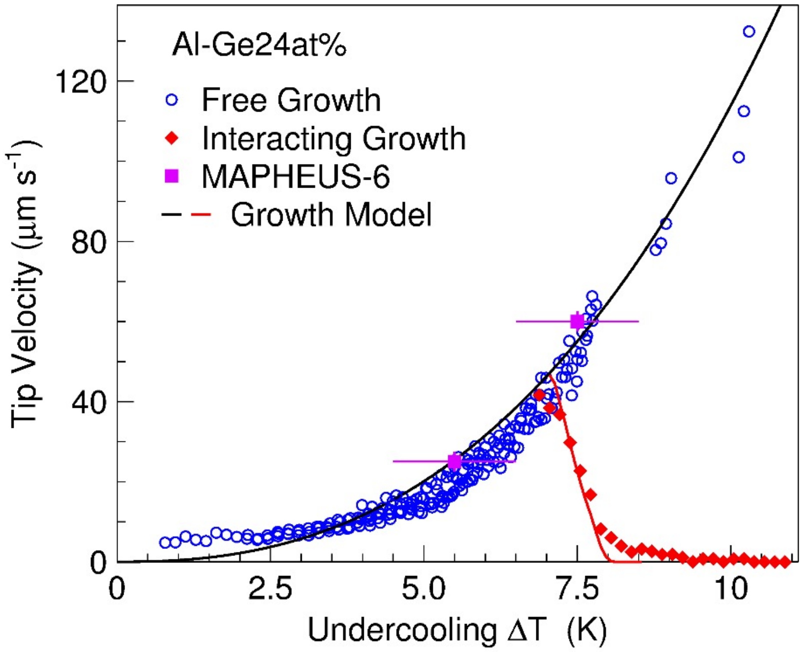
- microgravity data on MAPHEUS *confirm* ground-based experiments
- diffusion in Al-rich Al-Ni constant up to 14 at. % Ni
  → *Darken*‘s relation of self- and interdiffusion: *oversimplified* even at small concentrations

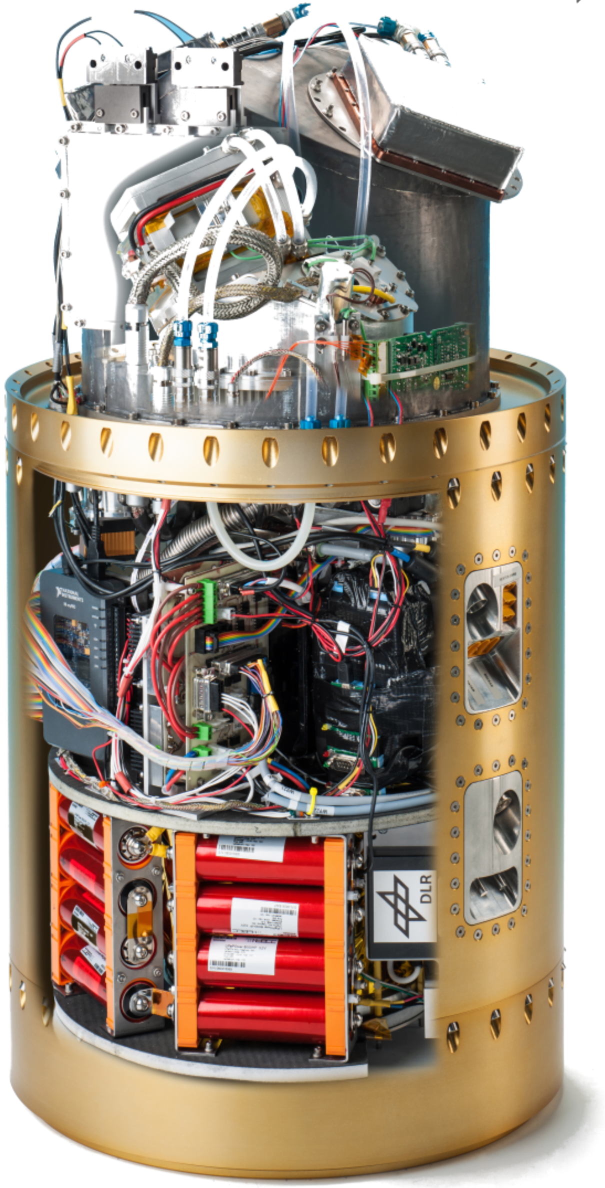
Sondermann, Neumann, Kargl, Meyer, Phys. Rev B (2016); Sondermann, Jakse, Binder, Mielke, Heuskin, Kargl, Meyer, Phys. Rev. B (2019);
Sondermann, Kargl, Meyer, Phys. Rev. Lett. (2019)

Equiaxed Dendritic Growth in Al-Ge alloys

solutal field evolution and neighbor interacted growth







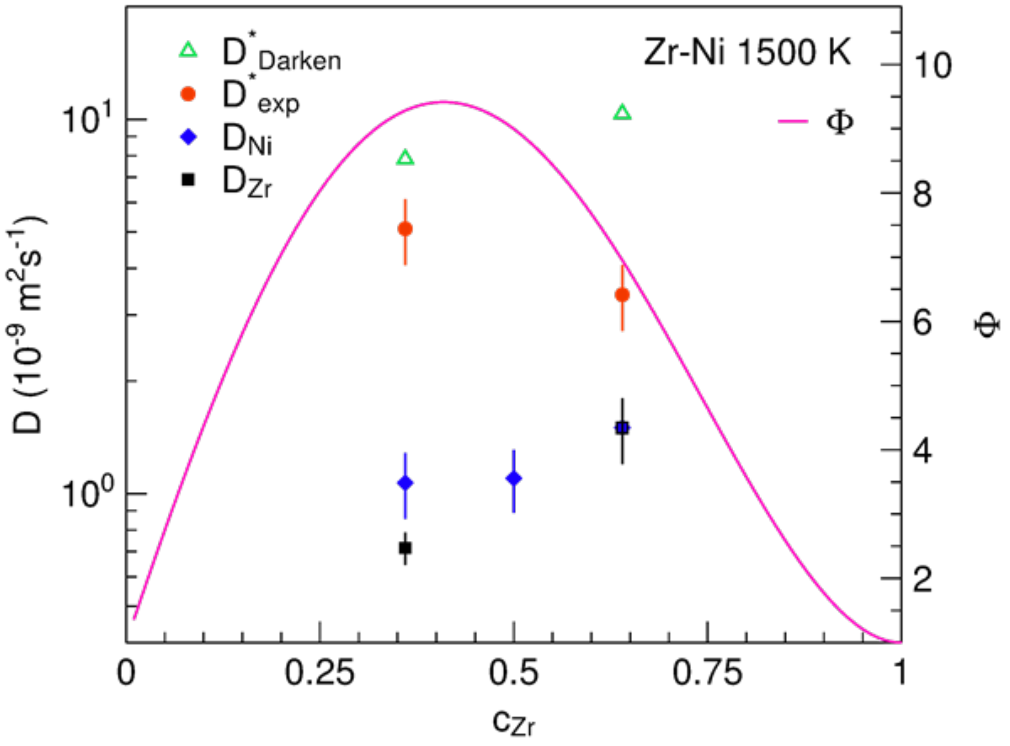
MAPHEUS X-ray module

- MAPHEUS-06: *diffusive conditions* in horizontal samples in 1g
- *quantitative determination* of solutal field evolution enabled
  test of general accepted dendrite growth models
- tip velocities for large undercoolings compare well with 3D models

Becker, Klein, Kargl, Phys. Rev. Materials (2018); Becker, Kolbe, Steinbach, Kargl, Scripta Mater. (2022)

Self- and Interdiffsuion in Zr-Ni melts

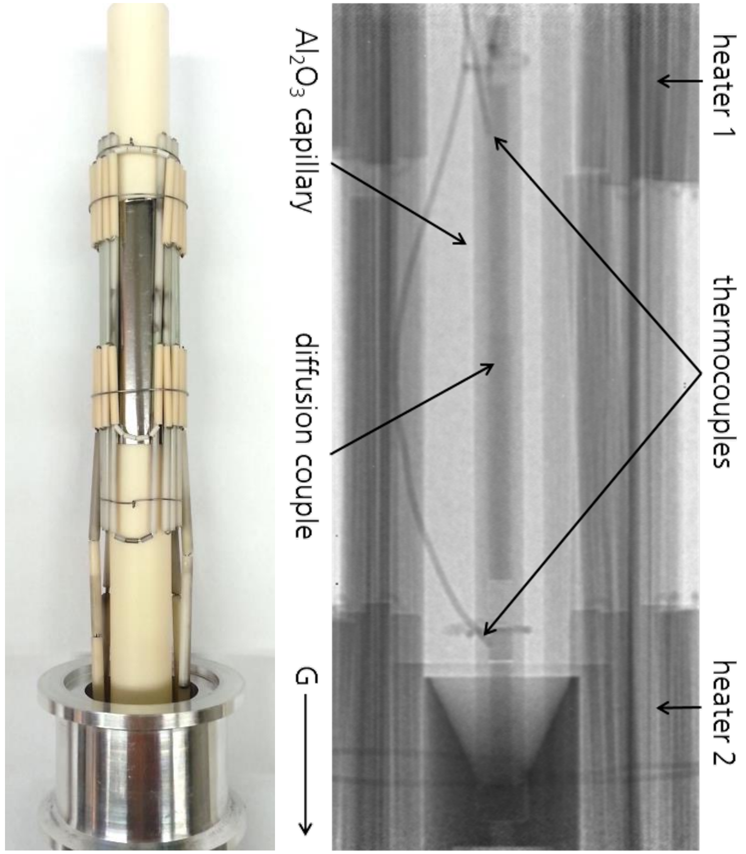
in-situ measurements with X-ray and neutron radiography, and radio tracer experiments



Darken:

$D_{AB} = (N_A D_B + N_B D_A)\ \Phi$

thermodynamic factor *Φ* (Ghosh 1994)



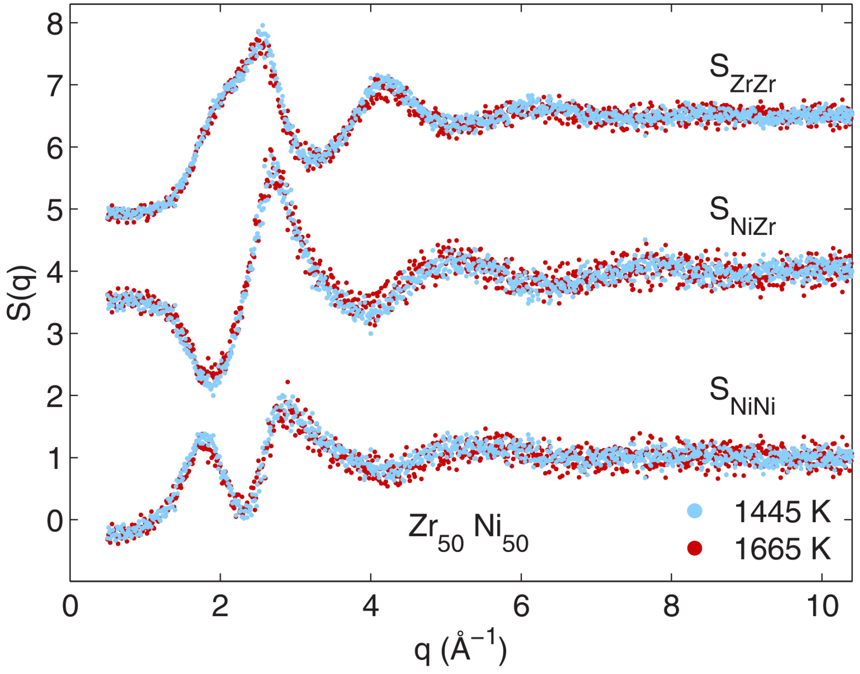
neutron radiography at ANTARES

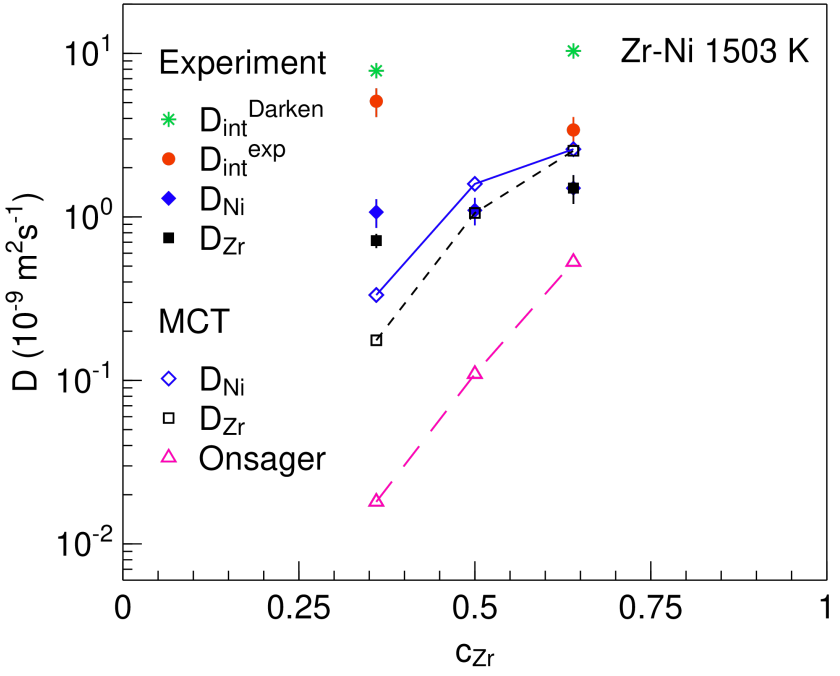
- *Darken overestimates* interdiffusion values by about factor 2 – 3
- interdiffusion slightly larger than Ni self diffusion
- ratio of self- and interdiffusion in *good agreement with MCT predictions*

Yang, Heintzmann, Kargl, Binder, Nowak, Schillinger, Voigtmann, Meyer, PRB (2018); Basuki, Yang, Gill, Rätzke, Meyer, Faupel, PRB (2017)

Predicting Diffusion Coefficients from Microscopic Structure

Mode Coupling Theory (MCT) using experimentally measured structure factors





compact, robust, plug&play ESL for neutron/X-ray scattering

- neutron diffraction (D20) + isotopes → partial structure factors = input to theory
- MCT predicts *structure–dynamics relations:* relations between diffusion and inter-diffusion coefficients
- rationalize *mixing effects* between dynamics and thermodynamics, test empirical relations (Darken)

Nowak, Holland-Moritz, Yang, Voigtmann, Kordel, Hansen, Meyer, Phys. Rev. Mater. (2017)
Yang, Heintzmann, Kargl, Binder, Nowak, Schillinger, Voigtmann, Meyer, PRB (2018)

Institute of Materials Physics in Space

Electromagnetic Levitation in Parabolic Flight

TEMPUS

Parabolic Flight Campaign, Paderborn September 2020

Parabolic Flights – Airbus 300/320 operated by NOVESPACE

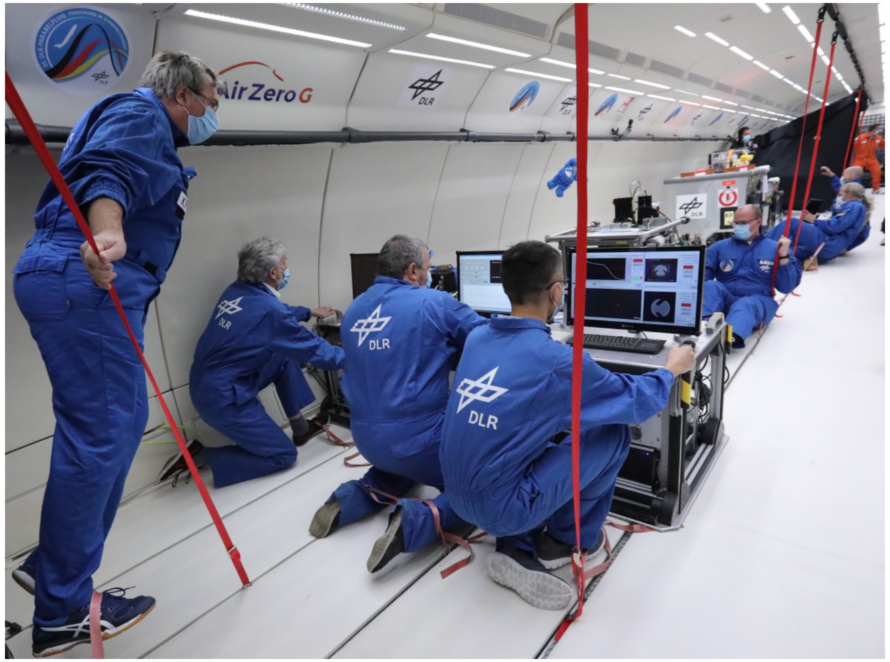
24^{th} DLR Parabolic Flight Campaign
TEMPUS, ESL, X-RISE

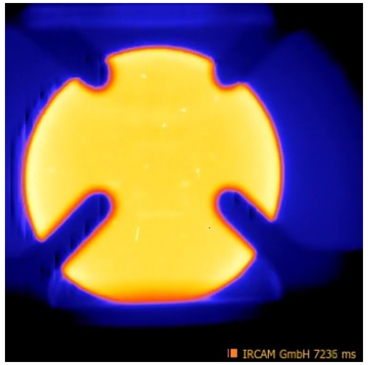
ESL positioning system tests for MAPHEUS
Neumann, Jonas, Meyer

- pull-up from 6100 m above sea-level to 8500 m
- 21 seconds of *free fall*, 31 parabolas per flight day
- 10 – 15 experiments, about 40 participants

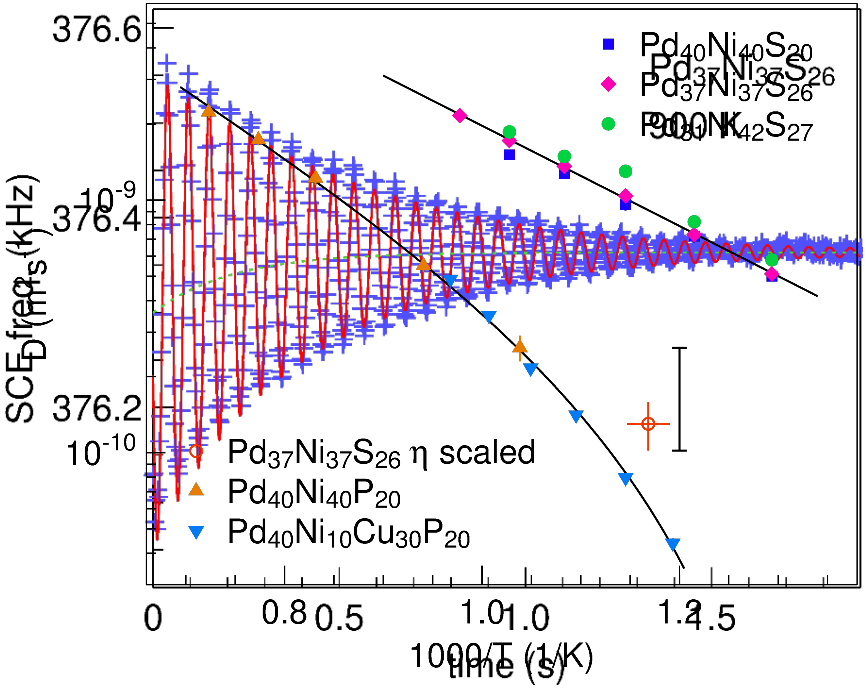
Accurate Thermophysical Property Data of Liquid Alloys

chemically reactive metallic melts, deep undercooling





freely suspended
oscillating droplet



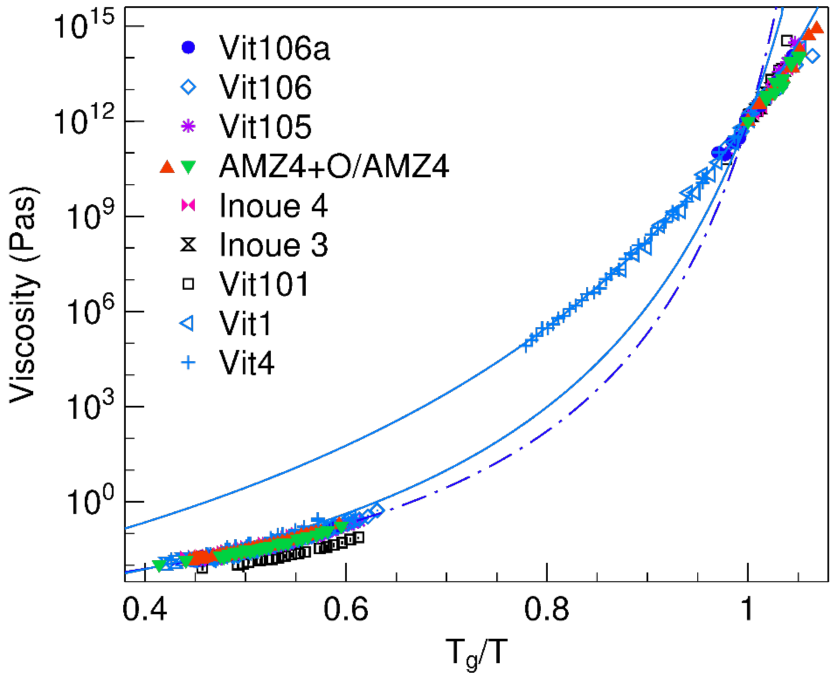
- TEMPUS with SCE (inductice sample coupling electronic): measurement of various melt properties
- oscillating droplet technique: *benchmarking* ground-based viscosity measurements
- Novell Pd-Ni-S alloys:
  comparison to diffusion data (QENS - NEAT) indicates a strong decoupling of component diffusivities

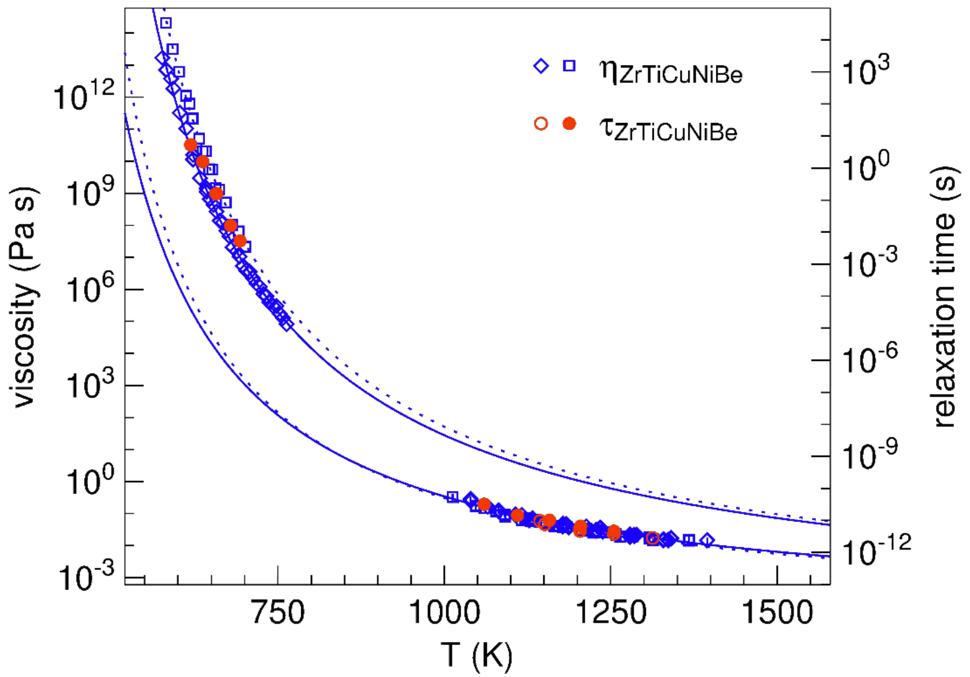
Wilden, Yang, Bochtler, Busch, Günther, Russina, Meyer, J. Phys. Cond. Mat. (2021)

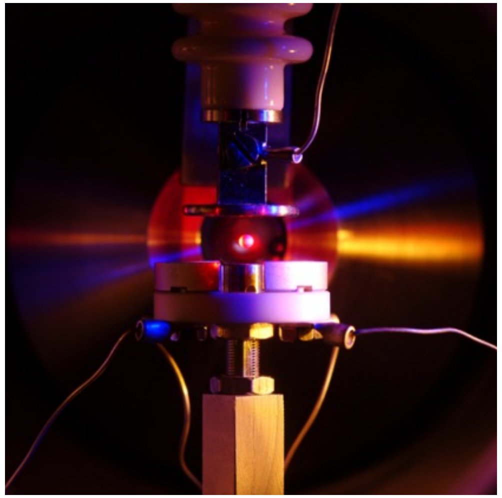
HELMHOLTZ RESEARCH FOR GRAND CHALLENGES DLR

Structure – Property Relations in Glass-Forming Metals

origin of dynamics mismatch in Zr-based alloys







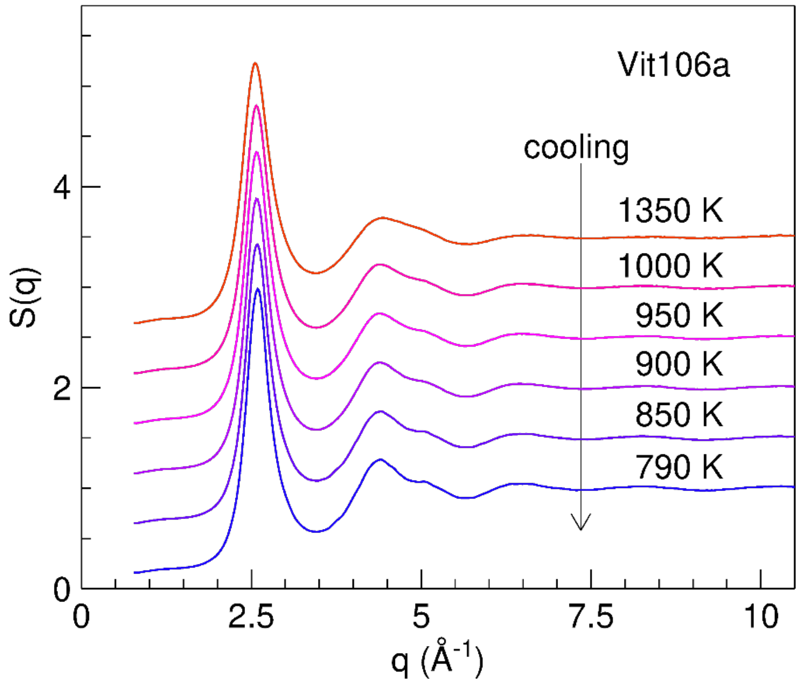
ESL: oscillating droplet technique

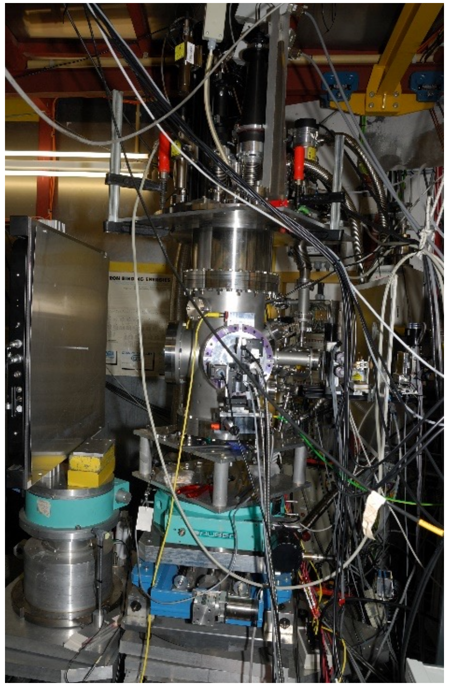
- measurements of viscosity from the liquid to the glass transition with various techniques; relaxation times with XPCS (P10 PETRA-III) and QENS (FRM-II)
- in undercooled melts: *mismatch* found in Zr-based glass-forming alloys: ~1 order of magnitude
- *structural origin* and mechanism? liquid-liquid transition?

Jonas, Hembree, Yang, Busch, Meyer, (submitted); Amini, Yang, Pineda, Ruta, Sprung, Meyer, Phys. Rev. Mater. (2021)

Structure – Property Relations in Glass-Forming Metals

origin of dynamics mismatch in Zr-based alloys





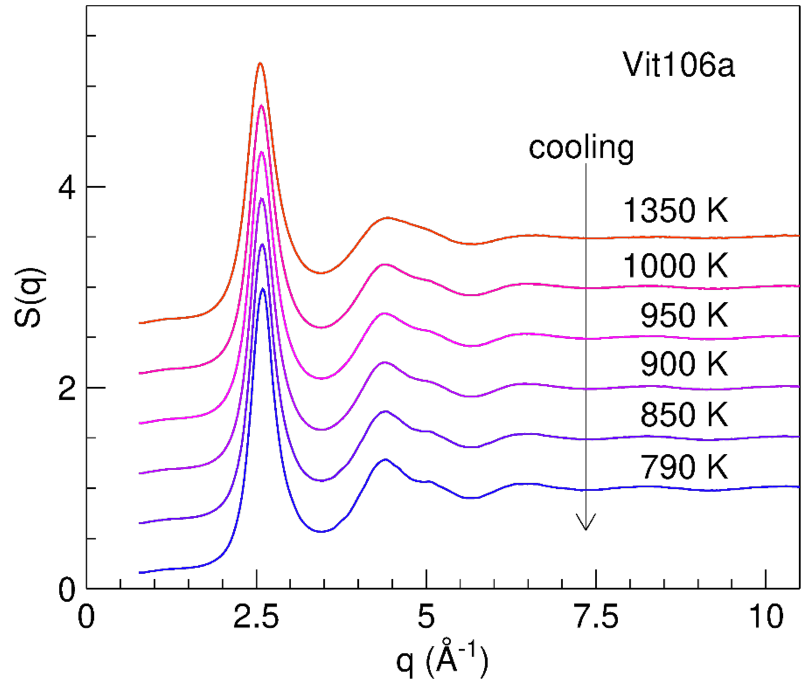
- electrostatic levitation *enables* measurements from the liquid to the glass on Zr-based BMG's
- structural changes in the undercooled melts *revealed* by X-ray diffraction at PETRA III
- consistent, corresponding feature in the heat capacity

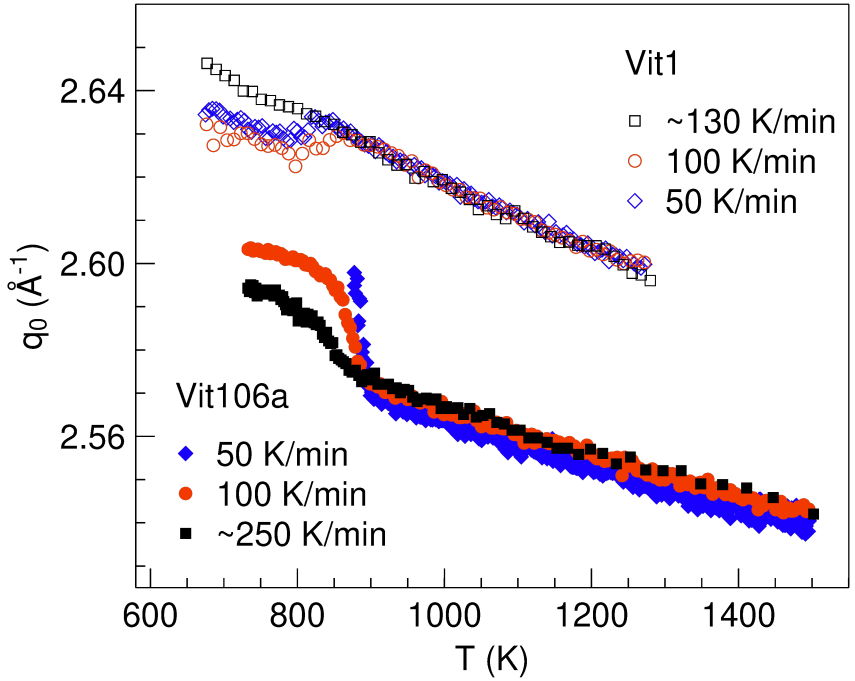
Jonas, Yang, Meyer, Phys. Rev. Lett. (2019); Wei, Yang, Bednarcik, Kaban, Shuleshova, Meyer, Busch, Nature Commun. (2013)

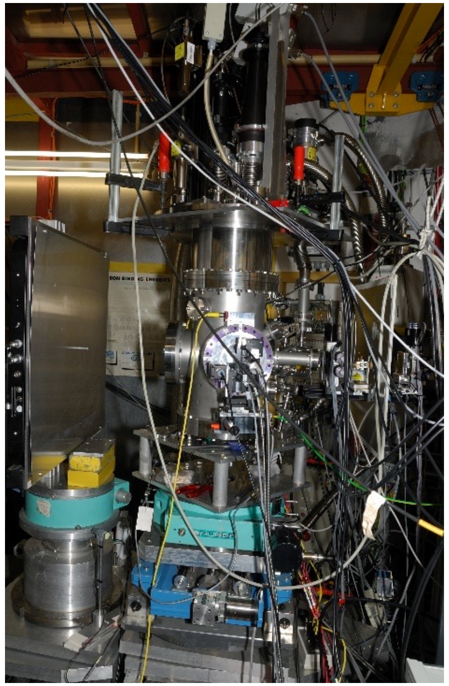
HELMHOLTZ
RESEARCH FOR GRAND CHALLENGES
DLR

Structure – Property Relations in Glass-Forming Metals

origin of dynamics mismatch in Zr-based alloys



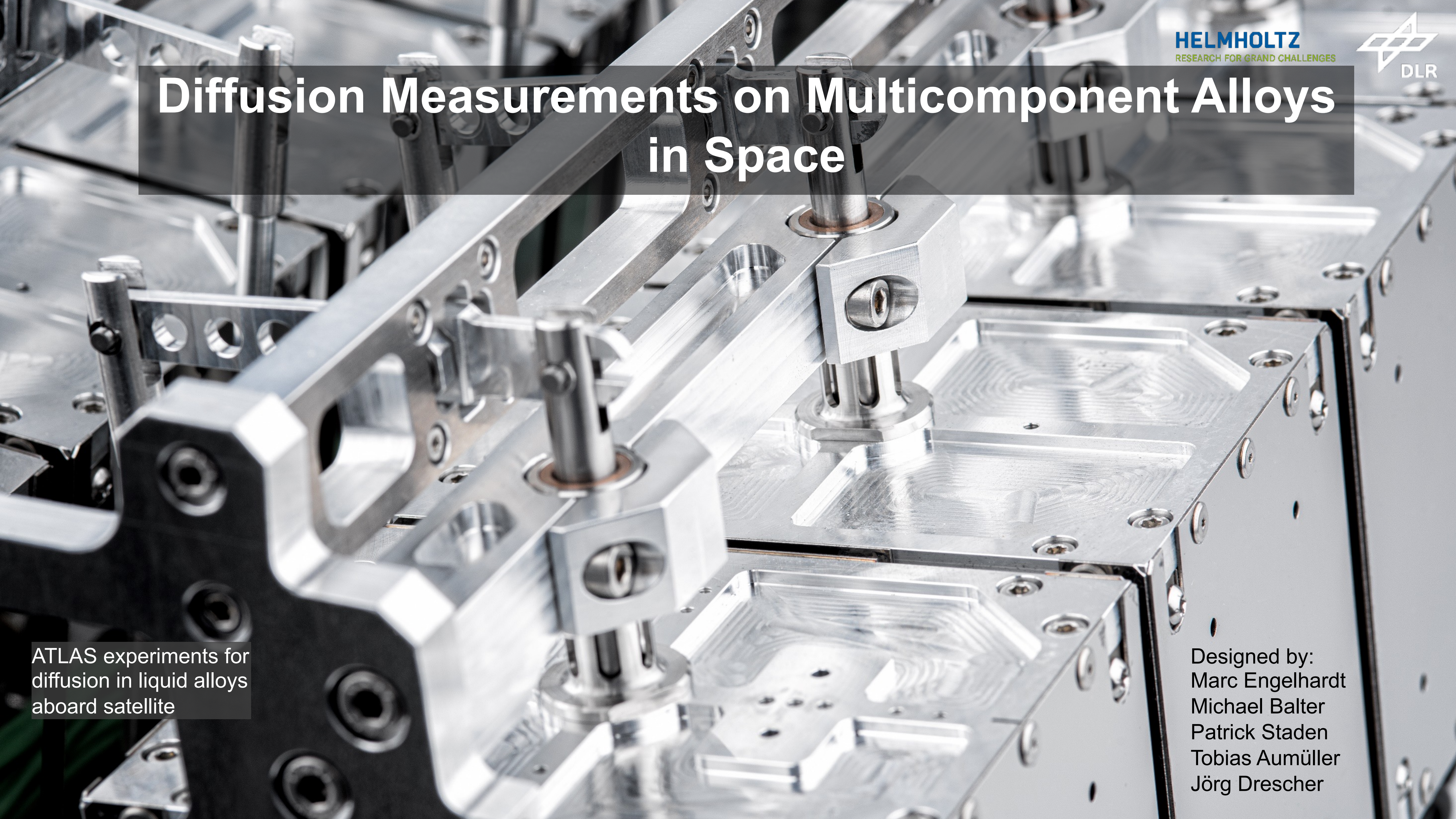




- controlled heating/cooling during synchrotron ESL levitation experiment
- no heating rate dependent transition temperature, transition time ~30s; *diffusion* length of 10-100 nm
- *requires* long range mass transport and significant structural changes;
  recently confirmed by SAXS at P21.2 PETRA -III

Jonas, Yang, Meyer, Phys. Rev. Lett. (2019); Wei, Yang, Bednarcik, Kaban, Shuleshova, Meyer, Busch, Nature Commun. (2013)

Diffusion Measurements on Multicomponent Alloys in Space



ATLAS experiments for diffusion in liquid alloys aboard satellite

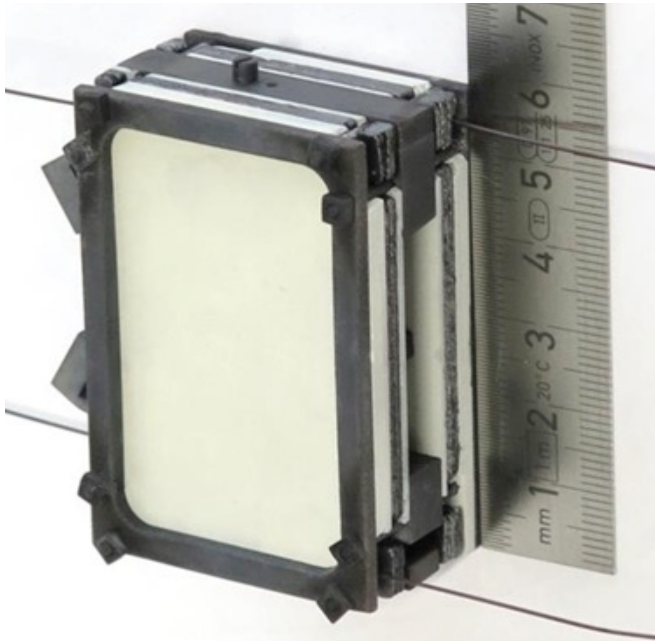
Designed by:
Marc Engelhardt
Michael Balter
Patrick Staden
Tobias Aumüller
Jörg Drescher

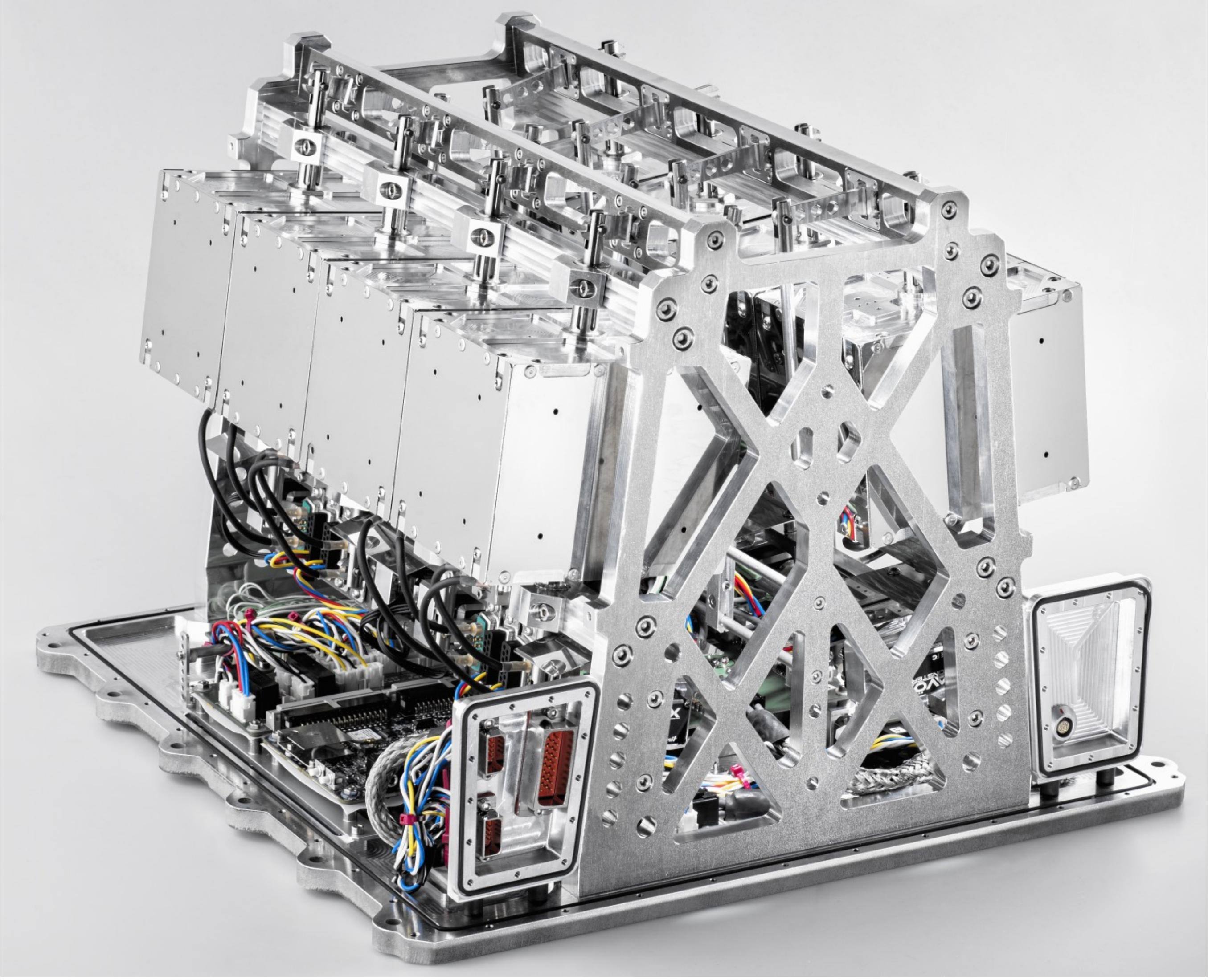
ATLAS – Atomic Transport in Liquid Alloys in Space

on orbital platform

multi-slice shear-cell furnace:

- long term annealing up to 1000°C
- thermodiffusion, inter- and, self-diffusion measurements
- multicomponent materials





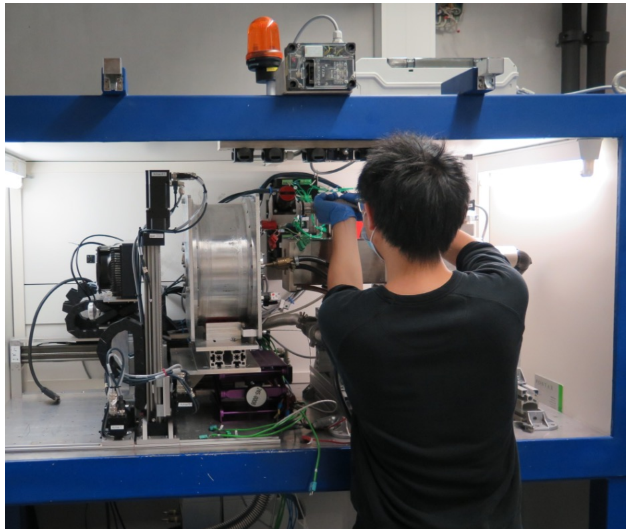
Conclusions

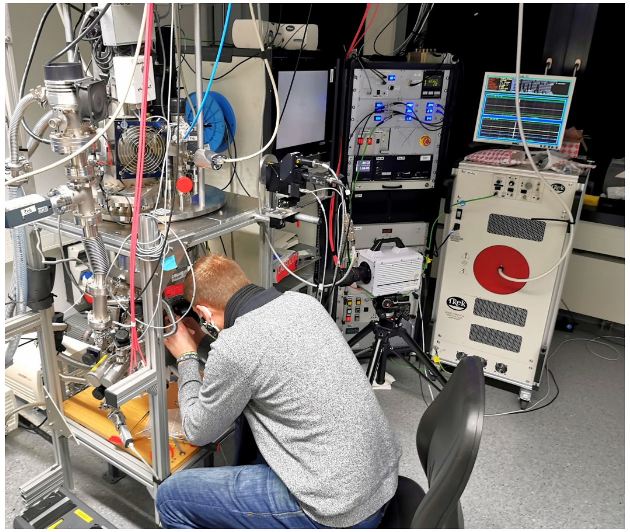
accurate measurements of liquid properties:

- self diffusion by quasielastic neutron scattering
- interdiffusion by X-ray and neutron radiography
- viscosity by levitation techniques
- *benchmark experiments in microgravity*
- (partial-) structure factors – levitated droplets

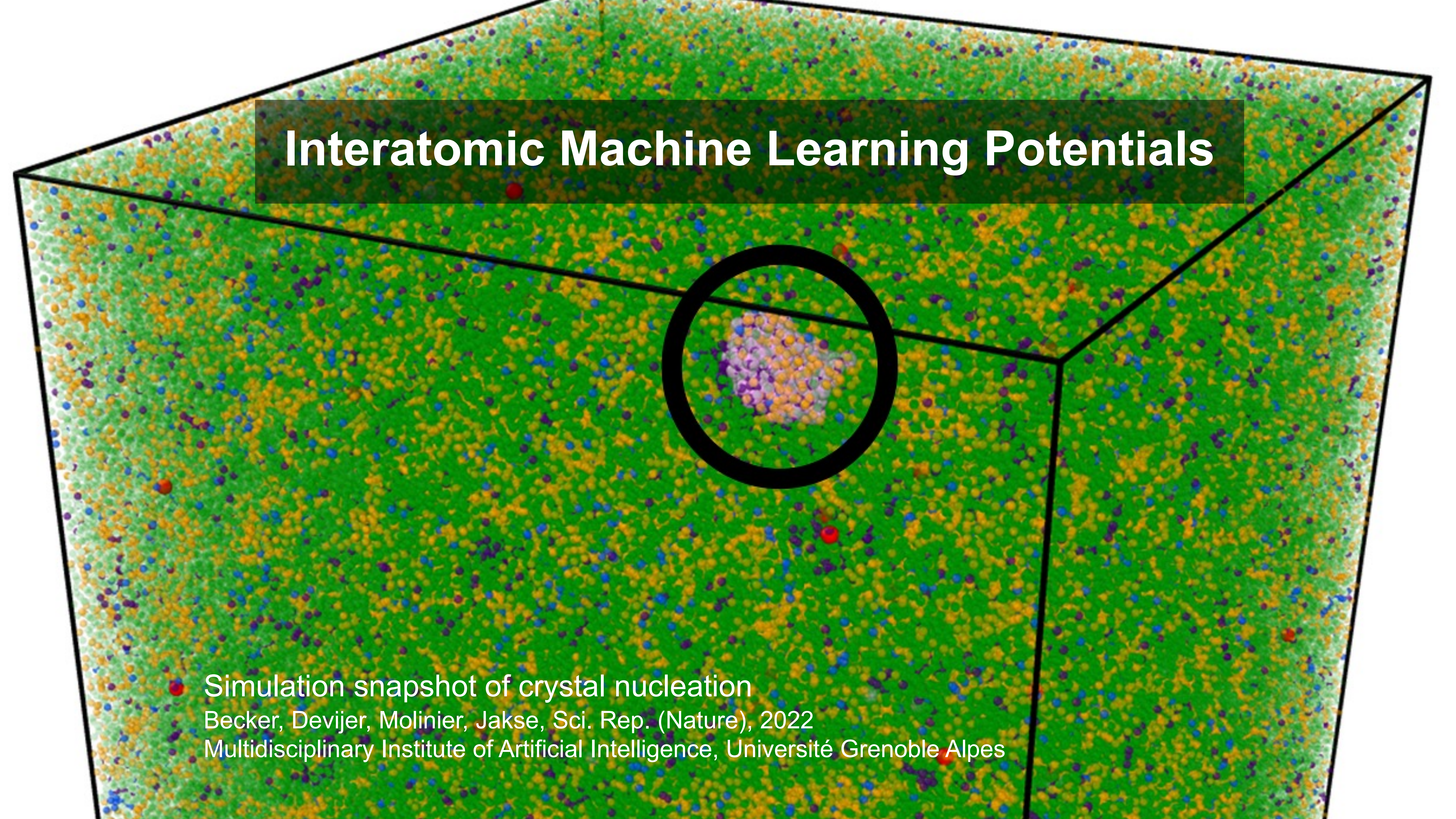
diffusion in liquid metals and alloys

- Stokes-Einstein *not valid* for dense systems
- Darken relation is *oversimplified*: cross terms significant
- *kinetics governs; highly collective* transport mechanism
- accurate coefficients of mass transport are key to an understanding of solidification phenomena





Interatomic Machine Learning Potentials



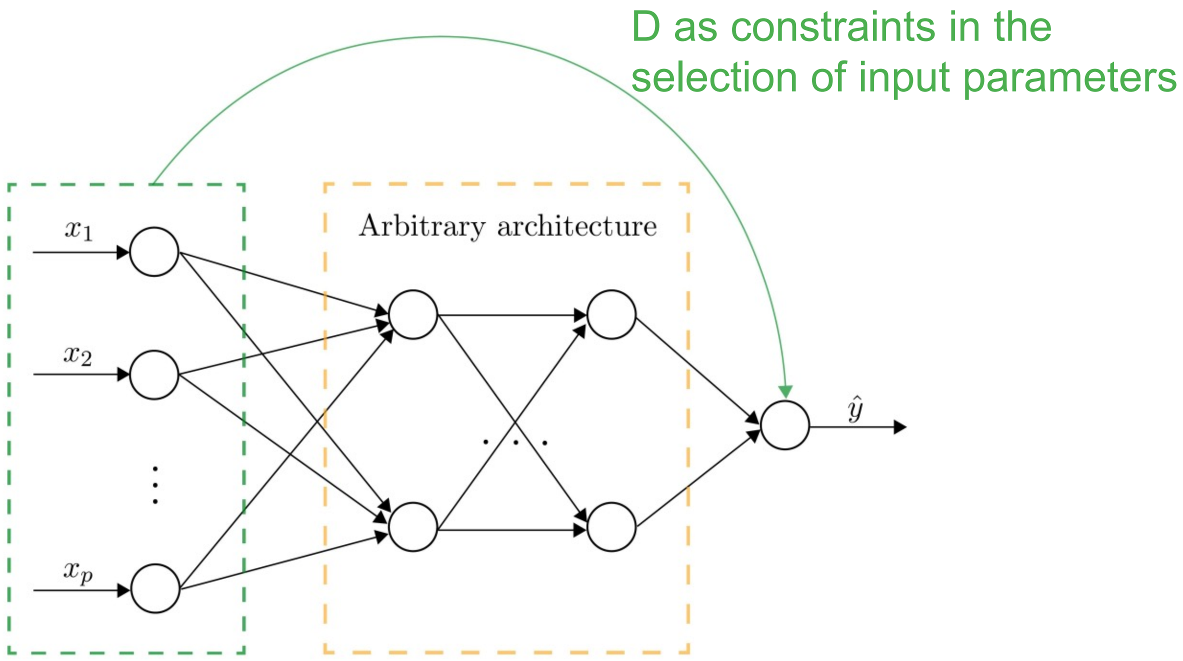
Simulation snapshot of crystal nucleation

Becker, Devijer, Molinier, Jakse, Sci. Rep. (Nature), 2022

Multidisciplinary Institute of Artificial Intelligence, Université Grenoble Alpes

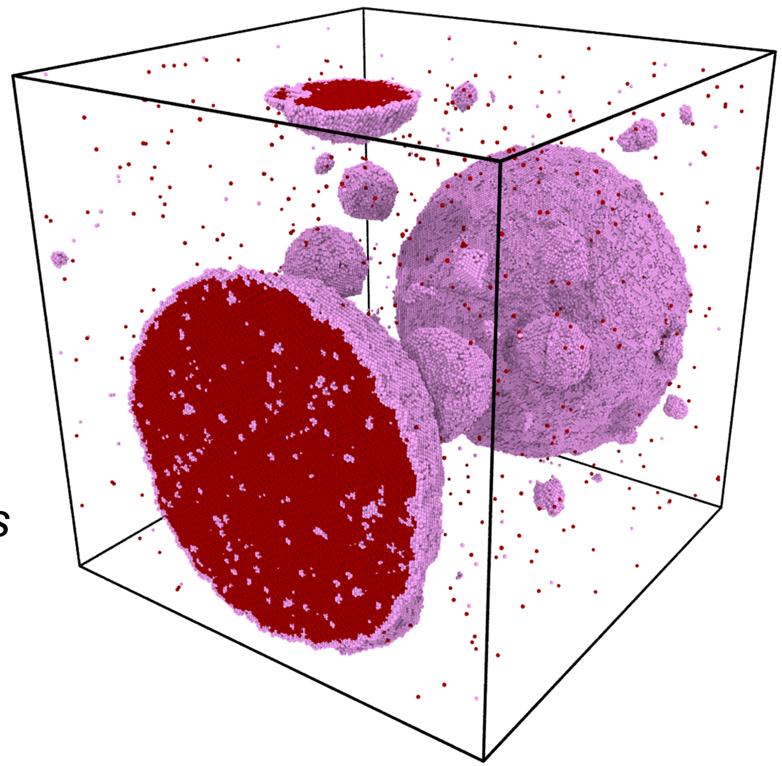
Interatomic Machine Learning Potentials for Metallic Melts

introduce diffusion coefficients D in the training procedure



Ab initio database:
x_i : *input v*ariables describing the local atomic environment
ŷ : output energies
Architecture: High Dimensional Neural Network to train with optimal number of input variables

LassoNet : Lemhadri et al., J. Machine Learning Res. 22, 1 (2021).
Adaptative Lasso : Dinh et al., (NeurIPS 2020)



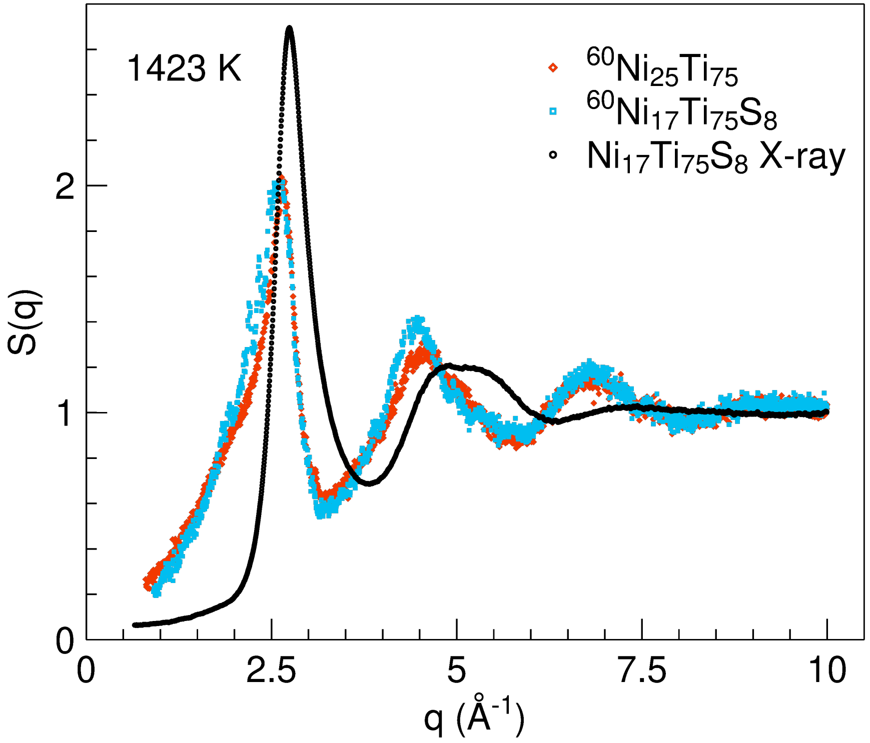
- large-scale simulations up to 10^7 atoms at large time-scales (10^{-6} s): solidification phenomena reachable
- objective: training procedure gauged by *experimental diffusion coefficients*
  ➡ *reliable potentials* for MD simulation to model *liquid and solidification in realistic systems*

Jakse, Sandberg, Saliou, Jarry, Devijver, Voigtmann, Horbach, Meyer, submitted (2022)

HELMHOLTZ RESEARCH FOR GRAND CHALLENGES DLR

Ground-based Research with X-rays and Neutrons

partial structure factors and phase formation



- X-ray / neutron diffraction; Ni isotopes
- melt structure and solidification behavior of novel glass-forming alloys *containing Sulfur*

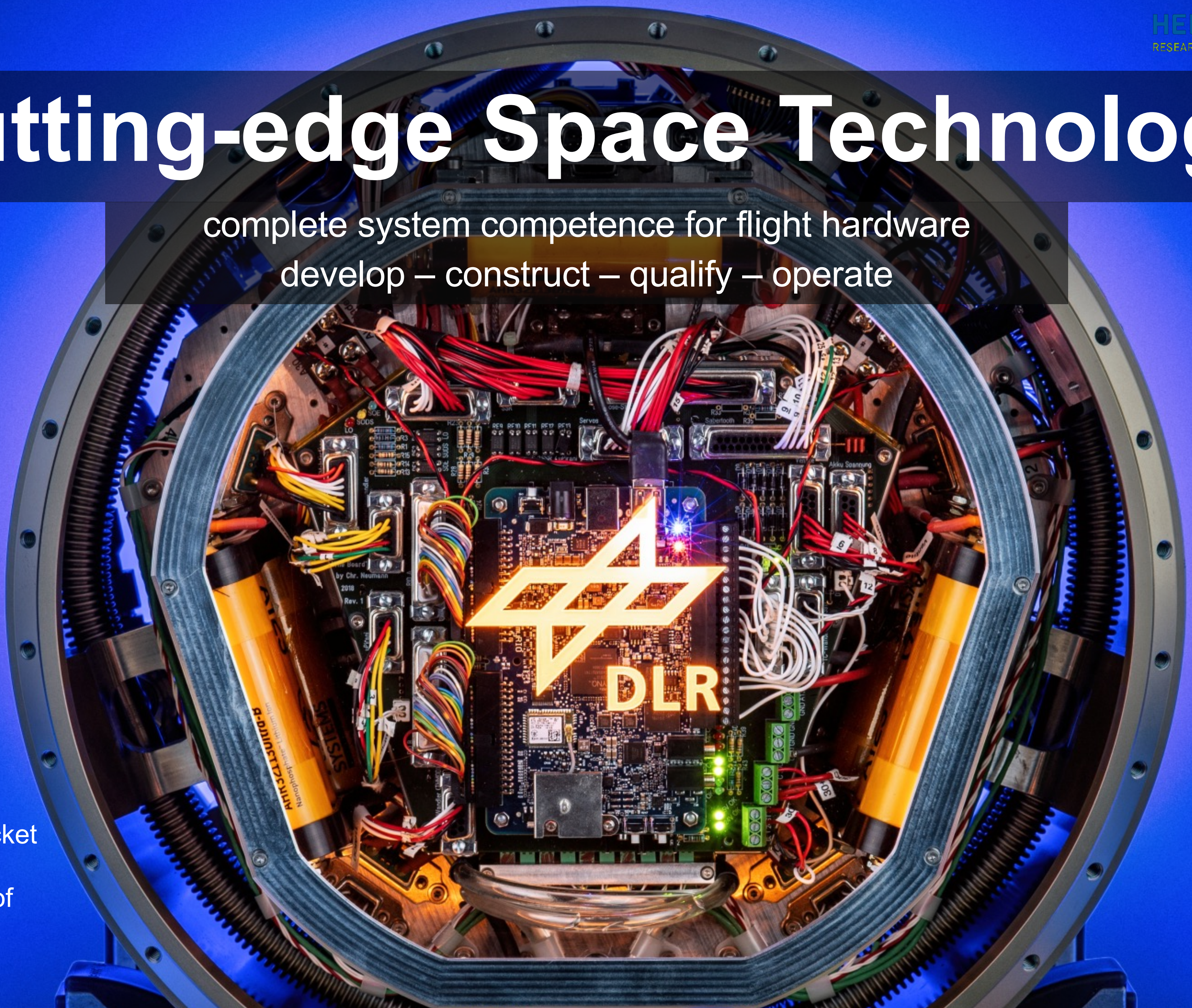
Spin-off from MAPHEUS:
Compact plug+play levitator (Synchrotron/Neutron Sources)

Wilden, Yang, Kolbe, Holland-Moitz, Kuball, Bochtler, Buslaps, Busch, Meyer (submitted); Wilden, Yang, Holland-Moritz, Hansen, Meyer et al. (in preparation)

Cutting-edge Space Technology

complete system competence for flight hardware

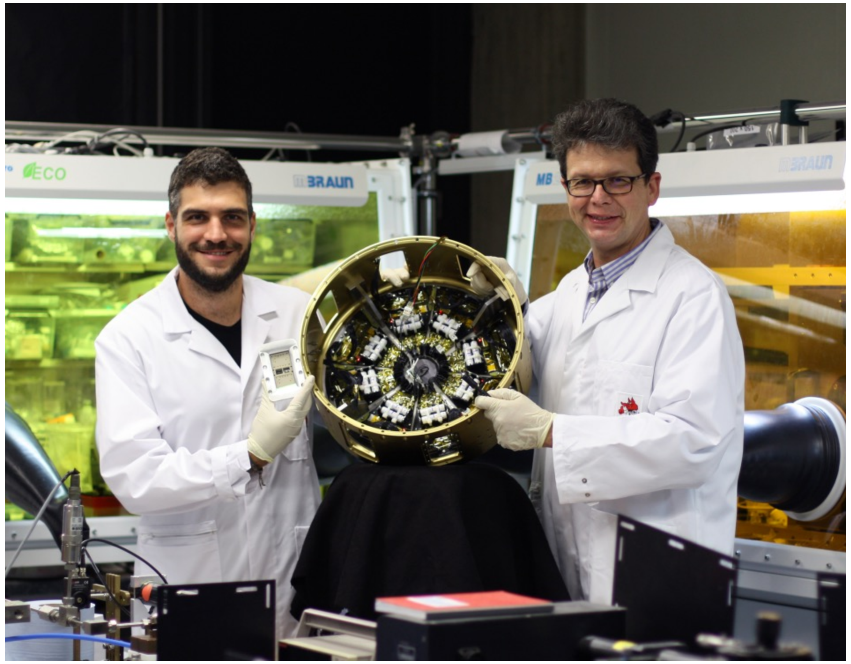
develop – construct – qualify – operate



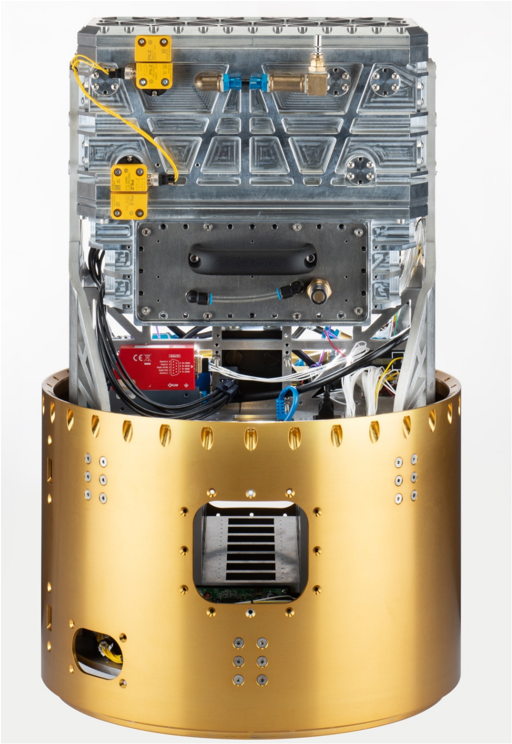
MAPHEUS sounding rocket
ARTEC experiment for
directional solidification of
Aluminum cast alloys

Materials Physics in Space

- *integrated ground-based program* in combination with *key experiments under space conditions*
- *continuous development* of novel measurement techniques and advanced processing technologies
- *unprecedented experimental capabilities* in the upcoming years



joint organic solar cell experiment with TUM



3d printing in space
and in-situ with X-rays

experiment qualification at CAST

Research with Neutrons

- *key experiments with neutrons* in combination with an *integrated in-house program*
- *continuous development* of novel technologies and advanced sample environments
- *unprecedented experimental capabilities* in the upcoming years

ILL7 guide hall - Chartreuse

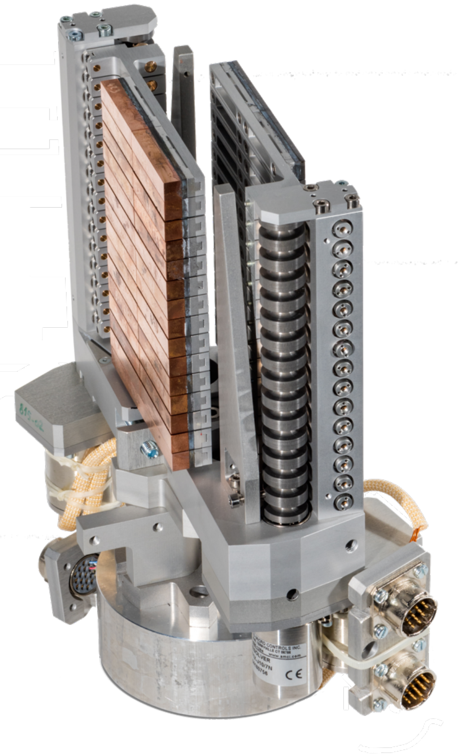
ILL7 guide hall - Vercors

ILL22 guide hall - NEXT

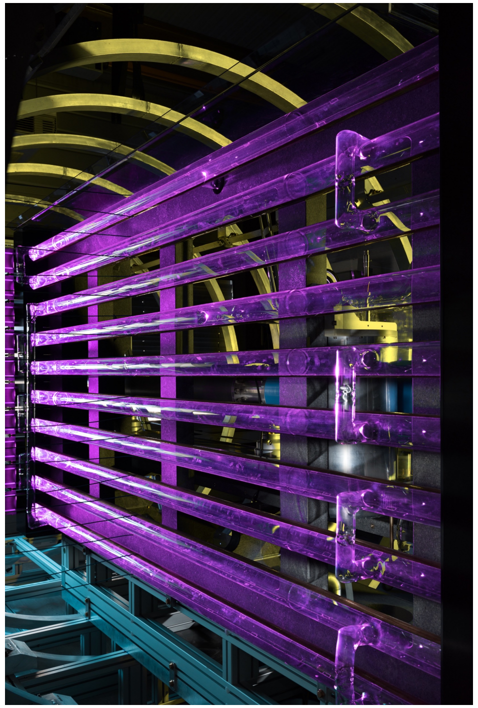
ILL5 H1H2

Research with Neutrons

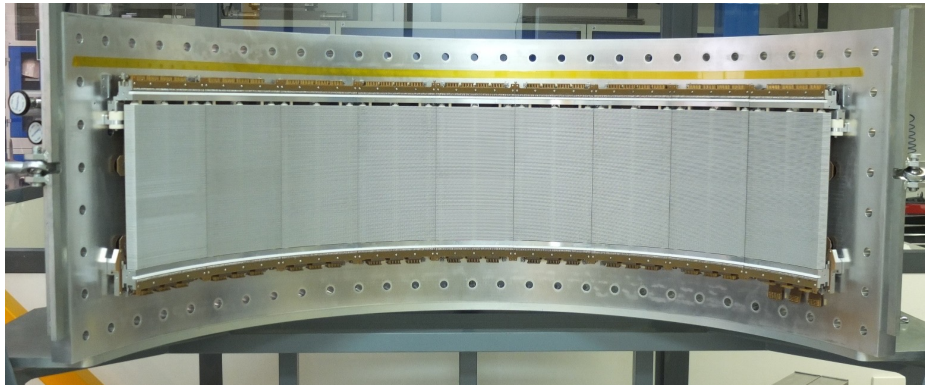
- *key experiments with neutrons* in combination with an *integrated in-house program*
- *continuous development* of novel technologies and advanced sample environments
- *unprecedented experimental capabilities* in the upcoming years



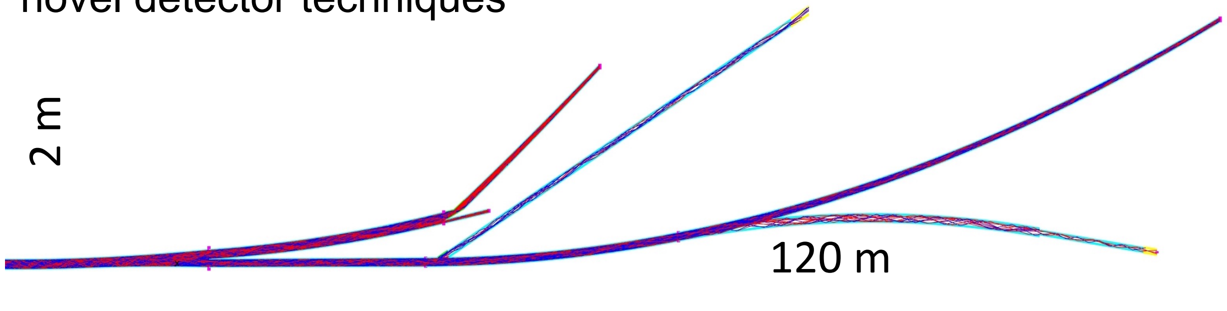
new monochromators



enhanced polarization capabilities



novel detector techniques



advanced guides

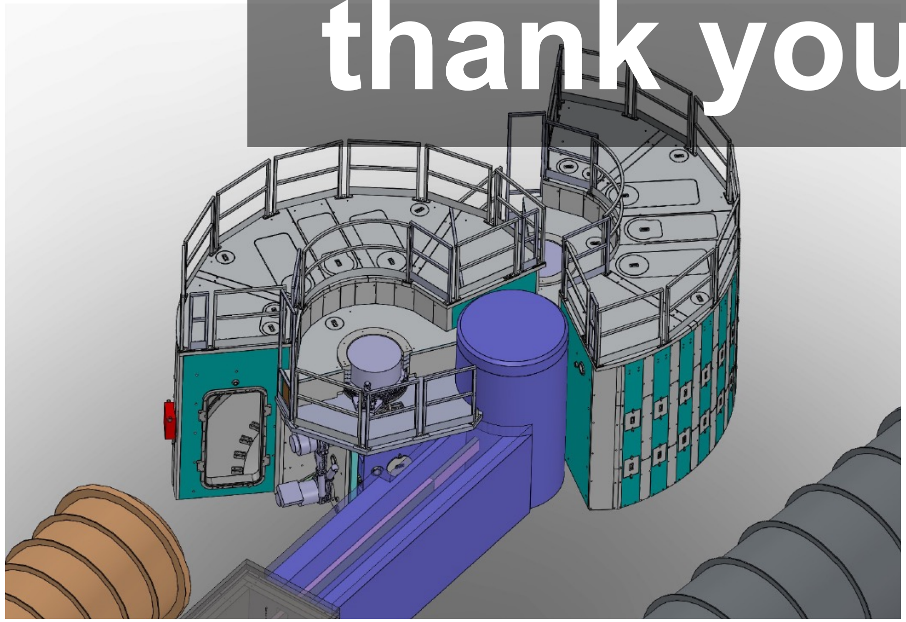
NEUTRONS
FOR SOCIETY

Research with Neutrons

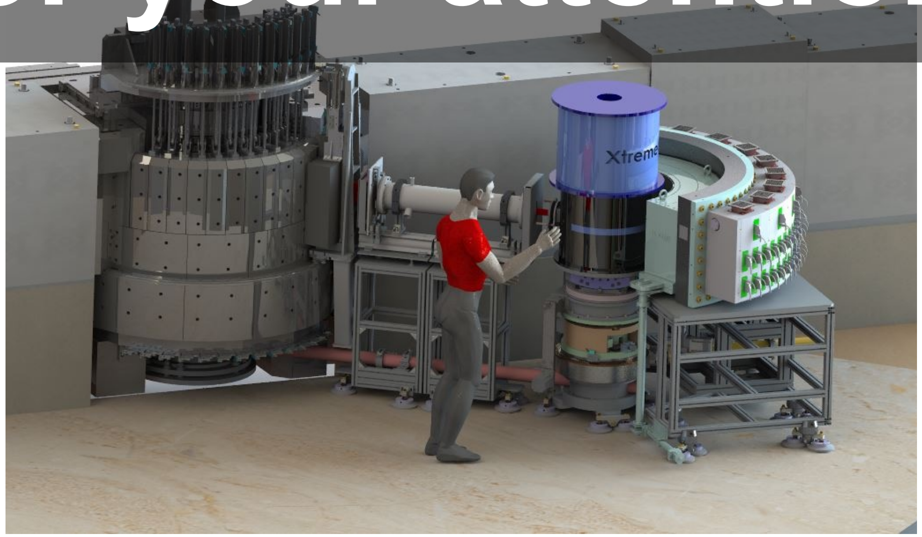
- *key experiments with neutrons* in combination with an *integrated in-house program*
- *continuous development* of novel measurement techniques and advanced sample environments
- *unprecedented experimental capabilities* in the upcoming years

Numerous new / upgraded instruments

thank you for your attention



SHARP+ ToF spectrometer



XtremeD difractometer