2-1. IROAST Distinguished Professors

No.	Name	Project Title
2-1-1	**Dmitri Aleks MOLODOV** RWTH Aachen University, Germany	Mechanics and dynamics of grain boundaries in Al and Mg bicrystals
2-1-2	**László PUSZTAI** HUN-REN Wigner Research Centre for Physics, Hungary	Nanoscale assemblies in hydrogen-bonded liquids and in amorphous materials
2-1-3	**Yufeng ZHENG** Peking University, China	Development of Zn based biodegradable metals

No. 2-1-1	Mechanics and dynamics of grain boundaries in Al and Mg bicrystals			
Name	Dmitri A. MOLODOV	Title	Professor	
Affiliation (home)	Institute of Physical Metallurgy and Materials Physics (IMM), RWTH Aachen University, Germany Email: molodov@imm.rwth-aachen.de			
Research Field	Advanced materials			
Period of appointment	April 1, 2024- March 31, 2025			
Host Professor	Sadahiro TSUREKAWA			
Affiliation	Faculty of Advanced Science and Technology Email: turekawa@kumamoto-u.ac.jp	Title	Professor	

Details of Activities

1. During the last year, our research focused on the investigation of grain boundary dynamics and local mechanical behavior in the close vicinity of grain boundaries in metallic bicrystals. The experiments using the nanoindentation technique were performed on Al and Fe-3%Si bicrystals with different grain boundaries. Further experimental and computational studies were carried out to establish and analyze the relationship between the inclination dependence of the grain boundary energy and the curvature driven grain boundary migration. Also, the mechanisms of shear-coupled grain boundary migration were addressed in the simulation study of the motion of different twin boundaries in fcc (Cu) and hcp (Mg) structures. The research work outlined above is intended to be continued and further developed in the next year.

2. In the reporting period, the nanoindentation studies on the mechanical properties of the specific grain boundaries in aluminum were continued and previously planned additional experiments on Al bicrystals were carried out. The measurements were performed on bicrystals with the 70.5°⟨110⟩ tilt boundary having the geometry of the coherent Σ3 twin boundary. The experiments were accompanied by atomistic simulations performed in the group of Prof. L. Barrales-Mora of the George W. Woodruff School of Mechanical Engineering Georgia Tech Lorraine in Metz, France. The draft of the paper in which the results obtained for all Σ3 grain boundaries in Al bicrystals investigated in this project are reported and analyzed is currently being worked on.

The nanoindentation technique was also applied to investigate the local mechanical properties of grain boundaries in bicrystals of Fe-3% Si, focusing on the propagation of plastic deformation across the boundary. With this aim ⟨110⟩ twist boundaries with different misorientations were examined and compared with symmetric tilt grain boundaries having the same misorientation angles. The bicrystals of Fe-3%Si with Σ3, Σ9, Σ27a and Σ33c ⟨110⟩ twist boundaries, as well as {111}Σ3/⟨110⟩ and {221}Σ9/⟨110⟩ symmetric tilt boundaries were prepared using the floating zone method and characterised by EBSD technique. To investigate the deformation mechanisms, the nanoindentation tests were conducted on and near the grain boundaries. The results obtained allowed to deepen our understanding of the role of different grain boundaries in the plastic deformation of the crystalline solid. In particular, these results indicate that the propagation of plastic deformation across a grain boundary in Fe-3%Si is primarily governed by dislocation transfer rather than dislocation generation at the boundary.

The results of this study are planned to be introduced in a presentation at the International Conference on Processing & Manufacturing of Advanced Materials (THERMEC'2025) in Tours, France, from June 30 to July 4, 2025:
Yoshitake Ichimura, Dmitri A. Molodov, Seiichiro Ii, Sadahiro Tsurekawa: Plastic deformation propagation across grain boundaries in Fe-3%Si bicrystals: A comparative study of twist and tilt grain boundaries.

In the other long-term project, which was completed in the reference year, the migration behaviour of low angle ⟨100⟩ tilt and mixed tilt-twist grain boundaries as well as of different grain boundaries with misorientations close to the Σ3 CSL orientation relationship was investigated in high purity Al bicrystals. An in-situ technique was used for observing and measuring grain boundary migration and shape. The experiments were accompanied by molecular statics simulations to compute the structure and energy of the boundaries having a similar geometry to those examined in the experiments. 2D vertex modelling as well as molecular dynamics simulations were applied to evaluate the effect of inclinational energy anisotropy on grain shape evolution and grain growth kinetics. In contrast to pure tilt boundaries, which remain straight in their original position or form immobile facets during annealing and therefore cannot move, the mixed low angle boundaries were found to readily assume a continuously curved shape and move steadily under the capillary force. Furthermore, the capability of nearly Σ3 60°⟨111⟩ incoherent {110} and {112} twin boundaries to migrate during annealing was found to depend crucially on their initial inclination. The results of these experimental and simulation studies performed to determine the influence of the inclination anisotropy of grain boundary energy on the migration of grain boundaries under capillary force are reviewed and discussed in the paper submitted for publication in Journal of Materials Science:
Dmitri A. Molodov, Jann-Erik Brandenburg, Luis A. Barrales-Mora, Sadahiro Tsurekawa: On the effect of inclination dependence of grain boundary energy on grain boundary migration in Al bicrystals, J. of Materials Science, 2024, submitted

In the further part of our research in the reporting period, the focus was put on the role of sulfur for grain growth in Ni, which is suspected to be the cause of the observed abnormal grain growth in Ni, but so far remains poorly understood. To clarify this issue, in this work the temperature dependence of the grain size change in pure Ni and S-doped Ni with 50 ppm sulfur was measured. In contrast to pure Ni, S-doped Ni showed anomalous grain size variations with temperature, characterized by a significant growth in the range between 823K and 973K, a decrease of grain size above 973K and a renewed growth above 1173K. The observed anomaly is assumed to be related to the phenomenon of grain boundary wetting and the formation of a liquid (or quasi-liquid) film of S-rich phase at the grain boundaries. For a better understanding and proof of this interpretation, the temperature dependence of grain growth in the alloy of the Al-Sn system was also measured and analyzed, where the wetting transition at different grain boundaries was previously experimentally determined. These results and their interpretation are presented in the article submitted recently for publication in Journal of Materials Science:
Sadahiro Tsurekawa, Misaki Teraura, Takateru Yamamuro, Dmitri A. Molodov: Anomalous grain growth in S-doped Ni and comparative insights from the Al-Sn alloy exhibiting grain boundary wetting transition, J. of Materials Science, 2025, submitted.

In cooperation with the group of Dr. A. Ostapovets from the Institute of Materials Physics in Brno, Czech Republic, we have continued the computational and theoretical investigation of grain boundary migration - shear coupling, one of the possible mechanisms of metal plasticity. There are two basic approaches in the literature to describe the shear-coupled migration of grain boundaries and to predict the coupling factor. One is based on solving the Frank-Bilby equation and considering the dislocation content of the grain boundary, the other on considering the disconnection glide along the grain boundary. Although opinions exist that these approaches are incompatible, we demonstrate the internal correlation between them by considering the shear-coupled migration of the Σ3(111) boundary in fcc Cu. The results of our analysis show that there are no contradictions between the approaches and their

combination can help to thoroughly understand the physical mechanisms of shear-coupled boundary migration. The results of this work are presented in the paper submitted for publication in Journal of Materials Science:
Ritu Verma, Dmitri A. Molodov, Andriy Ostapovets: On the mechanism of shear coupled migration in FCC crystals, J. of Materials Science, 2024, submitted.

During the stay in Kumamoto in May 2024, Prof. Molodov has delivered two lectures on "Grain Boundary Migration" (“Fundamentals and measurement methods” and “Key dependencies and relationships”) for graduate students as part of the regular course of Professor Tsurekawa "Materials Interface Science".


Furthermore, the results of our research were presented in two talks at the International Conference on Intergranular and Interphase Boundaries in Materials”, July 8-12, 2024 in Beijing:
(i) Jann-Erik Brandenburg, Luis A. Barrales-Mora, Sadahiro Tsurekawa, Dmitri A. Molodov: On the effect of inclination dependence of grain boundary energy on capillary-driven grain boundary migration
(ii) Andrej Ostapovets, Ritu Verma, Dmitri A. Molodov: On the models of shear coupled migration of twin boundaries

No. 2-1-2	Nanoscale assemblies in hydrogen-bonded liquids and in amorphous materials			
Name	László PUSZTAI	Title	Scientific Advisor	
Affiliation	HUN-REN Wigner Research Centre for Physics, Hungary Email: pusztai.laszlo@wigner.hun-ren.hu			
Research Field	Advanced materials / Data science and AI			
Period of appointment	April 1, 2024- March 31, 2025			
Host Professor	Yoichi NAKAJIMA			
Affiliation	Faculty of Advanced Science and Technology Email: yoichi@kumamoto-u.ac.jp	Title	Associate Professor	

1. Research outline and its perspective

My primary research goal in general may be described in short as '***understanding disordered structures*'.** Accordingly, my main activity (still, in general) is the investigation of the microscopic structure of liquids, amorphous materials and disordered crystals. We combine experimental data, such as total scattering structure factors (TSSF) from X-ray and neutron diffraction (XRD and ND, respectively) and EXAFS spectra, with computer modeling tools, such as Reverse Monte Carlo (RMC) and molecular dynamics (MD) simulations. As a result of such an approach, large sets (containing tens of thousands) of atomic coordinates ('particle configurations') in simulation boxes are provided that are consistent (within errors) with experimental data. These configurations are then subjected to various geometrical analyses, so that specific questions concerning the structure of a material may be answered.

2. Research progress and results in the fiscal year

(i) *The intermediate range structure of a prototype covalent glass.* – In order to investigate the relationship between the partial structures and the stiffness transition in As_xSe_{1-x} glasses, anomalous X-ray scattering (AXS) and X-ray and neutron diffraction (XRD and ND) experiments were carried out. For the AXS experiments, anomalous terms near the absorption edges were experimentally obtained instead of the theoretical values with large ambiguities. The results were analyzed by reverse Monte Carlo (RMC) modeling to obtain partial structure factors, $S_{ij}(Q)$, partial pair distribution functions, $g_{ij}(r)$, and three-dimensional atomic configurations. The $S_{ij}(Q)$ and $g_{ij}(r)$ functions gradually vary with *x*; however, an important change was observed in the intermediate-range element-selective atomic structures near the stiffness transition composition. With decreasing x across the so-called intermediate phase compositions, a rapid decrease of the As–As wrong bonds is visualized. However, the other anomalies found in Ge–Se glasses are not clearly observed, such as a rapid decrease in pre-shoulder position in SSeSe(Q), a rapid decrease in the number of edge-sharing connections, and an exclusion tendency of the connections between the As(Ge) atoms sharing two Se atoms, which may be related to the anisotropic pyramidal atomic configurations around the As atoms in the As–Se glasses in contrast to the isotropic tetrahedral ones around the Ge atoms in the Ge–Se glasses.

Related publication: Hosokawa, S.; Stellhorn, J.R.; Boudet, N.; Blanc, N.; Magome, E.; **Pusztai, L.**; Kohara, S.; Ikeda, K; Otomo, T.; Local- and Intermediate-range Partial Structure

Study of As-Se Glasses; *J. Phys. Soc. Jpn*., Vol.**93**, No.1, Article ID: 014601 (2024); DOI: https://doi.org/10.7566/JPSJ.93.014601

(ii) On the hydration structure of simple sugars. – Ab initio molecular dynamics (AIMD) simulations have been performed on aqueous solutions of four simple sugars, α-D-glucose, β-D-glucose, α-D-mannose, and α-D-galactose. Hydrogen-bonding (HB) properties, such as the number of donor- and acceptor-type HB-s, and the lengths and strengths of hydrogen bonds between sugar and water molecules, have been determined. Related electronic properties, such as the dipole moments of water molecules and partial charges of the sugar O atoms, have also been calculated. The hydrophilic and hydrophobic shells were characterized by means of spatial distribution functions. β-D-glucose was found to form the highest number of hydrophilic and the smallest number of hydrophobic connections to neighboring water molecules. The average sugar−water H-bond length was the shortest for β-D-glucose, which suggests that these are the strongest such H-bonds. Furthermore, β-D-glucose appears to stand out in terms of the symmetry properties of both its hydrophilic and hydrophobic hydration shells. In summary, in all aspects considered here, there seems to be a correlation between the distinct characteristics of β-D-glucose reported here and its outstanding solubility in water. Admittedly, our findings represent only some of the important factors that influence the solubility.

Related publication: Bakó, I.; **Pusztai, L**.; Pothoczki, Sz.; Outstanding Properties of the Hydration Shell around β-d-Glucose: A Computational Study; *ACS Omega* **9**, 20331–20337; DOI: https://doi.org/10.1021/acsomega.4c00798 (2024)

(iii) A peculiar structural feature in some metallic glasses: the rejuvenation effect. -- To experimentally clarify the changes in structural and dynamic heterogeneities in a metallic glass (MG), $Gd_{65}Co_{35}$, by rejuvenation with temperature cycling (cryogenic rejuvenation), high-energy X-ray diffraction (HEXRD), anomalous x-ray scattering (AXS), and inelastic x-ray scattering (IXS) experiments were carried out. By repeated temperature changes between liquid nitrogen and room temperatures 40 times, we observed tiny but clear structural changes by HEXRD even in the first-nearest neighbor range. The partial structural information obtained by AXS revealed that slight movements of Gd and Co atoms occur in the first- and second-nearest neighbor shells around the central Gd atom. The composition heterogeneity in nanometer size is markedly increased for the Gd atoms by temperature cycling, whereas the other heterogeneities are negligible. A distinct change was detected in microscopic elastic property by IXS: the width of longitudinal acoustic excitation broadens by about 20%, indicating an increase in the elastic heterogeneity of this MG induced by thermal treatments. These static and dynamic results explicitly clarify the features of the cryogenic rejuvenation effect experimentally.

Related publication:
Hosokawa, S.; Stellhorn, J. R.; **Pusztai, L.**; Yamazaki, Y.; Jiang, J.; Kato, H.; Ichitsubo, T.; Magome, E.; Blanc, N.; Boudet, N.; Ohara, K.; Tsutsui, S.; Uchiyama, H.; Alfred Q.R. Baron, A.Q.R.; Structural and dynamical changes in a Gd-Co metallic glass by cryogenic rejuvenation; *Acta Materialia* **284**, 120616 (2025); DOI: https://doi.org/10.1016/j.actamat.2024.120616

3. Research plan for the next fiscal year

During FY 2024, I've collaborated mostly with my former host professor, Dr. Hosokawa (now at Shimane University), and Dr. Nakajima from the Department of Physics, KU.

Follow-up publications with Prof. Hosokawa on the structure of amorphous materials will continue to appear for a while (at the moment, there is one publication accepted and scheduled to appear in 2025, as well as one submitted and another one under preparation). Also, further research is planned concerning the structure and dynamics of covalent and metallic glasses.

The joint research work with Dr. Nakajima, on high pressure diffraction measurements on various liquid mixtures, has evolved into a joint KAKENHI proposal with him, which proposal was approved in FY2022. We will work on Reverse Monte Carlo simulations on high temperature, high pressure iron containing liquids that may be present in the interior of the Earth.

4. List of journal papers in international journal as of the end of December; (excluding conference papers and proceedings)

1) issued with volume numbers from April 2024 to December 2024

Bakó, I.; **Pusztai, L**.; Pothoczki, Sz.; Outstanding Properties of the Hydration Shell around β-d-Glucose: A Computational Study; *ACS Omega* **9**, 20331–20337; DOI: https://doi.org/10.1021/acsomega.4c00798 (2024)

2) scheduled to be issued with volume numbers from January 2025 to March 2025 and confirmed to be issued thereafter as of the end of December

Hosokawa, S.; Stellhorn, J. R.; **Pusztai, L.**; Yamazaki, Y.; Jiang, J.; Kato, H.; Ichitsubo, T.; Magome, E.; Blanc, N.; Boudet, N.; Ohara, K.; Tsutsui, S.; Uchiyama, H.; Alfred Q.R. Baron, A.Q.R.; Structural and dynamical changes in a Gd-Co metallic glass by cryogenic rejuvenation; *Acta Materialia* **284**, 120616 (2025); DOI: https://doi.org/10.1016/j.actamat.2024.120616

Symposiums/seminars for graduate students/undergraduate students at KU:

I have delivered a 90-minute presentation as an *IROAST Seminar* (Kumamoto University, 22 November 2024), titled '*Neutron scattering methods in condensed matter physics, chemistry and materials sciences*'.

(See https://iroast.kumamoto-u.ac.jp/symposiumsseminars/the-136th-iroast-seminar/)

<table>
<tr><td>No. 2-1-3</td><td colspan="3">Development of Zn based biodegradable metals</td><td rowspan="2">  </td></tr>
<tr><td>Name</td><td>Yufeng ZHENG</td><td>Title</td><td>Distinguished Professor</td></tr>
<tr><td>Affiliation</td><td colspan="4">School of Materials Science and Engineering, Peking University, China
Email: yfzheng@kumamoto-u.ac.jp</td></tr>
<tr><td>Research Field</td><td colspan="4">Biotechnology & healthcare technology /Advanced materials</td></tr>
<tr><td>Period of appointment</td><td colspan="4">April 1, 2024- March 31, 2025</td></tr>
<tr><td>Host Professor</td><td colspan="4">Yoji MINE</td></tr>
<tr><td>Affiliation</td><td colspan="2">Faculty of Advanced Science and Technology
Email: mine@msre.kumamoto-u.ac.jp</td><td>Title</td><td>Professor</td></tr>
</table>

[Details of activities]

1. Research outline and its perspective

Zn based biodegradable metals and alloys are one of the new research direction in the field of metallic biomaterials, and with the research framework of IROAST, we aims to develop new kind of Zn alloys with high mechanical strength and good ductility, which may have the potential usage as bone implant and cardiovascular stent materials.

2. Research progress and results in the fiscal year

I collaborated with Prof. Mine, Prof. Kwak and Prof. Takashima on the mechanical property of Zn based biodegradable metals, and we studied the fatigue behavior of Zn-Li alloys with different addition of the Li element. I also started to collaborate with Prof. Xu on the international collaboration research grant application, with the topic to regulate the degradation behavior of the Zn alloys with the surface modification technique by polymers.

3. Research plan for the next fiscal year

I will continue the research work with Prof. Mine, Prof. Kwak and Prof. Takashima, and expect to explore new collaboration with Prof. Otani and Prof. Arima on possible topic to combine the nanoCT technique with the in vivo testing of the biodegradation products on the surface of the Zn alloy cardiovascular stent.

4. List of awards, grants, and patents

I have been selected as one of the “Highly Cited Researchers 2024,” in the field of materials science by Clarivate.

5. List of journal papers in international journal as of the end of December (excluding conference papers and proceedings)

Siqi Jin, Yameng Yu, Ting Zhang, Daping Xie, Yufeng Zheng, Chunming Wang, Yunsong Liu, Dandan Xia, Surface modification strategies to reinforce the soft tissue seal at transmucosal region of dental implants, Bioactive Materials 42 (2024) 404-432

He Huang, Liudang Fang, Zhipei Tong, Gencheng Gong, Hui Yu, Olga Kulyasova, Ruslan Z Valiev, Dandan Xia, Yufeng Zheng, Dong Bian, Nanostructuring of Zn–Li-based alloys through severe plastic deformation: Microstructure, mechanical properties, and corrosion behaviors, Nano Materials Science in press https://doi.org/10.1016/j.nanoms.2024.07.004