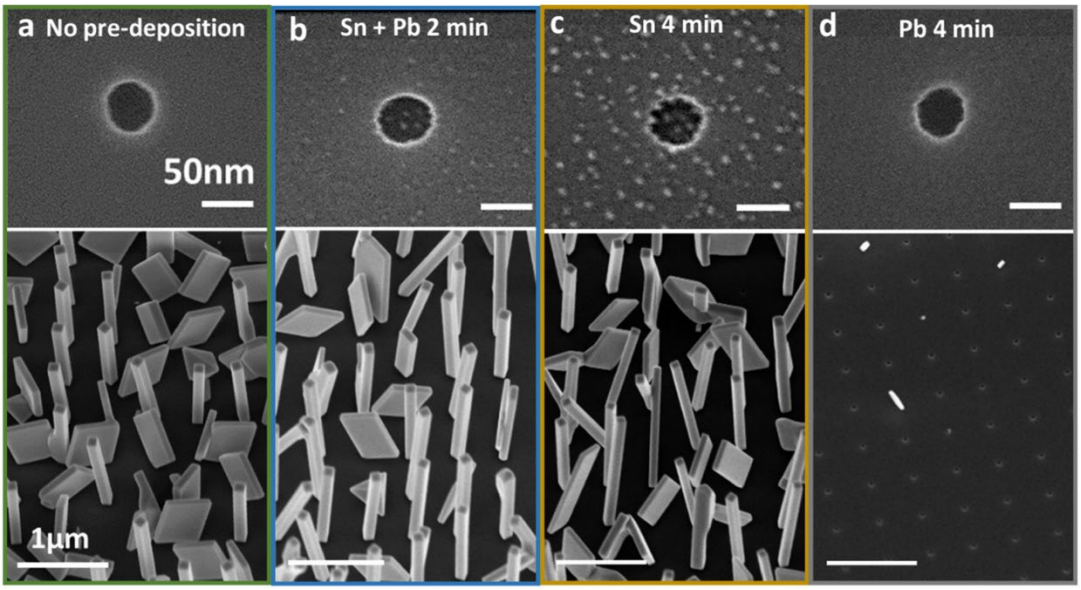
DUTCH ASSOCIATION FOR CRYSTAL GROWTH

nederlandse vereniging voor kristalgroei

FACET

DACG Newsletter

Newsletter of the Dutch Association for Crystal Growth (DACG), section of the KNCV and the NNV.



FACET Issue 2/2024
December 2024
Editor: Joop H. ter Horst (Info@dacg.nl)

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DACG board
President: Dr. Carmen Guguta (Technobis)
Secretary: Prof. Dr. Antoine van der Heijden (TUD)
Tresurer: Dr. Hans te Nijenhuis (Malvern Panalytical)
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Elias Vlieg (Universiteit Nijmegen)
Electable:
Anne Claude (University of Amsterdam)

If you are interested in a role within the DACG just contact us: info@dacg.nl.

Cover photo
Representative SEM images of the PbSnTe growth substrate before (top panels) and after growth of PbSnTe nanostructures (bottom panels)

Mientjes, M. G. C., Guan, X., Lueb, P. J. H., Verheijen, M. A. & Bakkers, E. P. A. M., Catalyst-free MBE growth of PbSnTe nanowires with tunable aspect ratio, *Nanotechnology* 35(32) (2024) 325602. https://iopscience.iop.org/article/10.1088/1361-6528/ad47c8

From the editor

Dear all,
We had an excellent spring symposium at Maastricht University on the 31st of May 2024, organized by the group of Dr Giuditta Perversi. On the 17th of January 2025 we will start the year well with the delayed fall symposium. You can find the agenda of this symposium at the University of Nijmegen in this Facet. We will hand out the Piet Bennema Crystal Growth Award 2024, among others!
Your ideas for the Facet are welcome! It is easy to contribute to the FACET. Just send an email with your contribution to the editor via info@dacg.nl.
Looking forward to seeing you at the symposium in January!
Joop ter Horst

DACG Fall Symposium January 17, 2025

📢 **Join Us for the DACG Fall Symposium 2024!**

We are thrilled to announce that the **DACG Fall Symposium 2024** will take place on **Friday, January 17, 2025**, at the prestigious **Radboud University Nijmegen**.

This year's symposium promises an engaging program filled with:

🎤 **Expert Keynote Speakers** sharing the latest insights and trends

🧑‍🤝‍🧑 **Inspiring Presentations** from leaders in the field

🤝 **Networking Opportunities** with professionals and researchers

💡 **Interactive Discussions** to shape the future of nucleation, crystallization, crystal growth, and self-assembly

This is a unique opportunity to connect with peers, exchange ideas, and gain new perspectives.

📍 **Date:** January 17, 2025

📍 **Location:** Radboud University Nijmegen

We look forward to welcoming you to a day of knowledge sharing and collaboration. Mark your calendar now—you won't want to miss it!

Registration: https://dacg.nl/event/dacg-fall-symposium-2024-general-members-meeting/

09:30 – 10:00	**Registration and coffee**
10:00 – 10:10	**Welcome and introduction** Carmen Guguta (DACG Chair)
10:10 – 10:40	**Crystal growth in confined spaces: A study of crystallization pressure in a potash alum model system** Anton Tuluk (RU Nijmegen)
10:40 – 11:10	**Crystal orientation using the azimuthal scan method** Dirk Kok (Malvern Panalytical)
11:10 – 11:40	**New insights into the mechanism of Non-Photochemical Laser Induced Nucleation *(tentative title)*** Burak Eral (Delft University of Technology)
11:40 – 12:10	**Biocrystallization for resource recovery** Annemerel Mol (Wageningen University & Research)
12:10 – 13:00	**Lunch**
13:00 – 14:00	**General Members Meeting DACG**
14:00 – 14:30	**Directing sequential self-organization with self-assembled nanocrystals *(tentative title)***

	Ariane Mader (AMOLF)
14:30 – 14:45	**Coffee break**
14:45 – 15:00	**Piet Bennema Crystal Growth Award 2024 – Award Ceremony** Bart Zwijnenburg (Nobian), Chair of the Jury
15:00 – 15:45	**Co-crystal prediction using network science and machine learning** Jan-Joris Devogelaer (Johnson & Johnson Innovative Medicine), prize winner of the Piet Bennema Crystal Growth Award 2024
15:45 – 16:45	**Lab tour**
16:45 – 17:30	**Drinks / nibbles**

Venue: Radboud University Nijmegen, Huygens Building, Heyendaalseweg 135, 6525 AJ Nijmegen

Agenda for the DACG Annual Meeting, 17 January 2025

1. Opening
2. Finalize agenda
3. Minutes Annual meeting 30 November 2023
4. Documents sent / received: report of the financial audit committee (see agenda item 6)
5. Annual Report Oct 2023 – Sep 2024
6. Financial

(a) Annual Financial Report Oct 2023 – Sep 2024
(b) Report Financial Audit Committee
(c) Budget Oct 2024 – Sept 2025

7. Status Stichting
8. Young-DACG
9. DACG website
10. Board

Role	2023 – 2024	Appointment deadline
President	Carmen Guguta	Step down Oct 2024
Secretariat	Antoine van der Heijden	Step down Oct 2025
Treasurer	Hans te Nijenhuis	Step down Oct 2024
FACET	Joop ter Horst	Step down Oct 2026
Member	Elias Vlieg	Step down Oct 2026

Since 2023 Anne Claude (PhD student UvA) succeeded Marloes Bistervels to support the board as a representative of the Young-DACG.
The board proposes to elect Anne Claude as a regular board member as representative of Young-DACG.

Both Carmen Guguta and Hans te Nijenhuis will step down from the board and are not re-electable. DACG-members who would like to apply for a board membership, can announce themselves by sending an e-mail to the DACG secretariat (a.e.d.m.vanderheijden@tudelft.nl), **ultimately by 31 December 2024**.

11. Activities 2024 – 2025
    a. NNV Advisory Board Meeting, early 2025 (section boards will be invited)
    b. April 2025: spring symposium DACG, location and date to be decided
    c. October 2025: DACG annual meeting + fall symposium, Nobian, Enschede, date to be decided
12. Questions before closure of meeting
13. Adjourn

Report DACG-NVK Spring symposium

On 31 May, 2024, the DACG and NVK (Nederlandse Vereniging voor Kristallografie) organized a joint spring meeting hosted at Maastricht University. The local organizer was **Giuditta Perversi**, who was supported by a team of students. The symposium was preceded by an Early Career Event on 30 May. **Jacco van der Streek** and **Jules Harings**, presented their work and shared their experiences, from both industry and academia, with MSc students, PhD students and post-docs. A poster session was also part of this Early Career Event and allowed the participants to present and discuss their work.

Giuditta Perversi (right) and Jessica Steinlechner (left)

ETPathfinder

The symposium on 31 May consisted of a balanced program of speakers from both the crystallographic and crystallization communities in The Netherlands and Belgium. Belgium was represented by three keynote speakers: **Ken Haenen** (Hasselt Univ), **Sandra Van Aert** (Univ of Antwerp) and **Shehab Ismail** (UCLouvain), presenting on properties of CVD diamond films, recent advances in 3D atomic structure quantification using scanning transmission electron microscopy and structural studies on ciliary trafficking, respectively. The other three speakers **Gertjan Koster** (Twente Univ), **Olga Chukhutsina** (VU Amsterdam) and **Albert Guskov** (Univ of Groningen) presented on the structure and properties of low dimensional epitaxial oxides, advanced crystallographic approaches to resolve how lazy photoreceptors work and Vitamin B12

scavenging and processing in bacteria, respectively. Unfortunately, due to time constraints, the lecture of Giuditta Perversi was cancelled; the presentation will be shifted to one of the next DACG symposia. The symposium ended with an interesting visit to the Einstein Telescope Pathfinder (ETPathfinder), after an introduction by **Jessica Steinlechner** (Maastricht Univ) on this prestigious project focused on the detection of gravitational waves.

The DACG symposium was attended by ca. 60 participants. On behalf of the DACG and NVK board, we would like to acknowledge Giuditta Perversi and her supporting team, for organizing this successful symposium and for hosting the DACG & NVK communities in Maastricht.

Antoine van der Heijden

A selection of recent publications

F. Valls Mascaró, M.T.M. Koper, and M.J. Rost, Quantitative Study of Electrochemical Adsorption and Oxidation on Pt(111) and its Vicinal Surfaces, *Electrochim. Acta*, 506, 145014 (2024). https://doi.org/10.1016/j.electacta.2024.145014

F. Valls Mascaró, M.T.M. Koper, and M.J. Rost, Step Bunching Instability and its Effects in Electrocatalysis: Pt(111) and its Vicinal Surfaces, *Nature Catalysis*, (2024). https://doi.org/10.1038/s41929-024-01232-2

Rovaris, F., Peeters, W. H. J., Marzegalli, A., Glas, F., Vincent, L., Miglio, L., Bakkers, E. P. A. M., Verheijen, M. A. & Scalise, 2H-Si/Ge for Group-IV Photonics: on the Origin of Extended Defects in Core-Shell Nanowires, *ACS Applied Nano Materials*. 7(8) (2024) 9396–9402. https://doi.org/10.1021/acsanm.4c00835.

Mientjes, M. G. C., Guan, X. , Lueb, P. J. H., Verheijen, M. A. & Bakkers, E. P. A. M., Catalyst-free MBE growth of PbSnTe nanowires with tunable aspect ratio, *Nanotechnology* 35(32) (2024) 325602. https://iopscience.iop.org/article/10.1088/1361-6528/ad47c8

Peeters, W. H. J., van Lange, V. T., Belabbes, A., van Hemert, M. C., Jansen, M. M., Farina, R., van Tilburg, M. A. J., Verheijen, M. A., Botti, S., Bechstedt, F., Haverkort, J. E. M. & Bakkers, E. P. A. M. , Direct bandgap quantum wells in hexagonal Silicon Germanium, *Nature Communications* 15(1) (2024) 5252. https://doi.org/10.1038/s41467-024-49399-3.

Rossi, M., van Schijndel, T. A. J., Lueb, P., Badawy, G., Jung, J., Peeters, W. H. J., Kölling, S., Moutanabbir, O., Verheijen, M. A. & Bakkers, E. P. A. M. , Stemless InSb nanowire networks and nanoflakes grown on InP, *Nanotechnology* 35(41) (2024) 415602. https://iopscience.iop.org/article/10.1088/1361-6528/ad61ef.

Mattinen, M., Chen, W., Dawley, R. A., Verheijen, M. A., Hensen, E. J. M., Kessels, W. M. M. & Bol, A. A. , Structural Aspects of MoSx Prepared by Atomic Layer Deposition for Hydrogen Evolution Reaction, ACS Catalysis. 14(13) (2024) 10089-10101. https://doi.org/10.1021/acscatal.4c01445.

Krabben, L.M. van der; Gruginskie, N.; Eerden, Maarten van; Gastel, Jasper van; Mulder, Peter; Bauhuis, G.J.; Khusyainov, Dinar; Afanasiev, D.; Vlieg, Elias; Schermer, J.J., Reduced Surface Recombination in Extended-Perimeter LEDs toward Electroluminescent Cooling, *ACS Applied Electronic Materials*, **2024**, 6(2), 1483-1492. https://doi.org/10.1021/acsaelm.3c01816

Pinetre, Clement; Ritou, Loic; Gerard, Charline J.J.; Cercel, Hugo; Leeman, M.; Kellogg, R.M.; Tinnemans, P.T.; Dupray, Valerie; Horst, J.H. ter. Rare Case of Polymorphism in the Binary System of Enantiomers of a Praziquantel Derivative, *Organic Process Research & Development*, **2024**, 28(4), 1224-1232. https://doi.org/10.1021/acs.oprd.4c00035

Vries, Tom E. de; Eert, Evi van; Weevers, Lucas; Tinnemans, P.T.; Vlieg, E.; Meekes, H.L.M.; Gelder, R. de, Optimizing Link Prediction for the CSD Cocrystal Network: A Demonstration Using Praziquantel, *Crystal Growth & Design,* **2024**, 24(12), 5200-5210. https://doi.org/10.1021/acs.cgd.4c00438

Rapeenun, Peerapon; Gerard, Charline J.J.; Pinetre, Clement; Cartigny, Yohann; Tinnemans, Paul; Gelder, Rene de; Flood, Adrian E.; Horst, J.H. ter, Searching for Conglomerate Cocrystals of the Racemic Compound Praziquantel, *Crystal Growth & Design* **2023**, 24(1), 480-490. https://doi.org/10.1021/acs.cgd.3c01158

Nagaraj Nagalingam, Aswin Raghunathan, Vikram Korede, Edwin F.J. Overmars, Shih-Te Hung, Remco Hartkamp, Johan T. Padding, Carlas S. Smith, Huseyin Burak Eral, Low-cost fluorescence microscope with microfluidic device fabrication for optofluidic applications, HardwareX **2023**, 14, e00415, ISSN 2468-0672, https://doi.org/10.1016/j.ohx.2023.e00415

Nagaraj Nagalingam, Vikram Korede, Daniel Irimia, Jerry Westerweel, Johan T. Padding, Remco Hartkamp & Hüseyin Burak Eral, Unified framework for laser-induced transient bubble dynamics within microchannels. Sci Rep 14, 18763 (2024). https://doi.org/10.1038/s41598-024-68971-x

Recent Theses

Francesc Valls Mascaro, Platinum Surface Instabilities and their Impact in Electrochemistry
PhD defense: 5 September 2024, Leiden University
Promotors: Prof. dr. M. T. M. Koper, Dr. M. J. Rost

Hydrogen fuel cells are expected to be pivotal for the energy transition towards renewable energy sources. However, their economic feasibility is severely hindered by the high cost and degradation rate of the platinum electrode, which forms the key component for the catalytic reaction. Addressing this challenge necessitates designing improved catalysts, which requires a better fundamental understanding of both their reactivity and their degradation mechanism. In this thesis, we investigate the stability of model platinum surfaces submerged in electrolyte under an applied voltage, thus simulating the operational conditions of fuel cells. For this we used a home-build Electrochemical Scanning Tunneling Microscope (EC-STM), which allows us to observe the surface structure at the atomic scale in real time during its operation. Our findings surprisingly show that several instabilities lead to significant structural changes. At high voltages, nanoislands with dendritic shapes form during the reduction of the surface oxide, which is the origin of the surface roughening and thus linked to the catalyst degradation. Moreover, we demonstrate that closely-spaced atomic steps are unstable and bunch together, resulting in steps with multi-atomic heights. This structural change has a significant effect on the catalyst reactivity as well as on its potential of zero charge, as we explain in detail. Finally, we investigate vicinal platinum surfaces to better understand the reactivity of facets on nanoparticles: again we find an instability leading to structural changes. Overall, this thesis sheds light on the fundamental, atomic processes that drive platinum surface restructuring as well as their implications for reactivity.
https://scholarlypublications.universiteitleiden.nl/handle/1887/4054933

Vikram B. Korede, Taming Crystallization with Light
PhD defense: 7 October 2024, Delft University of Technology
Promotor: Dr. H.B. Eral, Prof. dr. J.T. Padding

Crystallization is one of the most widely used purification and separation processes applied in a multitude of industries such as pharmaceuticals, food & beverages, agriculture, and fine chemicals. However, the initial step of the crystallization process, nucleation, is still poorly understood and highly stochastic. As a result, most crystallization processes lack proper control over the properties of the crystals produced. Among many techniques for achieving better control over the nucleation process, the application of non-photochemical laser induced nucleation (NPLIN) has gathered significant interest. This is because of its potential to improve product quality in crystallization processes by directly controlling the nucleation rate, both spatially and temporally. Additionally, NPLIN can induce crystallization in solutions that would otherwise take a long time to nucleate, offering a unique advantage over traditional methods. However, despite its promising capabilities, NPLIN is not widely used in practice yet. The fundamental mechanism behind NPLIN is not fully understood, making it unclear how it should be applied effectively in practice and for which systems NPLIN could be beneficial. This Ph.D. project aims to delve into the fundamental mechanisms of NPLIN, by examining how specific laser and solution parameters influence nucleation kinetics, leveraging innovative experimental

setups. Laser parameters being studied include laser-exposed volume, laser irradiation position, laser intensity and laser wavelength, and solution parameters include supersaturation levels, solution filtration, and the presence of impurities or dopants, particularly nanoparticles.
The thesis begins with a comprehensive review of the experimental and computational literature on NPLIN. It then presents a detailed study on the effect of the laser-exposed volume and laser irradiation position on the nucleation probability within partly illuminated supersaturated aqueous potassium chloride solutions. An increase in the laser-exposed volume resulted in a higher nucleation probability and a higher number of crystals per nucleated sample. Furthermore, laser irradiation, particularly through the air/solution interface, not only enhances nucleation probability but also influences the formation of different crystal morphologies. These observations are partly explained by the Nanoparticle Heating mechanism and the Dielectric Polarization model (Chapter 2). The research then transitions to a microfluidic platform, which allows for high-throughput and crystallization detection using the deep learning method. This innovative approach addresses the need for large data sets in NPLIN research, which has been a significant challenge due to the manual nature of traditional experiments. The study examines the effects of laser intensity, wavelength, supersaturation, solution filtration, and intentional doping on nucleation probability in supersaturated potassium chloride solutions. Higher laser intensities and increased supersaturation significantly enhance nucleation probabilities. The laser wavelength effect was only observed for 355 nm at higher laser intensities. Solution filtration suppresses the NPLIN effect, whereas the addition of nanoparticles as dopants into the solution not only increases the NPLIN probabilities but also affects the crystal morphology. The results highlight the importance of impurities in the solution and support the hypothesis that nanoparticle or impurity heating could be the key mechanism in understanding NPLIN (Chapter 3).
The study finally investigated the effects of solution filtration, laser intensity, and nanoparticle properties including nanoparticle concentration and material on NPLIN probability in supersaturated aqueous urea solutions. The study highlights the significant role of impurities in NPLIN, demonstrating that doping with different nanoparticle materials leads to varied nucleation probabilities. In particular, gold nanoparticles were found to enhance nucleation more effectively than silica nanoparticles. Additionally, it was observed that NPLIN probabilities followed a Poisson distribution to changes in nanoparticle concentration and laser intensity respectively. The findings in this chapter enhance our understanding of the critical role of impurities in comprehending the NPLIN mechanism (Chapter 4).
https://repository.tudelft.nl/record/uuid:7c197383-2adc-49d6-9b28-cfd93114142b

Nagaraj Nagalingam, Laser-Induced Cavitation for Controlling Crystallization from Solution
PhD defense: 14 October 2024, Delft University of Technology
Promotor: Dr. H.B. Eral, Prof. dr. J.T. Padding

Primary nucleation control is crucial for obtaining crystals with specific properties, such as purity, size, morphology, and polymorphic form. Non-photochemical laser-induced nucleation (NPLIN) has attracted interest due to its ability to control these properties without chemical reactions, using non-invasive methods, and allowing spatio-temporal precision. However, the exact mechanism underlying NPLIN remains debated in the literature.

This dissertation explores how micron-sized vapor bubbles, formed by laser interaction with supersaturated aqueous solutions, can trigger crystal nucleation. Despite aqueous solutions generally being transparent to laser wavelengths of 532 nm and 1064 nm, transient bubbles can still form due to energy absorption by impurities or by focusing the laser. The research begins by examining the crystallization of KCl in aqueous solutions, initiated by bubbles formed using focused laser light with nanosecond pulse width. Findings show that solute accumulation at the bubble surface exceeds the saturation limit, leading to localized supersaturation. A finite element method model, validated by experimental bubble size data, is used to estimate solute transfer and supersaturation levels. The model demonstrates a concentrated solute boundary layer around the bubble, driven by high solvent evaporation rates associated with bubble growth. The experimental results for crystallization probability and crystal count align with classical nucleation theory predictions based on the numerically estimated supersaturation at the vapor-liquid interface.
The bubble formation mechanism proposed for NPLIN is extended to other solutes like NH_4Cl, NaCl, KBr, and CH_4N_2O. Experiments with NH_4Cl and NaCl yield a general analytical relation for supersaturation in the liquid surrounding the bubble, explaining NPLIN activity for these solutes when an unfocused laser is used. The predicted bubble sizes, based on Mie theory, correlate with the minimum nucleation rate necessary for crystal formation, indicating that the bubble-driven mechanism is a key factor in NPLIN.
Since isolated bubbles are rare in irradiated volumes due to the random distribution of impurities, the study also investigates bubble-bubble interactions and their effect on crystallization. The dynamics of single laser-induced bubbles in microchannel geometries are analyzed, revealing a unified theory for bubble size and lifetime as a function of laser energy. This analysis also uncovers a transient flow instability, rare in low Reynolds number flows, which originates from the channel walls and is characterized by the Womersley number and flow timescale. The research further demonstrates crystallization using bubble pairs in microchannels with KMnO4 as a model salt. The interaction between bubbles produces microjets that alter nucleation kinetics through induced shear, enabling crystallization at lower laser energies and solution supersaturation compared to single bubbles. A numerical model based on the boundary integral element method is used to predict microjet velocities and the resulting shear, correlating these factors with crystallization probabilities.
Overall, this work advances understanding of NPLIN, suggesting that bubble formation and interactions can be harnessed to achieve targeted crystallization with lower energy inputs and reduced supersaturation, paving the way for more efficient laser-induced crystallization processes.
https://repository.tudelft.nl/record/uuid:17b93f25-8f3f-4ab4-b5f8-ba446d923b6d

Eline Grothe, Classification of Stereoisomerism and Multicomponent Systems in Crystallographic Databases

PhD defense:	1 November 2024, Radboud Universiteit, Nijmegen
Promotor:	Prof. dr. Elias Vlieg
Co-promotors:	Dr. Hugo Meekes, Dr. R. de Gelder

The magnitude and growing complexity of the data in crystallographic databases such as the Cambridge Structural Database (CSD) can pose a challenge when assembling the right data set

to address a research question. This is complicated further by the limitations of available search possibilities and the complexity of problem statements. This thesis describes the development of methods that are needed to assemble a dataset based on chirality of the components and based on the composition of multicomponent crystals.
https://repository.ubn.ru.nl/handle/2066/311641

Marloes Bistervels, Light-controlled self-assembly | Crystallization in the spotlight
PhD defense: 29 November 2024, University of Amsterdam
Promotor: Prof. dr. Wim Noorduin
Co-promotor: Prof. dr. Fred Brouwer

Precise control over biotic and abiotic self-assembly processes is of fundamental interest, with practical implications for developing simple and scalable routes toward complex three-dimensional (3D) architectures with advanced functionalities. While techniques for achieving either local and static or dynamic and global control have been developed, local and dynamic control remains challenging. In this thesis, we harness light to achieve spatiotemporal control over the self-assembly of biorelevant crystals.
By utilizing photochemical reactions, we create local gradients of precursors that drive the nucleation and growth of bioinspired co-precipitation of barium carbonate nanocrystals and amorphous silica. Using a custom-built optical setup and optimizing the UV irradiation with the reaction conditions, we control self-assembly in terms of time, position, nucleation rate, and morphology.
By employing near-infrared (NIR) laser light, we demonstrate spatiotemporal control over the crystallization of retrograde soluble compounds induced by locally heating water. Modulating the NIR light intensity enables to start, steer, and stop crystallization of carbonate minerals with micrometer precision.
By integrating ion exchange into the light-driven self-assembly process, we introduce a two-step strategy based on self-organization and conversion reactions, shaping a diverse range of chemical compositions into user-defined designs. Considering thermodynamic stability and chemical reactivity, we design orthogonal conversion reactions for the sequential positioning of different metal chalcogenide semiconductors and the integration of various compositions into the same hybrid architecture.
This work opens previously unimaginable opportunities for light-directed self-assembly of functional composites, forging a genuine collaboration with self-assembly processes: simple hands-off autonomy when possible, and precise hands-on command when necessary.
https://dare.uva.nl/search?identifier=bc9d1913-8abf-47a9-b25e-6a7a38a4a3b5