Prof Kenneth S. Suslick

School of Chemical Sciences, University of Illinois at Urbana-Champaign, US

Sonofragmentation and Sonocrystallization: How Solids Break and Make in Cavitating Liquids

Fundamentally, chemistry is the interaction of energy and matter. Surprisingly, there are relatively few ways of putting energy into molecules. Through the process of acoustic cavitation, high intensity ultrasound has found numerous applications in driving chemical reactions and in the preparation of unusual materials, i.e., "sonochemistry". We do not yet understand, at a molecular level, how things break. What happens to macroscopic solids (crystalline or not) as stresses are applied and how those stresses create breakage of chemical bond breakage and weaker inter-molecular interactions remain remarkably mysterious. Mechanochemistry (i.e., the chemical consequences of grinding, milling and breakage of bulk materials) is an area with a long (but not well-known) history that is currently undergoing vigorous renaissance and rapid expansion of interest for chemists, physicists, theoreticians and industrial researchers. Mechanical action can produce dramatic physical and mechanochemical effects when the energy is spatially or temporally concentrated. An important example of such phenomena in solids is the mechanical initiation of explosions, which has long been speculated to result from 'hot spot' generation at localized microstructures in the energetic material. Direct experimental evidence of such hot spots, however, is exceptionally limited; mechanisms for their generation are poorly understood and methods to control their locations remain elusive. Here we report the generation of intense, localized microscale hot spots in solid composites during mild ultrasonic irradiation, directly visualized by a thermal imaging microscope. These hot spots, with frictional heating rates reaching >106 K/s, nucleate exclusively at interfacial delamination sites in composite solids. In related work, the application of ultrasound to crystallization (i.e., sonocrystallization) can dramatically affect the properties of the crystalline products. Sonocrystallization induces rapid nucleation, generally yields smaller crystals of a more narrow size distribution compared to quiescent crystallizations, and has become increasingly important in the pharmaceutical industry for the preparation of APIs (active pharmaceutical ingredients). The control of morphology of the crystallization process is critical to reproducible dose response for APIs and is under increasing scrutiny in pharmaceutical manufacturing by the FDA. Ultrasound can induce significant improvement in the uniformity of crystallite size and rates of crystallization. We have developed a mechanistic understanding of the origin of these phenomena and begun to separate the details of the effects of ultrasound on nucleation, mass transport, shockwave fragmentation of crystallites, and inter-particulate collision. Decoupling experiments were performed to confirm that interactions between shockwaves and crystals are the main contributors to crystal breakage. We have discovered a mechanochemical extension the Bell–Evans–Polanyi principle: activation energies for solid fracture correlate with the binding energies of the solids.

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Prof Carlo Massimo Casciola

Dept. of Mechanical and Aerospace Engineering Sapienza University of Rome, Italy

Bubble Nucleation in Flowing Liquids

Bubble nucleation is a ubiquitous phenomenon whose prediction proved a formidable task, particularly in the case of water. Here a self-contained model is discussed which is shown able to accurately reproduce data for bulk water over the most extended range of temperatures for which accurate experiments are available [1]. The computations are based on a Ginzburg-Landau model which, as only inputs, requires a reliable equation of state for the bulk free energy and the interfacial tension of the water-vapor system. Rare event techniques borrowed from statistical mechanics allow the determination of the free-energy barrier and the nucleation rate. By consistently including thermal fluctuations [2] in the spirit of Fluctuating Hydrodynamics, the approach is extended to dynamic conditions in presence of solid walls of different wettability [4] to allow coupling with fluid motion [4]. The talk will focus on the wall wettability in compliance with the fluctuation-dissipation balance, a crucial point in the context of the fluctuating hydrodynamics theory. New results concerning the coupling of nucleation and fluid flow, the effect of micro-confinement, and time-changing thermodynamic conditions will also be addressed.

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Dr David Fernandez-Rivas

University of Twente, Netherlands

Light and Electrons to make Bubbles and Droplets

We are developing new ways to use bubbles in two fields: biomedical and green technologies. 1) Heating a liquid contained in microfluidic channels with lasers can lead to an increase in temperature that causes a bubble to appear. As the bubble grows, it can push the liquid out of the channel, and generate high velocity micro droplets. These droplets can overcome challenges of needle free drug delivery, since they interact with biological tissues presumably causing minimal damage. 2) The presence of bubbles on electrodes during electrochemical processes can be seen as evidence that a desired product, e.g., hydrogen has been produced. However, if the bubble blocks the active electrode surface, undesired effects can negatively affect the overall efficiencies of chemical reactors. We have been working on ways to decouple the generation of hydrogen and bubble growth, using microfabrication tricks, careful experiments, and modelling work. I will share our latest results and potential new avenues of our research.

Dr Sigmund T. Thoroddsen

King Abdullah University of Science and Technology (KAUST)

Laser-cavitation in superfluid and solid helium

Liquid helium (He-II) becomes superfluid at temperatures below 2.17K and if the pressure is then increased above 25 atmospheres it becomes a solid. The interface between this solid and the superfluid can exhibits intriguing wavelike oscillations, similar to those at a free surface. Herein, we use an optical cryostat and ultra-high-speed video imaging at frame rates up to 7 million fps to experimentally investigate laser-induced interfacial dynamics in these two exotic phases. The cryostat has been modified from that described in Speirs *et al.* [1] and can reach temperatures down to 1.2 K and pressures as high as 39 atm. We direct a pulsed Nd:YAG laser onto a parabolic mirror to produce sufficient power to achieve laser-cavitation. We observed the dynamics of cavitation bubbles in the superfluid and characterize melting and resolidification for the solid. We also track interface instabilities and shockwaves inside the different phases.

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*Funding for this research was provided by King Abdullah University of Science and Technology (KAUST) under grant URF/1/4075-01-01.

Dr Simo A. Mäkiharju

Assistant Professor and Director of the FLOW Laboratory, Department of Mechanical engineering
University of California Berkeley

Development of in-lab X-ray Particle Velocimetry and Multispectral CT for Multiphase Flows

The ability to measure fluid flow properties non-intrusively is often crucial for understanding of fluid dynamics, and subsequently to enable improving industrial and medical processes and equipment for the benefit of society. Particle image velocimetry utilizing visible wavelengths is one of the most useful tools we presently have for the study of single-phase flows. However, many multiphase flows are opaque at visible wavelengths due to the refractive interfaces between the phases. Fortuitously, at X-ray wavelengths the change in refractive index between most fluids of interest is very small; therefore, X-ray based Particle image (or tracking) Velocimetry (XPV) can measure fluid flow properties in multiphase flows. Using a combination of first principle-based modelling and experiments, we have been working towards practical in-lab XPV techniques and have explored the physical and present technical capabilities and limitations. The numerical modeling utilized both established Monte Carlo codes and simpler inhouse codes based on Beer-Lambert law. The experiments utilized both scintillator-based, and photon counting X-ray detectors, and used traditional and our custom developed 50 µm hollow carbon tungsten-coated particles to enable velocity measurements. We present results of Hz to kHz in-lab XPV in canonical flows. Complementing XPV we also tackle the challenge of quantitative measurement of composition. For multiphase flows it is challenging to non-intrusively quantify the spatial distribution of gas, liquid and solids - especially near solid boundaries even in a time-average sense, yet alone time-resolved. For this challenge multispectral photon counting Computed Tomography (mpCT) is being developed and has potential to yield otherwise unattainable data.

Dr Patrik Vagovic

Center for Free Electron Laser Science, DESY, Hamburg, Germany, European XFEL, Schenefeld, Germany

Megahertz X-ray Microscopy, a novel tool for the characterization of stochastic dynamics at European XFEL

The launch of the European XFEL (EuXFEL), the world's first megahertz X-ray Free Electron Laser (XFEL) source, has created new avenues for investigating dynamic processes on microscopic spatial and sub-nanosecond temporal scales. With its capability to record fast X-ray videos at a rate of up to 4.5 MHz, and to produce each frame with fs illumination and exceptional phase contrast at photon energies up to 24 keV. MHz X-ray microscopy has been demonstrated to be a valuable tool that was applied in a pilot demonstration for studying laser-driven explosions in capillaries [1]. In response to this, we have created a specialized setup that can be utilized by a wider range of scientific and industrial users to study such phenomena. As an example, Fig. 1 displays the use of the technology in exploring fluid dynamics in a Venturi tube. Additionally, the XFEL's high repetition rate (MHz) and high number of photons per pulse (10^{12}-10^{13}) make it possible to produce 3D images with a single pulse, and to record them with MHz sampling. This technology is currently being developed through the EIC-Pathfinder MHz-TOMOSCOPY project [2]. I will present the latest advancements, provide recent examples, and examine future prospects for this exciting field of research.

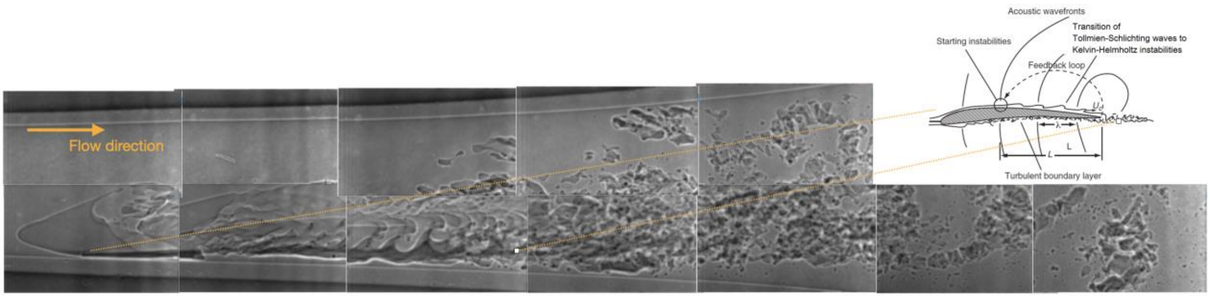

Figure 1 The stochastic dynamics in Venturi tube filmed with MHz X-ray microscopy

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Dr Jin Wang, Qing Zhang, Ya Gao, Pice Chen, Miaoqi Chu, Qinteng Zhang

Advanced Photon Source, Argonne National Laboratory, 9700 S. Cass Ave., Lemont, IL 60439

Precision Structure Tracking for Understanding the Interplay of Hydro- and Thermodynamic Parameters in Ultrafast Multiphase Micro-Sized Flows [1]

Liquid microjets have found industrial, commercial, and technological applications such as machining, cooling, printing, and additive manufacturing. In internal combustion engines, high-pressure liquid-fuel injection plays the most crucial role in the energy conversion process to improve combustion efficiency and emission. To interrogate the complex turbulent and multiphase flows, we developed an ultrafast X-ray near-field speckle imaging method ideally suitable for visualizing the multiphase turbulent and cavitating flows emanating from direct-injection nozzles

[1] This research used resources of the Advanced Photon Source, a U.S. Department of Energy (DOE) Office of Science User Facility operated for the DOE Office of Science by Argonne National Laboratory under Contract No. DE-AC02-06CH11357.

with unprecedented spatiotemporal resolution. The ultrafast liquid-fuel dynamics are dominated by injection pressure as well as fuel temperature through cavitation, an important thermodynamic parameter but often difficult to control in engine combustion. The fast near-nozzle dynamics, measured in wide-range operating conditions and with a realistic injection nozzle, are sensitive to the hydrodynamic parameters, such as injection pressure, and surprisingly, their interplay with the fluid temperature, an important thermodynamic parameter. With the most direct and quantitative measurement, we discovered that the near-nozzle fuel-jet dynamics could be perfectly scaled by a single dimensionless parameter, cavitation number [1]. This universal scaling shows that cavitation can be harnessed to elevate the pneumatic-hydraulic to kinetic energy conversion efficiency, which is critical for promoting fuel atomization and engine combustion performance. This enhancement effect will have even more impact on engine combustion using alternative low-emission fuels with higher saturated vapor pressure. Conversely, the X-ray imaging based high-precision structure tracking method is also ideally suitable for understanding the turbulence in the multiphase flows by quantifying turbulence fluctuation and intensity. The results bring greater impetus to developing realistic models and simulations of multiphase turbulent and cavitating flows to understand how they affect atomization prior to combustion processes.

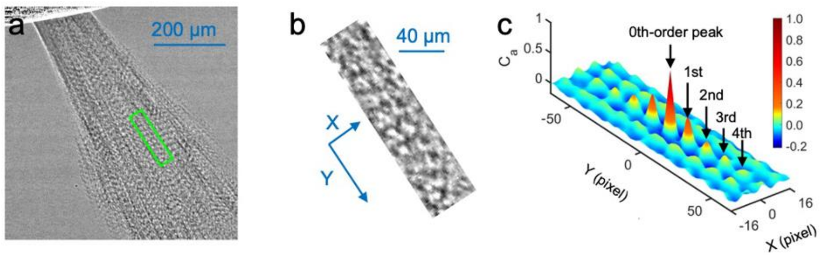

***Figure 1**. Autocorrelation of ultrafast multi-exposure x-ray near-field speckle a) multi-exposure spray image, b) Region-of-interest (ROI) selected, c) 3D auto-correlation map in the ROI up to the 5th order for determining high-precision velocity.*

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Prof Gretar Tryggvason

Department Head of Mechanical Engineering and Charles A. Miller Jr. Distinguished Professor, Johns Hopkins University, US

Simulations and characterization of complex gas-liquid flows

While bubbly two-phase flows have been studied extensively, they occur only for a relatively limited void fraction range. For high void fractions, particularly if the flow is turbulent, we expect to see a complex dynamic interface whose topology changes repeatedly as fluid masses coalesce and break apart. We present recent simulations of such flows and discuss the challenges of finding the appropriate statistical descriptions. Approaches drawn from studies of heterogeneous solids, rheology, and premixed combustion provide some guidance, but do not cover the full complexities of a dynamic interface and fluid turbulence. In order to generate coarse or reduced order models it is important to determine what such model looks like and how fully resolved results can be coarsened in order to both inform and validate coarse models. We present coarsening strategies where the flow is simplified by diffusion but a sharp interface is retained, as well as skeletonization to form simplified liquid structures in a formal way. Examples are drawn from recent simulations of complex turbulent buoyant two fluid flows in vertical channels, undergoing massive topology changes, and the breakup of unstable interface. We end by discussing preliminary efforts to construct coarse models based on these coarsening approaches and speculate about the general nature of such models.

Prof Stefan Hickel

TU Delft, Netherlands

Turbulence resolving simulation of fuel mixing and combustion with accurate multiphase-thermodynamics models for transcritical pressures

We will present novel physical and numerical models for the large-eddy simulation (LES) of inert and chemically reacting multiphase flows at very high operating pressures, with particular attention to highly non-ideal fluid states and other high-pressure effects. This comprehensive real-fluid multiphase thermodynamics framework represents supercritical fluid states as well as condensation and evaporation at transcritical multi-phase fluid states very high accuracy and unprecedented computational efficiency through a new family of fast and robust vapor–liquid equilibrium calculation methods. The new methodologies also include real-gas transport and chemistry models that consistently account for real-gas properties and multiphase states, combined with fast time integration methods for detailed, reduced and global mechanisms. We will give a rationale for our modelling choices and discuss selected applications.

Dr Outi Tamisola

KTH, Sweden

Direct numerical simulation of non-Newtonian two-phase flows

Many multiphase flows in everyday life have non-Newtonian carrier fluids, both in various industries (food, process, construction, chemical) and in nature (human body, natural disasters). In this talk, we present how non-Newtonian constitutive equations can be efficiently included in direct numerical simulation algorithms of turbulent and laminar multiphase flows. We will then present simulation results for non-Newtonian turbulent flows, such as a turbulent bubbly flow with and without surfactants. Non-Newtonian effects are crucial to be able to determine the drag and bubble behaviour.

Prof Jos´e M. Gordillo

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On the fast, inertial jets, ejected after the implosion of cavities

The fast, axisymmetric jets, observed after the collapse of a cavity can be described as a two-stage process: indeed, the pressure jump existing between the atmospheric and the vapor pressures for the case of cavitating bubbles, or the pressure difference established by capillarity or by gravity for the case of bursting bubbles, initially accelerates the liquid inwards towards the axis of symmetry, inducing a far field flow rate per unit length, $Q_\infty \propto VL$ with V and L a characteristic radial velocity and L a characteristic length. The second stage, during which the fast jet is ejected, is driven by a purely inertial process which ensures that the value of the flow rate per unit length induced during the acceleration stage remains constant in time while viscous, capillary or gravitational forces can be neglected in a first approach. Then, making use of theory and of numerical simulations, we [1, 2] first analyze the case of a conical bubble with a half-opening angle β when the value of the far-field dimensionless flow rate per unit length q_∞ is fixed to a constant, finding that this type of jets converge towards a purely inertial β-dependent self-similar solution of the Laplace and Euler-Bernoulli equations in which the jet width and velocity are respectively given, in the limit $\beta \ll 1$, by $r_{jet} \approx 2.25 \tan\beta\sqrt{q_\infty \tau}$ and $v_{jet} \approx 3q_\infty/(2\tan\beta\sqrt{q_\infty\tau})$ respectively, with τ indicating the dimensionless time after the jet is ejected. For the case of parabolic cavities with a dimensionless radius of curvature at the plane of symmetry r_c, our theory predicts that $r_{jet} \propto (2r_c)^{-1/2}(q_\infty\tau)^{3/4}$ and $v_{jet} \propto q_\infty (2r_c)^{1/2}(q_\infty\tau)^{-3/4}$, a result which is also in good agreement with numerical simulations. Our physical description is further confirmed once the theoretical predictions are compared with the results of numerical simulations describing the ejection of the jets produced when a bubble bursts at a free interface: indeed, our results reveal that the scalings for the jet width and velocity are given by $r_{jet} \propto \sqrt{q_\infty\tau}$ and $v_{jet} \propto \sqrt{q_\infty/\tau}$, a result which notably differs from the common belief that the jet width and velocity follow the inertio-capillary scaling $r_{jet} \propto \tau^{2/3}$ and $v_{jet} \propto \tau^{-1/3}$ [3]. In fact, our description reproduces the time evolution of the jet width and velocity for over three decades in time, obtaining good agreement with numerical simulations from the instant of the inception of the bubble-bursting jet until the jet width is comparable to that of the initial bubble. The present results might also find applications in the description of the very fast jets, with velocities reaching up to 1000 m s^{-1} when a bubble cavitates near a wall for small values of the stand-off parameter [4, 5], and in the quantification of the so-called bazooka effect.

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<u>Prof Christine Contino-Pépin</u>[1], Stéphane Desgranges[1], Charlotte Berard[2], Noé Dumas[2], Anthony Novell[3], Benoit Larrat[4], Florian Correard[2], Nicolas Taulier[5], Marie-Anne Estève[2]

Ultrasound-activatable perfluorocarbon nanodroplets and resulting formulations: Potential for brain drug delivery

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[2] Institut de NeuroPhysiopathologie (INP), CNRS UMR7051, AMU, Marseille, France
[3] BioMaps, CEA, CNRS, Inserm, Université Paris-Saclay, Orsay, France
[4] NeuroSpin / BAOBAB, CEA, CNRS, Université Paris Saclay, Gif-sur-Yvette, France
[5] Equipe LIB, CNRS UMR 7371 – INSERM U1146, Sorbonne Université, Paris, France

Besides their well-known and wide use in diagnostics, the therapeutic use of ultrasounds has recently emerged.[1] In this field, perfluorocarbon (PFC) emulsion nanoparticles are increasingly investigated as ultrasound (US) contrast agents and ultrasonically enhanced drug delivery vehicles.[2] Within this framework, our team has been working for several years on the production of stable perfluorocarbon droplets optimized for both early detection of tumor development and controlled therapy. These "theranostic tools" consist of perfluorooctyl bromide (PFOB) droplets stabilized and dispersed in water thanks to a shell resulting from the self-assembling of tailor-made fluorinated surfactants called “F-TAC” [3] and “Dendri-TAC”.[4] Due to the fluorophilic property of perfluorocarbons, it is not possible to encapsulate any drug, even hydrophobic, within the droplet core. To do so, we used a mixture of PFOB/biocompatible oil in different ratios to prepare our nanoemulsions (NEs). Playing on several parameters we produced nanodroplets with an interesting mean diameter (Do < 80 nm) for medicinal applications. We also succeeded in limiting the nanodroplets growth by a freeze-drying step, affording dry formulations of PFOB.[4] Once optimized, a fluorescent dye was encapsulated onto the PFC/oil nanodroplets in order to allow their in vitro monitoring and visualization of tumor accumulation after intravenous injection in mice. In a recent project (*BubDrop4Glio project*, Inca Plan Cancer) we have demonstrated the ability of these nanoformulations to cross the Bood-Brain Barrier (BBB) after ultrasound-assisted opening (Fig.1).[5] This presentation will cover all the NEs optimization, drug or dye encapsulation and biological validation (*in vitro* and *in vivo* studies) of these new ultrasound-sensitive nanodroplets and their potential for brain drug delivery.

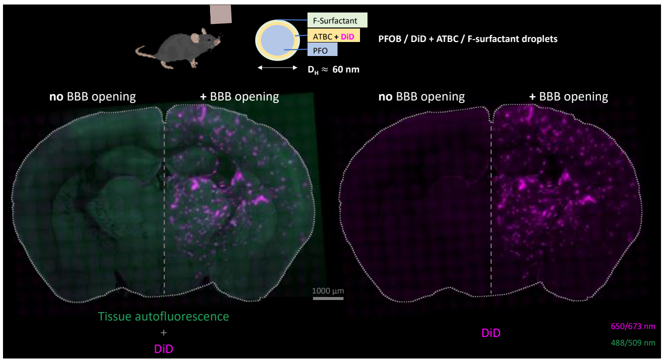

Figure 1. Fluorescence microscopy of mouse brain after i.v. injection of DiD-labelled droplets

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Prof Nicola Taulier
University of Sorbonne, France

Ultrasound Induced Vaporization of Perfluorohexane Droplets and Perfluorohexane/Water Droplets

Evidence of a heterogeneous nucleation Acoustic droplet vaporization (ADV) is a mechanism where a liquid perfluorocarbon droplet undergo a phase transition and transform into microbubbles when triggered by ultrasound of intensity beyond a critical threshold. ADV is still far to be understood as numerous experiments can not be correctly predicted by a model. I will show measurements of acoustic pressure threshold at which the vaporization of liquid perfluorohexane (PFH) occurs for three systems produced by microfluidics: plain PFH droplets, PFH droplets containing many nanometric water droplets and droplets made of a PFH corona encapsulating a single micrometric water droplet. For all the three systems, the probability to observe a vaporization event was measured as a function of pressure. Since our experiments were performed on solutions of droplets, we developed a statistical model to extrapolate, from our experimental curves, the ADV pressure thresholds in the case where only one droplet would be insonified. We observed that the value of ADV pressure threshold decreases as the radius of the plain PFH droplets increases. This value was further reduced when PFH droplets encapsulate a micrometric water droplet, while the encapsulation of many nanometric water droplets did not modify the threshold. We demonstrated that this behavior is not due to superharmonic focusing nor homogeneous nucleation. However, it can be explained by heterogeneous nucleation when using a model developed on the assumption that the surface, on which the nucleus appears, is soft. This new model correctly predicts the variation of ADV pressure threshold for plain PFH droplets when their radius decreases. In addition, this model shows that vaporization is due to a nucleus appearing on the internal surface when the encapsulated water droplet radius is micrometric, but on the external surface for nanometric encapsulated water droplets.