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Project Description - Project Proposals

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Switchable drops on laser-structured substrates

Project Description

1 State of the art and preliminary work

Dynamics of water drops has been intensively studied during the last century on different materials: from natural and artificial porous clothing surfaces (to avoid the falling rain drops to penetrate the structure) [1-4] to heterogeneous/chemically patterned substrates [5-14], and, more recently, on gradient and soft surfaces [15-18]. The main aim is to create at the liquid-air, liquid-solid interfaces the physico-chemical conditions in such a way that, tending to minimize its surface energy, the droplet experiences a well-defined, controlled motion. This droplet manipulation has large applications in micro-fluidic and, eventually, nano-fluidic devices without power supply, laboratory-on-a-chip, surface cleaning or ink-jet printing technologies.

Recent developments in areas like microelectronics or 3D printing have demonstrated a pressing need to understand cases in which (de)wetting hydrodynamics and substrate dynamics are strongly coupled. The Priority Programme 2171 aims at establishing a deeper understanding of the fundamental physics behind the dynamic (de)wetting of flexible, adaptive, and switchable substrates, combining experimental and theoretical perspectives. Dependence of the wetting characteristics on surface roughness and surface morphology is inherent in everyday life. In particularly, they can strongly influence the characteristic time scales of fluid flow in thin films during spinoidal dewetting and of spreading drops [19, 20]. Thus, the control of surface roughness and morphology would allow regulated motion of small portions of liquid (or drops) on structured substrates.

Different techniques of surface treatment can be utilized to modify the roughness and the morphology of the substrate. Among them are well established methods of chemical or plasma treatments, but also novel approaches based on surface structuring by high energetic beams, are applied. We suggest focused femtosecond laser pulses to prepare individually micro-/nanostructured substrates for the control of wettability [21-23]. The method is universal for structuring of any solids (including dielectrics, semiconductors, and metals) [24, 25], contactless, and non-toxic. In comparison to other techniques, by femtosecond laser structuring we can achieve very high spatial and temporal resolution, precise control over experimental parameters, high reproducibility of results, minimal thermal modifications and a high degree of chemical purity at the structured area. The laser-induced surface morphologies range from very regular periodic structures of a few hundred nanometers to complex multiscale patterns with a feature size about a few tenth of micrometers depending on the applied irradiation dose [26, 27]. Moreover, due to the Gaussian intensity profile the feature size of laser-induced structures can be varied on the scale of the focused laser beam about of 100 μ m.

In the frame of the Priority Programme 2171, we plan to study systematically - through a combination of experiment, theory, and numerics switchable drop behavior on laser-structured substrates. We will examine the dynamics of water/alcohol drops on laser-nanostructured micro-channels, and investigate the mechanism of controlled coalescence over gradient surfaces. The periods and the amplitude of laser-induced periodic surface structures (LIPSS, ripples) generated at the bottom of micro-channels will be checked as control parameters for the switchable drop behavior. Then, as external key factors for the switching



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behavior, we will investigate the role of the temperature gradient, electric and light-fields. As theoretical tool, a phase field approach will be used. Computer simulations will support the experiments from this project as well as from the other experimental SPP 2171-groups.

Lehrstuhl Experimentalphysik und Funktionale Materialien, BTU-Cottbus-Senftenberg (LEP):

The irradiation of solid surfaces with intense ultra-short laser pulses at fluence around the ablation threshold leads to a modification of surface morphology and to the formation of laser-induced periodic surface structures (LIPSS, ripples) with feature size in the range between a few 100 nanometers and a few 10 micrometers [21-23, V_et_al_17, VBR13]. Though, in principle, any material can be processed to show similar surface morphologies, we focus our research on metals and silicon targets.

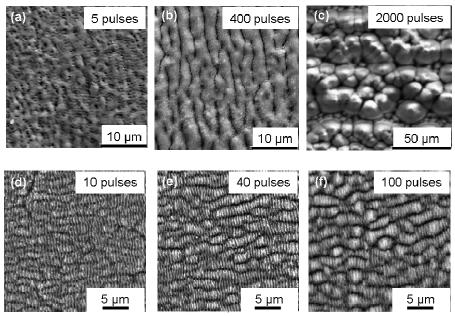


Figure 1: SEM images illustrate dose dependence of LIPSS: (a)-(c) silicon irradiated at1.12J/cm² [V_et_al_17], and (d)-(f) stainless steel irradiated at 1.50 J/cm².

Stainless steel is a commonly used metal in various technological applications. As all metals, it may be conveniently textured in large, homogeneous areas that offer a great potential for modification and control of such surface properties as wettability, tribology, and more. A comprehensive review of the state of the art has been given by Vorobyev and Guo [21], presenting an abundant list of more than 220 references to the field. About silicon (Si), typical applications for LIPSS structured surfaces may range from silicon-based devices in photovoltaic and optoelectronic to novel biomedical applications [22, 24].

LIPSS structured surfaces and wettability

Though the physical mechanisms of LIPSS formation are still disputed between a lithography-like ablation by a modulated field distribution [25] and self-organized structure formation from a laser-driven thermodynamic surface instability [VRVB15], in all experiments a large variety of features and feature size has been observed, depending on irradiation fluence and/or number of pulses [23, 26, V_et_al_17, VBR13, R_et_al_18] (cf. Fig. 1).

A promising application of LIPSS is the possible control of wettability [21-23], derived from the similarity of the modified surface profile with examples in nature, e.g. the lotus leaf. In a large number of investigations (cf. [21, 22] and references therein) it has been shown that it is

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possible to either make a surface highly hydrophilic or to increase its hydrophobicity. Obviously, here multi-scale topologies play an important role [26], consisting of a superposition of nanostructures on larger periodicities (Fig. 2), which are readily formed by multiple-pulse irradiation [V_et_al_17, R_et_al_18].

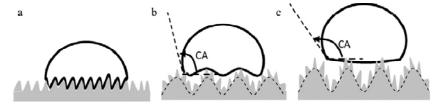


Figure 2: Scheme of a water drop on textured surface (image from [26]): (a) Wenzel model [1, 2]; (b) and (c) Cassie–Baxter model [3, 4].

Interestingly, the transition between hydrophobic to hydrophilic textures can be achieved by increasing either the local fluence (Fig.3) [27] or the number of pulses (Fig. 4). To the best of our knowledge, only surfaces with one or the other type of textures have been produced so far. In the present project, we will systematically study surfaces consisting of areas with different wettability.

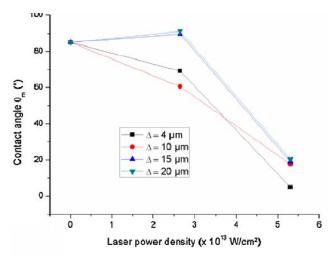
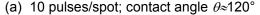
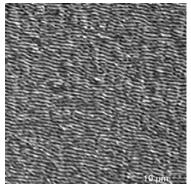


Figure 3: Transition from hydrophobic to hydrophilic behavior with increasing local fluence (the diagram from [27]).





(b) 200 pulses/spot; contact angle θ < 20°

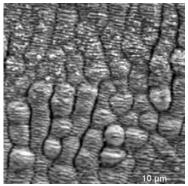


Figure 4: SEM images demonstrate dose/structure dependence of wettability (stainless steel; irradiation at 2 J/cm²). (a) 10 pulses/spot: \approx 600-nm nano-ripples, hydrophobic; (b) 200 pulses/spot: complex multi-scale pattern of \approx 600-nm nano ripples and \approx 3.5 µm grooves; hydrophilic [Varlamova and Sarker, unpublished results, 2015]

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Electrical properties of LIPSS structured silicon surfaces

The AFM micrograph in Fig. 5(a) with the corresponding profile line on the right side of the figure ((c)*Topography*), presents the topography of laser-structured p-Si targets. The ablation patterns produced with circularly polarized light (1.1 TW/cm²×1000 pulses) consist of round nano-spheres with an average diameter ~130 nm and with the height between 20 and 40 nm. A map of the contact potential (CP) on the structured surface is presented in Fig. 5(b) with the corresponding profile line ((d) *CP difference*). A comparison between Figs. 5(a) and 5(b) shows the CP drop by about 50 mV within the nanospheres and around each sphere, one can see an increased CP compared to the substrate. Astonishingly, the contact potential difference on the structured surface is more than 250 mV (see scales in Figure 5(b) and in Figure 5(d) the CP difference).

The observed variation of CP can be caused by dopant segregation during the ablation process and phase transformation of the silicon [VCRR07].

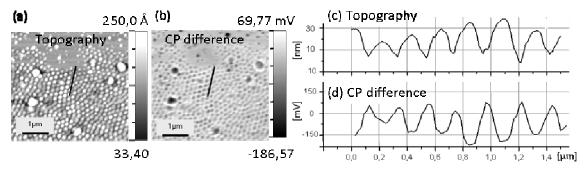


Figure 5: Scanning Probe Microscope micrographs from an ablated spot generated on p-Si (100) with 1000 pulses of circularly polarized light at 1,1 TW/cm² [VCRR07]. (a) Topography of the investigated area and (b) Distribution of the contact potential with corresponding cross-sections (c) and (d) of the areas indicated with thick black lines.

Novelty and excellence of the method: switching character of laser structured substrates

Despite numerous studies on laser-induced surface structuring and wetting behavior of laser-structured surfaces [21-24, 26, 27], these studies and applications can only be considered as a preliminary stage of the investigations intended in the present proposal. They are based on homogeneous structuring of the full surface. We start from the preliminary observations that

- (a) the detailed surface morphology is strongly dependent on the irradiation conditions, with an important influence of the irradiation dose [VBR13];
- (b) the wettability is considerably determined by the detailed features of the morphology, with the possibility of both an increase as well as a decrease of the contact angle [26];
- (c) the laser-induced modifications affect internal material properties, resulting, for instance, in the creation of nanoscaled potential patterns on the structured silicon surface [VCRR07].

In our project, we will produce substrates consisting of areas with heterogeneous wettability: macro and micro-sized hydrophilic channels or centers surrounded by hydrophobic boundaries or areas. We will systematically study dynamic behavior of liquid drops on these surfaces and investigate the mechanism of controlled coalescence over structured areas with a gradient wettability. The realization of wettability gradients in one step with a femtosecond laser, by making use of the fluence gradient inside the beam profile, is a real scientific and technologic challenge which would allow new applications to generate a complex functionalization of the material and a decrease of the duration of the surface treatment.

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Lehrstuhl Theoretische Physik II / Statistische Physik und Nichtlineare Dynamik, BTU-Cottbus-Senftenberg (LTP):

Our collaboration has been started with studies on the origin of laser-induced surface structures. Laser generated structures has been described in the frame of a self-organization model, resulting in a rich number of common publications, see for example, VRVB15 and R et al 18.

In our project, we investigate theoretically / numerically drops and thin liquid film on laser structured substrates. In this aim, phase field theories are very suitable. At LTP we have also developed phase field models already validated for various situations [28-34, 3B09, BB09, 35-39, BB14a,b, B-et-al17]. The phase field approach delivers a continuum thermodynamical model able to treat multi-phase problems. An auxiliary variable - the phase field- is added to the usual set of state variables in order to provide an explicit indication of the thermodynamics. This parameter may take different values for different phases and undergoes a rapid but smooth variation in the interface region. In mono-component systems the density ρ (scaled by the liquid density) is the natural order parameter: ρ =1 designates the liquid regions of the system and $\rho\approx0$ the gaseous one.

In the phase field model the interface is introduced by gradients of the phase field (see, e.g. Refs. 40-42). The formalism is based on a **free energy**:

$$F(\rho) = \int_{V} \left[f(\rho) + \frac{K}{2} \left(\frac{\partial \rho}{\partial z} \right)^{2} \right] dV$$
 (1)

where the first term represents the free energy for the homogeneous bulk phases. For a system in equilibrium and without interfacial mass exchange the free energy density has the form of a symmetric double-well potential with two minima corresponding to the two alternative phases. We take:

$$f(\rho) \sim \rho^2 (\rho - 1)^2. \tag{2}$$

The second term in (1) is a "gradient energy" which is a function of the local state. The specific interfacial free energy σ is, by definition, the difference between the actual free energy (per unit area)of the system and that which it would have if the properties of the phases were continuous throughout. Hence the free energy excess of the interface takes the form:

$$\sigma = \int_{-\infty}^{+\infty} K \left(\frac{\partial \rho}{\partial z} \right)^2 dz \tag{3}$$

which gives a direct connection between the surface tension coefficient σ and the gradient energy term $K\left(\frac{\partial\rho}{\partial z}\right)^2$ (where K is the square gradient parameter).

Being free of interface conditions, the phase field models are very attractive and effective to describe spatially and temporally varying interfaces with complicated geometries. They achieved considerable success in modelling solidification phenomena [43, 44], dendritic growths [45], two phase flow with variable density [46, 47], static contact angles, dynamic wetting [34, 3809, 48], coalescence of drops with different miscible liquids: fast-, delayed- and partial-coalescence [35-38, BB14a], and control drop motion under vibrations [BB14b].

The physical processes such as surface tension phenomena are incorporated in the Navier-Stokes equation with the help of the Korteweg stress tensor and substitutes into the phase field model the classical boundary condition for tangential stresses at the liquid -vapor interface. The components of the capillary tensor *T* are given by [40-43, 49, 50]:

$$T_{ij} = K \left(\rho \Delta \rho + \frac{1}{2} (\nabla \rho)^2 \right) \delta_{ij} - K \frac{\partial \rho}{\partial x_i} \frac{\partial \rho}{\partial x_j}$$
(4)

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where δ_{ij} is the Kronecker symbol. Thus, the extended Navier-Stokes (momentum) equations reads:

$$\rho\left(\frac{\partial \vec{v}}{\partial t} + (\vec{v} \cdot \nabla)\vec{v}\right) = -\nabla p + \nabla \cdot \underline{T} + \nabla \cdot (\eta \nabla \vec{v}) + \nabla\left(\frac{\eta}{3}\nabla \cdot \vec{v}\right) + \rho \vec{g}. \tag{5}$$

In equation (5), $p=\rho\frac{\partial f}{\partial\rho}-f$ denotes the thermodynamic pressure, η the dynamic viscosity, \vec{g} is the gravitational acceleration. The momentum equation is completed by the mass conservation equation:

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \vec{v}) = 0. \tag{6}$$

In earlier work we have studied the stability and break up of thin liquid films for several boundary values ρ_S (see Fig. 6). For a certain range of depths, the liquid is unstable and breaks up into numerous separated droplets. Via coarsening, in the late state of evolution only one single drop remains [34]. Coarsening is achieved by liquid transport along the precursor film and/or by evaporation and condensation. The contact angle of the remaining droplet is controlled through the density at the solid substrate ρ_S (Dirichlet condition). With this boundary condition one takes into account the associated van der Waals long-range interactions at the liquid-solid interface.

Pismen & Pomeau [42] assumed that for short-ranged solid-fluid interactions compared to the thickness of the diffuse interface, a supplementary energy term (a polynomial function of density) can be locally added in the free-energy functional in the vicinity of the wall. By minimizing this free-energy density, the (Dirichlet) boundary condition ρ_S has been obtained.

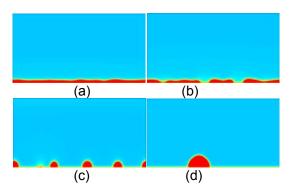


Figure 6: Transition in a thin unstable liquid layer under microgravity conditions: (a) t=950; (b) t=1400; (c) t=2800; (d) t=75000 ($h_0=40$, $\rho_S=0.5$, static contact angle $\theta_S=90^\circ$).

Thus, a flat solid surface with variable wettability can be realized by varying ρ_S in the range 0 < ρ_S <1 (partial wetting). The case ρ_S =0 describes non-wetting phenomena and for the case ρ_S ≥ 1 the unbalanced molecular forces produce a precursor liquid film which covers the solid surface (complete wetting). Note that all interfaces (liquid/gas,solid/gas, and solid/liquid) are diffuse due to the existence of the precursor.

We have numerically verified the relation found by Pismen and Pomeau [42] between the **contact angle** and ρ_S ,

$$\theta_{\rm S} = -1 + 6\rho_{\rm S}^2 - 4\rho_{\rm S}^3$$
.

We have also proposed a scheme for studying dynamic contact angles and drops running down on an inclined partially wetting substrate under gravity effects [34]. Figure 7 shows a **liquid droplet** of millimeter dimensions rolling down from left to right on a sloped solid support (α =30°). In these pictures the flow lines inside and around the drop refer to the comoving frame of reference. The drop loses reflection symmetry and two different contact angles occur. Contact angles were measured from phase field simulations using contour plots (level 0.5) of the phase field variable in which tangent lines are drawn at the macroscopic foot of the droplet.

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We found a very good agreement with the Voinov law [3B09] which claims the proportionality between the difference $\theta_a^3 - \theta_s^3$ (θ_a , θ_a is the advancing contact angle) and the drop velocity U along the inclined plane (Fig. 8). Preliminary simulations on chemically patterned substrates provide quite beautiful examples of pattern formation at the solid substrate, if the liquid film lying on the substrate becomes unstable. If the patterned surface contains a code (Fig. 9), the unstable liquid layer over the substrate can be used to decode this information [BB09].

Coalescence of two liquid drops with a body of different but perfectly miscible liquids shows interesting behaviour if the contact angle at the solid substrate is small enough. In this case the **coalescence** is assisted by **Marangoni** flows at the droplet interfaces, flowing from the drop with lower surface tension to the drop with higher surface tension. For small contact angles at the solid substrate, the lateral contact between the approaching droplets is realized through the precursor film at the solid substrate. The thin liquid channel connecting the droplets does not

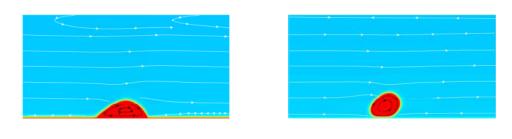


Figure 7: The drop formation on an inclined plane: full liquid-gas system with corresponding flow lines for (a) ρ_S =0.1, static contact angle θ_S =161° and, respectively, (b) ρ_S =0.7, static contact angle θ_S =55°.

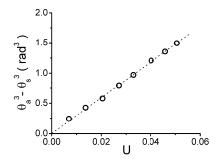


Figure 8: Code validation of Voinov law: $\theta_a^3 - \theta_s^3 \sim U$ (θ_a -the advancing contact angle, θ_s - the static contact angle, U- the drop velocity) [3B09].



Figure 9: 3D-Time evolution of a liquid on a patterned substrate found by the phase field method, from [BB09], illustrated through iso-density surface snapshots.

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allow a rapid mixing of the two miscible liquids, maintaining for longer time the gradient in surface tension between the droplet interfaces. The Marangoni flows can delay and also prevent droplet coalescence, a fact that allows motion control of tiny droplets/thin liquid films in microfluidic devices [35-39, BB14a, B et al 17].

Some 3D pictures illustrating the self-propulsion of twin sessile sub-millimetric drops by surface tension gradients at the droplet interface are depicted in Figure 10. The twin drops have the radius R = 4 X 10^{-4} m and the static contact angle at the solid substrate is θ_S = 15° [BB14a]. The shear flow acting along the droplet interfaces, from the droplet with lower surface tension (left) to the other one having higher surface tension deforms the fused drop and pushes it in the direction of the Marangoni force along the solid substrate (namely to the right in Figure 10). After mixing and homogenization (after t = 6 s), the surface tension gradient vanishes and the drop propulsion stops.

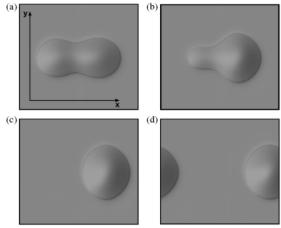


Figure 10: 3D-Time evolution of the coalescence of twin drops with different surface tensions: σ_1 = 0.05 N/m (left-side-droplet): σ_2 = 0.053 N/m (right-side-droplet) seen from above (into (x,y) plane). The snapshots correspond to: (a) t = 0.6 s, (b) t = 1.35 s; (c) t = 1.95 s and (d) t = 6 s [BB14a].

1.1 Project-related publications

1.1.1 Articles published by outlets with scientific quality assurance, book publications, and works accepted for publication but not yet published.

[V_et_al_17] O. Varlamova, K. Hoefner, M. Ratzke, J. Reif, D. Sarker, "Modification of surface properties of solids by femtosecond LIPSS writing: comparative studies on silicon and stainless steel", Applied Physics A **123**, 725 (2017).

[VBR13] O. Varlamova, M. Bounhalli, J. Reif, "Influence of irradiation dose on laser-induced surface nanostructures on silicon", Applied Surface Science **278**, 62 (2013).

[R_et_al_18] S. Razi, O. Varlamova, J. Reif, M. Bestehorn, S. Varlamov, M. Mollabashi, K. Madanipour, M. Ratzke, "Birth of periodic micro/nano structures on 316L stainless steel surface following femtosecond laser irradiation; single and multi scanning study", Optics and Laser Technology **104**, 8 (2018).

[VRVB15] O. Varlamova, J. Reif, S. Varlamov, M. Bestehorn, Chapter "Self-organized surface patterns originating from laser-induced instability" in Progress in Nonlinear Nano-Optics. Springer (2015).

[VCRR07] O. Varlamova, F. Costache, M. Ratzke, J. Reif, "Control parameters in pattern formation upon femtosecond laser ablation", Applied Surface Science **253**, 7932 (2007).

[B_et_al_17] R. Borcia, I. D. Borcia, M. Helbrig, M. Meier, Ch. Egbers, M. Bestehorn, "Dancing drops over vibrating substrates", European Physical Journal Special Topics **226**, 1297 (2017). [BB14a] R. Borcia, M. Bestehorn,"A phase field description of self-propulsion of twin sessile drops induced by surface tension gradients", Fluid Dynamics Research **46**, 041405 (2014). [BB14b] R. Borcia, I.D. Borcia, M. Bestehorn, "Can vibrations control drop motion?", Langmuir **30**, 14113 (2014).

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[3B09] R. Borcia, I. D. Borcia, M. Bestehorn, "Static and dynamic contact angles: A phase field modeling", European Physical Journal Special Topics **166**, 127 (2009). [BB09] R. Borcia, M. Bestehorn, "Controlled pattern formation in thin liquid layers", Langmuir **25**, 1919 (2009).

- 1.1.2 Other publications
- 1.1.3 Patents
- 1.1.3.1 **Pending**
- 1.1.3.2 Issued
- 2 Objectives and work programme
- 2.1 Anticipated total duration of the project

36 months

2.2 Objectives

The goal of our Joint Project is to study systematically - through experiment/ theory/ numerics - switchable drop behavior on laser-structured substrates. We will investigate the following open issues of great importance for the technological horizons of the material functionalization:

- A) Switchable drops on micro-channels controlled by morphological features of laser-induced patterns.
- B) Controlled drop coalescence on gradient substrates.
- C) Realization of a switching behavior on laser-structured substrates, by temperature gradients, electric, and light fields.

Both, experimental and theoretical/numerical methods will be applied and shall complement each other. Experimental work is provided by LEP group, theory and numerics will be performed by LTP group.

2.3 Work programme incl. proposed research methods

2.3.1 Surface structuring (LIPSS formation) (LEP)

General. All laser processing will be performed in air with an amplified Ti: Sapphire laser system (Ti³+:Al₂O₃), delivering pulses of ≈0.3 mJ energy at 1 kHz repetition rate and of ≈100 fs duration at full width at half maximum. The output beam with a diameter of about 5 mm is linear (horizontal) polarized and has a Gaussian spatial intensity profile and energy pulse-to-pulse stability < 3%. After a careful control of intensity and polarization, the beam is focused at normal incidence onto the front side of the target to a spot size about of 100 μ m diameter. The target is mounted on motorized computer controlled translation stages allowing high precision displacement in x- and y-directions. For each direction the target may be moved with a scanning/writing velocity from 0.1 to 100 mm/s, resulting in an effective pulse number (dwelling time) per spot from 1,000 to 1, correspondingly.

2-D structures (large area structuring) are written by placing parallel lines with adjustable overlapping pitch, thus allowing to vary the irradiation dose not only via the scanning speed but also via adjusting the pitch of adjacent lines (in y-direction) between 70% and 0% overlap.

After structuring the surface topography will be inspected ex-situ by surface sensitive microscopy techniques, such as Optical Microscopy (OM), Scanning Electron Microscopy (SEM), Scanning Force Microscopy (AFM, EFM), and Digital Holographic Microscopy (DHM).

2.3.1.1 Extended LIPSS areas

We start our experimental investigation with preparing/writing long (several millimeters) single lines on stainless steel and silicon targets by focused Gaussian laser pulses. Via variation of

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laser intensity and scanning velocity different types of surface morphologies can be generated in dependence on the applied irradiation dose, from high periodic linear sub-wavelength structures, known also as ripples, to hierarchical nano/micro-patterns, consisting of nanostructured micro-islands separated by deep microgrooves (cf. Fig. 1 and Fig. 4). We will systematically investigate the effect on the laser generated surface morphologies of multi-pulse irradiation, of applied fluence as well as of local intensity variation because of the Gaussian beam profile. It will be clarified whether already the dose variation across the beam profile allows creating hydrophobic/hydrophilic areas within a single ablation line/spot.

As next step, we perform the large area structuring on surfaces of investigated targets. Generally, 2-D structured areas present a regular arrangement of long channels covered with complex nano/micro-patterns in the center and linear periodic structures at the edge that reflect, of course, the Gaussian beam profile. A more intriguing situation we observed by increasing of the separation distance between adjacent lines. From preliminary experiments, formation of coherent, i.e. phase-locked LIPSS upon multi-pulse/spot irradiation occurred on silicon even if the modified spots barely overlap [51]. This effect is very pronounced on metallic surfaces. For uniform surface structuring we do not really need any overlap between adjacent lines, which may significantly minimize the processing time for large surface structuring. We will systematically study this phenomenon on stainless steel and on silicon targets and define the optimal separation distance, at which the interspace between adjacent lines is still filled with regular LIPSS.

All investigations performed on stainless steel will be carried out at the same manner on other metals.

2.3.1.2 Engineering of surfaces with heterogeneous wettability

Surfaces with heterogeneous wettability, the gradient surfaces, will be generated at various geometries. As first, we prepare a matrix of hydrophilic dots on a hydrophobic field and vice versa. As next, we produce hydrophilic macro- and micro-channels on the hydrophobic background.

Then, two approaches for the formation of hydrophilic channels or centers will be pursued:

- (a) Writing individual lines at lower speed/higher fluence than the main surface
- (b) Superimposing spots or lines on a homogeneously structured area by over-writing.

The goal is an optimal engineering of a controlled heterogeneity by determination of all impact parameters.

2.3.2 Surface morphology and wettability studies (LEP)

In parallel to the femtosecond laser surface structuring, we will investigate the substrate morphology of each of the fabricated surface by using optical, Atomic Force and Scanning Electron Microscopy. To measure the statistical roughness of structured areas, we analyze the surfaces with Digital Holographic Microscope. We quantify the periodicity and the feature size as well as the anisotropy of the generated patterns and decompose these morphologies at different scales as it has been described in reference [52]. Relevant topographic parameters at the relevant scale will be looked for to describe the morphology of the surface with regard to wettability behavior. Optimization of experimental parameters will be given from simulation.

We propose to study the wettability behavior of these structured surfaces by different methods:

- Determination of static and dynamic contact angles.
- Determination of hysteresis of contact angle (advanced and receded contact angle by increasing and decreasing volume of liquid method and inclined substrate method).

The objective is to determine the relations between the wettability behaviors, surface topography and the laser conditions used to elaborate these surfaces.

As LIPSS features and irradiation conditions are strongly correlated, a series of experiments is planned to optimize the structures in strong feedback with the theoretical simulations.

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2.3.3 Studies on switchable behaviour of the system (LEP+LTP)

The investigated system will consist of a laser-nanostructured surface (conducting and semiconducting substrates) and drops of a polar fluid (water, methanol, ethanol, and propanol).

Control of switchable behavior via surface topography

We start with numerical predictions for the experiments. Based on preliminary results, the theory/ numerics of the laser-structured surfaces will be elaborated on the phase field model presented in Sec. 1. The laser-structured surfaces will be modelled by assuming the substrate noisy around a given density ρ_S :

$$\rho(x,y)|_{z=0} = \rho_S + A\vartheta(x,y)$$

with ϑ a zero-mean noise in the interval $0 \le x \le x_1, 0 \le y \le y_1$:

$$\vartheta(x,y) = \frac{1}{\sqrt{M_0 N_0}} \sum_{m=-M_{0/2}, m \neq 0}^{M_0/2} \sum_{n=-N_{0/2}, n \neq 0}^{N_0/2} \exp(ik_m x) \exp(ik_n y) \exp(i\varphi_{m,n}), \tag{7}$$

$$k_m = \frac{2\pi}{x_1} \text{ m}$$
 , $k_n = \frac{2\pi}{y_1} \text{ n}$, $\varphi_{m,n} = -\varphi_{-m,-n}$

where φ is an uniformly random-distribution in the interval $[0, 2\pi]$, x_1, y_1 , are, respectively, the lateral and transversal lengths of the computational domain, and A denotes the noise amplitude. With this choice, the random distribution (7) has the **correlation length** for isotropic distributions at the solid substrate [55]

$$\delta = \frac{2x_1}{M_0} = \frac{2y_1}{N_0}$$

which can be used as control parameter in our simulations. For $M_0 \to \infty$, one obtains a Gaussian white noise. For example, Figures 11-a, b, c show 2D-patterns produced with (7) for three different correlation lengths.

Our goal is to study numerically the role of the correlation length, and of noise amplitude (roughness) on drop behavior on noisy surfaces. We will use Eqs. (5), (6) in numerical codes and we will implement the density at the solid substrate given by (7). The problem will be investigated in two and three spatial problems. The changes of wettability properties at the solid substrate (drop curvature) will be studied. The numerical results will be used as input for the parameter range in the experiment. Based on these results, we will experimentally examine the dynamics of water/alcohol drops on laser-nanostructured channels, and investigate the mechanism of controlled coalescence over gradient surfaces. The periods and the amplitude of laser-induced periodic surface structures generated at the bottom of channels as well as the statistical roughness of the structured area will be checked as control parameters for the switchable drop behavior. In turn, the experimental results will be applied to optimize the parameter used by numerics. The continuous exchange between LEP and LTP has been taken into account in our GANTT chart.

Control of switchable behavior via temperature gradient

We will investigate at LEP the effect of temperature gradient on the droplet dynamics by the controlled heating and cooling of the structured substrates. Switchable drop behavior induced by temperature fields will be studied as well at LTP.

For non-isothermal systems, the Marangoni effects created by the unbalanced surface forces at the free liquid interface and along the solid surface will be incorporated by assuming the square gradient parameter K (in Navier-Stokes equation (5)) and the density at the solid boundary ρ_S linear functions on temperature T. The temperature field T obeys the heat (energy) equation:

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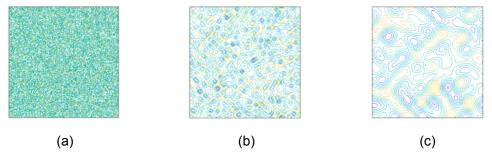


Figure 11: 2D-patterns (isotropic distributions) produced with (7) for three different correlation lengths: (a) $\delta = 2$; (b) $\delta = 10$; (c) $\delta = 40$.

$$\rho C_{v} \frac{dT}{dt} = \nabla \cdot (\varkappa \nabla T) \tag{8}$$

where C_v represents the heat capacity at constant volume of the liquid, and \varkappa its thermal conductivity.

Control of switchable behavior via external electrostatic and light fields

Laser-structured silicon surfaces (p/n doped Si (100)) reveal a spatial variation of the surface potential correlated with the surface morphology [VCRR07]. This effect is caused by dopant segregation during the ablation process and phase transformation of the silicon. By applying of an external electrostatic field, we will modify the contact potential on the structured surface and influence the behavior of water/alcohol drops.

In the case of laser-structured metals, we illuminate the structured area with a light source (a lamp or/and laser) to excite a surface-plasmon-polariton (SPP) field [53, 54]. The effect of the external electrostatic and the induced SPP field on the (de)wetting dynamics will be studied. Switchable drop behavior induced by electric fields will be studied at LTP by incorporating in the Navier-Stokes equation (5) the electrohydrodynamic (EHD) force exerted on a dielectric fluid (water, for example) per unit volume [56, 57]:

$$\vec{f}_e = \rho_e \vec{E} - \frac{1}{2} E^2 \nabla \epsilon + \nabla \left[\frac{1}{2} \rho \left(\frac{\partial \epsilon}{\partial \rho} \right)_T E^2 \right]$$

where \vec{E} is the electric field and E is its magnitude. The free electric charge density and the permittivity are denoted by ρ_e and ϵ , respectively. The permittivity ϵ varies linearly with the temperature, for high frequency of the applied field (much larger than the frequency of the charge relaxation) the free electric charge ρ_e do not appear.

In this theoretical frame, we will study not only the switchable drop behavior on LIPSS-substrates under electric fields but also the photoresponsive molecular switches of the arylazopyrazole (AAP), for the SPP2171-group Prof Bart Jan Ravoo, Westfälische Wilhelms-Universität Münster. The photoisomerization will be modelled as a change in the electrical field, reflecting the increased dipole moment of the cis-AAP in comparison with the trans-AAP.

2.3.4 Risk management (LEP)

As has been shown in previous sections, the LEP team has extensive expertise concerning of scientific and technical problems of the project. A temporary unavailability of the laser system can be overcome by subcontracting with neighbouring laboratories (BAM, MPI Berlin) or within SPP2171 collaboration partners (Prof. Evgeny Gurevich & Prof. Jeanette Hussong, Ruhr Universität Bochum and Prof. Frank Müller, Otto-Schott-Institut für Materialforschung, Jena Universität). Such a collaboration also gives us access to fs laser systems with different pulse durations/shapes and, thus, additional degrees of freedom that can be explored in the project. Another problem can concern the performance of wettability measurements. Here, we will cooperate with Prof. Stephane Vallette from Ecole Centrale de Lyon, LTDS, Lyon, France. The

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LTDS group has extensive knowledge and expertise as well as laboratory facilities to study wetting behavior even with very small drops.

During the model elaboration, experiments and results evaluation, the two joint groups will be in closed collaboration in order to discuss open questions/issues and to prepare common publications. The proposed topic will be worked out by both partners, as follows:

EXPERIMENT: O. Varlamova (LEP)

No	Objectives and estimated times	
01	Preparation of extended LIPSS areas with hydrophobic/hydrophilic wetting properties.	3 months
O2	Experimental studies on morphological features of the laser-induced patterns and their effect on the wettability of the structured substrate.	3 months
O3	Preparation of surfaces with heterogeneous wettability/Preparation of surfaces with wettability gradient (gradient surfaces).	6 months
04	Investigation of the geometry of gradient surfaces as controlling parameter for the switchable droplets behavior.	6 months
O5	Experiments on controlled coalescence on gradient surfaces. Study on drop motion on sloped gradient surfaces: realization of switchable drops on microchannels, for example, for different drop sizes or viscosities.	6 months
O6	Study of external key factors for the switching behavior: the role of the temperature gradient, electric E-and light-fields.	9 months
07	Comparison with the results from other SPP2171-groups and writing up the results.	3 months

THEORY/NUMERICS: R. Borcia (LTP)

No	Objectives and the estimated time	1					
R1	Development of the numerical code for 3 months noisy substrates. Tests in two spatial dimensions						
R2	Study of the role of the correlation length and amplitude of the roughness at the solid substrate. Investigation of different droplet behaviors in 2D on sloped surfaces	id					
R3	Extension in three spatial dimensions. Creating of noisy stripes with different correlation lengths. Statistical studies on drop motion on sloped noisy striped substrates: To realize switchable drops on microchannels, for example, for different drop sizes or viscosities	6 months					
R4	Importance of the multiscale for the functionalization of the surface	6 months					

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R5	Controlled coalescence on sloped gradient patterned surfaces	6 months
R6	Influence of temperature and electric fields on switchable behavior of drops on noisy substrates	6 months
R7	Comparison with the results from other SPP2171-groupsand writing up the results	3 months

GANTT chart

	1st year				2nd year			3rd year				
Months	1-3	4-6	7-9	10-12	13-15	16-18	19-21	22-24	25-27	28-30	31-33	34-36
Research	01	O2	03			04		O5+R3		06+R	6 (07+R7
Objectives	R	R1	R2	R3		R4			R5	00+K	0	77 TK7

Parallel to the objective described above, continuously we will be network with the groups presented in Sec. 2.5.

2.4 Data handling

We plan peer-reviewed publications in high-level journal articles and conference books from the workshops, schools and International Conference organized within our SPP. In order to improve the transparency and the scientific result circulation, we will be oriented to open access publications. The measured data will be secured on the department servers with daily backup.

2.5 Other information (Networking within SPP2171)

Please use this section for any additional information you feel is relevant which has not been provided elsewhere.

Within SPP2171 we will deliver also numerical simulations to the LIPSS experimental group Prof Frank Müller, Otto-Schott-Institut für Materialforschung, Jena Universität which are interested in studying molding phenomena.

We will perform phase field simulations for the experimental group Prof Bart Jan Ravoo, Westfälische Wilhelms-Universität Münster and we will investigate the dynamics of a thin liquid film on chemically patterned switchable substrates, *i.e.* photoresponsive molecular switches of the arylazopyrazole (AAP). The photoisomerization will be modelled as a change in the electrical field, reflecting the increased dipole moment of the cis-AAP in comparison with the trans-AAP

We will validate our phase field simulations with the help of the numerical simulations given by the particle models performed in the group Prof Thomas Speck, Johannes Gutenberg-Universität Mainz.

A benchmark study will be done on droplet impact on a hard substrate covered by a thin liquid film, project-part of Dr Gregory Lecrivain, Helmholtz-Zentrum Dresden-Rossendorf, Dresden.

We will also offer support for the numerical validation / verification for the group Prof Tatiana Gambaryan-Roisman, Institute of Technical Thermodynamics and Center of Smart Interfaces, Technical University Darmstadt for droplet spreading on structured substrates and for the group PD Dr Svetlana Gurevich & Prof Andreas Heuer, Westfälische Wilhelms-Universität Münster, which are using kinetic Monte Carlo and Molecular Dynamics simulation to study dynamical behavior of liquids on chemically structured surfaces. For the group PD Dr Svetlana Gurevich & Prof Andreas Heuer we will be prepare laser structured substrates as well.

Wetting of surfaces can be influenced by LIPSS, which lamellae angle is controlled by magnetic field. We plan to test for the group Prof Evgeny Gurevich & Prof Jeanette Hussong, Ruhr Universität Bochum whether it is possible to make the LIPSS in magnetic nanocomposites by direct laser processing.

To complement our experimental facilities with other femtosecond laser sources (shorter pulses, higher fluences, etc.), we plan intensive collaborations with the research group Prof Evgeny

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Gurevich & Prof Jeanette Hussong, Ruhr Universität Bochum and with the LIPSS experimental group Prof Frank Müller, Otto-Schott-Institut für Materialforschung, Jena Universität.

2.6 Descriptions of proposed investigations involving experiments on humans, human materials or animals as well as dual use research of concern

Not applicable

2.7 Information on scientific and financial involvement of international cooperation partners

We will organize scientific discussions for clarification of open issues, exchange of data and scientific results, short visits and, of course, we will participate in the SPP workshops/meetings.

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4 Requested modules/funds

Explain each item for each applicant (stating last name, first name).

4.1 Basic Module

4.1.1 Funding for Staff

LEP: We apply for one postdoctoral position - Temporary Position for Principal Investigator (EG 13 TV-L, 100 %) - and for one student position (Hiwistelle) for 3 years. Dr Olga Varlamova in close collaboration with her younger colleague will be employed in the project for the whole application period, being responsible for the points outlined in Sec. 2.3 (objectives O1-O7).

LTP: We apply for one postdoctoral position - Temporary Position for Principal Investigator (EG 13 TV-L, 100 %) - for 3 years. Dr Rodica Borcia will be employed in the project for the whole application period, being responsible for the points outlined in Sec. 2.3 (objectives R1-R7).

4.1.2 Direct Project Costs

4.1.2.1 Equipment up to €10,000, Software and Consumables

Requested amount: € 21,000 (7,000 p.a.) For the experiments, consumables like

samples/targets2000,00€polarization optics1000,00€ultrafast optics (mirrors, lenses)500,00€mount, optomechanical components1000,00€AFM tips2000,00€SEM filaments500,00€

will be yearly required.

4.1.2.2 Travel Expenses

Requested amount: € 21,600 (€ 7200 p.a.). 2 international conference (one week) for 2 persons, 4000€/year SPP-meetings (4-5 days) for 2 persons, 1200€/year DFG form 53.01– 03/18 page 18 of 20

Research visits of the SPP or/and international collaborators: 2000€/year

Within the SPP2171, we plan intensive collaborations with the research group Prof Evgeny Gurevich & Prof. Jeanette Hussong, Ruhr Universität Bochum and with the LIPSS experimental group Prof Frank Müller, Otto-Schott-Institut für Materialforschung, Jena Universität. This requires several research stays at Bochum/Jena for Dr O. Varlamova (each of about one weeks duration). This requires several research stays at Bochum/Jena for Dr O. Varlamova (each of about one weeks duration).

Total 4.1.2: 42600€

- **4.1.2.3 Visiting Researchers**, (excluding Mercator Fellows)
- 4.1.2.4 Expenses for Laboratory Animals
- 4.1.2.5 Other Costs
- 4.1.2.6 Project-related publication expenses

The results of the project will be published in open access journals: 1000€/year

- 4.1.3 Instrumentation
- 4.1.3.1 Equipment exceeding Euro 10,000
- 4.1.3.2 Major Instrumentation exceeding Euro 50,000
- 4.2 Module Temporary Position for Funding
- 4.3 Module Replacement Funding
- 4.4 Module Temporary Clinician Substitute
- 4.5 Module Mercator Fellows
- 4.6 Module Workshop Funding
- 4.7 Module Public Relations Funding
- 5 Project requirements
- 5.1 Employment status information

Dr. rer. nat. Olga Varlamova is postdoctoral researcher at Brandenburg University of Technology Cottbus-Senftenberg, Germany. With expertise in laser processing and functionalization of solid surfaces by using of femtosecond laser pulses and controllable variation of wetting properties of solids on micro/nanoscale.

<u>Scientific output</u>: 28 peer-reviewed articles and more than 20 contributions at national (DPG Tagung) and international conferences (EMRS, PIERS, Nano SEA, GADEST, IWAN, IMA) and workshops. h-index: 13. Reviewer for the following journals: Applied Surface Science, Applied Physics A, Optics and Laser Technology, Optics and Lasers in Engineering, Journal of Materials Processing Technology, Chinese Optics Letters.

PD Dr. rer. nat. habil. Rodica Borcia is Privatdozent at Brandenburg University of Technology Cottbus-Senftenberg, Germany. With expertise in mathematical physics, waves and instabilities, plasma physics, fluid dynamics, Dr Rodica Borcia has developed in the last decade a significant research on phase-field modelling in two-phase-systems. In addition the elaboration of the theoretical models into a thermodynamical description by using phase field models, the elaboration of the numerical codes and the performance of computer simulations were

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important tasks of her research for describing Marangoni convection in two-layer-systems, drops and bubbles, contact line dynamics, thin liquid films, coating and wetting phenomena, spreading and coalescence, controlled motion under vibrations, problems occurring everywhere in daily life in a variety of microfluidic applications.

<u>Scientific output</u>: 42 peer-reviewed articles, 5 invited lectures, h-index: 10, Chairman of IMA-10 - 10th Conference of the International Marangoni Association Interfacial Fluid Dynamics and Processes, June 2020, Iasi, Romania.

For more details see the attached CVs for each applicant.

5.2 First-time proposal data

Not applicable

5.3 Composition of the project group

List only those individuals who will work on the project but will not be paid out of the project funds. State each person's name, academic title, employment status, and type of funding.

LEP:

- -Prof. Jürgen Reif, University Professor, BTU Cottbus-Senftenberg, Cottbus, Germany (retired since April 2018)
- -Juergen Bertram, Technician, BTU Cottbus-Senftenberg, permanent position, university funding
- -Prof. Inga Anita Fischer, University Professor, Department of Experimental Physics and Functional Materials, BTU Cottbus-Senftenberg, permanent position, university funding

LTP:

- -Prof. Michael Bestehorn, University Professor, Department Statistical Physics/Nonlinear Dynamics, BTU Cottbus-Senftenberg, Cottbus, Germany, permanent position, university funding
- -Sebastian Richter, PhD student, university funding
- -Tillmann Rosenow, PhD student, university funding

5.4 Cooperation with other researchers

5.4.1 Researchers with whom you have agreed to cooperate on this project (outside the collaborations within SPP)

Prof. Alexander Oron

Faculty of Mechanical Engineering, Technion Israel Institute of Technology, Haifa, Israel

Prof. Stephane Vallette

Ecole Centrale de Lyon, Laboratoire de Tribologie et Dynamique des Systemes, Lyon, France

Prof. Jürgen Reif

Department Experimental Physics: Materials Science

BTU Cottbus-Senftenberg, Cottbus, Germany (retired since April 2018)

Dr. Florenta Adriana Costache, Group leader "Smart Micro-Optics"

Fraunhofer Institute for Photonic Microsystems, Dresden, Germany

5.4.2 Researchers with whom you have collaborated scientifically within the past three years

Prof. Jürgen Reif

Department Experimental Physics: Materials Science

BTU Cottbus-Senftenberg, Cottbus, Germany (retired since April 2018)

Prof. Michael Bestehorn

Department Statistical Physics/Nonlinear Dynamics

BTU Cottbus-Senftenberg, Cottbus, Germany

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Prof. Harald Schenk
Chair Micro- and Nanosystems
BTU Cottbus-Senftenberg and
Fraunhofer Institute for Photonic Microsystems, ISS, Cottbus, Germany

Prof. Uwe Harlander
Department of Aerodynamics and Fluid Mechanics
BTU Cottbus-Senftenberg, Cottbus, Germany

Prof. Christoph Egbers
Department of Aerodynamics and Fluid Mechanics
BTU-Cottbus-Senftenberg, Cottbus, Germany

PD Dr. rer. nat. habil. Hans Riegler, Project Leader Max-Planck-Institute of Colloids and Interfaces Potsdam/Golm, Germany

Dr. Stefan Karpitschka, Project Leader Max-Planck-Institut für Dynamik und Selbstorganisation, Göttingen, Germany

Prof. Reiner Schmid
Enviroment and Natural Sciences
Arbeitsgebiet Leichtbaukeramik
BTU-Cottbus-Senftenberg, Cottbus, Germany

5.5 Scientific equipment

List larger instruments that will be available to you for the project. These may include large computer facilities if computing capacity will be needed.

LEP possesses at BTU Cottbus-Senftenberg:

- -Titan:Sapphire Laser with CPA (chirped pulse amplification)
- -3D micro-positioning system
- -Scanning probe microscope (AFM)
- -Scanning Electron Microscope (SEM)
- -Digital Holographic Microscope (collaboration with Prof. Harald Schenk)
- -Equipment to measure static and dynamic contact angle (collaboration with Prof. Reiner Schmid)

LTP possesses at BTU Cottbus - Senftenberg two Linux Cluster for performing the numerical simulations. As the state of Brandenburg is a member of the North-German Supercomputing Alliance (Norddeutscher Verbund zur Förderung des Hoch- und Höchstleistungsrechnens - HLRN), all members of the BTU Cottbus - Senftenberg have also access to the HLRN-III Cray System located in Berlin and Hannover. If the Linux Clusters are broken, could be used the access to the HLRN-III Cray System located in Berlin and Hannover (Cray XC30: 1488 nodes X 24 cores, Cray XC40: 2064 nodes X 24 cores). Very recently at LTP a new Linux Cluster has been achieved (for parallel computing) having 56 Processors 4 Hz, 128 GB RAM, 3 TB storage.

5.6 Project-relevant cooperation with commercial enterprises

If applicable, please note the EU guidelines on state aid or contact your research institution in this regard.

not applicable

5.7 Project-relevant participation in commercial enterprises

Information on connections between the project and the production branch of the enterprise

not applicable

6 Additional information