ERC Starting Grant 2020
Part B2[1]
(not evaluated in Step 1)

Section a. Context, State-of-the-art and objectives

Context: Mastering thermal transport at solid/liquid interfaces in a multiscale context becomes a major challenge for a plethora of engineering applications due to miniaturization of different components in nowadays systems. Specifically, it is increasingly important for both industrial and social needs, e.g., energy harvesting and conversion, cooling applications in datacenters and mobile devices, biomedical and chemical industries.

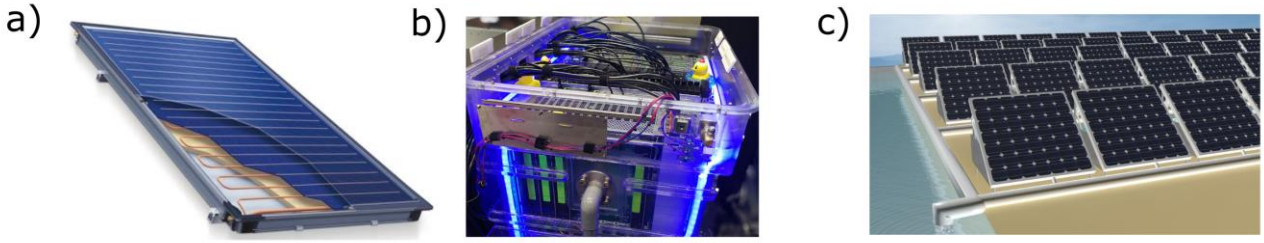

Fig. 1. Several examples where the solid/liquid thermal transport is crucial: a) solar collectors[1]; b) liquid cooling in datacenters[2]; c) combination of the solar cells with water purification systems[3]

As a part of the strategic challenges related to the climate change and the green energy transition, we need to convert and re-use energy sources more efficiently. Therefore, the next generation high technologies require much higher degrees of performance, sobriety, and compactness. For many of those technologies, heat and mass transport phenomena are often present. In this context, a mastering and tailoring interfacial thermal transport will surely overcome several issues for the improvement of the current technologies.

One of the convenient ways to enhance thermal transport between a solid and a liquid is based on nanostructurations[4] of the surface to increase interfacial area, and, respectively, surface to volume ratio. However, this approach has some limitations that are intently related to the presence of an interfacial boundary resistance between two different species. The latter can significantly reduce energy transport across the interface. Additionally, the presence of the nanostructured features at the interface induces mechanical stresses such as the bending of the interfacial region, and the impact of this curvature on the liquid properties can be crucial. Therefore, for improving energy exchanges, one needs to make a balance between different phenomena in order to optimize thermal transport across the interface. Historically, macroscale energy process optimization was an engineering issue. Now, it is clear, that this subtle design of the solid/liquid interface must be found based on a clear understanding of the interacting medium properties in vicinity of the interface that can be nanostructured.

From microscopic point of view, thermal transport across the solid/liquid interface is generally considered in frames of an interfacial thermal resistance, but such considerations could not address a significant enhancement of thermal transport properties of the systems with well-developed solid/liquid interfaces[5]. Up to now, the physical nature of this phenomena has not been studied in detail. However, a comprehensive understanding of such interaction is extremely important to master and tailor the transfer efficiency. This information is largely unexploited because we lack “clear pictures” of the mechanisms that explain heat carriers transfer across the interface. While, it will be necessary to keep on further examination of the impact of various parameters on the interfacial conductance, a breakthrough can only come from physical insight of the dynamical interaction of the phases that meet at the interface, which is the core of this proposal.

Thus, the main objective of the project is the development new pathways to control thermal transport close to the solid/liquid interface. Such program requires to proceed to the clear identification of the physical mechanisms that rule heat carriers transfer across the interface.

[1] Instructions for completing Part B2 can be found in the ‘*Information for Applicants to the Starting and Consolidator Grant 2020 Calls’*.

First, we briefly summarize the recent advances regarding to the thermal transport across the interface. Using this knowledge and improving it above the current state-of-the-art, the PI intends to provide answer the question – "**What are the original mechanisms that drive the flow of energy through multiphasic interfaces? Why are they happening? And finally, how can we control and enhance these mechanisms?**"

State of the art; Heat transfer in the absence of mass flow at the macroscopic level can be described with well-known Fourier law:

$$\vec{J} = -K\vec{\nabla}T$$

where $\vec{J}$ is the heat flux, K is the thermal conductivity, $\vec{\nabla}T$ is the temperature gradient. Its solution is the linear temperature profile between the point-like heat source and the heat sink in one dimensional case (see Fig. 2A).

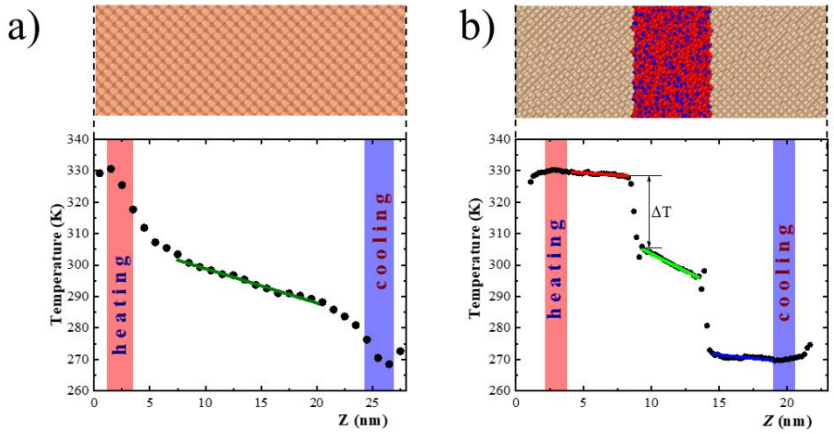

Fig. 2. An atomistic view of temperature profiles in a homogeneous material (left) and a three-layer "solid-fluid-solid" system (right) simulated by molecular dynamics. Thermal resistances to the heat flow clearly rise at the solid/liquid interface in the multilayer system.

In case of multiphase systems an additional temperature drop (ΔT) appears (see Fig. 2B) at the interface separating the two-phases media. This temperature drop is usually defined in terms of an interfacial thermal resistance (R) known as the Kapitza thermal resistance:

$$J_i = \frac{-\Delta T}{R},$$

where J_i is the heat flux across the interface with respect to its normal n_i. The magnitude of this resistance significantly influences the thermal transport in systems where there is an important surface-to-volume fraction. At the microscopic scale the value of the interfacial resistance depends on the local interactions between atoms of a solid and a fluid[6,7]. It was found well-pronounced dependence of the interfacial thermal resistance on the wetting angle[8,9] of the fluid in contact with the solid. However, if the latter aspect is simple to perceive a more detailed spectral analysis of the heat flow in the silicon-based systems with the solid/liquid interface has shown that the scaling law with wetting angle is not universal[10,11]. For instance, it was demonstrated that the features of thermal transport across the solid/liquid boundary at such scales is more likely depicted considering a liquid density depletion layer[12,13]. In frame of elastic-wave propagation theory (i.e. phonons at microscale), C. Ulises Gonzalez-Valle and Bladimir Ramos-Alvarado[12] showed that out-of-plane modes (perpendicular to the surface) of the substrate contribute more significantly than in-plane-modes (parallel to the surface) to the thermal transport across the interface. It was also shown the dependence of boundary resistance on the confined liquid slab thickness[14,15] and the curvature of the interface[16].

Only a limited number of studies are starting to consider microscopical features but mainly from the solid side[7,11,12]. However, it is of urgent need to develop additional cutting-edge approaches from both solid and liquid sides as well as new models to fully understand their interactions. In the current project, we will pay a significant attention to the impact of the specie's property modification arising at the contact of the solid and liquid interface. One of the examples, which can lead to such modifications, is the "solid-like" behavior of the adsorbed water nanofilm[17] particularly due to hydrogen-bond network appearing.

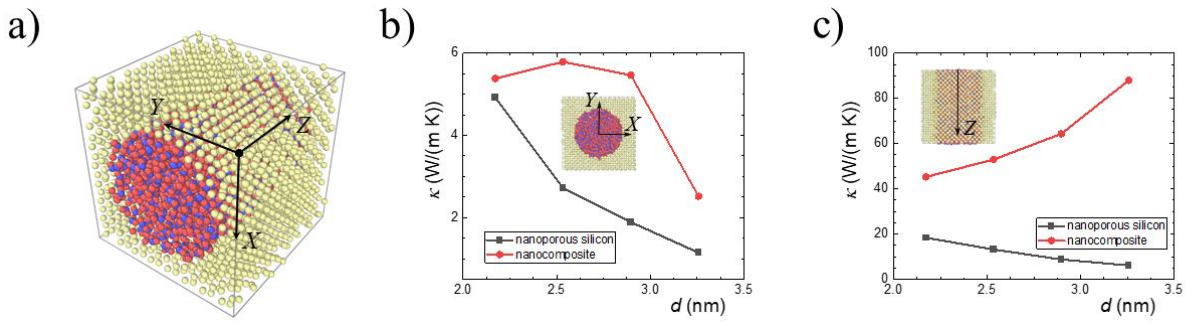

Fig. 3. a) an atomic configuration in the nanocomposite system "porous silicon with cylindrical pore filled by liquid"; thermal conductivity of empty porous silicon and nanocomposite system in the perpendicular (b) and in the parallel (c) directions of the cylinder axis calculated with molecular dynamics.

In this context, recent studies[18,19] show interesting results considering the anisotropy of thermal conductance of interfacial layer between a solid and a liquid. Similar work has independently been achieved by PI while considering the thermal transport in porous media with cylindrical morphology of the pores (see **Fig. 3**). Specifically, an outstanding enhancement of about 100% of thermal transport is observed along the solid/liquid interface. This gives additional degrees of freedom regarding the engineering of the interface in order to maximize/minimize thermal transport across the interface with the aim to achieve the largest efficiency for energy applications in actual systems.

Objectives: The above paragraph presents a short overview of the state-of-the-art. It also shows that significant efforts are paid for inductive observation. Yet, such approaches are not always relevant and may fail to explain unexpected heat transport behavior. For example, an interfacial boundary resistance by its nature reduces thermal transport, and the current microscopic studies could not address thermal transport enhancement in nanofluids where the adding of small quantities of nanoparticles lead to a significant (more than 20 % for 1-5 % volume fraction of nanoparticles) enhancement of pristine fluid thermal conductivity, and porous systems filled by liquids. The origins of such anomalous variations are unknown and represent one of the most important challenges to solve in ThermInAtom. With such misunderstandings of microscopical phenomena we totally lose any possibility of mastering the thermal transfer through interfaces just when it appears to be urgent.

The groundbreaking concept of ThermInAtom project is to understand microscopical mechanisms that characterize thermal transport across the liquid/solid interface. Those mechanisms are significant for paving new paths to control the interfacial thermal transport. We will go in deep for mining of the physical insight of the considered processes taken into account significantly different nature of thermal transport in solids and liquids that is essential for the descriptions of heat transfer across the interface. Specifically, the heat carriers in solids are elementary excitations – quasiparticles (such as phonons, vibrons, electrons, etc.) without mass transfer. On the other hand, in liquids thermal transport arise mainly due to mass transport via diffusion, convection, boiling. Therefore, a natural question is "What is the behavior of the interface?". Is it induced by the collective movements of atoms or by individual ones? To address these questions, we will firstly consider the modification of the bulk solid/liquid properties in vicinity to the interface, and we will further examine the response of the interface subject to external force source.

As stated above, the project is based on the precise microscopic considerations of the transfer processes to find previously unconsidered features of interface behavior. This will be done by considering numerous factors that can lead to a significant thermal conductivity variation at the interface. Among them, the main hypotheses that influence thermal transport close to the interface are: i) the presence of a liquid film on a solid substrate that improves specular scattering of phonons, and thus increases their mean free path[20]; ii) the solid/liquid interface that supports surface waves which propagates energy and contribute to thermal transfer; iii) the confined liquid close to the interface acts as "nanochannel" which increases the diffusivity of water molecules; iv) the adsorbed layer of a liquid on a solid substrate has significantly larger local thermal conductivity because of the crystal-like behaviour due to the local augmentation/depletion of the liquid density. We will investigate the contribution of all these mechanisms and others to the thermal transport. Besides, they should be considered simultaneously to capture all the physic.

A Junior ERC project is now the perfect instrument to move knowledge in this field into new directions. It will give us the possibility to perform investigations of such complex phenomena starting from the numerical simulations, moving through experimental measurements, toward to the prediction for "real-life applications". These successive steps require multidisciplinary skills on both theoretical and experimental aspects.

Considering the multidisciplinary approach that involves the study of liquid behavior at the nanoscale, solid state physics and nanosystems simulations are parts of innovative nature of the project. The microscopic description of the water dynamics together with material processing/characterization experience will give us the possibility to achieve the original and important results for improvement of heat transfer across/along the solid/liquid interface. Eventually, the enrichment of the simulations and theoretical analysis with the data provided by experiments, on different space and time scales, underlines the ambitions to improve existing models through multiple test/validation steps.

It is clear that for the project implementation we chose unconventional and uncharted paths. When state-of-the-art methodologies are mainly based on the finding of scaling laws using parameters (work of adhesion, wetting angle, depletion length, etc.) for heat transport modelling the ThermInAtom project aims to find the physical principles, in a general framework, to understand thermal transport across the solid/liquid interface. This route is based on the decomposition of the equimolar and surface of tension impact on the thermal transport that represents a new fundamental approach. Additionally, in this context the modification of the liquid and solid properties in a vicinity to the interface will be examined in both equilibrium and out-of-equilibrium approaches.

The main objectives of the project can be expressed as follows:

1) To establish a comprehensive understanding of the interface separating solid and liquid phases, and to find the contribution of the different surfaces on thermal transport.
2) To manipulate interfacial properties by variation the liquid/solid properties and their interactions in atomistic simulations.
3) To develop strategies to control the thermal transport across the solid/liquid interface by tuning of interfacial properties and morphology of the interface.
4) To develop new experimental techniques devoted to study the thermal transport properties across the solid/liquid interface based on the photothermal methods and scanning thermal microscopy.
5) To formulate new models describing the thermal transport across the solid/liquid interface based on the experimental results and results of simulations.
6) To design the optimal configuration of the solid/liquid interface based on the experimental and theoretical results to enhance thermal transport.

Achieving of these objectives is certainly challenging but doable and requires significant skills in the different domains such as materials research, thermal transport, liquids, and interfaces. Nevertheless, latest results and the multiple expertise in these various domains of PI make such challenges realistic. We have already obtained outstanding results in both experimental and simulation approaches. For example, we have shown the significant impact of the three-phase contact line on a droplet shape. We have proposed an efficient technique for the parametrisation of interaction potential due to the minimisation of the line tension impact[21]. We have also shown the significant impact of the adsorbed layer of liquid on the droplet volume, and the Gibbs adsorption impact on the droplet shape in external field based on the analytical modelling was stated[22]. The significant enhancement of the thermal conductivity of the porous silicon filled by the liquid compared to the empty one were both studied experimentally[23] and with the use of numerical simulation approaches[24]. Different tendencies in the size dependence of the surface tension of the water droplet for different morphology of the interface were found[25]. Recently, we made a breakthrough advance in experimentally developing of photoacoustic approaches for the characterisation of (i) thermal transport in nanofluids[26], (ii) heat fluxes across the nanostructured solid/liquid interfaces[27], and (iii) interfacial boundary resistance in nanostructured materials[28]. Other technique such as SThM in vacuum environment for further examination of the heat fluxes at the nanoscale due to the presence of adsorbed liquid film[29] was tested. Eventually Raman spectroscopy was carried out for the study of the anisotropy of thermal transport in nanomaterials[30]. All these research investigations, which have been done for the last few years by the PI, prove that he will be able to tackle both theoretical and experimental aspects of the present project.

Section b. Methodology

The above-mentioned results are the building blocks or initial steps for further work going beyond the current state-of-the-start in the examination of thermal transport across the solid/liquid interface. The new experimental and simulations techniques will be further developed to achieve this goal. For this purpose, we have gathered a unique group of competences with the best experts in the field that are not available anywhere

else. Our research strategy is divided into 4 work packages to tackle the 6 objectives mentioned in the first section of this project. The main tools will now be detailed. *Tool 1 - Molecular dynamics*

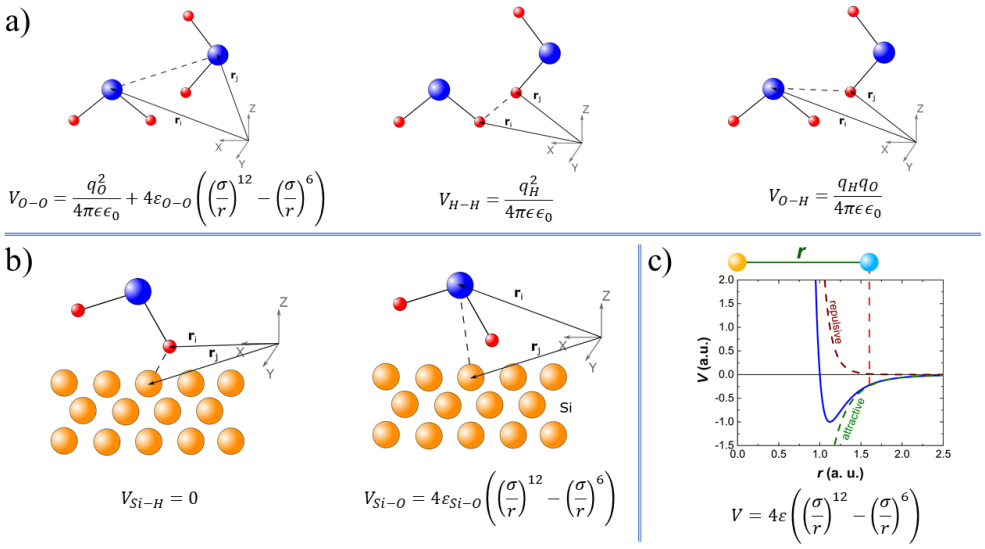

Fig. 4. Examples of interactions: a) a model representation of the water molecule based on the three sites model (SPC and SPCE/); b) an example of interaction of a water molecule with the uncharged solid substrate; c) example of the Lenard-Jones interactional potentials.

For numerical modelling of the solid/liquid interaction close to a nanostructured interface, the main tool that we plan to use is atomistic modelling with molecular dynamics (MD). Molecular dynamics methods are the best suited approaches for the description of heat and mass transfer across interfaces at nanoscales. As a powerful tool to elucidate heat transport processes in multiphase systems it will bring new and valuable contributions to the before mentioned problems in connection with the description of heat and mass transfer. In principle, any type of gas, liquid, solid states, and inter-phase phenomena can be solved without the knowledge of macroscopic "thermo-physical properties" such as thermal conductivity, viscosity, latent heat, saturation temperature and surface tension. The numerical data such as temperature, energy, atom motion, structural factor, etc. evaluated with MD technique will provide a basis for the further micro- and mesoscopic simulations. In addition, consideration of “micro-geometry” of the interface is a promising way to improve ultimately macroscopic behaviour of efficient systems and devices.

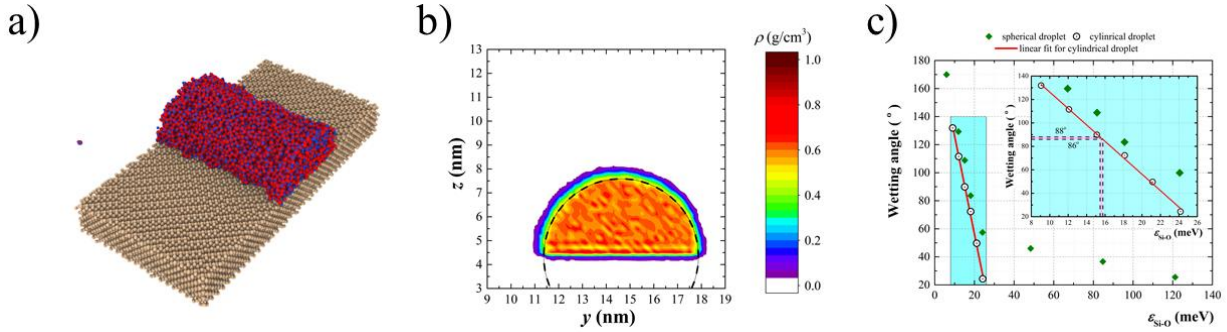

Fig. 5. a) molecular dynamics snapshot of a water droplet on silicon substrate: cylindrical droplet morphology was chosen to minimise impact of the line tension; b) density map of the liquid inside droplet; c) dependence of the wetting angle of the considered droplet and a spherical one as a function of Lenard-Jones interactional parameter[21]

Principle: MD uses classical Newton’s equations of motion to describe the system evolution with a large number of degrees of freedom. Because of its specificity one needs to initialize simulations by setting an initial configuration for the positions of atoms and molecules. Then construction of geometry and morphological nanoscale features of contacting interfaces can be performed. It is therefore possible to simulate different kinds of structuration of the solid surface. Evolution of the system based on the atoms movements with respect to the interatomic forces (see **Fig. 4**) and external fields in presence gives an inside regarding numerous processes arising at the nanoscale[31]. More specifically, implementation of the wave-packet formalism gives us the

information regarding to phonon scattering at the interfaces[32] which is crucial to understand energy transfer. These data will relate to the parameters that define interaction between solid and liquid in the same way as PI did for wetting angle (see **Fig. 5**).

Equilibrium and non-equilibrium Molecular Dynamics:

As it was mentioned previously, one can evaluate heat and mass transfer in the heterogeneous solid-fluid composite media featuring the interface with the use of MD approaches. As an example, non-equilibrium MD (NEMD) and equilibrium MD (EMD) for thermal conductivity evaluation can be mentioned[33]. Both approaches can be used to study diffusion and viscosity effects of liquid layer adjusted to solid substrate, heat transfer coefficients from solid to fluid, boundary thermal resistance of interface between solid and fluid, etc. These parameters are critical to apply conservation of mass, momentum, and energy in *classical* macroscopic models of *heat and mass transfers, and fluid mechanics.* MD was successfully applied for the study of thermal and mass transport across the solid/fluid interface[34–36]. However, almost all approaches presented in the literature were limited to the case of smooth interfaces. The main goal of the project will be focused on the investigation of the influence of heterogeneity of solid surface on molecular and thermal transport across the interface. Additionally, in the project the influence of the nano-impurities in liquid (nanoparticles) contacting with solid substrate on the thermal and mass transport coefficients will be considered.

Below we present a list of MD methods that will be used during the project implementation.

Both equilibrium (EMD) and non-equilibrium (NEMD) are based on the time-reversible equations of motion. The NEMD shows a consistent macroscopic basis for the irreversible macroscopic second law of thermodynamics[37,38]. The main idea for using NEMD is to apply spatial distribution of the controlled variables like temperature (heating and cooling distinct regions as seen in Fig. 2), constraint, pressure, or heat flux[37]. On the other hand, when using EMD, we consider fluctuations of the system around equilibrium. As a well-known example, for calculation of thermal conductivity in the system one can use the following equation based on Green-Kubo formalism:

$$\kappa_{ij} = \frac{V}{k_B T^2} \int_0^\infty dt \langle J_i(t) J_j(0) \rangle,$$

where κ_{ij} is the thermal conductivity, V is the volume of considered system, T is the temperature, $J_i(t)$ is the heat flux in "i^{th}" direction. The main advantage of this approach is the possibility to obtain directly the anisotropy of thermal transport in the studied system at equilibrium. This has a significant importance for the investigations of hybrid systems with liquids, hence temperature gradients imposed in NEMD can modify the liquids properties. However, there are still debates regarding implementation of this approach for the study of heterogeneous system. Therefore, one of the important challenges of the project will be to verify the applicability of the equilibrium-based methods to hybrid systems. If no we will propose the necessary modifications.

A wave-like approach:

One of the assumptions that may lead to the significant variation of thermal transport at the solid/liquid interface is the wave-like behaviour of interfacial layer and the presence of density fluctuation close to the interface due to the surface tension. To study these features more properly, we will carry the investigation of the static ($S(q,0)$) and the dynamic structure factors ($S(q,t)$). Such quantities appear to be an ideal tool to investigate the anisotropic properties of the system and will lead to a step forward towards a clearer description about heat transfer behaviour near the interface. Moreover, studying the dynamical properties of correlation between particles positions such as dynamical structure factor, $S(q,t)$[39–41], or density-density correlation function, $C(q,t)$ [42,43]continues to be a challenge and a topic of an active discussion in the soft and hard matters fields. Specifically, structure factor insists the information regarding density fluctuations arise in the system under consideration. Thus, it will be indispensable tool for the understanding of the nature of thermally induced excitations at the interface.

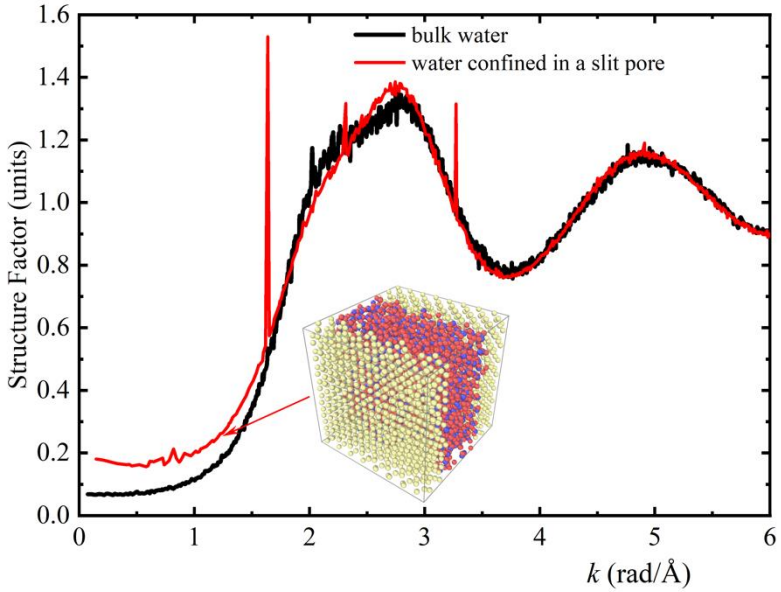

Fig. 6. The static structure factor $S(q,0)$ for a bulk water and the bulk water confined in a pore with slit morphology for T=300 K.

Fig. 6 presents our preliminary results of the static structure factor calculations for the bulk water and for the water slab confined in a pore with slit morphology. It can be seen from **Fig. 6** that the shape of $S(q,0)$ for the bulk water exhibits well-known features already discussed in the literature[44]. However, the confined water manifests some features such as a deviation from the well-known trend in low-wavelength range. It can be used as a starting point for the further implementations by keeping in mind that its behavior should be analyzed more precisely in the hydrodynamic limit.

For this part of the project that is related to the theoretical development and numerical simulations we are going to develop the program solution in C++ that can be integrated into the molecular dynamics package LAMMPS[45]. This will allow us to perform the calculations of given quantities on the fly. Specifically, we will obtain the information about the wavelength of the excitation raised at the interface from the static structure factor while dynamic $S(q,t)$ will give us the insight about the relaxation-time and frequency in the considered systems.

Phonon-interface scattering effect:

MD simulations are also efficient in the providing detailed information on the phonon-interface scattering. The main concept of such simulations is to construct a phonon wave packet from a single branch of the phonon dispersion curve with a narrow frequency range and a well-defined polarization[32]:

$$u_{il\mu} = A\epsilon_{i\mu\lambda}(\boldsymbol{q}) \exp[\mathrm{i}\boldsymbol{q}(z_1 - z_0)] \exp[-\eta^2(z_1 - z_0)^2],$$

where A is the amplitude of the wave, $\epsilon_{i\mu\lambda}(\boldsymbol{q})$ is the polarization vector for the band λ at the wavevector $\boldsymbol{q}$, z is the direction where the wave packet will be created, and η is the parameter that can be selected based on the considering system. Such technique allows us to visualize the wave packet splitting into reflected and transmitted parts, that can propagate through and backward an interface.

Tool 2 – Photoacoustic techniques

Effective thermal conductivity of the investigated systems will be considered by using several experimental approaches. Firstly, with the use of the photoacoustic approach, where a thermal perturbation is induced in the system by modulated or pulsed laser irradiations. The monitored response corresponds to the pressure perturbation that arises due to thermal elastic coupling. The main advantage of this technique is the possibility to carry out measurements without performing a direct contact with the excitation source. This is particularly important for the samples considered in this project where a microscale surface and volume features do not allow us simple direct instrumentation. In the project we will use the gas-microphone and the piezoelectric detection principles about which the PI has already performed pioneer works.

Principle: In the gas-microphone configuration the response is monitored as the pressure fluctuations in the gas chamber where the studied sample is set. The magnitude and the phase shift of the pressure's perturbation are directly related to the thermophysical and the optical properties of the sample. In the piezoelectric

configuration, the thermoelastic response in the sample is induced by the incoming light pulse. This response is measured with the use of piezoelectric transducer. Such approach is also sensitive to the thermophysical and optical properties of the sample. Additionally, for hybrid systems the recorded signal contains the information about elastic properties of the considered systems and the movements of the liquids inside the nano-channels of porous network that is important for the calculations of correlations with MD simulations. Combination of both configurations (gas-microphone and piezoelectric) will provide accurate evaluations of effective parameters of the systems with large solid/liquid interfaces. These effective values will be corelated with the theoretical ones calculated using the simulation inputs (interfacial boundary resistance, thermal conductivity of the components, etc).

Despite all its advantages (simple set up, robust post-processing, versatile type of a considered sample, etc.), the main weakness of both photoacoustic approaches lies in intrinsic difficulties to analyse very thin samples (with the thickness less than 5 μm). Such methods are also limited to investigate, in one shot, the anisotropy of thermal transport as the set signal, and the recorded response along one direction are recorded.

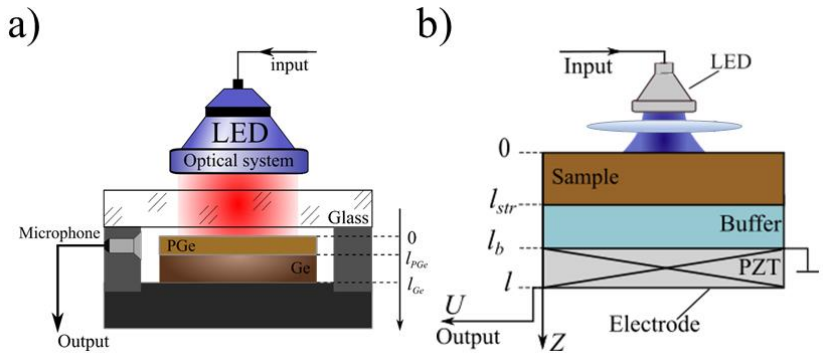

Fig. 7. A schematic sketch-view of a) gas-microphone photoacoustic cell; b) piezoelectric photoacoustic cell;

Tool 3 - Raman spectroscopy

The typical sketch-view of the Raman setup is presented in **Fig. 8 a**. In this case the laser beam is focused on the sample surface, and the reflected (or transmitted for transparent samples) light is collected by the same optical system. Then reflected light is filtered by the notch or edge filter to cut the excitation laser wavelength, and in a such way scattered light is further analysed by spectrometer.

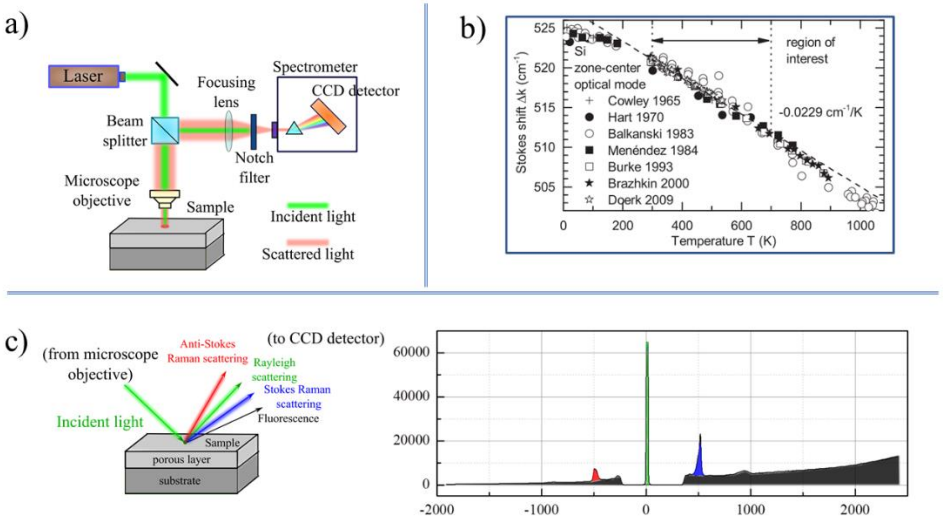

Fig. 8. a) A schematic sketch-view of the Raman set-up; b) dependence of the optical phonon peak of silicon on temperature (extracted from[46]); c) Stokes/Anti-stokes peaks for silicon

Raman scattering is the inelastic scattering of a light by a matter. It takes place when phonons transfer (for Stokes process) a portion of energy to the molecular vibrations. Specifically, the peaks in the Raman spectra correspond to the high frequency phonons for solids. Therefore, the positions of the peaks are sensitive to the strains in the material. Because of thermoelastic property, it can be used for the temperature measurements in the light beam focusing area. As an example, the dependence of the silicon peak of the optical phonons position on the temperature is presented in **Fig. 8 b**.

Another possibility of the temperature sensing is based on the measurements of the Stokes/Anti-stokes ratiothat is also sensitive to the temperature (see Fig. 8 c):

$$\frac{I_{AS}}{I_S} = C\left(\frac{\lambda_S}{\lambda_{AS}}\right)^4 exp\left(\frac{\hbar\omega}{kT}\right),$$

where I_S and I_{AS} are the intensities of the Strokes and Anti-stokes peaks, C is the constant that depends on the set-up configuration, λ_S and λ_{AS} are the wavelengths of the scattered light due to the Stokes and Anti-stokes processes, ω is the frequency corresponded to the Raman peak.

Thus, Raman spectroscopy allows us to investigate the temperature response of thin systems (with characteristic sizes ~1 μm), and simultaneously the laser light can be used as a heating source. This allows us to study the temperature rise in the sample as a function of the source power, and one can evaluate thermal conductivity of the samples based on the thermal problem resolution. Specifically, PI of the project proposed the method of thermal transport anisotropy study based on the laser beam shape modifications[47,48].

Tool 4 - Scanning thermal microscopy

Principle: Scanning thermal microscopy (SThM) is based on the Atomic Force Microscope (AFM) where a nanoscale tip interacts directly with a surface. Regular AFM experiments can give us the insightful information on the topography of the interfaces such as roughness or its periodicity or structuration at a nanoscale level. We will go further by using the SThM technique developed in the team. Briefly, in SThM we use a thermoresistive tip that heats up by Joule effect (when a small electrical current flow through the probe creating a nano-heat source as large as the tip (~100nm)). Raster scans along a surface of a sample give us the topographic information on the roughness or structuration of the interface and images of the heat transferred from the hot probe to the surface with a lateral resolution of ~100nm. The schematic representation of the basic principle of the SThM is shown in Fig. 9.

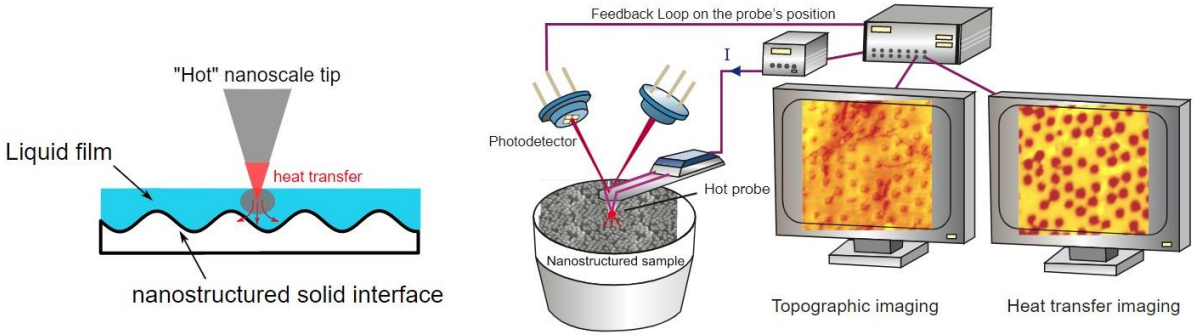

Fig. 9. Schematic sketch-view of the SThM measurements setup

Major improvements have been done to the SThM developed in the team in order to carry out quantitative information from the measurements. First, the system has been integrated in a controlled environment chamber where the pressure can be lowered to high vacuum (~10^{-6}mbar) in order to control humidity and/or to get rid of undesired convection losses.

Within the framework of this project we will use the introduced setup in an innovative manner by coupling the hot tip with a thin liquid layer in order to accurately study the energy transfer from the solid phase to the liquid as well as the interface effect. Moreover, such coupling has never been attempted before that makes our setup unique in this area.

Tool 5 - Time Domain Thermoreflectance (TDTR)

Principle: Time domain thermoreflectance by using the femtosecond laser is also a very powerful technique to study nanoscale energy transfers due to its unparalleled temporal resolution (less than 10^{-12}sec that can be easily reach by using the optical sampling). For example, the picosecond ultrasonic can be used in more precise measurements of extremely thin thickness, and in studying of the acoustic wave propagation. It has also been successfully applied in the field of nanoscale heat transfer to measure thermal conductivity of solid and liquid samples as well as Kapitza interface resistance between various materials.

The basic principle of the pump-probe TDTR is simple: a brief laser pulse is used to generate the acoustic or heat waves through the absorption of the laser energy by a thin metallic transducer. Then a second delayed laser pulse is used to monitore the variation of the optical properties of this transducer. The delay between the two laser pulses is obtained by a mechanical delay line. The acoustic or the electro-optic modulator is also

combined with a lock-in detection to improve the signal, to noise the ratio, and to detect tiny variations ($\Delta R/R \sim 10^{-5}$) of the transducer reflectivity.
An example of the typical femtosecond pump-probe TDTR and measured thermal/acoustical signal is given in the following figure.

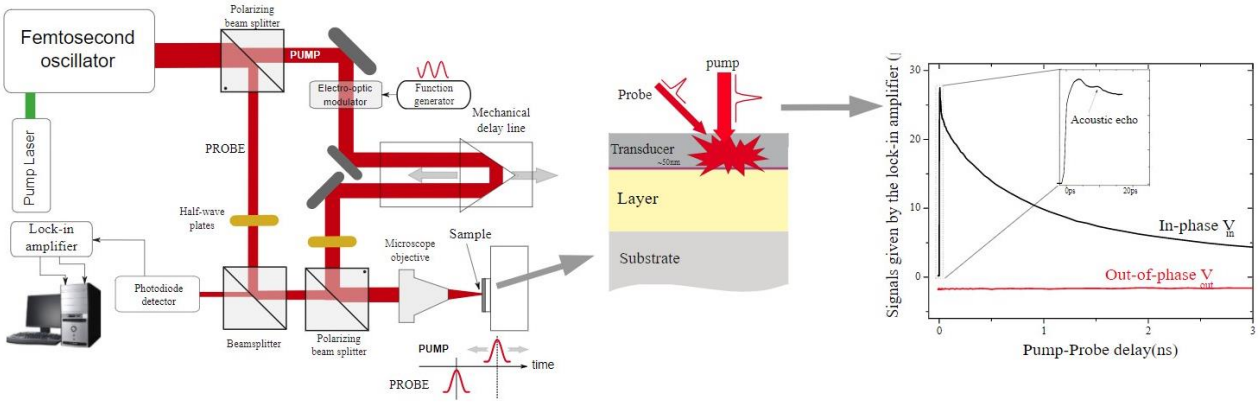

Fig. 10. Left: the classical femtosecond TDTR setup. Middle: the sample capped with a thin metallic transducer. Right: the typical thermoreflectance contained both heat transfer and acoustical wave propagation.

While this femtosecond pump-probe setup may be hardly built by novice, our team at LEMTA has a great experience in building such setup.

Materials and nanostructures

Our study will be focused firstly on the materials and the systems with a significant surface-to-volume ratio. Specifically, PI has proven experience with a various silicon based on the porous systems such as micro/meso porous silicon and the silicon nanowires arrays. These materials will be used as the starting points, and the methodology will be further expanded to the other nanostructured materials such as nanofluids (with silicon and carbon-based nanoparticles), nano/macro porous silicon, thermally expanded graphite. It is expected that such systems exhibit important change of thermal physical properties due to the solid/liquid interface. Besides, it will be possible to perform both experimental and theoretical/simulations studies of the effective thermal conductivity variation in the considered systems.
However, to go deeper insight the features of thermal transport across the solid/liquid interface, we will consider flat and nanostructured substrates elaborated with the atomic layer deposition techniques All facilities for the sample fabrication are available in the personal network of PI or in the regional cluster of laboratories of the CNRS/University of Lorraine.

Section c. Synthesis and WP description

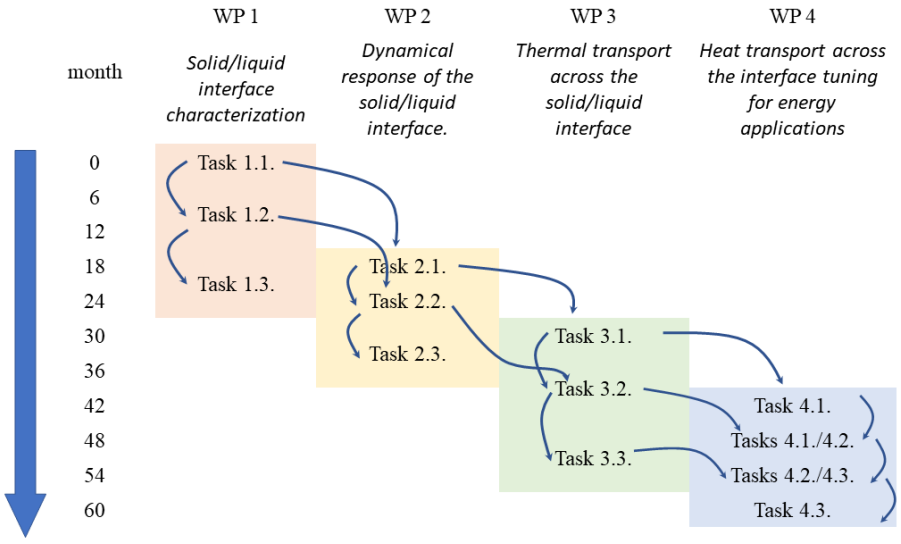

Fig. 11. The diagram that represents the timeframes of the project implementation.

All the above-presented theoretical and experimental methods are the corner stone for state-of-the-art studies going beyond classical methodologies used to investigate thermal transport across the solid/liquid interface. Each technique has its own specificities and limitations. Combining them into a top-level platform dedicated to microscale characterization of heat and mass transport will undoubtedly lead the PI and group working with him toward new understanding of this complex interaction. In order to achieve properly this ambitious research program, 4 interconnected (see **Fig. 11**) work-packages (WP) are defined hereafter:

WP 1. (0 – 24 months) *Solid/liquid interface characterization.*

This WP will be devoted to the study of the liquid and solid properties in vicinity to the interface, and it will be implemented in respect to the following tasks:

1.1. Modules will be developed for the calculation of the static and the dynamical structure factors. Radial distribution function, velocity autocorrelation function, mean square displacements, density fluctuation, etc. will be examined in equilibrium for different parametrization of the solid/liquid interaction.

1.2. Wetting angle, adsorption energy and forces, solid/liquid surface tension will be evaluated for different morphologies of the interface. Impact of the surface structuration and presence of the heterogenous regions on the solid substrate will be examined for the parameters mentioned above.

1.3. Experimental studies of the wetting angle of a droplet that is in contact with a solid with various surface morphologies and its functionalization. Additional attract-approach curves measurements with use of atomic force microscopy (AFM) built inside a controlled environment chamber, where the surrounding gas or pressure can be controlled, will be carried out to verify simulations received in Tasks 1.1 and 1.2.

WP 2. (18 – 36 months) *Dynamical response of the solid/liquid interface.*

This WP will be devoted to the investigations of the solid/liquid phases in response to dynamical actions:

2.1. MD simulations of the solid/liquid interface behavior will be examined under action of the pulsed and periodic force with different spatial distributions. Additionally, the transfer of phonon wave-packet across the interface will be examined. Spectral characteristic of the wave-packet will be chosen based on the data obtained in Task 1.1. This will give us the possibility to examinate precisely phonon scattering conditions at the solid/liquid interface. The received data will be important because they will highlight the process of transmission of the elementary excitations from the solid in the liquid.

2.2. Development of the analytical acoustical models that consider the adsorbed film presence with the input parameters from Tasks 1.2 and 2.1. This will give us the possibility to estimate interfacial boundary resistance based on the model.

2.3. Photothermal experiments, based on thermoreflectance and/or photoacoustic methods, with different excitation sources will be carried out to examine experimentally dynamical impact on the systems with significant solid/liquid interface. Photothermal in frequency domain can be used to precise measurements of the thermal conductivity of the sample under test (liquid or solid) and thermal interface resistance. The optical sampling by using ultrashort pulsed lasers will give access to vibrational modes and acoustical coupling between liquid and solid phases. The results will probe the elastic and thermophysical properties of the considered systems that can be compared with MD simulations.

WP 3. (30 – 54 months) *Thermal transport across the solid/liquid interface.*

This WP consists of several tasks:

3.1. Thermal transport studies across solid/liquid interface with the use of non-equilibrium molecular dynamics (NEMD). This task will give us the possibility to verify the acoustical-based model for the interfacial boundary resistance (Task 2.2).

3.2. NEMD simulations of the thermal transport across nanostructured the solid/liquid interface. During this task we will demonstrate dependence of the interfacial boundary resistance on the size (or curvature). SThM measurements of the thermal resistance between a nanoscale tip and a water adsorbed film. The obtained results will be correlated with the deliverables of Tasks 2.2 and 3.1.

3.3. Implementation of the equilibrium molecular dynamics methods for the investigation of the heterogenous systems (with the atoms of solids and liquids). Examination of anisotropy of the thermal transport at the solid/liquid interface. Raman measurements of the thermal transport anisotropy in porous silicon and silicon nanowires arrays that are empty/filled by liquid. Afterwards, the evaluation of thermal conductance

in interfacial boundary layer and further comparison with equilibrium molecular dynamics results will be performed.

WP 4. (42 – 60 months) *Heat transport across the interface tuning for energy applications.*

This work package will summarize all the results received in WPs 1-3, and the WP 4 implementation will significantly depend on the previous steps. Nevertheless, in frames of this WP:

4.1. As a result of the previous steps implementation, we will verify our hypotheses that explain thermal transport modification close to the solid/liquid interface. Some examples of such hypothesis: i) the presence of a liquid on a solid substrate modifies scattering condition of phonons at the interface, and thus impacts on their mean free path; ii) the solid/liquid interface supports surface waves that carry energy and contribute to thermal transport; iii) the pore acts as "nanochannel" that may change the diffusivity of water molecules; iv) the adsorbed layer of a liquid on a solid substrate has significant thermal conductivity because of the crystal-like behavior due to local increase of liquid density.

4.2. The analytical model will be developed to describe thermal transport modifications at the solid/liquid interface with respect to the identified physical insight in T. 4.1.

4.3. The optimal design of the solid/liquid interface will be proposed to enhance/reduce impacts of different mechanisms on the thermal transport across the solid/liquid interface.

The implementation of the project with respect of the presented WPs will give us the possibility to obtain a clear view about the phenomena that appear at the solid/liquid interface during thermal transport processes. This will allow us to control and tune processes related to the thermal transport (see **Fig. 12**). The main idea is to widespread the results, and to give the possibility to different teams reproduce and improve the approaches. Nevertheless, there are significant risks that the chosen methods and some routes may not work properly. Therefore, we have also developed fallback solutions for each of the identified risk.

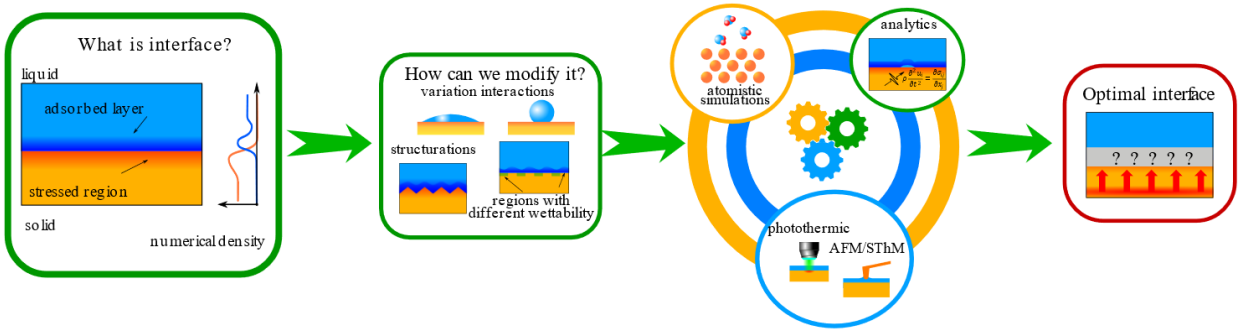

Fig. 12. The microscopic insight for the thermal transport management

Identified risks and back-up solutions

The main risk in the project is that chosen classical MD simulations will not be able to cover all the necessary spatial and time scales to reproduce the real-life behavior. In this case, the coarse-grained MD simulations[49] will be performed. For instance, our preliminary results[50] mitigates this risk, specifically, even representation of three-sites water molecule with a dipole (ELBA model[51]) gives the possibility to significantly reduce computational time.

Another risk relates to MD approach: in WP 1 the main results regarding to the solid/liquid interaction will be elaborated by using MD techniques. However, this approach is based on the usage of predefined potentials, and such potentials are not defined properly for all materials and theirs compounds. For instance, interatomic potential for the crystalline diamond-like Si is not the same as the potential for Si atoms in the silica (Si_2O). Additionally, for semiconductor and metallic materials surface band-bending of electronics states may perturb such interactions due to surface charges arising that could lead to problems with classical MD[52]. Therefore, as a *plan B*, in such situations we will use ab-initio technique that is (or was) developed in our team to tune the molecular dynamics input parameters. In this case, the forces are calculated from quantum mechanics, and the electronic effects are included naturally. We will tune potentials in our classical MD to represent the same properties at the interface as in ab-initio simulations. Specifically, we will match a density profile of the liquid at the interface, its radial distribution functions, and the structure factor.

In WP 2, there is a risk that the interfacial boundary resistance will not be accessible with photoacoustic experiments, which is a quite desirable value to be compared with MD simulations to achieve the project goals. This difficulty may arise because photoacoustic technique, which in general, is applied well to the evaluation of the effective averaged parameters (such as an effective thermal conductivity), and extraction of the microscopic parameters from such approach requires development effective media models. However, in this case we will use frequency and time domain thermoreflectance method (TDTR) which is currently under development in PI's team.

One of the main deliverables from WP 4 (the optimal interface to heat transfer) will significantly depends on the developed analytical models in WP 2 and WP 3. There is a risk that the model will be complicated, and it will be hard to perform optimization. To overcome such issues we will increase variation of configuration in atomistic simulations, and additionally we will use minimization procedures. If the number of parameters is reasonable (for example height and width of pillar, number of parameters in interactional potential) we will use radial basis function approximation[53]. Otherwise, if it will be necessary to vary significant number of parameters (for example, positions of atoms in the surface layer) low rank matrix method will be used[54]. The mitigation plan will have a less wide impact but will be valuable, and still will have a very high impact because it will give us the possibility to optimize morphology of the solid liquid interface to intensify heat exchange.

Since the project is uniquely positioned in the field of thermal transport across the solid/liquid interface, the results elaborated will be significant in numerous aspects of the energy applications. Particularly, they will give us the possibility to improve thermal transport in the system where a solid and a liquid met. The straightforward example that can be mentioned here is the situation when we need to harvest heat from the solid inside liquid and to further conserve and re-use it.

The key concept of this project is a completely new in the field of interfacial thermal transport, because we firstly concentrated on the microscopical features of the interface to identify the mechanisms of thermal transport across the solid/liquid interface. Detailed understanding of mechanisms will open the possibility to predict heat fluxes more precisely at the solid/liquid interface, and in a such way to propose the new routes to improve the efficiency of thermal management for energy applications.

Additionally, it should be noted that WP 1 is more general and deals with different aspects of interfacial science. Therefore, the implementation of this WP will also provide innovative solutions in other crucial domains such as intensified chemical reactors, membranes separation technologies, nanofluids, etc.

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