Thursday, November 25th

Session Nanoscale Heat Transfer - Measurement

10:20 – 10:50 Keynote

Stefan DILHAIRE, Bordeaux Univ. – LOMA, France

Abstracts

Keynote Speakers

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Biography

Stefan Dilhaire is Professor at University of Bordeaux. At LOMA (Laboratoire Ondes et Matière d'Aquitaine) Stefan Dilhaire's group studies mutual interaction of heat, light and electricity in micro-systems and nano-materials and its applications in renewable energy, in microelectronics, in nano-plasmonics, and biology.

ULTRAFAST ENERGY TRANSFER IMAGED BY TIME DOMAIN THERMOREFLECTANCE

The reduction of the size of nano-objects or nano-materials down to the nanoscale leads to strong modifications of its transport properties depending then on its size, shape, structure and obviously on its environment. Carrier confinement combined to interface effects gives rise to new transport properties. That is the case in absorption and emission of light where the new properties are given by electromagnetic near field coupling between the nano-objects included in the material. Concerning phonon transport, a frequency dependence of thermal conductivity can be observed. Plasmons confined in a tapered wave guide slow down producing hot carriers. This hot electron lifetime increases in a hot spot. All these processes occurring at time scales from femtoseconds up to nanoseconds are routinely accessible with ultrafast pump-probe techniques. i.e. heterodyne optical sampling allows to access to the energy transfer and understand the heat propagation into nano-objects themselves. The comprehension of energy transport mechanisms had been initiated by the study of a collection of nano-objects in solution without any coupling between them. We will describe different situations where the energy deposited by a femtosecond flash can be converted into phonons or plasmons traveling respectively at the speed of sound and speed of light in nano-materials. Our ultrafast imaging technique enables to record movies at 20 Tera image per second. Plasmons travelling at speed of light in metallic structures are revealed via the hot electron tail they leave behind them.

References
[1] Lalanne, P., Coudert, S., Duchateau, G., Dilhaire, S., Vynck, K. Structural Slow Waves: Parallels between Photonic Crystals and Plasmonic Waveguides, ACS Photonics, 2019, 6 (1), pp 4–17
[2] Lozan, O., Sundararaman, R., Ea-Kim, B., Rampnoux, J.-M., Narang, P., Dilhaire, S., Lalanne, P., Increased rise time of electron temperature during adiabatic plasmon focusing, Nature Communications, 8 (1), art. no. 1656, . (2017)
[3] D'Acremont, Q., Pernot, G., Rampnoux, J.-M., Furlan, A., Lacroix, D., Ludwig, A., Dilhaire, S., High-throughput heterodyne thermoreflectance: Application to thermal conductivity measurements of a Fe-Si-Ge thin film alloy library, Review of Scientific Instruments, 88 (7), art. no. 074902, (2017)
[4] Dehoux, T., Ghanem, M.A., Zouani, O.F., Rampnoux, J.-M., Guillet, Y., Dilhaire, S., Durrieu, M.-C., Audoin, B. All-optical broadband ultrasonography of single cells (2015) Scientific Reports, 5, art. no. 8650

This work was performed in the framework of the ANR SPIDERMAN project (No. ANR-18-CE42-0006) funded by the
French Agence Nationale de la Recherche and of the "Nano Thermal Imaging" project supported by the CRNA (Conseil Regional de Nouvelle Aquitaine, No. 2018-1R50303). This work was also supported by the Labex Laphia in the framework of the Nano-Imaging project (2018–2020).

Keywords: time domain thermoreflectance; hot electrons; plasmon; phonons.

Thematic Session: Nanoscale heat transfer and measurement
Keywords: two temperature model, thermo-reflectance, thin film, metal transducer, laser pulse heating

Investigating heat transparency of metal transducers in ThermoReflectance

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For the thermophysical characterizations of layered or nanostructured materials, Time Domain ThermoReflectance (TDTR) or Fourier Domain ThermoReflectance (FDTR) are very efficient techniques[1,2]. However, the new materials, which are often thin, lead to make measurements at short time scales or at high frequencies.
The TDTR technique named heterodyne or asynchronous optical sampling (HEOPS/ASOPS)[3] using femtosecond synchronized lasers can perform transient temperature measurements with a typical spectral response from 100 MHz up to 10 THz.
Nevertheless, the bandwidth limitation for the thermal analysis is not imposed by the optical sampling but by a metallic transducer deposited on top of the material of interest.
The role of this transducer is to convert the pump laser into a heat source and, also, to convert the surface temperature of the material into a reflectance variation measured by the probe laser.

In this presentation, we investigate the thermal transparency of several metal transducer thin films. An experimentally-suited two-temperature model[4] that describes the behavior of metals down to the short time scale, has been implemented and solved numerically. To obtain the optimal transducer film, some conditions must be checked such as a fast thermal equilibrium between the electron and phonon baths inside the transducer and a fast start of the phononic relaxation within the material of interest. In this scope, several parameters such as the transducer's thickness and the type of metal (Aluminimum, Gold, Silver …) are discussed.

References:

1. Pernot, G. *et al.* Precise control of thermal conductivity at the nanoscale through individual phonon-scattering barriers. *Nat. Mater.* **9,** 491–495 (2010).
2. Schmidt, A. J., Cheaito, R. & Chiesa, M. A frequency-domain thermoreflectance method for the characterization of thermal properties. *Rev. Sci. Instrum.* **80,** 094901 (2009).
3. Dilhaire, S., Pernot, G., Calbris, G., Rampnoux, J. M. & Grauby, S. Heterodyne picosecond thermoreflectance applied to nanoscale thermal metrology. *J. Appl. Phys.* **110,** (2011).
4. Hohlfeld, J. *et al.* Electron and lattice dynamics following optical excitation of metals. *Chem. Phys.* **251,** 237–258 (2000).

Acknowledgment:
This work was performed in the framework of the ANR SPIDERMAN project (ANR-18-CE42-0006) funded by the French Agence Nationale de la Recherche (ANR) and of the “Nano Thermal Imaging” project supported by the CRNA (Conseil Regional de Nouvelle Aquitaine, n°2018-1R50303). This work was also supported by the Labex Laphia in the framework of the NanoImaging project (2018-2020).

C'NANO 2020
The Nanoscience Meeting
TOULOUSE
Centre des congrès Pierre Baudis
December, 8, 9 and 10
C'NANO cnrs

Thematic Session: (Nanoscale heat transfer- Measurement)
Keywords: (Nanostructure, polymer, nanofiller, phonon scattering, thermal conductivity)

Synthesis and thermal properties of GO-TiO_2/PEDOT: PSS polymer nanocomposites

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It is well-known that the interdependence of thermal and electrical conductivities acts as a barrier to obtaining a high thermoelectric figure of merit. However, by using materials in which coexist nanoscale phases, it is possible to break the barrier interdependence by exploiting the phonon scattering at the nanostructured interfaces [1].

The aim of the current work is to decrease the lattice thermal conductivity of a polymeric matrix by introducing nano-assembled fillers of Graphene Oxide (GO) - titanium dioxide (TiO_2).

Nanocomposite materials have been prepared by mixing in aqueous media, sol-gel-prepared GO-TiO_2 as fillers into a polymeric matrix of Poly(3,4-ethylenedioxythiophene)-PEDOT and poly(styrenesulfonate)-PSS.

Thermal conductivitiy of the nanocomposites has been experimentally measured by using the Photothermal radiometry [2,3]. The acquired thermal conductivity of the nanocomposites showed a decreasing trend with the filler concentration, in agreement with the theoretical model proposed by Nan et. al. [4]. Adding just 5% GO-TiO_2 fillers leads to a thermal conductivity decrease from 0.39 $W.K^{-1}.m^{-1}$ for the polymeric matrix to 0.20 $W.K^{-1}.m^{-1}$ for the composite.

References:

[1] Tavkhelidze A. Large enhancement of the thermoelectric figure of merit in a ridged quantum well. Nanotechnology 2009;20:405401. https://doi.org/10.1088/0957-4484/20/40/405401.
[2] Garcia JA, Mandelis A, Farahbakhsh B, Lebowitz C, Harris I. Thermophysical Properties of Thermal Sprayed Coatings on Carbon Steel Substrates by Photothermal Radiometry n.d.:16.

[3] Depriester M, Hus P, Delenclos S, Hadj Sahraoui A. Study of thermal parameters' temperature dependence in solids using photothermal radiometry. Review of Scientific Instruments 2007;78:036101. https://doi.org/10.1063/1.2712937.

[4] Nan C-W, Birringer R, Clarke DR, Gleiter H. Effective thermal conductivity of particulate composites with interfacial thermal resistance. Journal of Applied Physics 1997;81:6692–9. https://doi.org/10.1063/1.365209.

Acknowledgments:
We are very grateful to the Université de Lille and the Université du Littoral Côte d'Opale for the financial support of this work.

Thematic Session: Nanoscale heat transfer - Measurement

Keywords: Raman spectroscopy, heat transport, porous silicon, two-phase nanocomposite

Evaluation of heat transport in «liquid-nanoporous Si» composite by µ-Raman spectroscopy

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During the last decade, the rapid evolution of elaboration methods for new nanomaterials and nano-architectured devices and systems requires deep understanding and technological mastering of fundamental mechanisms responsible for heat transport at nanoscale [1]. A Specific scientific interest is focused on thermal phenomena taking place in 2-phase (liquid-solid) nanocomposites. Behavior of liquids confined in nanopores of a solid-state material strongly affects energy transfers at nanoscale as well as global thermal properties of the nanocomposites.

Micro-Raman spectroscopy is known to be a non-destructive and highly sensitive multi-modal characterization technique widely used to study structural and thermal properties of various nanomaterials [2]. In order to evaluate the thermal conductivity of a studied material, laser light is used simultaneously for sample heating and for recording the temperature dependent Raman spectra. As for the structural analysis of nanomaterials, Raman spectral features (spectral position and shape) at room temperature allow to deduce characteristic size distribution and mechanical stresses.

In our work, we characterized the structural and thermal properties of nanoporous silicon layers by means of micro-Raman spectroscopy. The photo-induced temperature growth estimated from micro-Raman measurements was correlated with the steady-state heat transport finite element modeling (FEM) to estimate thermal conductivity values of nanoporous silicon. In particular, the impact of the layer porosity and the nanocrystallite size distribution, as well as the presence of oils in the nanopore networks on thermal transport were studied in details.

References:
[1] N. Zen et al., Nat. Commun. 5, 3435 (2014).
[2] Q. Li et al., Nanoscale, 9,10784-10793 (2017).

Acknowledgment: Financial support by the French National Research Agency (ANR) under grant No. ANR-19-CE09-0003

Thematic Session: Nanoscale heat transfer - Measurement
Keywords: Spin-crossover, molecular thin films, temperature mapping, thermal damping

Surface temperature mapping and thermal damping properties of spin-crossover molecules

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Molecular spin-crossover (SCO) materials, which exhibit a reversible solid-solid transition between the so-called low-spin (low-temperature) and high-spin (high-temperature) states,[1] constitute a promising class of phase-change materials for surface thermometry and thermal management applications.[2] These compounds exhibit a drastic change of their optical properties (absorption, refractive index) as well as an excess heat capacity accompanying the thermally induced phase transition.

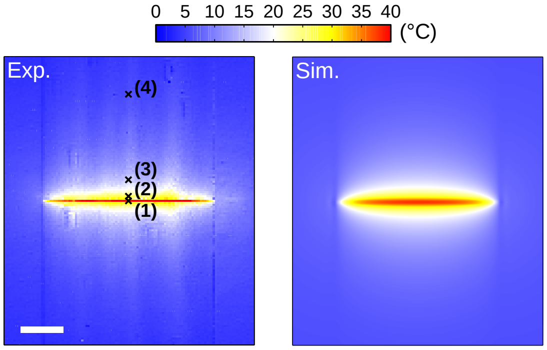

Figure 1. Experimental (left) and simulated (right) temperature maps of a Joule-heated (4 mA) gold nanowire on glass. Scale bar, 20 µm.

In this study, we show that the molecular SCO complex $[Fe(HB(tz)_3)_2]$ ($T_{transition}$ = 64 °C), recently synthesized in the form of high-quality, vacuum-deposited thin films, exhibits an exceptionally high switching endurance (> 10^7 thermal cycles) and long-term stability upon repeated thermal cycling,[3] opening the way towards real-world, technological applications.

This unprecedented reversibility allowed us to turn this SCO thin (200 nm) film into a high-performance, nanoscale temperature sensor, probed optically.[3] The capabilities of this SCO-based surface thermometer are quantified on a series of Joule-heated metallic nanowires, in which heat distribution could be mapped down to sub-µm spatial, µs temporal and 1 °C thermal resolution (*Figure 1*). Secondly, we experimentally assess the thermal damping effect of thicker (900 nm) films on the transient heating response of these wires.[4] A damping of the wire temperature, up to 10 %, is evidenced on a timescale of tens of microseconds due to the excess heat capacity associated with the switching of the molecular film.

[1] P. Gütlich, et al., *Angew. Chem., Int. Ed.* **1994**, 33, 2024 ; [2] G. Molnár, et al., *Adv. Mater.* **2018**, 30, 1703862
[3] K. Ridier, et al., *Nat. Commun.* **2020**, 11, 3611 ; [4] K. Ridier, et al., *Adv. Mater.* **2020**, 32, 2000987

Thematic Session: Nanophotonics
Keywords: Thermal radiation, nanoantennas, hybrid modes, plasmonics

Hybrid modes in a thermally excited isolated pair of antennas

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Infrared (IR) metamaterials made up of plasmonic resonators have been used in a wide range of applications, such as radiative cooling, photodetection, and solar cell design. In this regard, ensemble systems of metal-insulator-metal (MIM) resonators [1] have been studied extensively, and have proven their versatility. The study of coupling effects in an isolated pair of thermally excited MIM antennas, therefore, presents an essential step toward optimizing the far-field response of such array systems. However, this remains a major challenge in the mid-IR spectral range due to the difficulty of extracting the thermal radiation of an isolated subwavelength antenna. Thanks to a recently developed IR spatial modulation spectroscopy technique we have been able to characterize the intrinsic properties of a single subwavelength MIM nanoantenna [2]. Here we extend this study to a thermally excited, asymmetric, isolated pair of MIM antennas separated by a nanometric gap (Fig. 1(a)) [3]. Through thermal fluctuations, all the modes of the system are excited and hybrid bonding and anti-bonding modes (Fig. 1(b)) can be observed simultaneously. The manifestation of these modes occurs in the form of a splitting in the resonance peak of the thermal radiation spectrum of the system (Fig. 1(c), red curve). The effect of hybrid modes on the resonant behavior of the currently considered system may guide future efforts for realizing tunable optical and thermal systems for various applications, ranging from IR photodetection to multispectral biosensing.

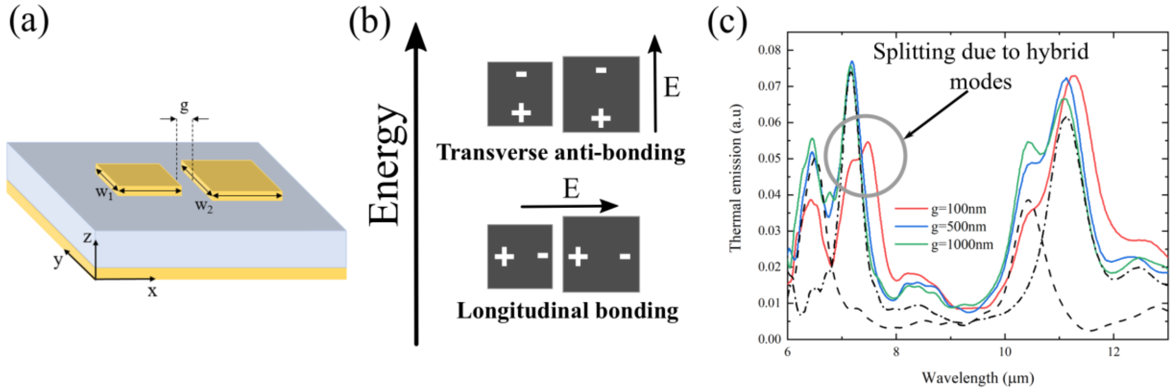

Figure 1. (a) Schematic of the coupled MIM system. (b) Sketch of the hybrid modes of the system. (c) Measured spectra of the MIM pair for various gap sizes (colored curves), along with spectra for the single MIM antennas making up the coupled MIM system (dashed and dash-dotted curves).

[1] P. Bouchon *et al. Optics Letters **37**, 1038 (2012).*

[2] C. Li *et al.* Physical Review Letters *121.24 (2018): 243901.*

[3] L. Abou Hamdan *et al. (submitted).*

Thursday, November 25th

Session Nanoscale Heat Transfer - Measurement

14:00 – 15:10 Keynote

Pierre-Olivier CHAPUIS, CNRS – CETHIL, France

Abstracts

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Biography

Pierre-Olivier Chapuis (Engineer & MS, 2004, and PhD, 2007, Ecole Centrale Paris) is a CNRS Research Scientist at CETHIL in the Microscale and Nanoscale Heat Transfer (MiNT) group since 2011. He was previously a post-doctoral researcher at the Catalan Institute of Nanotechnology (ICN2) in Barcelona (2008-2011). His interests deal with nanoscale heat transfer and conversion, in particular sub-wavelength thermal radiation and phonon heat conduction, including applications such as thermophotovoltaic energy harvesting or thermal management for electronics. His works involve both theoretical (Boltzmann transport equation, fluctuation electrodynamics) and experimental (scanning thermal microscopy, electro-thermal measurements) aspects.

SCANNING THERMAL MICROSCOPY: PROBING TEMPERATURE AND HEAT DISSIPATION OWN TO THE FEW-NANOMETER SCALE

Scanning thermal microscopy (SThM) [1], a technique derived from atomic force microscopy aiming at characterizing energy transfer at nanoscale, is applied with different thermoresistive tips, providing down to 10 nm spatial, few mK temperature, and pW.K-1 thermal conductance resolutions. Two main applications are highlighted: nanoscale thermal transport property determination and thermometry.

In ambient conditions, we demonstrate ballistic thermal transport in air. In vacuum, the tip-sample exchange before contact is mediated by means of near-field thermal radiation [2] and then by heat conduction across constrictions. SThM is found to be applicable for characterizing materials with thermal conductivity lower than ~3 W.m-1.K-1, but reduced sample area, as in the case of suspended phononic nanomembranes, can allow characterizing thermal conductivity in air up to ~50 W.m-1.K-1 [3]. Recent results obtained for various set of samples are underlined, including thin oxide amorphous films down to the native-oxide case [4]. Thermal transport mechanisms are discussed, in particular when ballistic phonon dissipation takes place.

We also highlight strategies for performing small-scale thermometry and discuss the link between the thermal signal and the actual sample temperature [5], taking examples from the electronics industry.

References
[1] S. Gomès *et al.*, Phys. Stat. Sol. A 212, 477 (2015).
[2] C. Lucchesi *et al*, submitted. ArXiV:1912.09394.
[3] A.M. Massoud *et al.*, Appl. Phys. Lett. 111, 063106 (2017).
4] E. Guen *et al.*, submitted.
[5] A. Pic *et al.*, submitted.

This work was performed with E. Guen[1], A.M. Massoud[12], A. Pic[13], C. Lucchesi[1], V. Lacatena[4], M. Haras[4] (PhD students), A. Alkurdi[1] (post-doc), J.F. Robillard[4], J.M. Bluet[2], S. Gallois-Garreignot[3], R. Vaillon[15], S. Gomes[1] (colleagues). [1]CETHIL, [2]INL, [3]ST Micro, [4]IEMN, [5]IES.

Support of projects TIPTOP & DEMO-NFR-TPV (ANR) and QuantiHeat & EFINED (EU) is acknowledged.

Keywords: nanoscale heat transfer; AFM; phonon; thermal radiation; ballistic ;near field.

Thematic Session: Nanoscale heat transfer - Measurement
Keywords: near-field thermal radiation, sub-wavelength radiative heat transfer

Near-field thermal radiation: material and temperature effects

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When two bodies at different temperatures are progressively brought closer, the radiative exchange increases strongly, especially because near-field interaction allows for photon tunneling in the sub-micrometric distance regime. Near-field radiative effects can be observed through two different manifestations. First, the distance dependence of the flux exchanged between two bodies at different temperatures is different from the prediction from the macroscopic theory. This is now well established for large bodies, such as parallel-surfaces and sphere-plane configurations, but a limited number of materials have been considered to date [1]. We have measured this dependence for materials such as SiO_2, graphite and InSb, with an experiment based on scanning thermal microscopy (SThM) in vacuum. Second, the temperature dependence of the flux exchanged is also expected to be different in the near field [2,3], as sizes and temperatures are tightly connected [4]. Only moderate temperature differences (up to $\Delta T \sim 400$ K) between emitter and receiver have been probed so far. We have focussed on near-field radiative heat transfer measurements between microspheres and planar substrates at various temperature differences [5]. Measurements for record temperature differences larger than 900 K, with the emitter being heated up to 1200 K, have been performed. The large temperature differences allow observing the temperature dependence of near-field radiative heat transfer. The measurements are compared with results of the proximity (also called Derjaguin) approximation. An analysis with different couples of materials confirms that lower conductances are found for asymmetrical emitter/substrate configurations.

References:
[1] C. Lucchesi *et al*., Nanoscale Horizons, *to be published*. [2] T. Kralik *et al.,* Phys. Rev. Lett. 109,224302 (2012). [3] Y. Tsurimaki *et al.*, J. Quantit. Spectrosc. Rad. Transf. 187, 310–321 (2017). [4] K.L. Nguyen *et al.,* Appl. Phys. Lett. 112, 111906 (2018). [5] C. Lucchesi *et al*., *to be published.*

Acknowledgments: Financial support by the French National Research Agency (ANR) under grant No. ANR-16-CE05-0013 and partial funding by the French "Investment for the Future" program (IDEXLYON ANR-16-IDEX-0005) are acknowledged.