China Scholarship Council (CSC)
PhD Thesis Proposal Form
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FIELD
(Physics)

Thesis subject title: **Theoretical and experimental investigation of plasmonic structures for applications in near-infrared and in mid-infrared ranges.**

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- **Thesis proposal (max 1500 words):**

Context: The physical phenomenon of surface plasmon resonance (SPR) is arisen by light/matter interaction at the interface of metallic and dielectric materials, especially in noble metals, e.g. silver and gold. This effect has attracted considerable interest in different domains: physics, chemistry, biology, etc. The plasmonic effect is distinguished into three categories: (i) surface plasmon polariton; (ii) localized surface plasmon resonance; and (iii) plasmonic nanostructures.

Localized surface plasmon resonance (LSPR) happens when collective oscillations of free electrons are confined to a finite volume, such as metallic nanoparticles (NPs). Generally, the LSPR in visible range is obtained with noble NPs with dimensions below 100 nm. The plasmonic properties of metallic NPs vary with their shape and size, and are also affected by the refractive index of the surrounding medium. It is also demonstrated that the SPR effect becomes much stronger when two or multiple metallic nano-objects are arranged very close to form the so-called plasmonic nanostructure (PNS). A prominent example for coupled SPR is a nano-hole array (NHA). In such a system, the surface plasmon polariton can propagate throughout the NHA surface thanks to the coupling of multiple nano-holes perforated appropriately in a metallic thin film. The properties of plasmon resonance of NHAs can be tuned by characteristic length scales and types of arrays such as periodic, quasiperiodic, and aperiodic structures. Such PNSs have great promise for many interesting applications, such as tunable filter, sensor, and color printer.

Recently, we demonstrated a robust technique based on the use of direct laser writing (DLW) method for realization on demand of plasmonic structures [1,2]. This allows obtaining plasmonic nanostructures in a single step without needing the preparation of polymeric template and lift-off process. By this direct fabrication technique, the nanoholes do not have circular shape as the laser focusing spot, due to the non-uniform heat transfer in a no-perfect flat Au film. It was theoretically and experimentally demonstrated that the properties of fabricated plasmonic nanoholes arrays are very close to those of ideal plasmonic nanostructures. Furthermore, it is theoretically demonstrated that the non-perfect circular shape of the

Au hole allows amplifying the electromagnetic field of the resonant peak by several times as compared to the case of perfect circular shape. The direct fabrication method thus paves the way to many interesting applications, such as color nanoprinter [3,4], and the rough holes of plasmonic structures could be an advantage for application in laser and nonlinear optics domains.

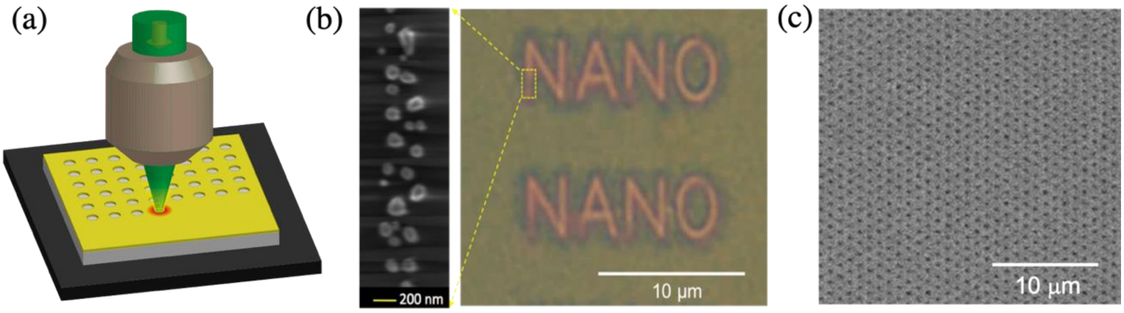

Figure 1: (a) Illustration of the fabrication technique based on the optically induced thermal effect via DLW technique. (b) Optical microscope images of plasmonic patterns consist of Au NPs: "NANO" letter. Left: SEM image of the Au NPs. (c) Scanning electron microscope image of a two-dimensional gold structure.

While the plasmonics-based applications in visible and near-infrared (NIR) use noble materials, such as silver and gold, the applications in mid-infrared (MIR) region require a combination of noble-metal and graphene or purely graphene, thanks to its exceptional electrical, optical and chemical properties. In particular, graphene can serve as plasmonic material for a large region (from MIR to THz) since its optical constant can be adjusted by apply a gate voltage, which cannot be achieved by conventional metal-based plasmonic materials.

Very recently, the Prof. E Wu's group in Shanghai has demonstrated the broadband MIR single-photon frequency upconversion spectroscopy based on the temporal-spectral quantum correlation of non-degenerate photon pairs as an excellent spectroscopy method at single-photon level. The system is featured with non-destructive and robust operation, which makes the single-photon-level MIR spectroscopy an appealing option in bio-chemical applications. Since the technique uses a nonlinear optical effect, namely, frequency upconversion process, a local high field is strongly demanded to have good conversion efficiency and to avoid unintended nonlinear noise caused by strong pump fields.

Thesis proposal: We therefore propose a joint PhD subject to realize PNSs (by using gold and/or graphene materials) by using the DLW technique and to exploit the use of fabricated PNSs for enhancement of nonlinear optical effects thanks to the strong field of the rough PNSs.

Goal 1: Demonstration of a direct fabrication of PNSs on gold and/or graphene thin film, by using DLW technique.

Goal 2: Demonstration of enhanced nonlinear optical effects, such second-harmonic generation, at NIR range, by using fabricated gold PNSs.

Goal 3: Realization of a sensitive detection of the plasmonic enhanced absorption by single molecules in MIR regime, by using fabricated graphene PNSs.

In order to achieve our aim, the work should deal with different aspects, from theoretical calculation and experimental fabrication to applications demonstration. A PhD study of 3 to 4 years is required. This joint

PhD subject will be co-directed and co-supervised by two laboratories, one French and one Chinese. The student will stay most time in French laboratory, and have only one or two short time stay in Chinese laboratory.

- **Publications of the laboratory (both) in the field** (max 5):

1. Q. C. Tong, M. H. Luong, J. Remmel, M. T. Do, D. T. T. Nguyen, N. D. Lai, "*Rapid direct laser writing of desired plasmonic nanostructures*", Opt. Lett. **42**, 2382- 2385 (2017).
2. F. Mao, G. L. Ngo, C. T. Nguyen, I. Ledoux-Rak, and N. D. Lai, "*Direct fabrication and characterization of gold nanohole arrays*", Opt. Express **29**, 29841-29856 (2021).
3. F. Mao, A. Davis, Q. C. Tong, M. H. Luong, C. T. Nguyen, I. Ledoux-Rak, N. D. Lai, "*Direct Laser Writing of Gold Nanostructures: Application to Data Storage and Color Nanoprinting*", Plasmonics **13**, 2285–2291 (2018).
4. Q. C. Tong, F. Mao, M. H. Luong, M. T. Do, R. Ghasemi, Q. T. Tran, T. D. Nguyen and N. D. Lai, "*Arbitrary Form Plasmonic Structures: Optical Realization, Numerical Analysis and Demonstration Applications*", Chapter 5, book "Plasmonics", INTECH (2018).
5. Y. Cai, Yu Chen, X. Xin, K. Huang, E Wu "*Mid-infrared single-photon upconversion spectroscopy based on temporal-spectral quantum correlation*", Photonics Research DOI 10.1364/PRJ.467695

Signature of the PhD director

Name and signature of the Laboratory director