LAM Special Issue call for papers │ Special issue on a 90-year journey towards light from the intramolecular universe

Guest Editors:

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The Reason:

With this special issue 'Nanospectroscopy, nanooptics and nanofabrication', we wish to highlight the rapid experimental and theoretical advances of this burgeoning interdisciplinary field, as well as to provide an account of its peculiar challenges and future prospects. Advances in understanding the fundamental mechanisms of optical resolution at the nanometer and Angstrom regime, novel simulation methods, and applications of these fundamental mechanisms are the main components. Furthermore, this special issue aims at promoting novel fabrication techniques which allow to reproduce the nano- or pico-scale environment for the near-field confinement, which deem to improve or open up enormous new horizontal of the research topics in the field of nanooptics, nanospectrosopy and picospectroscopy.

Topics of interest include but are not limited to

- New techniques and theories related to the tip-enhanced optical spectroscopy
- New techniques and theories related to the near-field confinement and excitation dynamics
- Novel technologies for ultrahigh-precision nano-fabrication
- Light-matter interactions in the nanometer, and quantum regimes

Historically, the intramolecular motions are pictorially illustrated using wiggling balls and connecting springs to represent atoms and bonds. Until recently these motions were optically revealed by their Raman scattered light, allowing us to literally 'see' how individual atoms vibrate within a chemical bound. Arriving at this point took us a journey of nearly 90 years, and what is presented in front of us is a wonderful intramolecular universe full of fascinations and surprises.

As early as 1928, Irish scientist E. Synge proposed a method to overcome the classical optical resolution limit. He exchanged several letters with A. Einstein in which he described two ideas. The first was to put a tiny metal particle in the focus of a lens and collect the particle's scattered light. The second suggestion involved a very fine hole in an otherwise opaque device. This aperture should be smaller than the light wavelength and it should be raster-scanned over the sample surface [1]. Synge even foresaw the use of piezo-electric devices for fast scanning with high lateral precision [2]. Yet in terms of a technical realization, Synge was ahead of his time. Only in the mid-1980s, his second idea began to be fulfilled in the form of aperture scanning near-field optical microscopy. With the combination of scanning probe microscope and optical microscopy, scanning near-field optical microscopy continuously pushed the optical resolution to new records, far beyond the diffraction limitation for the conventional optical microscopy [3-6].

Being able to provide molecular fingerprints, Raman spectroscopy were combined with scanning near-field optical microscopy in the early 2000, which further opened up the opportunity of identifying chemical information with nanometer spatial resolution [7]. Incessant outcomes from this combination appeared in the following years, whose technical and scientific progresses continuously stimulated and motivated us to further explore the ultimate spatial resolution of optical microscopy [8-24]. In 2013, a research group led by Zhenchao Dong and Jianguo Hou at University of Science and Technology of China (USTC) demonstrated sub-nanometer resolved single-molecule Raman mapping for the first time [25], pushing the spatial resolution with chemical identification capability down to ~5 Å. Since then, international researchers have been pursuing single-molecule Raman imaging technique to explore what is the ultimate limit of the spatial resolution and how this technique can be best utilized.

In 2019, two further milestone achievements were independently published by the CaSTL scientists entitled ‘Visualizing vibrational normal modes of a single molecule with atomically confined light’ [26], and by the USTC group entitled "Visually constructing the chemical structure of a single molecule by scanning Raman picoscopy" [27]. Both groups demonstrated the unprecedent spatial resolution down to the Angstrom regime at the single-chemical-bond level. The full spatial mapping of various intrinsic vibrational modes in the intramolecular universe were successfully revealed. The ability of such Angstrom-resolved spatial resolution to determine the chemical structure of unknown molecules arouse extensive interests of researchers in the fields of chemistry, physics, materials, biology, and stimulated active research to explore the underlying super-resolution mechanisms, to interpret the experimental data, and to mature the technique for wider applications. The combination of these factors raises questions for new theory and calls for further experimental investigations, which are the main motivation for the initiation of this special issue.

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