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Optical Modelling of DNA-Made Janus Nanoheaters

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Abstract: Nanostructures manufactured using DNA-based nanotechnology, especially DNA-origami, possess a DNA core that influences their thermoplasmonic performance. In this work, we identify key parameters that optimize its thermoplasmonic response by comparing them with their solid counterparts. We also explore the impact that random rotations have on their thermal performance.

The generation of Localized Surface Plasmon Resonances (LSPRs) is accompanied by a partial reemission of the incident electromagnetic energy (nanoantenna effect) but also by a partial absorption, which usually leads to substantial temperature increase. This behavior effectively converts the nanoparticles into both nanoantennae and miniaturized heat sources, commonly known in thermoplasmonics as nanoheaters [1].

Traditionally, researchers tried to minimize or avoid that absorptive nature of nanoantennae to reach strongly enhanced local fields and clean far field signals containing the information of the local targets [2]. However, plasmonic nanostructures (gold in particular), when acting as nanoheaters, have shown great potential in a wide range of applications such as drug delivery or photothermal therapies (PTTs). Hence, big efforts are being made to either find new nanoheaters or to improve the photoinduced thermal response of the existing metallic nanostructures [3-5].

Three factors are especially important when looking at designing nanoheaters for such applications: the biocompatibility of the nanostructure materials, the photothermal conversion efficiency of the applied agents and their stability under rotations with respect to the radiation source. These limiting factors provide a well-defined path for the design of nanoheaters oriented to PTTs. Firstly, nanoheaters must be made of biocompatible materials capable of providing a strong plasmonic response. Secondly, their geometry must support resonances within the so-called biological windows (NIR-I: 700-900 nm and NIR-II: 1000-1400 nm) where the penetration in the tissue and maximum permissible exposure (MPE) are maximum. Finally, they must present a reasonable tolerance to rotations [6]. It is well-known that anisotropic nanoheaters offer different thermal responses depending on their relative orientation with respect to the polarization of the incident beam [6-8]. This is of extreme importance in areas like PTT, where nanoheaters, either free in blood or attached to cells, present a random orientation distribution.

DNA-based nanotechnology, especially DNA origami techniques, provide feasible route to fabricate the aforementioned nanoheaters [9,10]. However, one of the main consequences of using DNA origami for the synthesis of metallic nanomaterials is that their structure is often not solid but has a core formed by DNA. This Janus-like composition can affect their optical behavior and have an impact in their thermoplasmonic response, which ultimately translates into practical variations of the photothermal efficiency.

Here, we present our most recent numerical results on the photothermal response of Janus nanoheaters based on DNA origami. We identify key parameters that optimize its thermoplasmonic response, comparing them with their solid counterparts, and exploring the impact that partial or complete metallic coatings have on their thermal performance. Finally, we study the behavior of these structures when they undergo random rotations and understand how this affects their averaged thermoplasmonic response.

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