Basic mechanisms of the femtosecond laser interaction with a plasmonic nanostructure in water

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ABSTRACT

This paper presents a complete partial differential equation based model to describe the interaction of an ultrafast laser with a plasmonic nanostructure in water. Apart from heating the structure itself, it is shown that this interaction also leads to the generation of a plasma in the water medium and to the production of a strong pressure wave and a nanobubble in the vicinity of the structure. Plasma collisions and relaxation are shown to be the main source of mechanical stress in the medium and the dominant factor for the pressure wave and bubble creation. An all-optical technique able to detect plasmonic enhanced bubble formation and pressure wave generation is also presented.

Keywords: Cell transfection, modeling, ultrafast laser, plasmonic nanostructures, cavitation, plasma

1. INTRODUCTION

Plasmonic enhanced laser cell nanosurgery has drawn a particular attention over the past few years, especially in the field of cell transfection. This technique consists in using the amplified near-field produced around plasmonic gold nanostructures when interacting with an ultrafast laser to induce a local disruption in the cell membrane, hence allowing the exchange of cellular material with the medium. The exact mechanisms leading to this disruption are still unknown. A complete model as well as a thorough characterization of the laser-structure interaction in water must be developed in order to get a deeper understanding of those mechanisms. Some efforts have been made in the past to model such a process where a laser heated nanostructure transfers its energy to a water medium. However, those models do not consider the effect of plasma generation and relaxation in the medium which can have major consequences on the state of the surrounding water. This paper presents a complete and general model of the laser interaction with a plasmonic nanostructure that includes this plasma dynamic. It also presents preliminary results concerning an all-optical technique to detect bubble formation as well as pressure wave generation in the medium.

2. MODELING THE INTERACTION

Absorption and scattering of the laser field by a plasmonic nanostructure in a cellular medium is the starting point of a long chain of events that ultimately leads to the disruption of the cell membrane. Because of the complexity of the cellular medium, all our modeling and experiments are conducted in a water medium which has proven to be well suited for this purpose. Following the laser irradiation, amplified near-field will first heat the conduction electrons of the gold of the nanostructure as well as generating a plasma in the water through a multiphoton ionization process. Electrons ejected from the nanostructure surface and electrons generated by impact ionization will also contribute to this plasma that diffuses in the medium. After some picoseconds, excited electrons and plasma transfer their energies to the gold lattice and the water molecules, hence creating a rapid rise in temperature that generates stress in the water and induce the emission of a strong pressure wave that propagates in the liquid. In some conditions, the energy transfer is strong enough to induce a phase transition in the liquid, thus creating a cavitation bubble. All those phenomena are summarized on figure 1 along with the time scale at which they occur. Chemical interaction with the free electrons of the plasma, pressure wave and cavitation bubbles have all been pointed out as potential damage mechanisms to cell membrane. It is thus important to develop a complete model that takes into account all those phenomena in order to get a better understanding of the cell membrane perforation process.

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2.1 Description of the model

The developed model is based on partial differential equations and is presented schematically in figure 2. It couples Maxwell’s equation with differential equations describing the dynamic of the different phenomena mentioned in the last section, including heat transfer equations for the electrons and lattice of the nanostructure, temperature evolution of the water, as well as compressible Navier-Stokes equations for the fluid dynamics and diffusion equation for the plasma density. A combination of IAPWS\cite{8} and SESAME equations of state (table 7150)\cite{9} is used to describe the water thermodynamic state. A total of 8 partial differential equations are solved in an auto-consistent manner. A finite element based software, COMSOL\textsuperscript{10}, is used to perform the calculation.

2.2 Simulation on gold nanorod

We have performed simulations on a 10\textit{nm} × 41\textit{nm} gold nanorod irradiated by a 133\textit{µJ/cm}\textsuperscript{2} 45\textit{fs} 800\textit{nm} laser pulse polarized linearly along its great axis. As shown in figure 3, this nanostructure is resonant at 800\textit{nm}, generating a strong amplified near-field as well as exhibiting a strong absorption of the field near its center. As it is difficult to simulate the interaction on a melted nanostructure, the fluence was chosen to ensure that the temperature of the nanorod stays below its melting point. Even at such low fluence, an appreciable pressure wave is created along with a highly localized bubble as shown on figure 4.
Since the nanorod generates plasma in the medium and undergoes a strong temperature increase, it is questionable whether heat transfer from the nanorod lattice to the water or the energy transfer from the plasma relaxation is dominant in creating stress in the medium, leading to the generation of the pressure wave and to the bubble creation. Figure 5 and figure 6 compares respectively the pressure wave generated 10nm away from the rod and the size of the nanobubble at its maximal extension following the laser pulse when the plasma contribution to the heat transfer is switched on and off. These results clearly show that the presence of the plasma strongly enhances stress-related phenomena in the medium. Indeed, plasma relaxation and collision occurs much faster than heat transfer through the water/gold interface, resulting in a higher stress generation in the medium. Plasma production is thus a dominant factor in the understanding of the basic mechanisms of cell membrane perforation.

Figure 3. a) Field amplification distribution around a 10nm × 41nm gold nanorod in water irradiated by a 800nm laser polarized linearly along its great axis. b) Absorption spectrum of the same nanorod.

Figure 4. a) Pressure wave moving away from a 10nm × 41nm gold nanorod in water 10ps after being irradiated by a 800nm laser polarized linearly along its great axis. Pressure is in MPa. b) Nanobubble formed around the same nanorod 120ps after the laser irradiation. The bubble is at its maximal extension. Density is in kg/m³.

Figure 5. Comparison of the pressure as a function of time at a point situated 10nm away from the nanorod with (blue line) and without (red line) the plasma mediated heat transfer. Pressure is in MPa.
3. PROBING THE INTERACTION

An all-optical technique has been developed to measure the evolution of both cavitation bubbles and pressure waves following a laser irradiation of a nanostructure. It consists of focusing a continuous He :Ne 633nm 2mW probe laser beam, colinear with the pump beam into a water quartz vessel containing nanoparticles. Probe beam is then filtered from the pump beam using a wavelength selective mirror and focused into a spatial filter consisting of a 10µm diameter pinhole. This spatial filter blocks the light that has been deviated while propagating through gradient of refractive index sample. Since pressure and density variation in the sample are known to create such gradient, it is thus possible using this technique to detect pressure waves and bubble formation. Figure 7 shows typical signals for pressure waves and nanobubbles detection following the irradiation of 100nm gold nanospheres with a 1ps 800nm laser. Note that no signal was observed for pure water sample. These results clearly demonstrate that the plasmonic nanostructures enhance the generation of nanobubbles and pressure waves, as was predicted by simulation.

4. CONCLUSION

Irradiation of a plasmonic nanostructure in a cellular medium with an ultrafast laser is the starting point of a series of phenomena that ultimately leads to the cell membrane disruption. A very complete model taking into account all those phenomena has been developed to get a better understanding of the overall process. Apart from heating the nanostructure, the plasmonic enhanced laser field also generates a plasma in the vicinity of the structure which, through electron-ion collisions and recombination, has been shown to be the main source of stress generation in the medium which leads to pressure wave generation as well as nanobubble formation. Finally, an experimental all-optical technique capable of detecting plasmonic enhanced pressure waves and nanobubbles has been introduced.
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