



Enhancement of confined femto-ablation at SiO₂/Si interface by embedded metallic nanoparticles

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ABSTRACT

Influence of doping an SiO₂/Si interface with metallic nanoparticles (MNPs) on confined laser ablation and resulting structural properties of the crystalline silicon (c-Si) substrate was investigated by irradiating the composed interface with a single, tightly focused femtosecond laser pulse. Confinement ablation regime was enforced by a 10 μm-thick SiO₂ layer capping the c-Si substrate. A mixture of gold (Au) and silver (Ag) nanoparticles was placed at the interface to take advantage of the presumed plasmon-induced enhancement of the incident field strength in a broad spectral range. The nano-plasmonic effect is visualised by numerical simulations utilising the mathematical apparatus of the finite-difference time-domain (FDTD) method. The structural transformations at the site of the laser-induced damage were investigated dominantly by the scanning (SEM) and high-resolution transmission (HRTEM) electron microscopes. A comparative analysis of the irradiation effects in the targets containing different combinations of the interface composing elements revealed clear and strong influence of the confinement and doping on the irradiation result. Character of the observed transformations (among others the crystal twinning) suggests dominant role of increased pressure in the process through the locally generated shock waves.

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1. Introduction

Microexplosion within the transparent dielectrics initiated by tightly focused ultra-short laser pulses proved to be a source of extreme thermodynamic conditions leading to unprecedented material transformations [1–3]. For the sake of clarity, similar, although noticeably weaker effect, one can also obtain by surface multiple-pulse irradiation of the materials [4,5]. It is obvious that confined ablation, i.e. ablation into a dense medium instead of vacuum, changes conditions of the material ejection from a surface. Frequently, the confined ablation was investigated in water plaxing the role of the dense medium [6]. In the meantime, it has been

shown that capping a substrate surface with e.g. a layer of the oxidised material (if possible) delivers noticeably more favourable conditions for this kind of ablation while keeping still technological simplicity [7].

The advantage of the confined regime relies primarily on increase in the pressure initiated by a high density of the deposited energy and reaction of the solid material surrounding the breakdown site. In such a geometry both the interface and the sub-interface area would be strongly influenced. Irreversible modification of the material occurs after exceeding the damage threshold of the sample by the intensity of irradiation. Femtosecond laser pulses tightly focused by a high-numerical aperture (NA) optics used to lead to the absorbed energy density in excess of the strength of the most of the materials [8]. A void surrounded by a shell of compressed, i.e. densified, material used to be formed as a result of the confined micro-explosion triggered in the focal area. A hot dense matter at pressures exceeding 1 TPa and temperatures of more than 10⁵ K were reported in the tabletop experiments, exceeding on the sub-micron scale the limits of high energy density

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and mimicking the conditions existing in the cores of stars and planets. Thus, studies of the confined micro-explosion open new research and application fields including astrophysical relevance [9], creation of exotic materials [10] and processing procedures for nanophotonics [11].

The big advantage of the confined microexplosion relies on keeping the laser-affected material in a limited space. Thus, it is readily available for the post-mortem investigation. It is natural that the material composition can also be used, independently of the energy delivery procedure, to intensify and control the laser-matter interaction. As a combination of metals and dielectrics on the microscale leads, under influence of optical wave, to the nanoplasmonic effects [12], it is reasonable to expect some field enhancement in the vicinity of the metallic NPs scattered over the interface and embedded in the capping SiO_2 layer. Material doping with nanoparticles has a very long history but, in the laser-matter interaction context, presence of MNPs was in the past considered dominantly as a damage precursor [19].

In this paper we suggest a simple extension of the existing methods supporting efficient transformation of the laser energy into exotic physical effects. First, the nanoparticles placed at an interface increase energy absorption, and then the confinement regime of a strong laser ablation strengthens the hydrodynamic effects accompanied by shock wave generation. Our main goal was to reveal what indeed happens below a doped interface under irradiation of the composite structure with a limited power and to find signatures of achieving the extreme thermodynamic conditions and the rare states of the matter.

2. Experimental set-up

The experimental set-up was in its irradiating part exactly the same as that previously described in detail in Ref. [3], while the interface geometry and its composition constituted the element of novelty. Laser pulses with a central wavelength of 800 nm and a duration of 40 fs were tightly focused (measured waist radius equal to $0.62\ \mu\text{m}$) with a high-numerical aperture microscope objective (MO, NA = 1.25) utilising an oil-immersion ($n = 1.515$) layer (see Fig. 1a). The SiO_2/Si interface, buried under a $10\ \mu\text{m}$ -

thick layer of SiO_2 was placed precisely in the focal plane of the irradiating flux. The cross-sectional view of the radiation-untreated (pristine) SiO_2/Si interface, doped with Au/Ag NPs is shown in Fig. 1b. In this view the nanoparticles quite regular in the top view can appear slightly distorted. It is very likely that some of them has been deformed during the cut performed with the focused ion beam (FIB) technique. Such a technique delivers additional energy to the particles of the ion beam way. Here, to illustrate the MNPs in the side view and at high magnification, we selected image with MNPs more distant from the cut plane. As a consequence, the nanoparticles are imperfectly visible but conserved their regular shape. The mix of MNPs embedded in a host dielectric material was used to extend spectral range of high absorption through generation of the relevant surface plasmons. Usually, plasmon resonances are spectrally narrow and the composition was used to assuage the influence of the fixed irradiation wavelength. Fig. 2 illustrates influence of the specific components on the effective absorptance/reflectance spectrum. The presented curves have been calculated according to the formula $A = 1 - R - T$ using the results of the recorded spectra (300–1600 nm) of the reflected and transmitted signals. Importantly, the applied in the measurement commercial spectrophotometer (Cary 5000, Varian, Palo Alto, CA, USA) was equipped with an integrating sphere and allowed for estimate of the absolute reflectance of the sample. The directly measured spectra were registered without the SiO_2 layer. The sample was placed on a computer-controlled nano-positioning stage. An imaging system working in the mode of the reflected light microscopy was installed for *in situ* inspection of the breakdown site.

The structure of Au/Ag NPs mixture was synthesized on a monocrystalline silicon (c-Si) substrate by the standard techniques based mainly on the temperature treatment and leading either to individual paraboloidal MNPs or metallic clusters. The dewetting process caused obtained mix of MNPs to be dominated by elongated (close to a paraboloidal shape) elements approximated in numerical modelling by a hemispherical form. Covering the structured interface with a $10\ \mu\text{m}$ -thick layer of SiO_2 by e-beam evaporation was the final step of the sample preparation. The mean diameters of Au and Ag NPs were 100.5 nm and

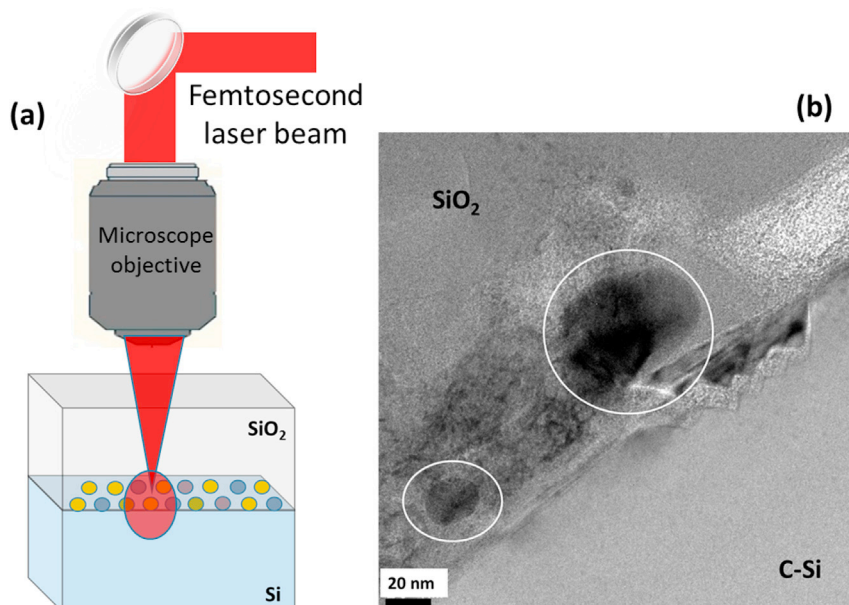


Fig. 1. a) Optical arrangement of the sample irradiation, b) cross-sectional HRTEM image of the interface of pristine SiO_2/Si with embedded Au/Ag NPs.

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