The effect of the substrate on the damage threshold of gold nano-antennas by a femtosecond laser

Monir Morshed, Md Ahasanul Haque and Haroldo T Hattori
School of Engineering and Information Technology, University of New South Wales at Canberra, ACT 2610, Australia
E-mail: monir.morshed@student.adfa.edu.au

Abstract
Gold nano-antennas with silica substrate may not be suitable for high power applications such as heat resisted magnetic recording, solar thermophotovoltaic, and nano-scale heat transfer systems. When a laser beam reaches to these nano-antennas, part of the light is absorbed by the metallic regions, leading to a temperature rise of the device. If these devices reach a temperature beyond its Tamman temperature (the temperature at which sintering of atoms or molecules start to occur), the antenna can be damaged. One strategy to allow the antenna to work at higher fluences (energy density) is to employ substrates that can quickly carry the heat away from the antennas. In this paper, we show that high thermal conductivity substrates, such as diamond, can allow the antenna to withstand 20 times higher fluence than a low thermal conductivity silica substrate.

1. Introduction
Nano-antennas can concentrate the incident light in a subwavelength gap by creating interference of surface plasmon resonances (SPRs) [1, 2]. Nano-antennas can strongly localize freely propagating optical radiation into a subwavelength gap [3], confining light beyond the diffraction limit [4], making them useful in single molecule imaging [5, 6], enhancement of the efficiency of photo-detectors [7, 8], near-field optical trapping [9], creation of nanoscale light sources [10], solar energy harvesting [11], heat transfer [12, 13], single algae cell detection [14], and enhancement of surface enhanced Raman spectroscopy (SERS) signals [15]. In recent years, different types of nano-antennas have been demonstrated such as dipole [16], bowtie [2], Yagi-Uda [17], spiral, log-periodic antennas [18], and phased-array [19] that can be used in these applications.

Plasmonic nano-antennas can enhance many photo-physical phenomena, but they also absorb light through intraband transitions and, consequently, can reach high temperatures when illuminated by lasers [20–22]. Although the localization of heat in small areas can be used for cancer treatment [23] and heat vapor generation [24], unexpected effects can be produced by the accumulation of heating and relaxation processes that can be detrimental in imaging, sensing, and spectroscopy applications [25]. Moreover, if the metal in the antenna reaches a temperature above its Tamman temperature, it will melt. If the antennas change their shape, their optical properties may be changed [26] and the electric field enhancement can be significantly reduced [27]. A gold nano-antenna with silica substrate can generally handle very low power (e.g. below 250 μW). However, this power handling capability can be increased by introducing alternative materials in the antenna or using different substrates. Mironov et al [28] stated that a proper choice of materials can produce devices that can handle higher fluences than standard gold antennas without melting.

Another aspect that influence the power handling capacity of nano-antennas is how fast the substrate can dissipate heat. In many applications, nano-antennas are fabricated on low conductivity substrates such as silica, meaning that most of the generated heat remains in the antennas for a long time. In this article, we examine the effect of different substrates on the fluence response of antennas, using different substrates such as silica and diamond. Diamond is well known for its very high thermal conductivity [29]. We experimentally show that antennas on a diamond substrate can handle 20 times higher fluence than on a silica substrate.
2. Device description and numerical analysis

A schematic diagram of a bowtie nano-antenna is shown in figure 1. The basic parameters of the bowtie are the length of each triangle $l$, width of the triangle $w$, apex angle $\alpha$, gap between two triangles $g$, and thickness $H$. The electric field enhancement factor of the nano-antennas is calculated by optimizing these parameters at the wavelength of 1053 nm. The optimum length and width for silica and diamond substrate nano-antennas are 190 nm and 380 nm, and 135 nm and 270 nm, respectively. The gap width, thickness and apex angle are always kept fixed to 50 nm, 100 nm and 90° for antennas. The nano-antennas in this article are made of gold, which is shown by the yellow regions in figure 1.

The device is analysed using commercial three-dimensional finite difference time domain (3D-FDTD) software with perfectly matching layers (PML) [30] at the computational boundaries. A plane wave is incident on the antenna from air. The grid sizes are chosen as $\Delta x = \Delta y = \Delta z = 5$ nm. The refractive index of SiO$_2$ is set to a value of 1.45 and the optical properties of gold (Au) are taken from Johnson’s article [31]. The enhanced electric field in the gap of nano-antenna is calculated as,

$$F_{rel} = \frac{|E_{gap,peak}|}{|E_{inc,peak}|}$$  \hspace{1cm} (1)

where $|E_{gap,peak}|$ is the magnitude of the electric field calculated in subwavelength gap of the nano-antenna, and $|E_{inc,peak}|$ is the magnitude of electric field of incoming light.

Figure 2 shows the electric field enhancement factor at the wavelength of 1053 nm in the middle of the air gap for silica and diamond substrate antennas, respectively. It also shows that the silica substrate antenna has electric field enhancement factor of 7.5 which is slightly higher than that of diamond which is 6.41.

The main limitation of gold/silica nano-antenna is that the continuous amount of heat due to optical illumination can eventually destroy the antennas since silica cannot dissipate the heat rapidly enough due to its low thermal conductivity. As a result, the surface temperature of antenna can exceed the Tamman temperature of gold which can lead to its destruction. In general, heat is produced due to absorption of light within the antenna [32]. The main parameter that leads to the melting of the metallic regions is the energy density or fluence [32]. The energy and fluence for a single pulse of a pulsed laser are given by,

$$W_{single-pulse} = \int_{t_1}^{t_2} P(t) dt = P_{peak} \tau_{eff}$$  \hspace{1cm} (2)

$$F_{single-pulse} = \frac{W_{single-pulse}}{\pi \phi_{spot}^2}$$  \hspace{1cm} (3)

where $P(t)$ is the instantaneous value of the power, $t_1$ and $t_2$ are the arbitrary instants when the pulse is not negligible and $\tau_{eff}$ is the effective duration of the pulse. $\phi_{spot}$ represent the spot size of the laser beam.

3. Fabrication and experiments

An electron beam evaporator (EBE) is used to deposit 100 nm gold (Au) on the top of both silica and diamond substrates at the rate of 5 nm s$^{-1}$. The bowtie antennas are fabricated using a FEI Helios NanoLab 600 dual beam
focused ion beam (FIB) system. Since 2 nm titanium layer has a small influence on the response of the structure compared with 100 nm gold, the optical properties of our designed nano-antenna are not greatly affected by the adhesion layer. However, it should be noted that since the adhesive layer titanium can boost-up the temperature in the structures (even for a 2 nm thickness) \[33\], it can lead to a significant heat accumulation in the gold region. The scanning electron microscope (SEM) images were taken using FIB before and after the laser exposure.

The scanning electron microscope (SEM) image of nano-antennas with no laser exposure are shown in figures 3(a) and (c) and images with laser exposure are in figures 3(b) and (d), for silica and diamond substrates, respectively. To assess the power capacity of the antennas, we have used a femtosecond pulsed laser with pulse duration of 300 fs and repetition rate of 20.8 MHz, with fluences changing from 12.8 J m\(^{-2}\) (100 \(\mu\)W from the femtosecond laser) to 2560 J m\(^{-2}\). As mentioned previously, all antennas are made of gold but they are fabricated on different substrates.
The initial tests are conducted on a silica substrate. The antennas were damaged at a fluence of 30.45 J m\(^{-2}\). Afterwards, the antennas on a diamond substrate were tested and were damaged at a fluence of 609.17 J m\(^{-2}\), therefore the diamond substrate allows the antennas to withstand 20 times higher fluence than in the silica substrate. The reason why the antennas on a diamond substrate can withstand higher fluence than on a silica substrate is because diamond has a much higher thermal conductivity than silica, i.e. 3320 W/(m.K) for diamond against 1.3 W/m.K for silica. The much higher thermal conductivity of diamond allows a rapid dissipation of heat generated in the antennas. Since the electric field is proportional to the square root of the fluence for a fixed spot size and repetition rate, it can be inferred that the nano-antennas on the diamond substrate can generate electric fields 4.22 times higher on a diamond substrate. Although the thermal conductivity of diamond is about 2554 times higher than silica, it has resulted in a much smaller improvement on the damage threshold of the antennas: this seems to be explained by the fact that because of the very small footprint of the nano-antennas, the temperature rise and subsequent melting happens very quickly (nano-structures can melt in picoseconds\([34]\),) while the heat transfer might take much longer time.

Lastly, we have analyzed the temperature effects for different substrates. In this experimental analysis, we have used a home-made setup where temperature dependent reflectance is measured. It is noted that we have only considered the specular reflectance, since the diffuse reflectance is small. A simple voltage dependent home-made heater with two electrodes is used to apply different temperatures in samples. To perform this experiment, 100 nm gold is deposited on top of both substrates (samples) by EBE system. The temperature in the surface of thin films is measured by an infrared thermo-meter. The reflectance of the films as a function of temperature is shown in figure 4. It is interesting that the reflectance changes more with temperature for a silica substrate when compared with a diamond substrate, probably because silica cannot dissipate heat quickly enough from the film.

Furthermore, we have theoretically analyzed the temperature distribution on both substrates, which is shown in figure 5. We can see from this figure that temperature is more uniformly distributed in the diamond substrate because of its high thermal conductivity whereas the heat cannot quickly propagate in the silica.
substrate because of its low conductivity. Therefore, when heat is transferred from the nano-antenna to the diamond substrate, it spreads heat to the diamond substrate which helps to dissipate the heat. However, the heating of the small antenna is quick and, if the temperature of the antenna exceeds the Taman temperature, it can be damaged. However, there is always a competition between heat generation and its subsequent temperature rise and the dissipation of heat: it seems that the substrate can help to dissipate heat but the time it takes for a small volume to heat is short, meaning that a high thermal conductive substrate will not be fully effective.

4. Conclusions

In conclusion, we have studied the effects of two substrates on the performance of gold nano-antennas: silica (low thermal conductivity) and diamond (high thermal conductivity). The theoretical results show that though the electric field enhancement factor for both substrate nano-antennas are similar, experimentally we can demonstrate that diamond substrate nano-antenna can withstand 20 times higher fluence than silica substrate and can generate 4.22 times higher magnitude of electric field than silica substrate. Consequently, diamond substrate can help to dissipate the heat generated by high fluence laser beams.

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ORCID iDs

Monir Morshed https://orcid.org/0000-0001-6664-9230

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