Nanometer-Scale Structuring of Gold Thin-Films and Graphene by Femtosecond Laser Bessel Beams

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Abstract: We report nanometer-size patterning of various thin films by femtosecond pulsed Bessel beams. Nanoslit arrays fabricated on gold films exhibit excitation of surface plasmon polaritons. We extend the approach to single-atomic-layer systems such as graphene.
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1. Introduction

The virtues of material processing by femtosecond (fs) laser pulses, such as minimal collateral damage and possibility of achieving sub-wavelength resolution make fs laser ablation a strong alternative or complimentary to lithographical techniques, especially when large-scale throughput is not of primary concern [1]. The deterministic nature of the ablation process allows for local ablation limited to the central region of the beam, where the fluence exceeds the threshold. As a result, effective exploitation of this method requires maximal pulse-to-pulse stability, shorter laser wavelengths and high numerical aperture (NA) focusing. On the other hand, high NA focusing, especially at short (near-UV) wavelengths, becomes tedious due to strict alignment constraints and aberrations. We have recently shown that Bessel beams, a particular class of diffraction-free beams [2], can be exploited to relax these constraints [3]. In particular, the focal depths of these beams can be orders of magnitude larger as compared to tightly focused Gaussian beams, thereby significantly relaxing sample-positioning constraints. The width of the beam stays constant throughout the line focus. Further, Bessel beams are conveniently generated by axicons, which do not suffer from increased aberrations at high NAs or short wavelengths. We exploit these advantages to generate nanometer-size structures on metal thin-films and atomically thin layered materials. For the former, semi-transparent gold films and for the latter, graphene is examined. These samples are of primary interest for many emerging applications [4]. We demonstrate the application of structures on gold to excitation of surface plasmon polaritons (SPP).

2. Experimental Approach

For the ablation experiments, we use a chirped-pulse amplification system producing 550-fs pulses at 1 kHz repetition rate at a central wavelength of 1030 nm. Second, third and fourth harmonics of the laser wavelength are also generated to improve the fabrication resolution. Gaussian laser output is converted to Bessel beam with a 40° base-angle axicon. At the fourth harmonic wavelength, the Bessel beam has a central diameter (separation of the first two zeroes of intensity) of 340 nm. Note that the multiple-ring nature of the Bessel beams is not of concern when near the ablation threshold, where only the central region of the main Bessel peak contributes to ablation. The samples are translated under laser illumination, on a three-dimensional piezo translation stage.

The samples used in gold thin-film experiments are deposited on glass substrate under 10^{-7} mBar pressure. Thickness of gold film is ~25 nm (5 nm chromium is used for adhesion). For graphene ablation experiments, we use commercial single-layer graphene (SLG) on Si/SiO_2 substrate (Graphene Supermarket). After ablation, the samples are imaged under optical, scanning electron (SEM) and Raman (for the case of SLG) microscopes.

3. Nanofabrication Results

Using the setup described above, and by gradually decreasing the laser average power, we obtain narrowing stripes on gold and SLG films. Typical SEM images for both cases are shown in Fig. 1. The best resolution, i.e. ablation width (without compromising stripe continuity) is ~125 nm for gold samples and ~400 nm for SLG. We also compare the ablation width to the calculated values from the intensity envelope of the Bessel beam, and obtain good agreement as shown in Fig. 1.

In the case of SLG, we observe that because the Si substrate has significantly stronger absorption and a lower damage threshold than SLG, with properly adjusted incoming pulse energy and sample translation speed, complete graphene removal can be achieved with no substrate damage. We attribute this observation to nonlinear-optical

shielding by the graphene layer. For verification, we perform a large set of experiments covering a range of speed and energy values, which clearly indicate that substrate protection happens when the pulses do not spatially overlap and when pulse energy after nonlinear absorption in graphene falls below ablation threshold of Si. The ablation status of sample layers is also verified by Raman microscopy measurements.

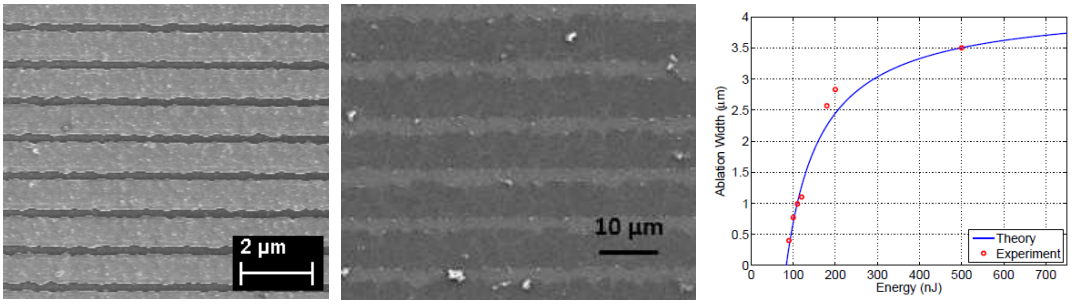

Fig. 1. SEM image of nanoslit arrays ablated on gold (left) and graphene (center) films. The evolution of ablation width (SLG case) follows the expectation from the Bessel function envelope (right).

4. Optical Characterization and Excitation of SPP

The ablation resolution obtained on gold thin films is sufficient for some plasmonics applications. In order to demonstrate this, we perform measurements of optical transmission through nanoslit arrays. We ablate arrays with various periodicities, over a 100 µm wide square region. Transmission spectroscopy measurements are done for transverse electric (TE) and transverse magnetic (TM) polarization of incoming light.

Fig. 2 shows the expected transmission behavior through such nanoslit arrays [5]. At each periodicity, two resonant excitations of SPP are expected, from air-metal and glass-metal interfaces. The resonances (minima in the optical transmission spectra) obtained in our experiments are in accordance with these calculations. Fig. 2 (right) shows some typical measured spectra, for two periodicities. First, we note that for each period, SPP excitation is observed only for TM polarization, as it should. Secondly, to demonstrate the double-resonance behavior, we drop glycerin on the metal and observe that the resonances at metal-air interface disappear.

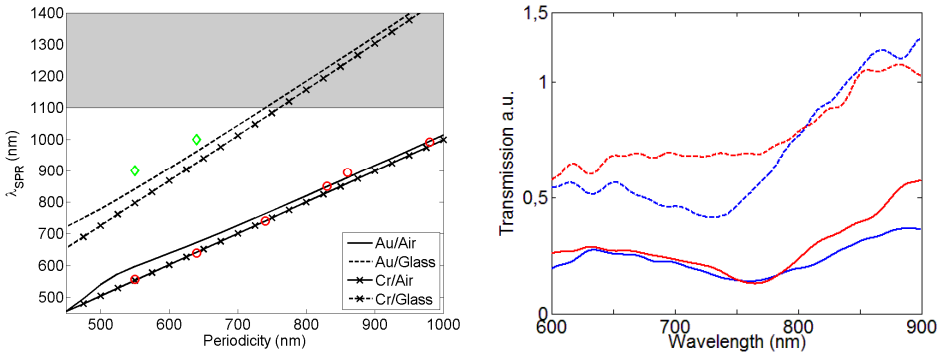

Fig.2. Left: Calculated and measured SPP resonance wavelengths for different nanoslit array periodicities. Right: Transmission spectra through slit arrays in air (solid) and glycerin (dashed), for TM polarized light. Blue and red curves correspond to 500 nm and 810 nm periods.

In conclusion, we show that high-resolution ablation by fs laser Bessel beams can be a very powerful approach for rapid fabrication of nanostructures. Especially thin-film type samples are very convenient and possess strong potentials for future photonics, materials science, and other applications.

References

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